

Volumetric Properties and Viscosities for Aqueous AMP Solutions from 25 °C to 70 °C

Amr Henni,^{*,†} Jonathan J. Hromek,[†] Paitoon Tontiwachwuthikul,[†] and Amit Chakma[‡]

Process Systems Laboratory, Faculty of Engineering, University of Regina, Regina, Saskatchewan, Canada S4S 0A2, and Department of Chemical and Materials Engineering, University of Waterloo, Waterloo, Ontario, Canada N2L 3G1

This paper reports the density and viscosity of aqueous 2-amino-2-methyl-1-propanol (AMP) solutions at five temperatures in the range 25 °C to 70 °C over the whole concentration range. The results are compared with data published in the literature. The derived excess molar volumes, the partial molar volumes, the partial molar volumes at infinite dilution, and the viscosity deviations were correlated as a function of composition.

Introduction

Recently, sterically hindered alkanolamines have been introduced as commercially attractive over conventional amines such as MEA, DEA, DIPA, and MDEA, and as rate promoters in amine blends. Sharma¹ proposed the use of 2-amino-2-methyl-1-propanol (AMP), a primary amine, as a solvent in carbon dioxide removal after observing the steric effect on the stability of the formation of the carbamate. The stoichiometry of the reaction allows loadings of CO₂ up to 1 mol of CO₂ per mol of AMP. As a hindered form of monoethanolamine (MEA), AMP [OH-(CH₂)C(CH₃)₂NH₂] has excellent absorption characteristics, superior stripping qualities, higher degradation resistance, and lower corrosion rate than conventional amines. Hindered amines have found wide applications in the area of selectivity toward H₂S in the presence of CO₂. Say et al.² and Goldstein et al.³ presented commercial applications of the hindered amine processes.

Measurements of the densities and viscosities of alkanolamines covering the whole concentrations range are scarce. Even though AMP is an important alkanolamine, to our knowledge, there are no published data of a comprehensive study of the densities and viscosities of aqueous AMP solutions. Xu et al.⁴ presented values of the density of aqueous AMP for six concentrations and the viscosity for two concentrations in the temperature range 20 °C to 90 °C. Li and Lie⁵ published a few values of the viscosity and density of pure and aqueous AMP in the temperature range 30 °C to 80 °C.

Measurements of the density and viscosity of the aqueous AMP solutions were performed at various temperatures, (25, 30, 40, 50, 60, and 70) °C, in order to cover a wide range of temperatures found in industrial applications. The excess volumes, the partial molar volume, the partial molar volumes at infinite dilution, and the deviation of the viscosity were derived.

Experimental Section

AMP (>97% pure) was purchased from Aldrich and was used without further purification. AMP was solid at 30 °C.

* To whom correspondence should be addressed. Telephone: (306) 585-4960. Fax (306) 585-4855. E-mail: amr.henni@uregina.ca.

[†] University of Regina.

[‡] University of Waterloo.

The solutions were prepared by mass on an analytical balance (model Ap 205 D, Ohaus, Florham Park, NJ) with ± 0.01 mg accuracy. The possible error in the mole fraction is estimated to be around ± 0.0001 . Densities of the binary mixtures were measured with an Anton Paar DMA-4500 density meter. Accuracies of our densities are about $\pm 5 \times 10^{-5} \text{ g}\cdot\text{cm}^{-3}$ when compared to the densities of pure MDEA published in the literature.⁶ Calibration was done using air and water at 20 °C. The density of water is then measured at several other temperatures (15–80 °C) and is compared to values provided by Anton Paar (Bettin et al.).⁷ The calibration is considered acceptable if the data are within $\pm 5 \times 10^{-5} \text{ g}\cdot\text{cm}^{-3}$ of published values. If necessary, a calibration at 40 °C and 60 °C can be initiated. The densities of water were measured using deionized water.⁸ Density measurements were reproducible to $\pm 2 \times 10^{-5} \text{ g}\cdot\text{cm}^{-3}$. In the viscosity measurement, the temperature was controlled by means of a digital controller (± 0.004 °C) in a well-stirred water bath to better than ± 0.01 °C as measured by a Cole-Parmer resistance thermometer (model H-01158-65, Anjou, Québec, Canada). Viscosities were determined with six different viscometers to cover, with precision, the range of temperature from 25 °C to 70 °C. A series of Cannon-Ubbelohde viscometers (0, 0B, 0C, 1, 1B, 2C—Cole Parmer) were used. The efflux time was measured with a hand-held digital stopwatch capable of measuring time to within ± 0.01 s. Experiments were repeated a minimum of four times at each temperature for all compositions. The equation for kinematic viscosity, according to Poiseuille's law, is

