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Density and Viscosity of Aqueous Solutions of (*N*-Methyldiethanolamine + Monoethanolamine), (*N*-Methyldiethanolamine + Diethanolamine), (2-Amino-2-methyl-1-propanol + Monoethanolamine), and (2-Amino-2-methyl-1-propanol + Diethanolamine)

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The densities and viscosities of aqueous blends of *N*-methyldiethanolamine (MDEA) and 2-amino-2-methyl-1-propanol with monoethanolamine (MEA) and diethanolamine (DEA) have been measured at (25, 30, 35, 40, 45, and 50) °C. The total amine strength in the solution was kept at 30 mass % in view of recent interest in using concentrated amine solutions in gas treating. Correlations for the density and viscosity of the ternary mixtures are presented.

Introduction

The removal of acid gas impurities such as CO_2 and H_2S from natural, refinery, and synthesis gas streams is a significant operation in gas processing. A wide variety of alkanolamines such as monoethanolamine (MEA), diethanolamine (DEA), di-2-propanolamine (DIPA), and *N*-methyldiethanolamine (MDEA) has been used for industrial gas treating.[1] A different class of amines, the sterically hindered amines, for example, 2-amino-2-methyl-1-propanol (AMP), has been suggested recently as attractive solvents for removal of acid gas impurities from sour gas streams. The use of mixed amine systems in gas treating processes is of increasing interest today. The mixed amine systems, which combine the higher equilibrium capacity of the tertiary amine with the higher reaction rate of the primary or secondary amine, can bring about considerable improvement in gas absorption and great savings in regeneration energy requirements. Blends of primary and tertiary amines (such as mixtures of MEA and MDEA) or secondary and tertiary amines (such as mixtures of DEA and MDEA) have been suggested for industrial gas treating processes.[1,2] As in MDEA, the CO_2 loading in AMP approaches a value of 1.0 mol of CO_2 per mol of amine, while the reaction rate constant for CO_2−AMP is much higher than that for CO_2−MDEA.[3] Since the sterically hindered amine, AMP, does not form a stable carbamate,[3] bicarbonate and carbonate ions may be present in the solution in larger amounts than the carbamate ions. Hence, the regeneration energy costs when aqueous solutions of AMP are used to absorb CO_2 may be lower, as in the case of using aqueous MDEA solutions. From these considerations (AMP + MEA + H_2O) and (AMP + DEA + H_2O) appear to be attractive new blended amine solvents in addition to (MDEA + MEA + H_2O) and (MDEA + DEA + H_2O) for the gas treating processes. The physical properties, for example, density and viscosity, of aqueous mixed amine solutions are essential for the rational design of gas treating units and for optimum gas treating processes. Knowledge of the physical properties of process solutions is necessary for the operation of process equipment such as pumps and heat exchangers in gas treating units. Solution density and viscosity are also important in the mass transfer rate modeling of absorbers and regenerators because these properties affect the liquid-film coefficient for mass transfer.

The density and viscosity data of some aqueous blended amine solutions have been reported in the literature, such as (MEA + MDEA + H_2O),[4−6] (MEA + AMP + H_2O),[5] (DEA + MDEA + H_2O),[7−10] and (DEA + AMP + H_2O).[9,10] However, published literature on the density and viscosity of (MEA + AMP + H_2O) and (DEA + AMP + H_2O) are sparse. Also, most of the density and viscosity measurements for the blended amine solvents have been done earlier over the temperature range (303 to 353) K. Hence, in this work the density and viscosity measurements have been done in the temperature range (293 to 323) K to cover the gap.

In this work the density and viscosity of aqueous solutions of MDEA + MEA, MDEA + DEA, AMP + MEA, and AMP + DEA were measured over the temperature range (20 to 50) °C. The total amine strength in the solution was kept at 30 mass %. The densities of the ternary mixtures have been correlated in this work using the Redlich−Kister equation. The viscosities of the ternary mixtures have been correlated successfully using the equation of Grunberg and Nissan.

Experimental Section

Reagent grade MEA, DEA, AMP, and MDEA of 98% purity were obtained from E. Merck. Distilled water degassed by boiling was used for making the amine solutions. The total amine contents of the solutions were determined by titration with standard HCl using methyl orange indicator.

