Densities of Aqueous Blended Amines

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The densities of aqueous mixtures of diethanolamine (DEA) + *N*-methyldiethanolamine (MDEA) + water, DEA + 2-amino-2-methyl-1-propanol (AMP) + water, and monoethanolamine (MEA) + 2-piperidineethanol (2-PE) + water were measured from 30 °C to 80 °C. A Redlich–Kister equation of the excess volume was applied to represent the density. Based on the available density data for five ternary systems: MEA + MDEA + H₂O, MEA + AMP + H₂O, DEA + MDEA + H₂O, DEA + AMP + H₂O, and MEA + 2-PE + H₂O, a generalized set of binary parameters were determined. The density calculations show quite satisfactory results. The overall average absolute percent deviation is about 0.04% for a total of 686 data points.

Introduction

Solutions of alkanolamines are an industrially important class of compounds used in the natural gas and synthetic ammonia industries and petroleum chemical plants for the removal of CO₂ and H₂S from gas streams. A wide variety of alkanolamines such as monoethanolamine (MEA), diethanolamine (DEA), di-2-propanolamine (DIPA), and Nmethyldiethanolamine (MDEA) have been used industrially for a number of years (Kohl and Riesenfeld, 1985). For CO₂ removal from gases by absorption in aqueous amine solutions and amine-promoted hot potassium carbonate, sterically hindered amines have been shown to have absorption capacity, absorption rate, selectivity, and degradation resistance advantages over conventional amines (Sartori and Savage, 1983; Say et al., 1984; Tontiwachwuthikul et al., 1991). 2-Amino-2-methyl-l-propanol (AMP) and 2-piperidineethanol (2-PE) are two of the sterically hindered amines.

Recently, the use of blended amines has been shown to produce absorbents with excellent absorption characteristics as well as superior stripping qualities (Chakravarty et al., 1985). The physical properties of aqueous solutions such as density are necessary for the design of acid gas treatment equipment. The density data of some aqueous solutions of blended amines have been studied in the literature: MEA/MDEA (Li and Shen, 1992; Li and Lie, 1994; Hagewiesche et al., 1995); MEA/AMP (Li and Lie, 1994); DEA/MDEA (Rinker et al., 1994; Teng et al., 1994). The density of DEA + AMP + H_2O and MEA + 2-PE + H₂O systems has not yet appeared in the literature. The objective of this work was to measure the density of ternary solutions of DEA + MDEA + H_2O , DEA + AMP+ H_2O , and MEA + 2-PE $+ H_2O$. Furthermore, a Redlich-Kister equation of the excess volume was to be applied to represent the measured density. Based on the available density data for five ternary systems, MEA + MDEA + H_2O , MEA + AMP + H_2O , DEA + MDEA + H_2O , DEA + $AMP + H_2O$, and $MEA + 2-PE + H_2O$, a generalized set of parameters will be determined.

Experimental Section

Alkanolamine aqueous solutions were prepared from distilled water. The distilled water was degassed by boiling. All alkanolamines are Riedel-de Haân reagent grade with the following purities: MEA, 99%; DEA, 99%;

Table 1.	Comparison of tl	e Densities o	of Aqueous DEA
Solutions	Measured in Th	is Study with	Literature
Values		v	

		$ ho/g\cdot cm^{-3}$					
	20 mass %	DEA	30 mass % DEA				
t/°C	Rinker et al. (1994)	this study	Rinker et al. (1994)	this study			
40	1.0142	1.0149	1.0272	1.0264			
60	1.0052	1.0051	1.0170	1.0172			
80	0.9931	0.9931	1.0037	1.0046			
		0.03 ^a		0.06 ^a			

^a AAD %.

MDEA, 98.5%; AMP, 98%; 2-PE, 98%. The densities of solutions were measured by using a 25-mL Gay-Lussac pycnometer. The measurements were performed in a constant-temperature water bath, in which the temperature could be held constant to ± 0.05 °C. The experimental errors were estimated to be equal to $\pm 0.05\%$ on the basis of comparisons with literature data.