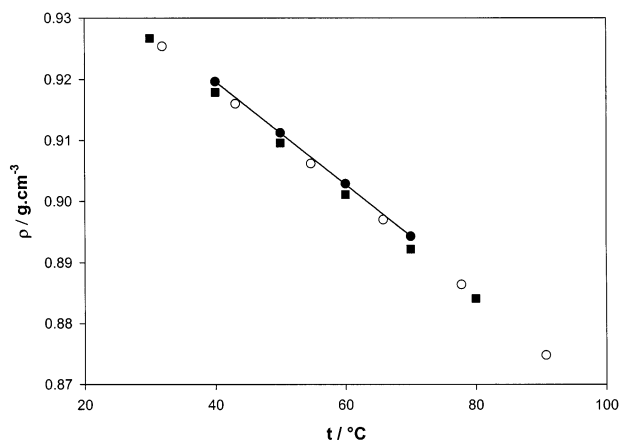
$$\nu = k_1 t - k_2/t \quad (1)$$

where t is the efflux time and k_1 and k_2 are the viscometer constants. The second term representing the correction due to the kinetic energy was found to be negligible.⁹ The value of the absolute viscosity (η) was obtained by multiplying the measured kinematic viscosity (ν) by the measured density.

Calibration constants for the two viscometers were checked using high-purity ethylene glycol (EG), diethylene glycol (DEG), and water. A review of published values of the kinematic and dynamic viscosities of EG and DEG, with new experimental data, was published by Lee and Teja.¹⁰

Table 1. Densities of Water (1) + AMP (2) Mixtures at Various Temperatures

x_2	$\rho/\text{g}\cdot\text{cm}^{-3}$ at the following $t/^\circ\text{C}$					
	25 °C	30 °C	40 °C	50 °C	60 °C	70 °C
0.0000	0.997 04	0.995 65	0.992 21	0.988 04	0.983 12	0.977 77
0.0503	0.996 94	0.994 70	0.989 66	0.984 18	0.978 15	0.971 66
0.0704	0.997 22	0.994 60	0.988 95	0.982 83	0.976 26	0.969 29
0.1005	0.997 05	0.993 88	0.987 50	0.980 60	0.973 43	0.965 83
0.2006	0.989 06	0.985 15	0.977 24	0.969 17	0.960 92	0.952 44
0.2939	0.978 77	0.974 76	0.966 66	0.958 38	0.949 92	0.941 28
0.4075	0.967 43	0.963 43	0.955 29	0.946 98	0.938 48	0.930 00
0.4982	0.959 61	0.955 61	0.947 43	0.939 08	0.930 56	0.921 85
0.5996	0.952 01	0.948 00	0.939 82	0.931 58	0.923 08	0.914 39
0.7028	0.945 34	0.941 38	0.933 10	0.924 70	0.915 97	0.907 00
0.8016	0.939 53	0.935 53	0.927 27	0.918 85	0.910 36	0.901 65
0.9001	0.934 80	0.930 70	0.923 35	0.914 78	0.906 31	0.897 77
1.0000			0.919 65	0.911 24	0.902 87	0.894 28

**Figure 1.** Comparison of the densities of pure AMP at various temperatures: ●, this work; ■, Li and Lie;⁵ ○, Xu et al.⁴

