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Volumetric Properties, Viscosities, and Refractive Indices for Aqueous 2-(Methylamino)ethanol Solutions from (298.15 to 343.15) K

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This paper presents the experimental data of densities and viscosities for aqueous 2-(methylamino)ethanol solutions at six temperatures in the range (298.15 to 343.15) K and refractive indices at seven temperatures in the range (293.15 to 323.15) K. Excess molar volumes, viscosity deviations, and molar refraction changes were calculated from the experimental results and correlated as a function of the mole fractions. The molar refraction changes were correlated as a function of the volume fractions. The partial molar volumes at infinite dilutions were determined from the apparent molar volumes. Negative values for both the excess molar volumes and the molar refraction changes were observed over the entire range of mole fractions, whereas the viscosity data exhibited positive deviations from linearity.

Introduction

Concerns about suppressing high concentrations of greenhouse gases in the atmosphere have encouraged research into different strategies to capture and store acid gases such as carbon dioxide (CO_2), sulfur dioxide (SO_2), carbonyl sulfide (COS), and hydrogen sulfide (H_2S) from industrial and natural gas streams. Solutions of alkanolamines are the most commonly used reactive compounds to remove such acidic gases.[1]

2-(Methylamino)ethanol (MAE) is a secondary amine often used in industrial operations. In the MAE molecule, a methyl group substitutes a hydrogen atom of the amino group of a monoethanolamine (MEA, a primary amine). However, the methyl group is supposed to enhance the reaction kinetics as it increases the basicity of the amine without appreciably increasing the hindrance around the nitrogen atom. In 2002, Ali et al.[2] reported that the electron-donating methyl group on the nitrogen atom increases the reaction rate between CO_2 and MAE. In addition, MEA has excellent regeneration characteristics if compared with MEA, as 20 % less regeneration energy was required.[3] Even though MAE is an important alkanolamine, to our knowledge, there is no published data on the densities, viscosities, and refractive indices for aqueous MAE solutions. These data are required for the development of the proper design of the absorption and stripping operations.

In this work, we have studied the density, viscosity, and refractive index of the binary systems of water (1) + 2-(methylamino)ethanol (2) over the entire mole fractions range and at various temperatures in order to cover a wide range of temperatures found in industrial applications. The experimental density data are used to calculate the excess molar volumes. The partial molar volumes at infinite dilutions are determined from the apparent molar volumes. Viscosity deviations and molar refraction changes were also calculated from the experimental viscosity and refractive index data. The calculated binary data were fitted to the Redlich−Kister equation.

Experimental Section

2-(Methylamino)ethanol was obtained from Sigma Aldrich with a stated purity of 98+ % by mass and was used without further purification. An analytical balance Ohaus (model Ap 205 D, Florham Park, NJ) was used to prepare gravimetrically the binary mixtures of MAE and deionized water with a precision of ± 0.1 mg. The overall possible uncertainty in the mole fractions was around ± 0.0001. Densities were measured for deionized water, MAE, and their binary mixtures using an Anton Paar DMA 4500 densimeter. The apparatus consists of a glass U tube with a PT100 platinum resistance thermometer inside a thermostatic jacket with a manufacturer stated accuracy of ± 0.01 K. The sample density is a function of the oscillation frequency. At each temperature, the relationship between the density and the oscillation frequency is

$$\rho = A + B\tau^2 \tag{1}$$

where ρ is the liquid density and τ is the period of oscillation. The calibration was done using air and water at ambient temperature. The densities of water were measured in the temperature range (279.15 to 353.15) K and were compared with the values measured by Bettin et al.[4] and with the values provided in the instruction manual of Anton Paar. The calibration was acceptable if the measurements were within $\pm 3 \times 10^{-5}$ g·cm^{-3} of the published values. The measurements were reproducible to 2×10^{-5} g·cm^{-3}. The uncertainty of our density data was estimated to be 5×10^{-5} g·cm^{-3}.

Viscosities were measured with seven different U-tube Glass Cannon−Ubbelohde viscometers to cover, with precision,[5] the range of temperatures from (298.15 to 243.15) K. To measure the viscosities of aqueous MAE at different temperatures, viscometers were placed in a well-stirred constant temperature bath (model CT 500, Cannon Instrument Company, State College, PA). The uncertainty of water bath temperature was ± 0.015 °C as measured by the Ertco-Hart RTD high-precision digital thermometer (model 850 C, West Patterson, NJ). The efflux time was measured manually using a digital stopwatch having an accuracy of 0.01 s. Each datum was an average of at least three runs with a maximum deviation in the kinematic viscosity of 0.1 % at each temperature for all mole fractions. The estimated uncertainty of the viscosity measurement was 0.5 %.