Density. The densities of the amine solutions were measured using a 25.76 × 10^{-6} m^3 Gay−Lussac pycnom-

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Table 1. Density, ρ, for MDEA (2) + MEA (1) + H_2O (3), MDEA (2) + DEA (1) + H_2O (3), AMP (2) + MEA (1) + H_2O (3), and AMP (2) + DEA (1) + H_2O (3) from 20 °C to 50 °C

	ρ/kg·m^{-3} at the following t/°C						
mass %/mass %	20	25	30	35	40	45	50
MDEA/MEA							
30/0	1032.2	1027.5	1023.1	1019.9	1018.2	1014.2	1011.2
28.5/1.5	1031.2	1026.0	1022.0	1019.1	1017.2	1013.5	1010.5
27/3	1030.2	1025.1	1021.0	1018.4	1016.1	1012.9	1009.7
25.5/4.5	1029.1	1024.4	1020.5	1017.8	1014.9	1012.1	1009.1
24/6	1028.4	1023.5	1019.6	1016.9	1013.6	1011.5	1008.1
22.5/7.5	1027.4	1022.7	1018.8	1016.2	1012.8	1010.7	1007.1
21/9	1025.9	1021.5	1018.0	1015.6	1012.1	1009.9	1006.0
0/30	1015.3	1012.1	1009.2	1004.9	1003.2	1001.0	997.76
MDEA/DEA							
28.5/1.5	1032.5	1028.0	1023.5	1020.2	1018.6	1014.8	1011.7
27/3	1033.0	1028.4	1023.9	1020.5	1019.0	1015.2	1012.1
25.5/4.5	1034.4	1028.8	1024.2	1020.9	1019.3	1015.7	1012.5
24/6	1034.8	1029.1	1024.6	1021.3	1019.7	1016.0	1012.9
22.5/7.5	1035.1	1029.5	1025.0	1021.7	1020.0	1016.4	1013.5
21/9	1035.5	1029.9	1025.5	1022.0	1020.6	1016.9	1014.1
0/30	1042.7	1037.4	1033.2	1029.1	1026.4	1022.7	1019.3
AMP/MEA							
30/0	999.85	997.41	994.62	991.22	988.72	986.24	983.13
28.5/1.5	1000.7	998.23	995.45	992.97	989.19	987.31	984.31
27/3	1000.7	998.81	996.21	993.56	990.08	988.01	986.01
25.5/4.5	1002.2	999.80	997.13	994.42	991.06	988.54	987.04
24/6	1002.7	1000.3	997.90	995.66	992.48	989.40	987.85
22.5/7.5	1003.5	1000.0	998.81	996.32	993.89	990.47	988.47
21/9	1004.4	1001.9	999.41	996.57	994.23	991.01	989.33
AMP/DEA							
28.5/1.5	1002.1	998.99	996.53	993.93	990.30	989.00	985.97
27/3	1003.9	1001.0	998.21	995.83	992.60	990.30	988.10
25.5/4.5	1005.7	1003.1	1000.2	998.81	993.85	992.71	990.15
24/6	1006.9	1004.6	1002.8	999.57	996.30	993.42	991.49
22.5/7.5	1008.8	1005.7	1004.8	1001.7	998.13	995.82	992.91
21/9	1010.4	1006.4	1006.4	1003.2	1001.1	997.60	994.32

eter. The pycnometer containing the amine solution was immersed in a constant-temperature bath. The bath temperature was controlled within ±0.2 K of the desired temperature using a circulator temperature controller (JULABO FP 55, FRG). Once the solutions reached the desired temperature, they were weighed to within ±0.0002 g with a Dhona 100 DS balance. Each reported value was the average of at least three measurements. The average absolute deviations (AADs) for the density measurements have been found to be within 0.04%.

Viscosity. The viscosity was measured using an Ostwald viscometer. The viscometer was immersed in a thermostated bath. The bath temperature was controlled within ±0.2 K of the desired level with a circulator temperature controller (JULABO FP 55, FRG). Each reported value was the average of at least three measurements. The average absolute deviations (AADs) for the viscosity measurements have been found to be within 0.03%.