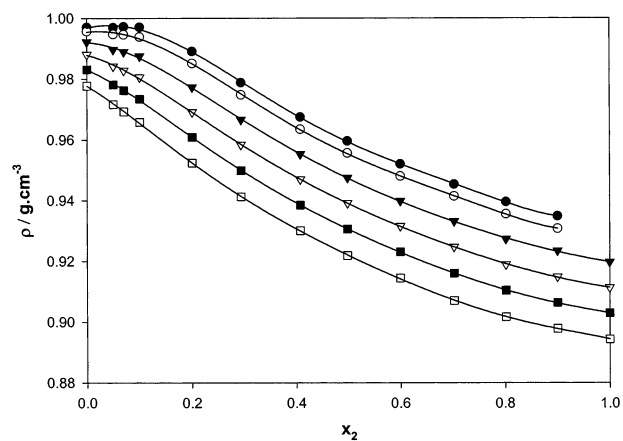
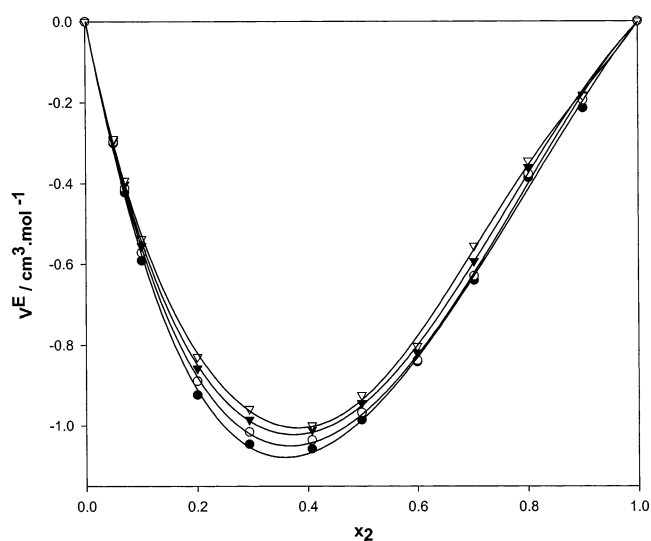
Values of the viscosity of water were taken from Stokes and Mills.¹¹ The values of the viscosities were reproducible to ± 0.003 mPa·s. From the overall average percent deviation of the means of the average efflux time and the accuracy of the density measurement, we estimate the uncertainty of the absolute viscosity to be less than 0.5%.

Results and Discussion

Experimentally measured densities of the aqueous AMP solutions at (25, 30, 40, 50, 60, and 70) °C throughout the whole concentration range are listed in Table 1. The pure values obtained are shown in Figure 1 and were consistently higher than the data published by Xu et al.⁴ but closer to them than to those of Li and Lie.⁵ Our values were on average 0.17% higher than those of Li and Lie, well below their reported accuracy of $\pm 0.5\%$. Both groups of authors used Gay-Lussac pycnometers and water baths regulated to ± 0.05 °C. Our values were also on average 0.24% higher than those of Aguila-Hernandez et al. (2001).¹² This group used a 95% purity AMP and an Anton Paar DMA-45 density meter in their measurements. It is possible that the purity of the AMP contributes to the discrepancy in density measurements found in the literature. The curves show an unexpected decrease in the density, for alkanolamines, as AMP is added to water, and the minimum occurs around the pure AMP value for all temperatures. The values of the measured density are shown in Figure 2.

The density values of the binary mixtures were used to calculate the excess molar volume, V_m^E , as

$$V_m^E = V_m - V_1^0 x_1 - V_2^0 x_2 \quad (2)$$

**Figure 2.** Densities of the water (1) + AMP (2) system at various temperatures: ●, 25 °C; ○, 30 °C; ▼, 40 °C; ▽, 50 °C; ■, 60 °C; □, 70 °C.**Figure 3.** Excess molar volumes of the water (1) + AMP (2) system at various temperatures: ●, 40 °C; ○, 50 °C; ▼, 60 °C; ▽, 70 °C.

where V_m is the molar volume of the mixture and V_1^0 and V_2^0 are those of pure water and pure AMP, respectively; x_1 and x_2 are the mole fractions of the pure components. Figure 3 displays the dependence of V_m^E on the composition at various temperatures. In all cases the V_m^E is negative, as is common for other completely miscible (water + polar organic) solvents with a minimum at around 35 mol % of AMP. These V_m^E values became less negative with increasing temperature, as is also common. Figures 2 and 3 show a sharp change in the density and V_m^E in the water-rich region. Negative values of V_m^E mean that there is a volume contraction and can thus be explained by the large difference in the molar specific volumes. Pal and Singh¹³ concluded that the contraction in the volume is due to the ability of the -OH group to form hydrogen bonds with water molecules. The second interpretation given was that such a marked change in V_m^E might also be due to the accommodation of the nonaqueous molecules occupying in the structured water lattice a void space. It is known that the number of cross-associated H-bonds decreases with increasing temperature, which leads to a positive contribution to V_m^E . As a consequence, V_m^E values became more negative with a decrease in temperature.