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Table 1. Densities of Water (1) + MAE (2) Mixtures from (298.15 to 343.15) K

	ρ/g·cm^{-3}					
x_2	T = 298.15 K	T = 303.15 K	T = 313.15 K	T = 323.15 K	T = 333.15 K	T = 343.15 K
0	0.99705	0.99569	0.99226	0.98805	0.98319	0.97778
0.0499	0.99645	0.99449	0.99011	0.98507	0.97947	0.97337
0.0657	0.99678	0.99458	0.98979	0.98441	0.97852	0.97217
0.1054	0.9979	0.99516	0.98939	0.98317	0.97655	0.96957
0.1987	0.99603	0.99248	0.98528	0.97779	0.97011	0.96217
0.3010	0.98825	0.98447	0.976815	0.96897	0.9609	0.95265
0.3834	0.98024	0.97643	0.96873	0.96083	0.95272	0.94443
0.5041	0.96881	0.96500	0.95732	0.94946	0.94141	0.93317
0.6000	0.96030	0.9565	0.94883	0.94099	0.93298	0.92480
0.6974	0.95364	0.94981	0.94212	0.93427	0.92628	0.91816
0.8009	0.94663	0.94276	0.93501	0.92712	0.91916	0.91100
0.9031	0.94086	0.93696	0.92916	0.92124	0.91322	0.90506
1	0.93618	0.93226	0.92442	0.91648	0.90846	0.90030

Table 2. Comparison of Density Data of Pure MAE between Alvarez et al.[7] and This Work from (298.15 to 323.15) K

T/K	Alvarez et al.[7]	this work
298.15	0.937683	0.93618
303.15	0.933789	0.93226
313.15	0.925948	0.92442
323.15	0.918024	0.91648
AD[a]/%		0.16

[a] AD, average deviation in percent.

Table 3. Comparison of Density Data for Pure MDEA between Alvarez et al.[7] and Others at Different Temperatures

	T/K				
	298.15	303.15	313.15	323.15	AD/%
ref 7	1.036832	1.033056	1.025447	1.017781	
ref 8		1.0315	1.0249	1.0174	0.08
ref 9			1.025	1.0096	0.43
ref 10			1.0246	1.0093	0.46
ref 11			1.0247	1.0099	0.43
ref 12	1.0359	1.032	1.02445	1.01666	0.08
ref 13	1.037863	1.0341	1.026523	1.018877	−0.1

The equation for kinematic viscosity, according to Poiseuille's law is

$$\nu = k_1 t - k_2/t \qquad (2)$$

where t is the efflux time and k_1 and k_2 are the viscometer constants. The second term in the equation represents the correction due to the kinetic energy and was found to be negligible.[6] The absolute viscosity (η) results from multiplying the time by the calibration constant of the viscometer and by the density of the sample.

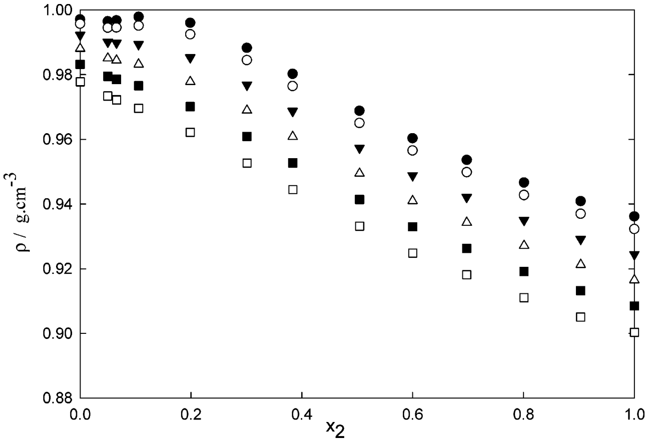

Figure 1. Densities of water (1) + MAE (2) system at various temperatures: ●, 298.15 K; ○, 303.15 K; ▼, 313.15 K; ▽, 323.15 K; ■, 333.15 K; □, 343.15 K.