Results and Discussion

Density. The measured densities of the solutions of (MDEA + MEA + H_2O), (MDEA + DEA + H_2O), (AMP + MEA + H_2O), and (AMP + DEA + H_2O) are presented in Table 1. The density measurements of this study are in good agreement with the literature results. For 30 mass % MDEA, over the temperature range (303 to 323) K, the experimental data of this study show 0.04% deviation from the experimental data of Li and Shen[4] and 0.05% from the experimental data of Li and Lie.[5] For the amine blends (28.5 mass % MDEA + 1.5 mass % MEA), (27.0 mass % MDEA + 3.0 mass % MEA), and (25.5 mass % MDEA + 4.5 mass % MEA) over the temperature range (303 to 323) K, the experimental data of this study show 0.03%, 0.04%, and 0.04% deviations, respectively, from the experimental data of Hagewiesche et al.[6] For 30 mass % DEA and the (24.0 mass % MDEA + 6.0 mass % DEA) blend, over the temperature range (303 to 323) K, the experimental data of this study show 0.19% and 0.06% deviations, respectively, from the experimental data of Hsu and Li.[9] For the 30 mass % AMP and the (24.0 mass % AMP + 6.0 mass % MEA) blend, over the temperature range (303 to 323) K, the experimental data of this study show 0.04% and 0.05% deviations, respectively, from the experimental data of Li and Lie.[5] For the (AMP + DEA + H_2O) system the experimental data of this study show a 0.19% deviation, from the experimental data of Hsu and Li[9] for the (24.0 mass % AMP + 6.0 mass % DEA) blend, over the temperature range (303 to 323) K. As shown in Table 1, densities of the ternary mixtures decrease with increasing temperature and decreasing mass % of MDEA and AMP in the mixture.

To correlate the density of liquid mixtures, a Redlich−Kister type equation for the excess molar volume is applied. For a binary system, the Redlich−Kister equation has the following expression:

$$V_{jk}^{E}/\mathrm{m^3\cdot kmol^{-1}} = x_j x_k \sum_{i=0}^{n} A_i (x_j - x_k)^i \qquad (1)$$

where A_i are pair parameters and are assumed to be temperature dependent:

$$A_i = a + b(T/\mathrm{K}) + c(T/\mathrm{K})^2 \qquad (2)$$

The excess volume of liquid mixtures for the ternary system is assumed to be

$$V^{E} = V_{12}^{E} + V_{13}^{E} + V_{23}^{E} \qquad (3)$$

Table 2. Binary Parameters, A_0, A_1, and A_2, of the Equation $V^{E}_{jk}/\text{m}^3\cdot\text{kmol}^{-1} = x_j x_k \sum_{i=0}^{n} A_i (x_j - x_k)^i$ for the Excess Volume for MDEA (2) + MEA (1) + H_2O (3)[a]

parameter		MEA + H_2O	MDEA + H_2O	MEA + MDEA
A_0	*a*	57.5000	295.790	5.88420
	b	−26.5203	−6.49510	−29.2633
	c	0.25580	−14.5927	−0.02920
A_1	*a*	−72.5441	−283.406	−0.05250
	b	−54.0621	−66.1991	−43.8061
	c	0.62970	−33.0817	0.99040
A_2	*a*	69.4650	238.139	0.32420
	b	−25.1257	−70.3239	13.8561
	c	0.38620	−18.7422	−0.38960

[a] AAD% = 0.1; no. of data points = 71.

Table 3. Binary Parameters, A_0, A_1, and A_2, of the Equation $V^{E}_{jk}/\text{m}^3\cdot\text{kmol}^{-1} = x_j x_k \sum_{i=0}^{n} A_i (x_j - x_k)^i$ for the Excess Volume for AMP (2) + MEA (1) + H_2O (3)[a]

parameter		MEA + H_2O	AMP + H_2O	MEA + AMP
A_0	*a*	137.363	148.188	5.83210
	b	13.6933	−30.0002	13.0217
	c	1.10680	−5.20980	−0.18560
A_1	*a*	−113.866	−113.932	0.51520
	b	13.0687	−28.1710	−38.3834
	c	2.89720	−12.5781	3.15840
A_2	*a*	95.4488	91.9730	0.77880
	b	−6.44560	8.84230	1.66520
	c	1.89620	−7.59000	−0.62640

[a] AAD% = 0.05; no. of data points = 71.