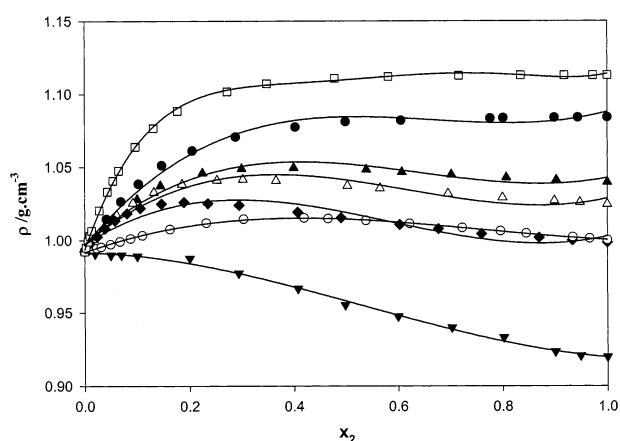
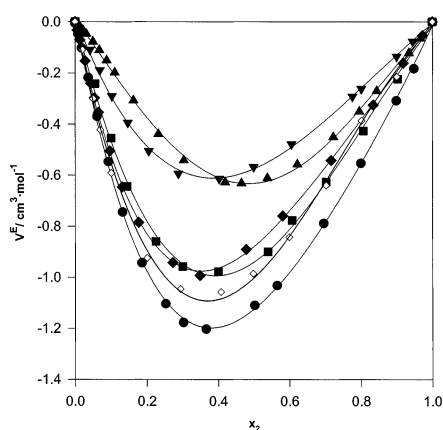
The values of the density of various alkanolamines are shown in Figure 4 for comparison. As shown, AMP has its

Table 2. Redlich–Kister Equation Fitting Coefficients of the Excess Volumes ($V_m^E/\text{cm}^3\cdot\text{mol}^{-1}$) for Water (1) + AMP (2) Mixtures at Various Temperatures

t °C	a_0	a_1	a_2	a_3	a_4	a_5	σ $\text{cm}^3\cdot\text{mol}^{-1}$
40	-3.9471	-1.9875	-0.3412	-3.7857	-0.5460	4.5557	0.009
50	-3.8912	-1.8561	-0.1142	-3.5617	-0.5315	3.8682	0.007
60	-3.8010	-1.8835	0.2777	-3.2002	-1.0114	3.4446	0.006
70	-3.7451	-2.0043	0.8955	-2.6455	-1.9426	3.2003	0.009

Table 3. Coefficients of the Polynomial $\rho/\text{g}\cdot\text{cm}^{-3} = \sum_0^n a_k x_2^k$ and the Standard Deviations for the Aqueous AMP Solutions at Various Temperatures

t °C	a_0	a_1	a_2	a_3	a_4	a_5	σ $\text{cm}^3\cdot\text{mol}^{-1}$
25	0.996 64	0.055 97	-0.718 89	1.442 00	-1.261 03	0.416 92	0.0005
30	0.995 78	0.008 62	-0.427 05	0.644 12	-0.299 92		0.0006
40	0.992 03	-0.013 89	-0.467 12	0.997 55	-0.879 03	0.290 21	0.0004
50	0.987 94	-0.050 63	-0.352 28	0.836 76	-0.779 54	0.269 08	0.0003
60	0.983 11	-0.082 56	-0.246 21	0.667 40	-0.650 25	0.231 48	0.0003
70	0.978 19	-0.130 97	0.004 77	0.080 77	-0.038 57		0.0004

**Figure 4.** Densities of various aqueous alkanolamine systems at 40 °C: \square , TEA;¹⁸ \bullet , DEA;¹⁸ \blacktriangle , DGA;²² \triangle , MDEA;⁶ \blacklozenge , EDEA;⁶ \circ , MEA;¹⁸ \blacktriangledown , AMP.**Figure 5.** Excess molar volumes of various aqueous alkanolamine systems at 40 °C: \bullet , MDEA⁶; \diamond , AMP; \blacksquare , DGA;²² \blacktriangle , TEA;¹⁸ \blacklozenge , MEA;¹⁸ \blacktriangledown , DEA.¹⁸

density decrease with the addition of the alkanolamine. The values of the densities of aqueous alkanolamines are the largest for TEA followed by DEA, DGA, MDEA, EDEA, MEA, and AMP.