Results and Discussion

For the density measurement, we have obtained data for the systems for which density data had been reported in the literature. Densities of 20 mass % DEA and 30 mass % DEA aqueous solutions have been reported by Rinker et al. (1994). The results of density measurements for temperatures ranging from 40 °C to 60 °C are presented in Table 1. The average absolute percentage deviations of the density measurements are 0.03% and 0.06% for 20 mass % DEA and 30 mass % DEA aqueous solutions, respectively. The density data obtained in this study are in good agreement with the data of Rinker et al. (1994).

The densities of three blended amine aqueous solutions, DEA + MDEA + H₂O, DEA + AMP + H₂O, and MEA + 2-PE + H₂O, for temperatures ranging from 30 °C to 80 °C were measured and are presented in Tables 2–4. The concentrations were selected as a total of 20 and 30 mass % alkanolamine aqueous solutions. The densities of 10 mass % MEA + 10 mass % MDEA and 10 mass % MEA + 10 mass % AMP aqueous solutions are also reported in Tables 2 and 3.

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 (Prausnitz et al., 1986) has the expression

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	Table 2.	Densities	(g·cm ⁻³) of DEA	+	MDEA	+	H_2	0
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	mass % DEA/mass % MDEA									MEA/MDEA
t∕°C	30/0	24/6	18/12	12/18	6/24	20/0	15/5	10/10	5/15	10/10
30	1.0306	1.0282	1.0265	1.0254	1.0240	1.0186	1.0172	1.0159	1.0143	1.0082
40	1.0264	1.0242	1.0222	1.0206	1.0188	1.0149	1.0132	1.0117	1.0101	1.0034
50	1.0225	1.0200	1.0175	1.0153	1.0132	1.0093	1.0087	1.0069	1.0055	0.9990
60	1.0172	1.0143	1.0120	1.0095	1.0071	1.0051	1.0031	1.0015	0.9997	0.9935
70	1.0112	1.0082	1.0060	1.0036	1.0012	0.9991	0.9975	0.9955	0.9935	0.9875
80	1.0046	1.0014	0.9991	0.9970	0.9948	0.9931	0.9910	0.9890	0.9872	0.9808

Table 3. Densities (g·cm⁻³) of DEA + AMP + H₂O

mass % DEA/mass % AMP								
24/6	18/12	12/18	6/24	15/5	10/10	5/15	10/10	
1.0227	1.0157	1.0086	1.0007	1.0120	1.0066	1.0003	0.9984	
1.0192	1.0113	1.0045	0.9960	1.0085	1.0021	0.9956	0.9952	
1.0138	1.0057	0.9982	0.9898	1.0032	0.9972	0.9904	0.9900	
1.0081	0.9996	0.9919	0.9825	0.9981	0.9914	0.9849	0.9844	
1.0024	0.9941	0.9853	0.9762	0.9925	0.9855	0.9787	0.9780	
0.9946	0.9867	0.9783	0.9692	0.9860	0.9794	0.9725	0.9712	
	24/6 1.0227 1.0192 1.0138 1.0081 1.0024 0.9946	24/6 18/12 1.0227 1.0157 1.0192 1.0113 1.0138 1.0057 1.0081 0.9996 1.0024 0.9941 0.9946 0.9867	mass % E 24/6 18/12 12/18 1.0227 1.0157 1.0086 1.0192 1.0113 1.0045 1.0138 1.0057 0.9982 1.0081 0.9996 0.919 1.0024 0.9941 0.9853 0.9946 0.9867 0.9783	mass % DEA/mas 24/6 18/12 12/18 6/24 1.0227 1.0157 1.0086 1.0007 1.0192 1.0113 1.0045 0.9960 1.0138 1.0057 0.9982 0.9898 1.0081 0.9996 0.9919 0.9825 1.0024 0.9941 0.9853 0.9762 0.9946 0.9867 0.9783 0.9692	mass % DEA/mass % AM 24/6 18/12 12/18 6/24 15/5 1.0227 1.0157 1.0086 1.0007 1.0120 1.0192 1.0113 1.0045 0.9960 1.0085 1.0138 1.0057 0.9982 0.9898 1.0032 1.0081 0.9996 0.9919 0.9825 0.9981 1.0024 0.9941 0.9853 0.9762 0.9925 0.9946 0.9867 0.9783 0.9692 0.9860	mass % DEA/mass % AMP 24/6 18/12 12/18 6/24 15/5 10/10 1.0227 1.0157 1.0086 1.0007 1.0120 1.0066 1.0192 1.0113 1.0045 0.9960 1.0085 1.0021 1.0138 1.0057 0.9982 0.9898 1.0032 0.9972 1.0081 0.9996 0.9919 0.9825 0.9981 0.9914 1.0024 0.9941 0.9853 0.9762 0.9925 0.9855 0.9946 0.9867 0.9783 0.9692 0.9860 0.9794	mass % DEA/mass % AMP 24/6 18/12 12/18 6/24 15/5 10/10 5/15 1.0227 1.0157 1.0086 1.0007 1.0120 1.0066 1.0003 1.0192 1.0113 1.0045 0.9960 1.0085 1.0021 0.9956 1.0138 1.0057 0.9982 0.9898 1.0032 0.9972 0.904 1.0081 0.9996 0.9919 0.9825 0.9981 0.9914 0.9849 1.0024 0.9941 0.9853 0.9762 0.9925 0.9855 0.9787 0.9946 0.9867 0.9783 0.9692 0.9860 0.9794 0.9725	