Table 4. Binary Parameters, A_0, A_1, and A_2, of the Equation $V^{E}_{jk}/\text{m}^3\cdot\text{kmol}^{-1} = x_j x_k \sum_{i=0}^{n} A_i (x_j - x_k)^i$ for the Excess Volume for MDEA (2) + DEA (1) + H_2O (3)[a]

parameter		DEA + H_2O	MDEA + H_2O	DEA + MDEA
A_0	*a*	34.5659	102.774	1.26600
	b	6.70150	7.06800	−23.8194
	c	6.83730	−10.5590	−0.01520
A_1	*a*	−31.7193	−100.591	0.22450
	b	22.6324	28.8465	−13.7526
	c	15.7516	−24.1089	10.4421
A_2	*a*	30.2581	90.4300	0.95990
	b	19.0276	24.4659	−2.04040
	c	9.06940	−13.7623	−0.58380

[a] AAD% = 0.07; no. of data points = 74.

Table 5. Binary Parameters, A_0, A_1, and A_2, of the Equation $V^{E}_{jk}/\text{m}^3\cdot\text{kmol}^{-1} = x_j x_k \sum_{i=0}^{n} A_i (x_j - x_k)^i$ for the Excess Volume for AMP (2) + DEA (1) + H_2O (3)[a]

parameter		DEA + H_2O	AMP + H_2O	DEA + AMP
A_0	*a*	55.6692	171.198	3.17040
	b	−16.1125	10.6782	22.4062
	c	−7.61780	5.07230	−0.24090
A_1	*a*	−50.0529	−141.524	−0.16940
	b	−4.72700	−12.8927	−18.5543
	c	−17.8144	12.1578	−8.11700
A_2	*a*	45.8656	129.198	0.21610
	b	17.7183	−31.5165	4.09050
	c	−10.4162	7.28710	−1.26730

[a] AAD% = 0.08; no. of data points = 74.

Table 6. Viscosity, η, for MDEA (2) + MEA (1) + H_2O (3), MDEA (2) + DEA (1) + H_2O (3), AMP (2) + MEA (1) + H_2O (3), and AMP (2) + DEA (1) + H_2O (3) from 20 °C to 50 °C

mass %/mass %	η/mPa·s at the following t/°C 20	25	30	35	40	45	50
MDEA/MEA							
30/0	3.46	3.02	2.68	2.27	1.92	1.72	1.49
28.5/1.5	3.44	2.85	2.55	2.27	1.88	1.72	1.47
27/3	3.44	2.84	2.53	2.26	1.86	1.71	1.46
25.5/4.5	3.43	2.81	2.51	2.25	1.84	1.69	1.44
24/6	3.42	2.78	2.49	2.23	1.82	1.68	1.43
22.5/7.5	3.38	2.75	2.46	2.21	1.80	1.67	1.40
21/9	3.33	2.72	2.45	2.19	1.78	1.65	1.39
0/30	2.63	2.20	2.10	1.85	1.60	1.50	1.29
MDEA/DEA							
28.5/1.5	3.61	2.95	2.62	2.21	1.85	1.67	1.46
27/3	3.68	2.91	2.59	2.19	1.86	1.62	1.40
25.5/4.5	3.79	3.05	2.67	2.24	1.94	1.68	1.49
24/6	3.69	2.94	2.58	2.17	1.89	1.65	1.44
22.5/7.5	3.73	2.99	2.61	2.19	1.86	1.63	1.43
21/9	3.85	3.03	2.69	2.26	1.98	1.66	1.47
0/30	4.22	3.26	2.57	2.51	1.97	1.72	1.41
AMP/MEA							
30/0	4.89	3.79	3.21	2.80	2.24	1.92	1.58
28.5/1.5	4.78	3.71	3.17	2.72	2.19	1.89	1.56
27/3	4.61	3.63	3.11	2.67	2.16	1.87	1.53
25.5/4.5	4.45	3.54	3.04	2.61	2.13	1.83	1.51
24/6	4.35	3.47	2.98	2.57	2.09	1.82	1.49
22.5/7.5	4.22	3.38	2.91	2.53	2.07	1.80	1.47
21/9	4.09	3.26	2.85	2.48	2.04	1.77	1.45
AMP/DEA							
28.5/1.5	4.84	3.76	3.18	2.76	2.21	1.90	1.57
27/3	4.82	3.75	3.16	2.74	2.19	1.89	1.55
25.5/4.5	4.78	3.73	3.15	2.71	2.18	1.88	1.54
24/6	4.77	3.70	3.13	2.70	2.16	1.87	1.52
22.5/7.5	4.75	3.69	3.12	2.68	2.15	1.86	1.49
21/9	4.72	3.68	3.10	2.67	2.13	1.84	1.48