In Figure 5, a comparison of the excess volumes of several alkanolamines shows that the addition of MDEA to water brings the largest decrease in the excess volume, followed by AMP, DGA, triethanolamine (TEA), then diethanolamine (DEA), and monoethanolamine (MEA). An interesting fact is that Chang et al.,¹⁴ using freezing point

depression measurement and total pressure data, found that, at 0 °C and 40 °C, MDEA and DGA solutions deviated the most from ideality.

A Redlich–Kister¹⁵ relation was used to correlate the excess volume data.

$$V_m^E/\text{cm}^3\cdot\text{mol}^{-1} = x_1 x_2 \sum_{f=0}^n a_f (x_1 - x_2)^f \quad (3)$$

The coefficients and the standard deviation (σ) are presented in Table 2. It is well-known that the Redlich–Kister relation does not correlate well unsymmetrical curves of excess volumes (and viscosity deviations). The introduction of a number of skewing factors did not reduce the number of coefficients used. We thus present the correlation of the densities of the solutions at each temperature with the following polynomial:

$$\rho/\text{g}\cdot\text{cm}^{-3} = \sum_{k=0}^n a_k x_2^k \quad (4)$$

The coefficients and the standard deviation (σ) are presented in Table 3. Hepler¹⁶ and Neal and Goring¹⁷ recognized the usefulness of thermal expansion data for revealing correlations between the molecular structure of solutes and their effects on water structure. 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 $(\partial^2 V_2^{\infty}/\partial T^2)_P$ to classify solutes as “structure makers” or “structure breakers”. A positive sign corresponds to a structure-making solute while a negative sign corresponds to a structure-breaking solute.

Both the partial molar volume of water at infinite dilution (V_1^{∞}) in AMP and the partial molar volume of AMP at infinite dilution (V_2^{∞}) in water were obtained using the method proposed by Maham et al.⁶ The apparent molar volume of water ($V_{\phi,1}$) and the apparent molar volume of AMP in water ($V_{\phi,2}$) were first calculated as

$$V_{\phi,1} = V_1^{\infty} + [V_m^E/(1 - x_2)] \quad (5)$$

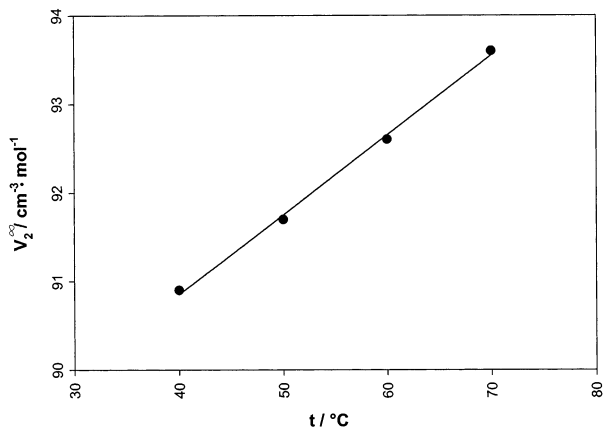
and

$$V_{\phi,2} = V_2^{\infty} + (V_m^E/x_2) \quad (6)$$

where V_1^{∞} and V_2^{∞} are the molar volumes are the mole fractions of pure water and AMP, respectively. An analyti-

Table 4. Partial Molar Volumes of AMP at Infinite Dilution in Water (V_2^∞) and of Water at Infinite Dilution in AMP (V_1^∞)

t °C	V_2^∞ cm ³ ·mol ⁻¹	V_1^∞ cm ³ ·mol ⁻¹
40	90.9	14.5
50	91.7	15.2
60	92.6	15.4
70	93.5	15.1