Table 4. Densities (g·cm⁻³) of MEA + 2-PE + H_2O

		mass % MEA/mass % 2-PE							
t∕°C	24/6	18/12	12/18	6/24	15/5	10/10	5/15		
30	1.0088	1.0087	1.0076	1.0065	1.0036	1.0037	1.0033		
40	1.0036	1.0026	1.0017	1.0001	0.9990	0.9992	0.9983		
50	0.9980	0.9969	0.9952	0.9937	0.9943	0.9936	0.9927		
60	0.9923	0.9906	0.9888	0.9873	0.9887	0.9879	0.9867		
70	0.9855	0.9839	0.9822	0.9807	0.9829	0.9820	0.9801		
80	0.9794	0.9769	0.9746	0.9739	0.9767	0.9752	0.9732		

$$V_{12}^{\rm E}/{\rm cm}^3 \cdot {\rm mol}^{-1} = x_1 x_2 \sum_{i=0}^n A_i (x_1 - x_2)^i$$
 (1)

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

$$A_i = a + b(T/K) + c(T/K)^2$$
 (2)

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

$$V^{\rm E} = V^{\rm E}_{12} + V^{\rm E}_{13} + V^{\rm E}_{23} \tag{3}$$

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

$$V^{\rm E} = V_{\rm m} - \sum x_i V_i^{\rm o} \tag{4}$$

where $V_{\rm m}$ is the molar volume of the liquid mixture and V_i° is the molar volume of pure fluids at the temperature of the system. The molar volumes of the liqud mixtures are calculated by

$$V_{\rm m} = \frac{\sum x_i M_i}{\rho_{\rm m}} \tag{5}$$

where M_i is the molar mass of pure component *i* and ρ_m is the measured liquid density.

In eq 1, A_i is the pair-interaction parameter; a generalized set of parameters can be, in principle, developed on the basis of the available density data in the literature. In this study we intend to develop a generalized set of parameters for the density calculation of blended amine aqueous solutions. The ternary systems selected for density calculation are MEA + MDEA + H₂O, MEA + AMP + H₂O, DEA + MDEA + H₂O, DEA + AMP + H₂O, and MEA + 2-PE + H₂O. The density data utilized in this study are from the following systems: pure fluids, water and alkanol-