Refractive indices of aqueous 2-(methylamino)ethanol solutions were measured by Atago RX-5000-α refractometer. Refractive indices of deionized water were measured and compared with the available literature data. The uncertainty in refractive index measurements was about 0.0001.

Results and Discussion

Table 1 presents the experimental density values for (MAE + water) at (298.15, 303.15, 313.15, 323.15, 333.15, and 343.15) K throughout the whole mole fraction range. As can be observed from Table 2, the density values of pure MAE were always lower by 0.16 % than the values published by Alvarez et al.[7] However, the pure MDEA densities of Alvarez et al.[7] were also higher than all others[8−12] (Table 3). The deviations varied from 0.08 % to 0.46 %; except for Berna-Garcia et al.[13] where the values of Alvarez et al. were lower by 0.10 %.

In Figure 1, the density curve increases as MAE mole fractions (x_2) decrease. The maximum values of the density occur in the water-rich region. The density values were used to calculate the excess molar volume as

$$V^{E} = V_{m} - (x_1 V_1 + x_2 V_2) \qquad (3)$$

with

$$V_{m} = (x_1 M_1 + x_2 M_2)/\rho \qquad (4)$$

where V_m is the molar volume of the mixture and x_i, V_i, and M_i (i = 1, 2) are mole fraction, molar volume, and molecular weight of the components water and MAE, respectively; ρ is the experi-

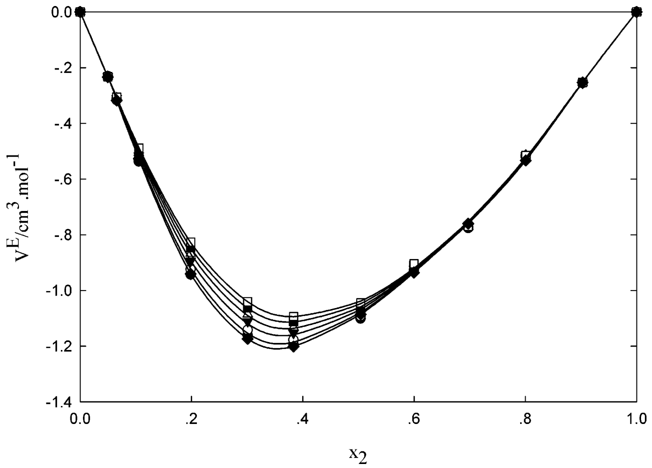

Figure 2. Excess molar volumes of water (1) + MAE (2) system at various temperatures: ●, 298.15 K; ○, 303.15 K; ▼, 313.15 K; ▽, 323.15 K; ■, 333.15 K; □, 343.15 K; −, eq 5.

Table 4. Redlich−Kister Equation Fitting Coefficients of the Excess Volumes (V^E/cm^3·mol^{-1}) for Water (1) + MAE (2) Mixtures from (298.15 to 343.15) K

T/K	a_0	a_1	a_2	a_3	a_4	a_5	σ/cm^3·mol^{-1}
298.15	−4.38	−2.40	−1.62	−0.37	2.81	2.36	0.01
303.15	−4.35	−2.32	−1.46	−0.24	2.60	2.07	0.01
313.15	−4.30	−2.17	−1.19	0.01	2.22	1.51	0.01
323.15	−4.25	−2.20	−1.10	1.13	2.12		0.01
333.15	−4.21	−2.01	−0.94	0.89	1.85		0.01
343.15	−4.17	−1.85	−0.78	0.69	1.58		0.01

Table 5. Partial Molar Volumes at Infinite Dilution V_1^∞ (Water) and V_2^∞ (MAE) from (298.15 to 343.15) K

T/K	V_1^∞/cm^3·mol^{-1}	V_2^∞/cm^3·mol^{-1}
298.15	15.3	76.7
303.15	15.3	76.9
313.15	15.4	77.3
323.15	15.5	77.8
333.15	15.8	78.3
343.15	15.9	78.9

mental density of the binary mixture. Figure 2 shows that the excess molar volume presents negative deviations from ideality at the temperatures selected in this work, as is common for other completely miscible solvents with a minimum value at around $x_2 = 0.4$. Negative deviations of the excess molar volume become less negative as the temperature increases, as is also common. This is due to the fact that the H-bonds of self-association and cross-association decrease with increasing temperature, which leads to a positive contribution to V^E. Figure 2 shows sharp changes in the excess volumes in the water-rich region. Negative excess molar volume values indicate that there was a volume contraction and can be explained by the large difference in the molar specific volumes. According to Pal and Singh,[14] that volume contraction was due to the ability of the -OH group to form hydrogen bonds with water molecules. However, a second explanation might be due to the accommodation of the nonaqueous molecules occupying in the structured water lattices, void spaces, so that such a marked change in V^E happened.

