

Measurement of Thermophysical Properties of Pure and Mixture of Alkanolamines from 288.15-323.15 K

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Density, viscosity and refractive index of pure, binary and ternary mixtures of diethanolamine (DEA) and methyl diethanolamine (MDEA) with water have been measured at different temperatures from 288.15-323.15 K at atmospheric pressure. Also, this study presents the effects of mixture composition and temperature upon these thermo-physical properties. To measure the density and refractive index, the amine mass percentages of binary and ternary aqueous solutions were between 5-40% and 40-80%, respectively. To determine the kinematic viscosity, the amine mass percentages for binary aqueous solutions were 5-40%. The densities, refractive indexes, and viscosities of 22 binary and ternary mixtures were finally measured.

Keywords: Density, Viscosity, Refractive index, DEA, MDEA

INTRODUCTION

Density, viscosity, and refractive index data for liquid mixtures are important for many industrial applications such as the design, operation, control and optimization processes as well as theoretical studies and modeling. For instance, Carbon dioxide (CO₂) and hydrogen sulfide (H₂S) should be removed from natural gas because removal of CO₂ increases the value of fuel and as H₂S is extremely toxic, CO₂ is regarded as one of the most important greenhouse gasses. Global climate changes due to global warming as a result of excessive emissions of greenhouse gasses is a crucial problem [1]. Thus, reducing greenhouse gas emissions has brought great attention to CO₂ capture. There are many techniques for capturing CO₂, among which chemical absorption and adsorption are especially effective.

From the environmental protection point of view, separating the acidic gasses from natural, refinery and synthetic gas streams has significant importance. It is

usually performed by gas-liquid absorption using different solvents like water, alcohols, amines and their mixtures [1-4]. Chemical or physical solvents can be used as sorbent liquids, however chemical solvents are more effective in removing CO₂ from fuel gas because of their higher absorption capacity [5,6]. Therefore, physicochemical properties of pure and mixture of the aforementioned solvents are crucial for purifying industrial gas streams [5-12].

Alkanolamine aqueous solutions are frequently used for capturing acid gasses during absorption processes [13]. For instance, the aqueous diethanolamine solution is a well-known solution for the selective removal of H₂S from gas stream [14,15].

The objective of the present work is to obtain new exact experimental data on the densities, viscosities, refractive indices and to provide derived thermodynamic properties of binary and ternary mixtures for the mentioned solvents at different temperatures and compositions and at atmospheric pressure.

Diethanolamine (DEA), methyl diethanolamine

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(MDEA) and their binary and ternary mixtures with water (W) are employed in Sarkhoon and Qeshm Gas Treating Company for gas sweetening by removal of CO₂ and H₂S. In this company, the amine weight mass percentages of binary and ternary aqueous solutions are from 5-40% and 40-80%, respectively. The removal of acidic gasses in this company is conducted over the temperature range from 288.15-323.15 K.

EXPERIMENTAL

Materials

The chemicals used in this study were diethanolamine (DEA, 99%) and methyl diethanolamine (MDEA, 99.5%) supplied by Merck. These chemicals were used without further purification. The molecular formula of DEA, molecular weight, and density at 298 K are C₄H₁₁NO₂, 105.14 g mol⁻¹ and 1.09 g cm⁻³, respectively. The molecular formula of MDEA, the molecular weight, and density at 298 K are C₅H₁₃NO₂, 119.16 g mol⁻¹ and 1.038 g cm⁻³, respectively. Doubly distilled water degassed by boiling was used for preparing the solutions. The total amine contents of the solutions were determined by titration with standard HCl using methyl orange indicator. Each test was repeated three times.

Apparatus

The density (ρ) and refractive index (n_D) of the pure samples and mixtures were measured using Mettler Toledo Combined Meter DR40. It is a compact and powerful device for determining these properties which provide an uncertainty of 5×10^{-2} kg m⁻³ and 1×10^{-4} for the density and refractive index, respectively. The temperature was held constant within ± 0.01 K. The density and refractive index of the samples were measured at 288.15, 293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15 K.