The excess volume of the liquid mixtures can be calculated from the measured density of the fluids

$$V^{E} = V_{m} - \sum x_i V_i^{\circ} \qquad (4)$$

where V_m is the molar volume of the liquid mixture and V_i° is the molar volume of the pure fluids at the system temperature.

The molar volume of the liquid mixtures is calculated by

$$V_{m} = \frac{\sum x_i M_i}{\rho_m} \qquad (5)$$

where M_i is the molar mass of pure component i, ρ_m is the measured liquid density, and x_i is the mole fraction of the pure component i.

A general set of temperature-dependent parameters has been developed using our own experimental data in the temperature range (20 to 50) °C and data from the open literature in the temperature range (60 to 80) °C for the four ternary systems (MEA + MDEA + H_2O), (DEA + MDEA + H_2O), (MEA + AMP + H_2O), and (DEA + AMP + H_2O). For convenience, we have considered MEA or DEA as the first component, MDEA or AMP as the second component, and H_2O as the third component in the ternary mixture. The densities of the pure fluids have been taken from the literature.[9] The determined parameters are presented in Tables 2−5.

Viscosity. The measured viscosities of the solutions of (MDEA + MEA + H_2O), (MDEA + DEA + H_2O), (AMP + MEA + H_2O), and (AMP + DEA + H_2O) are presented in Table 6. The viscosity measurements of this study are in good agreement with the literature data. For 30 mass % MDEA, (26 mass % MDEA + 6 mass % MEA), and 30 mass % MEA, over the temperature range (303 to 323) K, the experimental data of this study show 1.69%, 2.17%, and

Table 7. Parameters, G_{12}, G_{13}, and G_{23}, of the Equation $\ln \eta_m/\mathrm{mPa \cdot s} = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_3 \ln \eta_3 + x_1x_2G_{12} + x_1x_3G_{13} + x_2x_3G_{23}$ for MDEA (2) + MEA (1) + H_2O (3), MDEA (2) + DEA (1) + H_2O (3), AMP (2) + MEA (1) + H_2O (3), and AMP (2) + DEA (1) + H_2O (3)

parameter		ternary pair			
		MDEA + MEA + H_2O	MDEA + DEA + H_2O	AMP + MEA + H_2O	AMP + DEA + H_2O
10^3G_{12}	*a*	2061147.2	540402.42	−20188.704	183225.22
	b	−12381.298	−3432.7482	567.58227	−1239.5022
	c	18.523985	5.3820980	−1.5818676	2.0151020
10^3G_{13}	*a*	−23598.380	286385.54	−7125.3197	290505.08
	b	196.27770	−1639.8636	90.102332	−1666.1670
	c	−0.3298253	2.4226930	−0.1591033	2.4645170
10^3G_{23}	*a*	126845.80	223707.83	329243.12	317856.94
	b	−625.32640	−1223.2260	−1881.0553	−1806.9045
	c	0.8442153	1.7619542	2.7614435	2.6429190