The excess molar volumes were correlated by using the Redlich−Kister[15] equation:

$$V^E/\text{cm}^3\cdot\text{mol}^{-1} = x_1x_2\sum_{i=0}^{n} a_i(x_1 - x_2)^i \tag{5}$$

Table 4 shows the values of the parameters at each temperature together with their standard deviations from the fit. The standard deviation was calculated as:

$$\sigma = \left[\frac{\sum(y_i^{\text{exp}} - y_i^{\text{cal}})^2}{n - m}\right]^{1/2} \tag{6}$$

where y_i is the excess molar volume, n is the number of excess molar volumes, m is the number of adjustable parameters, and y_i^{exp} and y_i^{cal} represent the experimental and calculated values.

It is well-known that the Redlich−Kister[15] relation does not correlate unsymmetrical curves very well. The F test was used to determine the optimum number of parameters in this work. There is a usefulness for the thermal expansion data to reveal correlations between the molecular structure of solutes and their effects on water structure as explained by Hepler[16] and Neal and Goring.[17] They suggested using the sign of the second derivative of the partial molar volume at infinite dilution of the solute with respect to the temperature ($d^2V_2^o/\text{d}T^2$) to classify solutes as "structure makers" or "structure breakers". A positive value of $d^2V_2^o/\text{d}T^2$ indicated a structure-making characteristic of the solute while a negative value of that indicated the structure-breaking characteristic of the solute.

At infinite dilution the partial molar volumes of MAE (V_2^∞) in water were derived from the method proposed by Maham et al.[12] The apparent molar volume of water ($V_{\phi,1}$) and the apparent molar volume of MAE in water ($V_{\phi,2}$) were first calculated as

$$V_{\phi,2} = V_2^o + (V^E/x_2) \tag{7}$$

$$V_{\phi,1} = V_1^o + (V^E/(1 - x_2)) \tag{8}$$

where V_1^o and V_2^o are the molar volumes of pure water and MAE, respectively. An analytical extrapolation of $V_{\phi,1}$ to $x_2 = 1$ led to V_1^∞ and a similar extrapolation to $x_2 = 0$ led to (V_2^∞). Table 5 lists the partial molar volumes of water and MAE at infinite dilution. All the values of the molar volumes at infinite dilution (V_2^∞) were smaller than the corresponding molar values of pure MAE (V_2^o). The change in the excess volume is due to the MAE molecule fitting (partially) into the open or empty space in liquid water, so there is neither the structure making nor the breaking properties and is consistent with the observation that ($d^2V_2^o/\text{d}T^2$) values were equal to zero. Values of the apparent molar volumes of MAE in water (V$_{\phi,2}$) are shown in Figure 3 and increased with the increase in temperature.

The viscosities of the aqueous MAE solutions were measured at (298.15, 303.15, 313.15, 323.15, 333.15, and 343.15) K. Experimental data of the viscosities at different temperatures and mole fractions appear in Table 6. Figure 4 shows the S-shaped viscosity curves, and the sharp increase in the viscosity of the mixture after the addition of MAE with a maximum value occurred

Table 6. Viscosities of Water (1) + MAE (2) Mixtures from (298.15 to 343.15) K

	η/mPa·s					
x_2	T = 298.15 K	T = 303.15 k	T = 313.15 K	T = 323.15 K	T = 333.15 K	T = 343.15 K
0	0.8824	0.8047	0.6596	0.5507	0.4703	0.4074
0.0493	1.8011	1.5558	1.1950	0.9978	0.8170	0.6997
0.0657	2.2583	1.9265	1.4501	1.1330	0.9621	0.7903
0.1054	3.8045	3.1692	2.2720	1.6428	1.6428	1.0233
0.1987	8.7882	6.98675	4.6219	3.2417	2.3787	1.7470
0.2946	13.5053	10.5319	6.7482	4.5810	3.2609	2.4164
0.3880	18.2141	14.1082	8.8524	5.8856	4.1074	2.9857
0.5041	19.9760	15.4395	9.7187	6.4576	4.4994	3.2548
0.6000	19.1093	15.0474	9.4239	6.3192	4.4289	3.2267
0.7003	17.0241	13.4217	8.6520	5.8808	4.1768	3.0751
0.8019	14.3759	11.4316	7.5472	5.2289	3.7795	2.8273
0.8980	12.4636	10.0001	6.7158	4.7250	3.4635	2.6182
1	10.5106	8.5221	5.8331	4.1774	3.1079	2.3853