Table 5. Density of Six Pure Fluids Used in This Study

		no. of		
system	t/°C	data points	ref ^a	AAD $\%^b$
H ₂ O	22.89-80.04	5	(1)	0.025
	30 - 200	18	(2)	0.028
	15 - 60	10	(3)	0.035
	22.89 - 80.04	5	(5)	0.060
MEA	20.3 - 87.5	5	(7)	0.039
	30-80	8	(6)	0.028
	21.25 - 158.15	8	(5)	0.048
DEA	19.7 - 88.2	5	(7)	0.027
	24.05 - 159.75	8	(5)	0.036
MDEA	15 - 35	5	(3)	0.092
	19.7 - 88.2	5	(7)	0.037
	30-80	8	(6)	0.039
	23.15 - 197.75	8	(5)	0.055
AMP	31.9 - 90.7	6	(4)	0.035
	30-80	6	(9)	0.031
2-PE	30.6 - 84.2	5	(8)	0.012
overall		115		0.038

 a (1) Kell, 1975. (2) Perry and Chilton, 1984. (3) Al-Ghawas et al., 1989. (4) Xu et al., 1991. (5) Diguillo et al., 1992. (6) Li and Shen, 1992. (7) Wang et al., 1992. (8) Xu et al., 1992. (9) Li and Lie, 1994. b Calculated from eq 6.

 Table 6. Parameters of the Density Equation for Pure Fluids^a

pure fluid	a_1	a_2	a_3
H ₂ O	0.863 559	$1.21494 imes 10^{-3}$	$-2.57080 imes 10^{-6}$
MEA DEA	1.190 93 1 207 15	-4.29990×10^{-4} -1.51200 $\times 10^{-4}$	-5.66040×10^{-7} -7.66530 $\times 10^{-7}$
MDEA	1.228 64	-5.44540×10^{-4}	-3.35930×10^{-7}
AMP	1.156 32	-6.76170×10^{-4} -1.7650 × 10 ⁻⁴	-2.67580×10^{-7} -8.0842 × 10^{-7}
~-1 Ľ	1.120 30	1.7050×10	0.3042 × 10

^a Parameters of eq 6.

amines; binary systems, single-amine aqueous solutions; ternary mixtures, ternary aqueous solutions of blended amines.

The density data of the pure fluids applied are presented in Table 5. The temperature covers a wide range from 15 °C up to 200 °C. The densities of pure fluids are required to calculate the excess volume, as in eq 4, which is assumed to be the following expression:

$$a/g \cdot cm^{-3} = a_1 + a_2(T/K) + a_3(T/K)^2$$
 (6)

where a_i are parameters. The determined parameters are presented in Table 6. The average absolute percentage deviation (AAD%) is about 0.038% for 115 data points. The AAD% of density calculations for different investigators are also presented in Table 5. The results of density calculations of pure fluids are also shown in Figure 1. The agreement of density measurements among different investigators is quite good. Among six pure fluids, DEA has the highest density value and AMP has the lowest value of density. The densities of H₂O and MEA cross each other near 70 °C. At 50 °C, the densities of the pure fluids have the order DEA > MDEA > MEA > H₂O > 2-PE > AMP.

The density data of five single-amine aqueous solutions, MEA + H_2O , DEA + H_2O , MDEA + H_2O , AMP + H_2O ,



Figure 1. Density of the pure fluids: (points) experimental data; (solid lines) values calculated using eq 6.