Kinematic viscosity (ν) at atmospheric pressure was measured with a rotational automated Viscometer Anton Paar Stabinger SVM3000 over the temperature range of 293.15-353.15 K. The apparatus was calibrated by the manufacturer, using several viscosity standard reference fluids. It controls the temperature with an accuracy of 0.02 K and provides an uncertainty of $\pm 1\%$ for measuring the

viscosity. Each measurement was repeated three times.

RESULTS AND DISCUSSION

Table 1 demonstrates the experimental density and refractive index of pure solvents studied in this work at various temperatures between 288.15 K and 323.15 K. Table 2 shows the measured kinematic viscosity and derived dynamic viscosity of pure solvents in the temperature range from 293.15-353.15 K.

Also, the variation of densities with respect to the temperature for the pure solvents (DEA and MDEA) was measured in the mentioned temperature range. To evaluate the accuracy of the results, the values of density obtained in the present study were compared with those reported in the literature for the pure solvents [16-21], shown in Table 1. There is a good agreement between the present results and literature data.

Tables 3 to 9 as well as Figs. 1 and 2 provide the experimental values measured for the density, excess molar volume, viscosity and refractive index for binary and ternary mixtures at different temperatures and compositions. Binary mixtures are a blend of water with DEA or MDEA with mass percentages of DEA and MDEA in the range of 5-40%. Ternary mixtures are a combination of water with DEA and MDEA in the range of 20-40%.

Density

Experimental density values for aqueous binary and ternary mixtures are reported in Tables 3 and 4. Comparing the values indicates that the behavior of density is similar in all aqueous binary and ternary mixture systems. A decrease in the value of water causes an increase in the value of the density. When the temperature is raised, the density decreases smoothly. Figure 1 shows the effect of the temperature on the density for aqueous binary mixtures of DEA.

It is known that the density of liquid mixtures depends on some parameters like the size and the shape of molecules and also the intermolecular forces. The physical interactions comprise dipole-dipole and dipole-induced dipole interactions while the chemical contribution arises due to hydrogen bonding. The mixtures, which are formed by amines and water, have the interaction between the amine

Table 1. Comparison of the Experimental Density Values, ρ_{exp} , and Refractive Indices, n_D , of Pure Liquids with those Reported in the Literature at Different Temperatures

DEA				MDEA			
T (K)	Exp.	Lit. Ref.	Dev. (%)	Exp.	Lit. Ref.	Dev. (%)	Dev. (%)
$\rho_{exp} \times 10^{-3} \text{ (Kg m}^{-3}\text{)}$							
288.15	1.1005	1.0996 [16]	-0.08	1.0449	1.0445 [20]	-0.04	
293.15	1.0975	1.0974 [17]	-0.01	1.0412	1.0410 [20]	-0.02	
298.15	1.0939	1.0937 [18]	-0.02	1.0371	1.0374 [20]	0.03	
303.15	1.0907	1.0902 [17]	-0.05	1.0333	1.0337 [20]	0.04	
308.15	1.0870	1.0867 [19]	-0.03	1.0298	1.0302 [20]	0.04	
313.15	1.0841	1.0839 [17]	-0.02	1.0264	1.0265 [21]	0.01	
318.15	1.0808	1.0803 [19]	-0.05	1.0221	1.0227 [21]	0.06	
323.15	1.0775	1.0771 [17]	-0.04	1.0185	1.0189 [21]	0.04	
n_D							
288.15	1.4789	1.4794 [16]	0.03	1.4699	-	-	
293.15	1.4766	1.4770 [22]	0.03	1.4689	1.4693 [21]	0.03	
298.15	1.4759	1.4764 [16]	0.03	1.4671	1.4672 [21]	0.01	
303.15	1.4735	1.4742 [22]	0.05	1.4653	1.4653 [23]	0.00	
308.15	1.4726	1.4734 [16]	0.05	1.4625	1.4631 [21]	0.04	
313.15	1.4714	1.4720 [22]	0.04	1.4607	1.4607 [23]	0.00	
318.15	1.4695	1.4703 [16]	0.05	1.4589	1.4589 [23]	0.00	
323.15	1.4672	1.4679 [22]	0.05	1.4570	1.4571 [23]	0.01	