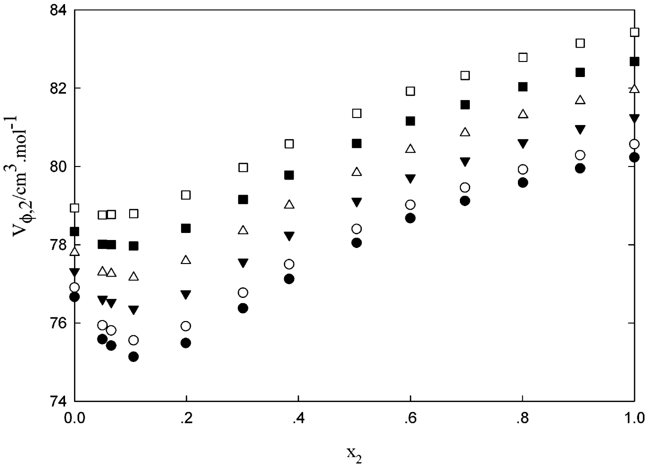

Figure 3. Apparent molar volumes of MAE in water at various temperatures: ●, 298.15 K; ○, 303.15 K; ▼, 313.15 K; ▽, 323.15 K; ■, 333.15 K; □, 343.15 K.

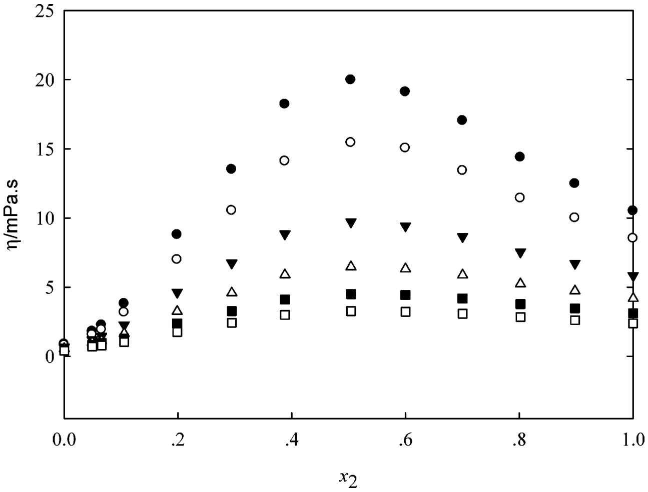

Figure 4. Viscosities of water (1) + MAE (2) system at various temperatures: ●, 298.15 K; ○, 303.15 K; ▼, 313.15 K; Δ, 323.15 K; ■, 333.15 K; □, 343.15 K.

at around 50 mol %. The S-shape of viscosity curves and the maximum in the viscosity deviations demonstrate the stable complexes formed in a ratio of 1 mol of water to 1 mol of alkanolamine according to the suggestions of Fort and Moore.[18] This was also the case for many other alkanolamine solutions. The viscosity deviations can be calculated from the MAE viscosity values using

$$\Delta\eta/\text{mPa}\cdot\text{s} = \eta - \eta_1 x_1 - \eta_2 x_2 \tag{9}$$

where x_1 and x_2 are the mole fractions of the pure components, and η, η_1, and η_2 are the viscosities of the mixture, pure water, and pure MAE, respectively. Figure 5 shows the dependence of the viscosity deviations on the composition and temperature. The viscosity deviation exhibits deviations from a rectilinear dependence of viscosity on the mole fraction. For all temperatures, the deviation is positive with a maximum around $x_2 = 0.5$.

$\Delta\eta$ values were correlated by using the Redlich−Kister[15] relation:

$$\Delta\eta/\text{mPa}\cdot\text{s} = x_1 x_2 \sum_{i=0}^{n} a_i (x_1 - x_2)^i \tag{10}$$

Table 7 lists the values of the parameters together with the standard deviations of $\Delta\eta$ for studied temperatures. The standard deviations were calculated by using eq 6 with $y_i = \Delta\eta_i$.