Table 7.	Density	of Binary	Alkanolamine Aqueous	
Solution	5	-		

		conc,	no. of		AAD
system	t∕°C	mass %	data points	ref^a	$\%^b$
$\overline{MEA + H_2O}$	30-80	30	8	(4)	0.040
	30-80	20	6	(6)	0.073
$DEA + H_2O$	20-100	10	5	(5)	0.029
	20-100	20	5	(5)	0.086
	20-100	30	5	(5)	0.070
	30-80	20	6	(8)	0.033
	30-80	30	6	(8)	0.050
$MDEA + H_2O$	15 - 60	10	10	(1)	0.070
	15 - 60	20	8	(1)	0.073
	15 - 60	30	8	(1)	0.064
	15 - 50	40	7	(1)	0.058
	15 - 60	50	10	(1)	0.064
	30-80	30	8	(4)	0.013
	30-80	20	6	(6)	0.015
	60-100	10	3	(5)	0.050
	60-100	20	3	(5)	0.094
	60-100	30	3	(5)	0.099
	60-100	40	3	(5)	0.023
	60-100	50	3	(5)	0.104
	29.85 - 49.85	30	3	(7)	0.068
	29.85 - 49.85	40	3	(7)	0.087
$AMP + H_2O$	20.3 - 88.7	9.05	7	(2)	0.116
	20.0 - 88.7	27.33	7	(2)	0.051
	20.3 - 88.7	50.94	7	(2)	0.097
	20.0 - 90.7	73.62	7	(2)	0.042
	20.0 - 90.7	88.32	7	(2)	0.035
	30-80	20	6	(6)	0.048
	30-80	30	6	(6)	0.084
$2 - PE + H_2O$	25 - 83.5	10	5	(3)	0.063
	25 - 83.5	30	5	(3)	0.168
	25 - 83.5	45	5	(3)	0.018
	25 - 83.5	60	5	(3)	0.044
	25 - 83.5	75	5	(3)	0.022
overall			259		0.053

 a (1) Al-Ghawas et al., 1989. (2) Xu et al., 1991. (3) Xu et al., 1992. (4) Li and Shen, 1992. (5) Rinker et al., 1994. (6) Li and Lie, 1994. (7) Hagewiesche et al., 1995. (8) This study. b Calculated from eq 1.

and 2-PE + H₂O, are listed in Table 7. The temperature ranges from 20 °C to 100 °C, and the concentration ranges from 9 to 88 mass % amine. The binary parameters, A_i in eq 1, for five pairs, MEA-H₂O, DEA-H₂O, MDEA-H₂O, AMP-H₂O, and 2-PE-H₂O, are determined from the density data of the single-amine aqueous solutions, and the

Table 8. Density of Blended Amine Aqueous Solutions

			c		
		conc,	no. of		
system	t/°C	mass 2%	nointe	rof ^a	
system	u c	1110352 70	points	Ter	70
MEA + MDEA +	30-80	6 + 24	8	(1)	0.012