Table 2. Experimental Results of Kinematic Viscosity ($\nu_{exp} \times 10^{-6} \text{ (m}^2 \text{ s}^{-1}\text{)}$) and Derived Dynamic Viscosity ($\eta \text{ (mPa s)}$) for Pure Solvents at Different Temperatures

T (K)	Kinematic	Dynamic	Lit. Ref.	Dev. (%)
DEA				
293.15	765.73	840.39	889.66 [24]	5.54
313.15	170.27	184.59	192.85 [24]	4.28
323.15	92.76	99.95	106.94 [24]	6.53
353.15	21.22	22.15	22.97 [24]	3.59
MDEA				
293.15	98.45	102.51	102.70 [25]	0.18
313.15	34.08	34.98	34.02 [25]	2.82
323.15	20.09	20.46	21.50 [25]	4.84
353.15	7.12	6.97	7.15 [25]	2.53

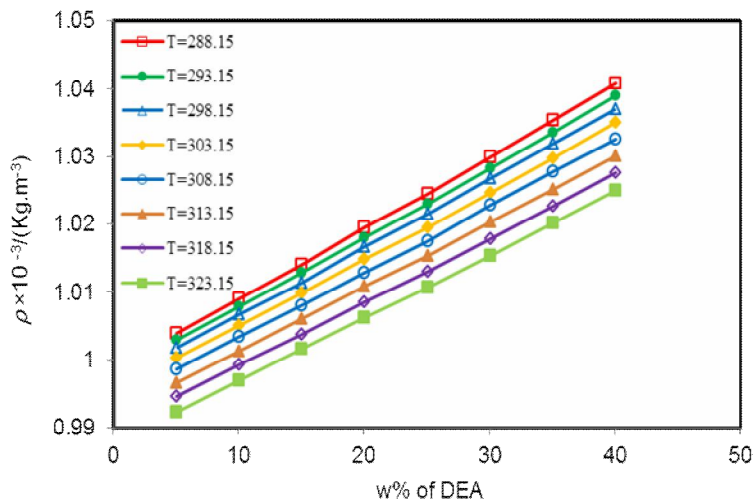


Fig. 1. Densities for the aqueous solutions of DEA as a function of mass % of DEA at different temperatures: 288.15-323.15 K and atmospheric pressure.

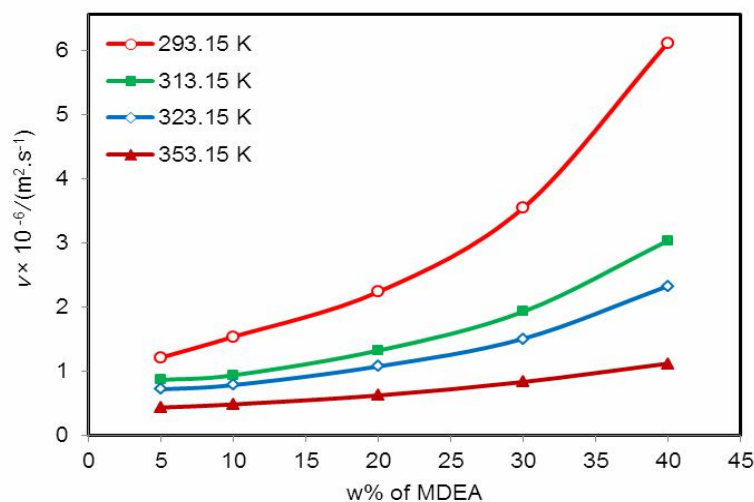


Fig. 2. Viscosities of the aqueous solutions of MDEA as a function of mass % of MDEA at different temperatures: 293.15-353.15 K and atmospheric pressure.

and water molecules and also the interaction of self-association. The polar nature of the N-H bond results in the formation of hydrogen bonds with other amine molecules or with other H-bonding systems. Excess molar volume, V_m^E , of a solution is a volumetric property that can be defined as the amount of volume which is in excess to that of an ideal solution in the same conditions of composition, temperature,

and pressure:

$$V_m^E = V_m - V_m^{ideal} \quad (1)$$

where V_m and V_m^{ideal} are the molar volumes of solution and pure components, respectively. Excess molar volumes of aqueous DEA or MDEA solutions with mole fractions can