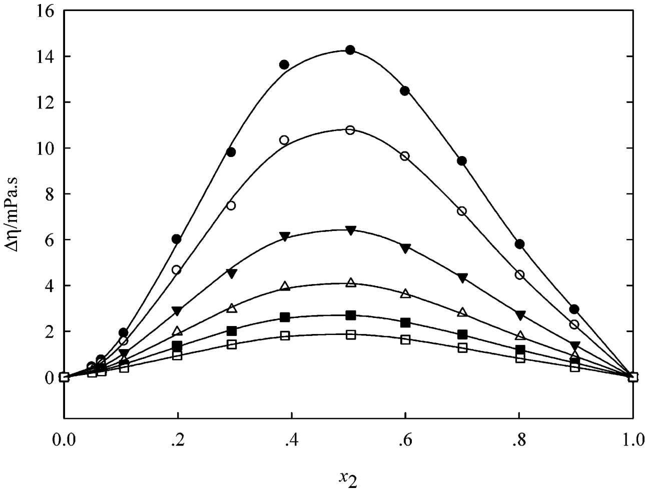

Figure 5. Viscosity deviations of water (1) + MAE (2) system at various temperatures: ●, 298.15 K; ○, 303.15 K; ▼, 313.15 K; Δ, 323.15 K; ■, 333.15 K; □, 343.15 K; −, eq 10.

Table 7. Redlich−Kister Equation Fitting Coefficients of the Viscosity Deviations ($\Delta\eta$/mPa·s) for Water (1) + MAE (2) Mixtures from (293.15 to 323.15) K

T/K	a_0	a_1	a_2	a_3	a_4	σ/cm^3·mol^{-1}
298.15	57.0201	10.9908	−66.5582	−29.6765	27.4029	0.20
303.15	43.2575	7.8523	−47.8031	−21.2104	17.9902	0.10
313.15	25.7191	5.1103	−24.9869	−12.6141	7.90121	0.10
323.15	16.3790	3.2598	−15.8548	−7.4181	6.3856	0.07
333.15	10.8349	2.2808	−9.5677	−4.4480	4.0838	0.05
343.15	7.5045	1.4866	−7.0402	−2.5579	4.1034	0.03

The experimentally measured refractive indices of the aqueous MAE solutions at (293.15, 298.15, 303.15, 308.15, 313.15, 318.15 and 323.15) K are listed in Table 8. The measured refractive indices values indicated a sharp increase in its values after the addition of MAE at all temperatures. The molar refraction changes (ΔR) were calculated at four different temperatures (298.15, 303.15, 313.15, and 323.15) K from the following relations employing refractive indices and molar volumes:[19]

$$\Delta R/\text{cm}^3\cdot\text{mol}^{-1} = R - \phi_1 R_1 - \phi_2 R_2 \tag{11}$$

$$R = \left(\frac{{n_D}^2 - 1}{{n_D}^2 + 1}\right)\left(\frac{x_1 M_1 + x_2 M_2}{\rho}\right) \tag{12}$$

$$R_i = \left(\frac{{n_D}^2 - 1}{{n_D}^2 + 1}\right)\left(\frac{M_i}{\rho_i}\right) \tag{13}$$

$$\phi_i = \frac{x_i V_i}{\sum_{i=1}^{2} x_i V_i} \tag{14}$$

where R and R_i are the molar refractions of the mixture and the pure component liquids, respectively. x_1 and x_2 are the mole fractions of water and MAE, respectively; ρ, ρ_1, and ρ_2 are the densities; and n_D and n_{Di} are the measured refractive indices of the mixture and the pure component liquids, respectively. M_1 and M_2 are the molecular weights of water and MAE, V_i is the molar volume of the ith component liquid. ϕ_1 and ϕ_2 are the volume fractions of water and MAE, respectively. There is no