H_2O	30-80	12 + 18	8	(1)	0.034
	30-80	18 + 12	8	(1)	0.023
	30-80	24 + 6	8	(1)	0.011
	30-80	5 + 15 15 + 5	6	(2) (2)	0.067
	30-80	13 ± 3 10 ± 10	6	(2) (6)	0.049
	29 85-49 85	10 + 10 15 ± 285	0 3	(0) (5)	0.000
	29 85-49 85	3.0 ± 27.0	3	(5)	0.040
	29 85-49 85	45 + 255	3	(5)	0.000
	29 85-49 85	2.0 + 38.0	3	(5)	0.015
	29.85 - 49.85	4.0 ± 36.0	3	(5)	0.004
	29.85 - 49.85	6.0 ± 34.0	3	(5)	0.057
MEA + AMP +	30-80	6 + 24	6	(2)	0.011
H ₂ O	30-80	12 + 18	6	(2)	0.029
2	30-80	18 + 12	6	(2)	0.034
	30-80	24 + 6	6	(2)	0.013
	30-80	5 + 15	6	(2)	0.088
	30-80	10 + 10	6	(6)	0.055
	30-80	15 + 5	6	(2)	0.083
DEA + MDEA +	25-80	1.94 + 30.24	7	(4)	0.011
H_2O	25-80	1.13 ± 29.68	7	(4)	0.015
	25-80	2.48 ± 27.76	7	(4)	0.016
	25 - 80	3.47 ± 26.71	7	(4)	0.014
	25 - 80	4.48 + 25.50	7	(4)	0.010
	25 - 80	5.64 + 24.44	7	(4)	0.013
	25 - 80	7.64 + 22.86	7	(4)	0.008
	25 - 80	1.04 + 24.00	7	(4)	0.029
	20-100	2.11 + 47.89	5	(3)	0.047
	20-100	9.04 + 40.96	5	(3)	0.025
	20-100	15.31 + 34.69	5	(3)	0.020
	20-100	18.52+31.48	5	(3)	0.030
	30-80	5 + 15	6	(6)	0.031
	30-80	10 + 10	6	(6)	0.022
	30-80	15 + 5	6	(6) (6)	0.022
	30-80 20-80	0 ± 24 12 \pm 19	6	(0) (6)	0.020
	30-80 20-80	12 ± 10 19 \pm 19	6	(0) (6)	0.000
	30-80	10 + 12 24 ± 6	6	(0) (6)	0.030
$\Delta E \Lambda \perp \Lambda M D \perp$	30-80	24 ± 0 5 ± 15	6	(0) (6)	0.040
	30-80	10 ± 10	6	(0)	0.038
1120	30-80	10 + 10 15 + 5	6	(0)	0.013
	30-80	6 + 24	6	(6)	0.020
	30-80	12 + 18	6	(6)	0.001
	30-80	12 + 10 18 + 12	6	(6)	0.024
	30-80	24 + 6	6	(6)	0.042
MEA + 2-PE +	30-80	5 + 15	6	(6)	0.100
H ₂ O	30-80	10 + 10	6	(6)	0.025
	30-80	15 + 5	6	(6)	0.052
	30-80	6 + 24	6	(6)	0.026
	30-80	12 + 18	6	(6)	0.054
	00 00				
	30-80	18 + 12	6	(6)	0.014
	30-80 30-80 30-80	$\begin{array}{c} 18 + 12 \\ 24 + 6 \end{array}$	6 6	(6) (6)	0.014 0.019