Table 3. Experimental Density ($\rho_{\text{exp}} \times 10^{-3} \text{ Kg m}^{-3}$) of Binary Mixtures (DEA or MDEA + Water) at Various Temperatures and Atmospheric Pressure

T (K)	288.15	293.15	298.15	303.15	308.15	313.15	318.15	323.15
DEA (w%)								
5	1.00387	1.00301	1.00171	1.00028	0.99862	0.99662	0.99464	0.99238
10	1.00903	1.00795	1.00680	1.00515	1.00350	1.00133	0.99929	0.99704
15	1.01401	1.01283	1.01142	1.00988	1.00814	1.00601	1.00383	1.00166
20	1.01948	1.01805	1.01672	1.01489	1.01290	1.01083	1.00859	1.00619
25	1.02445	1.02291	1.02146	1.01949	1.01748	1.01529	1.01303	1.01065
30	1.02992	1.02826	1.02676	1.02471	1.02282	1.02026	1.01779	1.01540
35	1.03536	1.03357	1.03196	1.02979	1.02781	1.02513	1.02261	1.02014
40	1.04088	1.03910	1.03701	1.03506	1.03260	1.03015	1.02755	1.02498
MDEA (w%)								
5	1.00358	1.0022	1.00034	0.99823	0.99636	0.99531	0.99408	0.99131
10	1.00804	1.00716	1.00605	1.00420	1.00250	1.00065	0.99798	0.99508
20	1.01849	1.01721	1.01579	1.01361	1.01160	1.00884	1.00566	1.00224
30	1.02890	1.02774	1.02540	1.02304	1.02047	1.01758	1.01410	1.01087
40	1.03978	1.03756	1.03478	1.03199	1.02905	1.02601	1.02289	1.01962

Table 4. Experimental Density ($\rho_{\text{exp}} \times 10^{-3} \text{ Kg m}^{-3}$) of Ternary Mixtures (DEA+MDEA+Water), at Various Temperatures and Atmospheric Pressure

T/K	288.15	293.15	303.15	313.15	323.15
DEA+MDEA (w%)					
20+20	1.01931	1.01871	1.01515	1.01092	1.00630
20+30	1.02463	1.02299	1.01901	1.01454	1.00972
20+40	1.03012	1.02864	1.02358	1.01903	1.01316
30+20	1.02801	1.02646	1.02278	1.01867	1.01353
30+30	1.03197	1.02997	1.02476	1.02084	1.01467
30+40	1.03643	1.03450	1.02997	1.02473	1.01909
40+20	1.03233	1.03117	1.02701	1.02269	1.01787
40+30	1.03684	1.03499	1.03133	1.02616	1.02066
40+40	1.03875	1.03621	1.03495	1.02963	1.02471

Table 5. V_m^E ($\text{cm}^3 \text{mol}^{-1}$) of Binary Mixtures (DEA or MDEA+Water), at Various Temperatures and Atmospheric Pressure

T (K)	288.15	293.15	298.15	303.15	308.15	313.15	318.15	323.15
DEA (w%)								
5	-0.0010	-0.0034	-0.0054	-0.0063	-0.0072	-0.0049	-0.0076	-0.0045
10	-0.0105	-0.0105	-0.0172	-0.0155	-0.0182	-0.0140	-0.0165	-0.0205
15	-0.0162	-0.0162	-0.0197	-0.0219	-0.0245	-0.0224	-0.0231	-0.0239
20	-0.0317	-0.0285	-0.0357	-0.0338	-0.0331	-0.0337	-0.0342	-0.0313
25	-0.0367	-0.0331	-0.0401	-0.0370	-0.0376	-0.0372	-0.0383	-0.0370
30	-0.0524	-0.0482	-0.0566	-0.0537	-0.0591	-0.0520	-0.0494	-0.0492
35	-0.0680	-0.0629	-0.0714	-0.0678	-0.0653	-0.0649	-0.0622	-0.0615
40	-0.0872	-0.0861	-0.0855	-0.0846	-0.0832	-0.0821	-0.0785	-0.0767
MDEA (w%)								
5	-0.0458	-0.0361	-0.0270	-0.0119	-0.0128	-0.0086	-0.0470	-0.0459
10	-0.0952	-0.1015	-0.1017	-0.0975	-0.0999	-0.0926	-0.0954	-0.0932
20	-0.2301	-0.2364	-0.2345	-0.2326	-0.2287	-0.2094	-0.2024	-0.1680
30	-0.3852	-0.3955	-0.3903	-0.3855	-0.3777	-0.3577	-0.3489	-0.3228
40	-0.5919	-0.5846	-0.5756	-0.5661	-0.5535	-0.5399	-0.5384	-0.5262