**Figure 6.** Partial molar volumes of AMP at infinite dilution in water at various temperatures: —, linear regression.

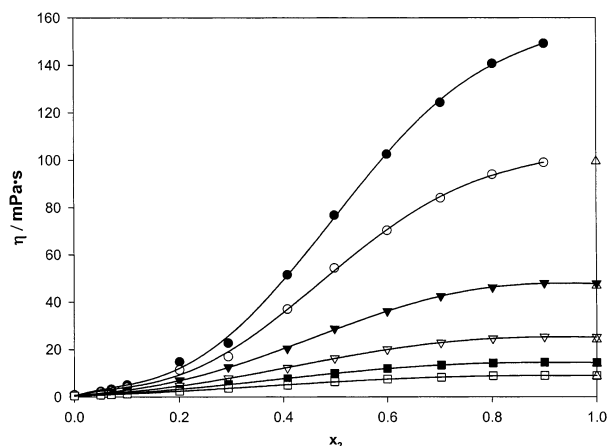
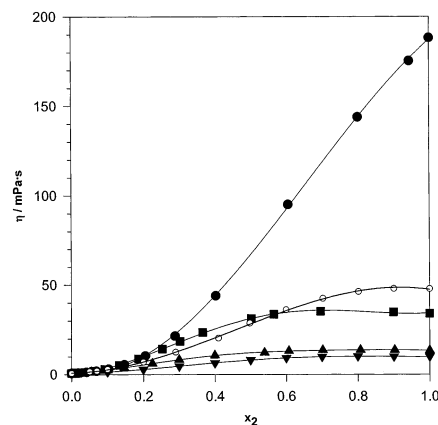
cal extrapolation of $V_{\phi,1}$ to $x_2 = 1$ led to V_1^∞ , and a similar extrapolation to $x_2 = 0$ led to V_2^∞ . Partial molar volumes at infinite dilution are listed in Table 4. V_2^∞ values varied linearly with temperature, as shown in Figure 6. According to the criterion described above, the solute (AMP) would be considered as having no net effect on the structure of water. This is consistent with the conclusion of Maham et al.^{6,18} concerning other alkanolamines (MEA, DEA, TEA, MDEA, and EDEA). All the values of the molar volumes at infinite dilution (V_2^∞) were smaller than the corresponding molar values of pure AMP (V_2^∞). The change in the excess volume can be explained by the AMP molecule fitting (partially) into the open, or empty, space in liquid water. This picture does not invoke either the structure-making or the structure-breaking properties and is consistent with the observation that $(\partial^2 V_2^\infty / \partial T^2)_P$ values are approximately equal to zero.

Having fewer values of V_m^E for x_2 near unity, the accuracies of V_1^∞ may be lower. It is still clear that $(\partial V_1^\infty / \partial T)_P$ was positive. All values of V_1^∞ were smaller than the corresponding molar volumes of pure water (V_1^∞) at the same temperature. As mentioned by Maham et al.,¹⁸ this is consistent with the idea that the molar volume of pure water is the sum of the actual molecular volumes plus the "empty" volume that arises from the hydrogen-bonded open structure of water.

Experimentally measured viscosities of the binary solutions of aqueous AMP solutions at (25, 40, 50, 60, and 70) °C are listed in Table 5 and shown in Figure 7. Data published by Li and Lie⁵ were also added to Figure 7 for comparison. Curves for the system water + AMP have an S-shape with a maximum in the AMP-rich region. Figure 7 shows a sharp increase in the viscosity of the mixture after the addition of AMP (40 mol %). The effect of the composition on viscosity decreases with an increase in temperature. The largest viscosity values were those of pure AMP at all temperatures. Aqueous MDEA, DGA, and MEA solutions were also shown to have this S-shaped form, as shown in Figure 8. Fort and Moore¹⁹ and Liler and

Table 5. Experimental Viscosities for Water (1) + AMP (2) Mixtures at Various Temperatures

x_2	η /mPa·s at the following t /°C					
	25 °C	30 °C	40 °C	50 °C	60 °C	70 °C
0.0000	0.890	0.805	0.653	0.547	0.466	0.405
0.0503	2.32	1.980	1.608	1.244	0.931	0.826
0.0704	3.23	2.67	1.931	1.466	1.154	0.933
0.1005	5.01	4.05	2.79	2.03	1.55	1.21
0.2006	14.82	11.32	7.00	4.66	3.27	2.40
0.2939	22.7	16.96	12.51	7.85	5.23	3.67
0.4075	51.5	36.9	20.37	12.17	7.63	4.94
0.4982	76.8	54.3	28.7	16.26	10.00	6.43
0.5996	102.5	70.3	36.1	20.0	11.97	7.50
0.7028	124.3	84.0	42.5	22.7	13.33	8.23
0.8016	140.7	94.0	46.2	24.6	14.22	8.88
0.9001	149.1	98.9	47.9	25.3	14.53	8.98
1.0000			47.8	25.1	14.40	8.91