 a (1) Li and Shen, 1992. (2) Li and Lie, 1994. (3) Rinker et al., 1994. (4) Teng et al., 1994. (5) Hagewiesche et al., 1995. (6) This study. b Calculated from eq 3.

values are listed in Table 9. The AAD % for the density calculation for each investigator are also presented in Table 7. For a total of 253 data points, the overall AAD % for the density calculations of binary systems is about 0.053%. Except for the systems of 50 mass % MDEA + H₂O (AAD % = 0.104%), 9.05 mass % AMP + H₂O (AAD % = 0.116%), and 30 mass % 2-PE + H₂O (AAD % = 0.168%), all the density calculations are below 0.1% which is a quite satisfactory result. From the results of the density calculations in Table 7, it has been shown that the density of single-amine aqueous solutions can be well-represented by the Redlich–Kister equation of the excess volume, i.e., eq 1. The results of density calculations for DEA + H₂O, MDEA + H₂O, and AMP + H₂O are also shown graphically

$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Table	9. Bina	ry Para	meters of the R	edlich–Kister Equat	tion of the H	Excess Vol	ume	
$\begin{array}{c c c c c c c c c c c c c c c c c c c $						bina	ry pairs		
$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	para	am	_	$\mathrm{MEA} + \mathrm{H_2O}$	$DEA + H_2O$	MDE	$A + H_2O$	$AMP + H_2O$	$2-PE + H_2O$
$ \begin{array}{c} b \\ c \\ c \\ c \\ -1.10780 \times 10^{-4} \\ c \\ -1.10780 \times 10^{-6} \\ b \\ 1.10780 \times 10^{-6} \\ 0.10780 \times 10^{-6} \\ 0.10780 \times 10^{-6} \\ -2.06623 \times 10 \\ -2.06623 \times 10 \\ -2.06623 \times 10 \\ -2.06623 \times 10^{-7} \\ -1.08578 \times 10^{-6} \\ -1.08578 \times 10^{-6} \\ -1.1325 \times 10^{-2} \\ -3.11374 \times 10^{-2} \\ -3.11374 \times 10^{-2} \\ -1.1325 \times 10^{-2} \\ -3.11374 \times 10^{-2} \\ -3.11374 \times 10^{-2} \\ -1.1325 \times 10^{-2} \\ -3.6877 \times 10^{-1} \\ -1.06202 \times 10 \\ -1.0600 \times 10 \\ -1.06202 \times 10 \\ -1.0600 \times 10 \\ -1.06202 \times 10 \\ -1.0600 \times 10 \\ -1.060$	A_0) (а -	$-5.92024 imes 10^{-2}$	-3.31562×10	-2.887	'74 × 10	-6.51042	-6.04991
$\begin{array}{cccccccccccccccccccccccccccccccccccc$			b -	$-1.77290 imes 10^{-4}$	$8.11654 imes 10^{-2}$	6.958	10×10^{-2}	$5.02584 imes10^-$	5.69162×10^{-3}
$\begin{array}{c c c c c c c c c c c c c c c c c c c $			с -	$-1.10780 imes 10^{-6}$	$6.15156 imes 10^{-6}$	-5.030	40×10^{-7}	$1.08578 imes10^-$	$-2.0967 imes 10^{-6}$
$\frac{b}{1.0385 \times 10^{-3}} = 9.75694 \times 10^{-2} = 6.36707 \times 10^{-2} -1.1325 \times 10^{-2} -3.11374 \times 10^{-2} -3.11374 \times 10^{-2} -3.11374 \times 10^{-2} -3.11374 \times 10^{-2} -1.1325 \times 10^{-2} -3.11374 \times 10^{-2} -3.111111111111100 \times 10^{-2} -3.111111100 \times 10^{-2} -3.11111100 \times 10^{-2} -3.1111100 \times 10^{-2} -3.1111100 \times 10^{-2} -3.11111100 \times 10^{-2} -3.11111100 \times 10^{-2} -3.1111100 \times 10^{-2} -3.1111100 \times 10^{-2} -3.1111100 \times 10^{-2} -3.1111100 \times 10^{-2} -3.111100 \times 10$	A_1	L •	a	2.17490	-3.52516×10	-2.066	23×10	5.55560	1.2181×10
$\begin{array}{c c c c c c c c c c c c c c c c c c c $			b	1.10385×10^{-5}	9.75694×10^{-2}	6.367	07×10^{-2}	-1.1325×10^{-2}	-3.11374×10^{-2}
$\begin{array}{c c c c c c c c c c c c c c c c c c c $						binar	y pairs		