Table 6. Excess Molar Volume (V_m^E ($\text{cm}^3 \text{mol}^{-1}$)) of Ternary Mixtures (MDEA+DEA+Water), at Various Temperatures and Atmospheric Pressure

T (K)	288.15	293.15	303.15	313.15	323.15
DEA+MDEA (w%)					
20+20	0.1969	0.1690	0.1492	0.1378	0.1199
20+30	0.2043	0.1947	0.1692	0.1525	0.1273
20+40	0.2104	0.1832	0.1724	0.1427	0.1374
30+20	0.2540	0.2443	0.2136	0.1891	0.1780
30+30	0.3217	0.3156	0.3145	0.2674	0.2784
30+40	0.3982	0.3757	0.3238	0.3066	0.2818
40+20	0.5901	0.4513	0.4199	0.3921	0.3573
40+30	0.5711	0.5682	0.4881	0.4746	0.4498
40+40	0.8863	0.8789	0.6292	0.6009	0.5197

Table 7. Kinematic Viscosity ($v_{\text{exp}} \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$) of Aqueous Solutions (DEA or MDEA + Water), at Various Temperatures

T (K)	293.15	313.15	323.15	353.15
DEA (w%)				
5	1.109	0.808	0.710	0.420
10	1.420	0.911	0.751	0.464
20	2.167	1.310	1.058	0.589
30	3.533	1.985	1.582	0.828
40	6.272	3.055	2.258	1.081
MDEA (w%)				
5	1.208	0.860	0.712	0.429
10	1.532	0.933	0.780	0.478
20	2.232	1.317	1.070	0.617
30	3.541	1.923	1.501	0.827
40	6.117	3.030	2.323	1.114

be expressed as:

$$V_m^E = \left[x_2 M_2 \left(\frac{1}{\rho} - \frac{1}{\rho_2} \right) + (1 - x_2) \left(x_1 M_1 \left(\frac{1}{\rho} - \frac{1}{\rho_1} \right) \right) \right] \quad (2)$$

where V_m^E is the excess molar volume, M_1 and M_2 are molar masses of water and DEA or MDEA, respectively. x_2 is the mole fraction of DEA or MDEA, ρ is the density of aqueous DEA or MDEA solutions, and ρ_1 and ρ_2 are densities of doubly distilled water and pure DEA or MDEA, respectively.

The excess molar volumes are reported in Tables 5 and 6. V_m^E for binary mixtures show the negative deviations from ideality. Instead, V_m^E values are positive for ternary mixtures at temperature and pressure conditions of this study. The large negative values of excess molar volumes reflect the compactness of binary solutions. This could be attributed to the strong hydrogen bonding interactions. In ternary solutions, the positive value of excess molar volume indicates that the non-specific interaction between MDEA and DEA components of the mixture is favorable. There is a

small difference between the size of MDEA and DEA molecules, and there is a steric hindrance of the components of the mixture. Therefore, in these solutions, positive values of V_m^E imply that the effect of the factors responsible for volume expansion outweighs those for volume contraction.

Kinematic Viscosity

The measured kinematic viscosity values for the pure and binary mixtures are shown in Tables 2 and 7, respectively. The shear rate was 200 s^{-1} for the measurements. The influence of the composition and temperature upon the value of kinematic viscosity was also analyzed. Viscosity increases with decreasing water and decreases with increasing the temperature. Figure 2 illustrates this dependency of viscosity for aqueous binary mixtures of MDEA.