**Figure 7.** Viscosities of the water (1) + AMP (2) system at various temperatures: ●, 25 °C; ○, 30 °C; ▼, 40 °C; ▽, 50 °C; ■, 60 °C; □, 70 °C; △, Li and Lie.⁵**Figure 8.** Comparison of the viscosities of various aqueous alkanolamine solutions at 40 °C: ●, DEA;²³ ○, AMP; ■, MDEA;²³ ▲, DGA;²² ▼, MEA.²⁴

Kosanović²⁰ state that an S-shaped viscosity curve and the presence of a maximum in the viscosity deviation curves indicate the formation of stable complexes. The viscosity of AMP at 40 °C was larger than that of MDEA, DGA, and MEA but was smaller than that of DEA.

Experimental viscosity values of the binary mixtures were used to calculate the viscosity deviation, defined by

$$\Delta\eta = \eta - \eta_1 x_1 - \eta_2 x_2 \quad (7)$$

where η is the viscosity of the mixture and η_1 and η_2 are those of pure water and pure AMP, respectively; x_1 and x_2

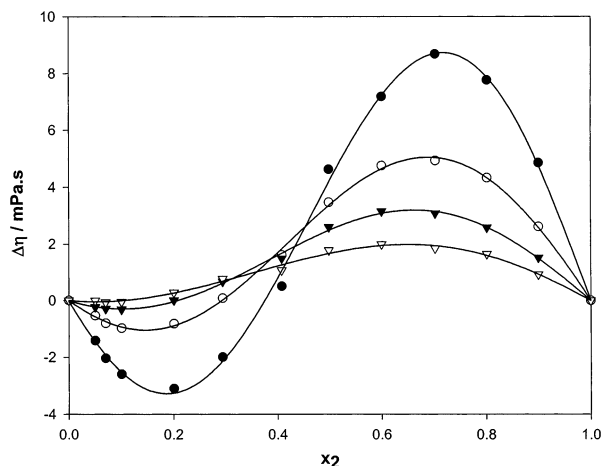


Figure 9. Viscosity deviations for the water (1) + AMP (2) system at various temperatures: ●, 40 °C; ○, 50 °C; ▼, 60 °C; ▽, 70 °C.

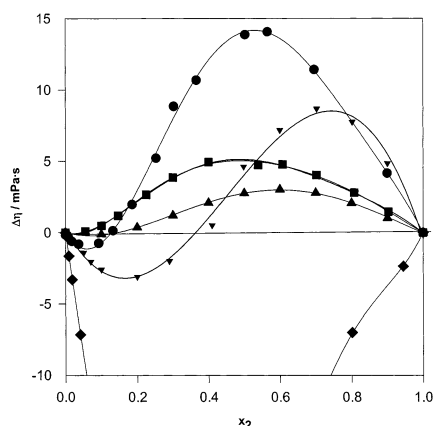


Figure 10. Comparison of the viscosity deviations of various aqueous alkanolamine solutions at 40 °C: ●, MDEA;²³ ■, DGA;²² ▲, MEA;²⁴ ▼, AMP; ◆, DEA.²³

are the mole fractions of the pure components. The viscosity deviation represents deviations from a rectilinear dependence of viscosity on mole fraction. Figure 9 displays the dependence of $\Delta\eta$ on the composition and temperature. Values of $\Delta\eta$ were all negative up to 40 mol % with a minimum at 20 mol % and then became positive throughout the whole concentration range with a maximum around 70 mol % for all temperatures. Figure 10 displays the different viscosity curves of aqueous solutions of two primary alkanolamines (MEA and DGA), a secondary alkanolamine (DEA), a tertiary alkanolamine (MDEA), and a primary hindered amine (AMP).

Note that the viscosity deviations of dilute DGA have a value of zero up to a mole fraction of 0.1. Both MDEA and MEA viscosity deviations are negative in the water-rich region (<0.2 mole fraction) at all temperatures and then become positive. DEA viscosity deviations calculated from Teng et al.²³ were negative for all compositions at all temperatures except at 70 °C and 80 °C, where they became positive in the DEA-rich region. In summary, all alkanolamines show a change in the sign of the viscosity deviations with a minimum in the water-rich region and a maximum in the amine-rich region. This remark applies to all classes of alkanolamines (primary, secondary, tertiary, and hindered alkanolamines).