Table 8. Refractive Index (n_D) for Water (1) + MAE (2) Mixtures from (293.15 to 323.15) K

	n_D						
x_2	T = 293.15 K	T = 298.15 K	T = 303.15 K	T = 308.15 K	T = 313.15 K	T = 318.15 K	T = 323.15 K
0.0000	1.3331	1.3326	1.3320	1.3314	1.3307	1.3299	1.3291
0.0493	1.3559	1.3552	1.3544	1.3536	1.3527	1.3517	1.3507
0.0657	1.3625	1.3617	1.3608	1.3599	1.3589	1.3579	1.3568
0.1054	1.3767	1.3754	1.3744	1.3732	1.3720	1.3708	1.3696
0.1987	1.3997	1.3983	1.3967	1.3956	1.3940	1.3921	1.3907
0.3010	1.4151	1.4137	1.4120	1.4105	1.4091	1.4074	1.4055
0.3834	1.4235	1.4218	1.4201	1.4186	1.4170	1.4151	1.4134
0.5041	1.4288	1.4272	1.4253	1.4239	1.4221	1.4204	1.4187
0.6000	1.4324	1.4306	1.4289	1.4272	1.4254	1.4238	1.4220
0.6974	1.4347	1.4329	1.4311	1.4294	1.4275	1.4257	1.4239
0.8008	1.4366	1.4348	1.4330	1.4313	1.4293	1.4274	1.4257
0.9031	1.4381	1.4363	1.4344	1.4328	1.4306	1.4288	1.4268
1.0000	1.4393	1.4375	1.4356	1.4339	1.4318	1.4298	1.4278

Table 9. Redlich−Kister Equation Fitting Coefficients of the Molar Refraction Changes, Δ*R* (eq 15), in Terms of Volume Fractions for Water (1) + MAE (2) Mixtures from (298.15 to 323.15) K

T/K	a_0	a_1	a_2	a_3	a_4	a_5	σ/cm^3·mol^{-1}
298.15	−29.13	18.86	−11.23	3.95	−6.26	9.96	0.014
303.15	−29.19	19.00	−11.42	3.54	−5.99	10.67	0.014
313.15	−29.29	19.07	−11.58	3.38	−5.89	11.41	0.015
323.15	−29.40	19.18	−11.57	3.42	−6.03	11.18	0.017

Table 10. Redlich−Kister Equation Fitting Coefficients of the Molar Refraction Changes, Δ*R* (eq 16), in Terms of Mole Fractions for Water (1) + MAE (2) Mixtures from (298.15 to 323.15) K

T/K	a_0	a_1	a_2	a_3	a_4	a_5	σ/cm^3·mol^{-1}
298.15	−29.2	−18.42	−10.97	−5.98	−7.25	−5.89	0.01
303.15	−29.27	−18.51	−10.94	−5.95	−7.42	−5.97	0.02
313.15	−29.37	−18.53	−11.04	−6.31	−7.64	−5.83	0.01
323.15	−29.44	−18.75	−11.12	−6.06	−7.71	−6.23	0.02

general rule for the calculation of function for the molar refraction changes. Therefore, for the sake of completeness, we calculated molar refraction changes (ΔR) as a function of volume fractions and as a function of mole fractions by using the Redlich−Kister expression:

$$\Delta R = \phi_1\phi_2 \sum_{i=0}^{n} a_i(\phi_1 - \phi_2)^i \tag{15}$$

$$\Delta R = x_1x_2 \sum_{i=0}^{n} a_i(x_1 - x_2)^i \tag{16}$$

The coefficients and the standard deviations are presented in Tables 9 and 10 for the expression in terms of volume fractions and in terms of mole fractions, respectively. Molar refraction changes were negative for the system studied over the entire range of mole fractions for all four temperatures with minimum values of ΔR at $x_1 = 0.3$ for all temperatures.

Conclusions

This work reports the densities and viscosities of the binary mixture of water (1) + MAE (2) from T = (298.15 to 343.15) K over the entire mole fractions range. The refractive indexes of water (1) + MAE (2) were also reported at temperatures between T = (293.15 to 323.15) K. The excess molar volumes (V^E) and the viscosity deviations ($\Delta\eta$) for this mixture show negative and positive deviations from ideality, respectively, for all temperatures and mole fractions covered in this work. Redlich−Kister type equations were used to correlate these properties.

The criterion proposed by Hepler[16] suggested that the addition of MAE to water had no effect on its structure. However according to Fort and Moore's[18] observations, the S-shaped viscosity curves and the presence of a maxima of the viscosity deviation curves (around $x_2 = 0.5$) of aqueous MAE solutions indicate a formation of stable complexes in the ratio 1:1.

Molar refraction changes were all negative for the system studied over the entire range of mole fractions for four temperatures with a minimum of ΔR occurring at $x_1 = 0.3$. The molar refraction changes exhibit negligible temperature dependency.

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