0.81% deviations, respectively, from the experimental data of Li and Lie.[5] For the amine blends (28.5 mass % MDEA + 1.5 mass % MEA), (27.0 mass % MDEA + 3.0 mass % MEA), and (25.5 mass % MDEA + 4.5 mass % MEA) over the temperature range (303 to 323) K, the experimental data of this study show 2.74%, 2.0%, and 2.17% deviations, respectively, from the experimental data of Hagewiesche et al.[6] For 30 mass % DEA and the (24.0 mass % MDEA + 6.0 mass % DEA) blend over the temperature range (303 to 323) K, the experimental data of this study show 2.40% and 2.67% deviations, respectively, from the experimental data of Hsu and Li.[10] For 30 mass % AMP and the (24.0 mass % AMP + 6.0 mass % MEA) blend over the temperature range (303 to 323) K, the experimental data of this study show 3.02% and 3.08% deviations, respectively, from the experimental data of Li and Lie.[5] For the (AMP + DEA + H_2O) system the experimental data of this study show 3.12% deviation, with the experimental data of Hsu and Li[10] for the (24.0 mass % AMP + 6.0 mass % DEA) blend over the temperature range (303 to 323) K. As shown in Table 6, viscosities of the ternary mixtures decrease with increasing temperature and decreasing mass % of MDEA and AMP in the mixture except for the ternary mixtures containing DEA.

For (MDEA + MEA + H_2O) and for (AMP + DEA + H_2O) the viscosity data for each of the ternary mixtures are almost identical for different molar ratios and may be taken to be equal to the viscosity data of 30 mass % MDEA and 30 mass % AMP, respectively, with little error.

The model of Grunberg and Nissan[5] for the viscosity of liquid mixtures was used to correlate the viscosity data of the amine mixtures. The equation of Grunberg and Nissan has the following form:

$$\ln \eta_m/\mathrm{mPa \cdot s} = \sum x_i \ln \eta_i + \sum\sum x_i x_j G_{ij} \tag{6}$$

where η_i is the viscosity of the *i*th pure fluid and x_i is mole fraction of the *i*th component, in the mixture. Viscosities of pure liquids are shown in Table 8.

For a ternary system

$$\ln \eta_m = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_3 \ln \eta_3 + x_1x_2G_{12} + x_1x_3G_{13} + x_2x_3G_{23} \tag{7}$$

G_{ij} in eq 7 are temperature dependent and are assumed to have the form

$$G_{ij} = a + b(T/\mathrm{K}) + c(T/\mathrm{K})^2 \tag{8}$$

The parameters of eq 7 are obtained by regression analysis of the experimental data of this work and those available in the open literature and are presented in Table 7.

Table 8. Viscosity, η, for Pure MEA, DEA, MDEA, AMP, and Water from 20 °C to 80 °C

	η^a/mPa·s				
t/°C	MEA	DEA	MDEA	AMP	water
20	24.10	890.5	104.5	164.5	1.050
25	18.98	566.3	77.19	132.3	0.900
30	15.11	383.9	57.86	99.48	0.845
35	12.28	262.4	44.14	69.98	0.800
40	10.02	188.2	34.31	46.93	0.668
45	8.455	145.6	26.53	32.17	0.650
50	6.972	119.5	21.67	24.21	0.559
60	5.047	57.69	14.39	13.99	0.476
70	3.779	35.09	9.987	8.640	0.412
80	2.912	22.43	7.088	5.649	0.363

[a] The viscosity of MDEA at 20 °C has been taken from ref 11. All other pure liquid viscosities have been taken from ref 5.

The calculated viscosities from the correlation (eq 7) are in excellent agreement with the experimental data of this study, the average absolute deviation between the correlated and the experimental data for the (MDEA + MEA + H_2O), (MDEA + DEA + H_2O), (AMP + MEA + H_2O), and (AMP + DEA + H_2O) systems being about 0.9%, 2.7%, 1.0%, and 2.1%, respectively.

Conclusion

The densities of eight (MDEA + MEA + H_2O) and (MDEA + DEA + H_2O) mixtures as well as seven (AMP + MEA + H_2O) and (AMP + DEA + H_2O) mistures and the viscosities of eight (MDEA + MEA + H_2O) and (MDEA + DEA + H_2O) mixtures as well as seven (AMP + MEA + H_2O) and (AMP + DEA + H_2O) mixtures were measured and correlated over the temperature range (20 to 50) °C. The correlated densities and viscosities of the ternary mixtures are in excellent agreement with the experimental data over the temperature and relative composition ranges studied.

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