In comparing water + ethylene glycol, water + 2-methoxyethanol, and water + MEA, Pagé et al.²¹ interpreted the results in a way that can explain the change in the sign of the viscosity deviation and the presence of a

Table 6. Redlich–Kister Equation Fitting Coefficients of the Viscosity Deviations ($\Delta\eta/\text{mPa}\cdot\text{s}$) for Water (1) + AMP (2) Mixtures at Various Temperatures

t °C	a_0	a_1	a_2	a_3	a_4	a_5	σ mPa·s
40	17.072	-72.174	-8.859	57.205	4.712	-37.998	0.25
50	13.598	-33.030	-9.949	24.577	5.737	-18.137	0.10
60	10.026	-17.287	-6.744	17.038	3.091	-15.354	0.08
70	6.691	-8.317	-2.376	6.644	0.143	-6.149	0.10

Table 7. Coefficients of the Polynomial $\ln \eta/\text{mPa}\cdot\text{s} = \ln \eta_0 + \sum_{k=1}^n a_k x_2^k$ and the Standard Deviations for the Water (1) + AMP (2) Mixtures at Various Temperatures^a

t °C	a_0	a_1	a_2	a_3	a_4	a_5
25	22.627	-67.322	137.110	-143.375	56.582	0.07
30	21.047	-61.664	125.419	-131.883	52.368	0.07
40	18.576	-45.781	74.028	-64.246	21.722	0.03
50	16.843	-41.624	66.893	-57.753	19.475	0.03
60	15.146	-36.545	56.947	-48.085	15.973	0.01
70	14.465	-38.175	63.103	-54.560	18.263	0.03

^a η_0 is the viscosity of pure water.

minimum in one region and a maximum in another region. This interpretation appears to be in contradiction with the structure neutrality picture resulting from the application of Hepler's criteria. Pagé et al.²¹ divided the concentration region into three parts: (i) $0 < x_2 < 0.1$, where the cosolvent disrupts the cooperative fluctuation units of liquid water; (ii) $0.1 < x_2 < 0.3$, where the cosolvent eliminates the extensive hydrogen-bonding connectivity between water molecules; and (iii) $x_2 > 0.3$, progressive replacement of water–cosolvent interactions by cosolvent–cosolvent interactions.

The calculated values of $\Delta\eta$ were correlated with a Redlich–Kister¹⁵ relation:

$$\Delta\eta/\text{mPa}\cdot\text{s} = x_1 x_2 \sum_{i=0}^n a_i (x_1 - x_2)^i \quad (8)$$

where x_1 is the mole fraction of water and x_2 is the mole fraction of AMP. The coefficients and the standard deviation (σ) are presented in Table 6. We also present the correlation of the viscosities of the solutions at each temperature with the following polynomial:

$$\ln \eta/\text{mPa}\cdot\text{s} = \ln \eta_0 + \sum_{k=1}^4 a_k x_2^k \quad (9)$$

where η is the viscosity of the binary solution, η_0 is the viscosity of pure water, and x_2 is the mole fraction of AMP. The values of the polynomial coefficients a_k are listed in Table 7.

Conclusions

This paper reports experimental data for the densities and viscosities of the aqueous AMP solutions over a range of temperature from 25 °C to 70 °C. The calculated V_m^E values for the aqueous AMP solutions were all negative at all temperatures and compositions. The criterion proposed by Hepler¹⁶ suggests that the addition of AMP to water had no effect on its structure, a conclusion similar to that of Maham et al.^{6,18} for MEA, DEA, TEA, MEA, and EDEA and Henni et al.²² for DGA. The viscosity deviations $\Delta\eta$ for AMP + water were negative in the water-rich region and then became positive in the amine-rich region at all

temperatures. This conclusion is valid for all alkanolamines studied thus far (MEA, DEA, TEA, MEA, DGA, and EDEA).

Aqueous AMP, DGA, MDEA, and MEA solutions exhibited S-shaped viscosity curves. Aqueous DEA viscosities did not have an S-shaped curve but still exhibited a change in the sign of the viscosity deviations from negative to positive in the DEA-rich region. The explanations given by Pagé et al.²¹ are in contradiction with the structure neutrality resulting from applying Hepler's criterion.

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