Experimental data reveal that the kinematic viscosities of the pure liquids have noticeable values, which may be the result of the effectiveness of self-association of the liquids. DEA is the most extensively self-associated liquid by H-bonding, followed by MDEA due to its increasing order of

Table 8. Experimental Values of Refractive Index (n_D) for Aqueous Solutions (DEA or MDEA+ Water), at Different Temperatures

T (K)	288.15	293.15	298.15	303.15	308.15	313.15	318.15	323.15
DEA (w%)								
5	1.3386	1.3381	1.3375	1.3369	1.3363	1.3356	1.3347	1.3339
10	1.3446	1.3440	1.3435	1.3428	1.3421	1.3413	1.3406	1.3396
15	1.3503	1.3497	1.3491	1.3485	1.3478	1.3469	1.3461	1.3455
20	1.3564	1.3557	1.3550	1.3543	1.3535	1.3527	1.3519	1.3509
25	1.3620	1.3612	1.3606	1.3598	1.3590	1.3581	1.3572	1.3563
30	1.3681	1.3673	1.3668	1.3659	1.3651	1.3641	1.3631	1.3622
35	1.3742	1.3734	1.3728	1.3718	1.3711	1.3701	1.3689	1.3680
40	1.3804	1.3796	1.3788	1.3780	1.3707	1.3760	1.3750	1.3740
MDEA (w%)								
5	1.3402	1.3385	1.3372	1.3351	1.3345	1.3368	1.3312	1.3303
10	1.3472	1.3470	1.3464	1.3456	1.3448	1.3436	1.3422	1.3402
20	1.3622	1.3622	1.3614	1.3606	1.3579	1.3578	1.3560	1.3538
30	1.3771	1.3773	1.3763	1.3753	1.3742	1.3724	1.3705	1.3681
40	1.3934	1.3924	1.3914	1.3903	1.389	1.3879	1.3866	1.3853

Table 9. Experimental Values of Refractive Index (n_D) for Aqueous Solutions (DEA+MDEA+Water), at different temperatures

T (K)	288.15	293.15	303.15	313.15	323.15
MDEA+DEA (w%)					
20+20	1.3531	1.3527	1.3514	1.3497	1.3479
20+30	1.3596	1.3591	1.3574	1.3554	1.3539
20+40	1.3676	1.3672	1.3657	1.3631	1.3615
30+20	1.3592	1.3581	1.3571	1.3552	1.3535
30+30	1.3667	1.3658	1.3642	1.3630	1.3604
30+40	1.3752	1.3738	1.3723	1.3702	1.3682
40+20	1.3676	1.3667	1.3651	1.3633	1.3612
40+30	1.3734	1.3735	1.3719	1.3699	1.3679
40+40	1.3639	1.3645	1.3631	1.3638	1.3593

strict effect. Also, the viscosity data show that the temperature impact on the DEA viscosity is more significant than that of MDEA viscosity, which might be due to their extent of H-bonding.

Dynamic viscosity can be calculated by the following relation:

$$\mu = \nu \cdot \rho \quad (3)$$

where η is the dynamic viscosity. The derived dynamic viscosities of pure fluids and their corresponding values reported in the literature [24,25] are shown in Table 2.

Refractive Index

Experimental refractive index data for pure, binary and ternary mixtures are given in Tables 1, 8 and 9, respectively. For the pure solvents, a comparison with the literature values [16,21-23] shows that the experimental values of the refractive index for either of the DEA and MDEA pure solvents are in acceptable agreement with the literature data for all temperatures, as depicted in Table 1.

The results of the experimental refractive index values indicate the same discipline for the density and viscosity. Changing the composition and temperature causes the similar influence on the value of the refractive index. In other words, refractive index values increase with increasing DEA or MDEA concentration and decrease with raising the temperature.

CONCLUSIONS

In this work, the experimental densities, excess molar volumes, viscosities and refractive indices of pure, binary mixtures of DEA+Water, and MDEA+Water, and also ternary mixtures of DEA+MDEA+Water were measured at different temperatures and different compositions at atmospheric pressure. This investigation was conducted using 22 binary and ternary mixtures with different compositions. The same discipline was found in composition and temperature dependency of density, viscosity, and refractive index. An increment in DEA and MDEA concentrations resulted in an increment in all of these three properties while increasing the temperature reduced all these properties. These data can be satisfactorily

applied to the design, operation, and optimization of gas sweetening processes in different chemical industries and in theoretical models.

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