$A_{0} = a - 2.42766 \times 10 5.53222 - 1.24706 \times 10 1 1.75649 \times 10^{3} - 1.0461 \times 10^{3} \\ b 1.89797 \times 10^{-1} 1.62914 \times 10^{-1} 1.00561 \times 10^{-1} - 1.6202 \times 10 \\ c - 2.8825 \times 10^{-4} - 6.4338 \times 10^{-4} - 1.62790 \times 10^{-4} 1.59224 \times 10^{-2} - 8.40241 \times 10^{-3} \\ b - 1.05682 - 9.86571 \times 10^{-1} 9.6347 \times 10^{-2} - 1.28358 - 6.67145 \\ c - 4.28233 \times 10^{-3} - 2.39399 \times 10^{-3} 1.02886 \times 10^{-5} - 4.86136 \times 10^{-3} 8.06681 \times 10^{-3} \\ b - 1.49472 \times 10 - 2.70341 \times 10 - 7.53195 \times 10^{-3} - 6.44203 \times 10 - 1.1186 \times 10^{2} \\ c - 1.52253 \times 10^{-2} - 5.68765 \times 10^{-2} - 3.651 \times 10^{-3} - 6.4203 \times 10 - 1.1186 \times 10^{2} \\ c - 1.52253 \times 10^{-2} - 5.68765 \times 10^{-2} - 3.651 \times 10^{-3} - 6.4203 \times 10 - 1.1186 \times 10^{2} \\ 1.02 - 0.99 - 0.$			N	IEA + MDEA	MEA + AMP	DEA +	- MDEA	DEA + AMP	MEA + 2-PE
$ \begin{array}{c} b & 1.89797 \times 10^{-1} & 1.62914 \times 10^{-1} & 1.00561 \times 10^{-1} & -1.06202 \times 10 & 6.16956 \\ c & -2.8825 \times 10^{-4} & -6.438 \times 10^{-4} & -1.62790 \times 10^{-4} & 1.59224 \times 10^{-2} & -8.40241 \times 10^{-3} \\ b & -1.05682 & 9.86571 \times 10^{-1} & 9.6347 \times 10^{-2} & -1.28358 & -6.67145 \\ c & 4.28233 \times 10^{-3} & -2.39399 \times 10^{-3} & 1.02886 \times 10^{-5} & 4.86136 \times 10^{-3} & 8.06681 \times 10^{-3} \\ A_2 & a & 0 & 0 \\ b & -1.49472 \times 10 & -2.70341 \times 10 & -7.53195 \times 10^{-3} & -6.44203 \times 10 & 1.11186 \times 10^2 \\ c & 1.52253 \times 10^{-2} & 5.68765 \times 10^{-2} & -3.651 \times 10^{-3} & 2.35996 \times 10^{-1} & 2.93065 \times 10^{-1} \\ \end{array} $	A_0) a	-2	2.42756 × 10	5.53222	-1.2470	06 × 10	1.75649×10^3	-1.0461×10^3
$\begin{array}{cccccccccccccccccccccccccccccccccccc$		b	1	1.89797×10^{-1}	1.62914×10^{-1}	1.0056	51×10^{-1}	-1.06202×10	6.16956
$A_{1} = a = 0 = 0 = 0 = 0 = 0 = 0 = 0 = 0 = 0$		С	-2	2.8825×10^{-4}	$-6.4438 imes 10^{-4}$	-1.6279	$90 imes 10^{-4}$	1.59224×10^{-1}	-8.40241×10^{-3}
$\begin{array}{cccccccccccccccccccccccccccccccccccc$	A_1	ı a	()	0	0	7 40-2	0	0
$A_{2} = \begin{pmatrix} c & 4.282.33 \times 10^{-3} & -2.33399 \times 10^{-3} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0$		b	- 1	1.05682	9.86571×10^{-1}	9.634	1×10^{-2}	-1.28358	
$\begin{array}{c} A_{2} \\ A_{2} \\ b \\ c \\ 1.52253 \times 10^{-2} \\ 1.52253 \times 10^{-2} \\ 0.58765 \times 10^{-3} \\ 0.58765 \times 10^{-3} \\ 0.551 \times 10^{-3} \\ 0.5596 \times 10^{-1} \\ 0.5596 \times 10^{-1} \\ 0.5596 \times 10^{-1} \\ 0.567 \\ 0.996 $	4	С	4	$\frac{1.28233 \times 10^{-3}}{2}$	-2.39399×10^{-3}	1.0288	36×10^{-5}	4.80130×10^{-1}	³ 8.06681 × 10 ⁻³
$ \begin{array}{cccccccccccccccccccccccccccccccccccc$	A_2	2 a b	(J 1 40479 v 10	U 9 70241 v 10	U 7 5 9 1 (05 v 10−3	U 6 44902 \vee 10	$0 \\ 1 11196 > 10^{2}$
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Figure 2. Density of diethanolamine aqueous solutions: (points) experimental data; (solid lines) values calculated using eq 1.

t/°C

in Figures 2–4. As shown in Figure 2, the density calculations for 10 to 30 mass % DEA aqueous solutions are quite satisfactory. For the MDEA + H_2O system, except for the data of 50 mass % MDEA (Rinker et al., 1994), all the density calculations are quite satisfactory. At the same temperature, the density of MDEA + H_2O increases with increases in the mass % of MDEA. The density data of AMP + H_2O cover a wide concentration range, i.e., from 9.05 to 88.32 mass % AMP. The density calculations for the AMP + H_2O are quite good, as shown in Figure 4. The density of AMP + H_2O decreases as the AMP mass % increases.

The density data of the ternary aqueous solutions of blended amines considered in this study are for MEA + MDEA + H₂O, MEA + AMP + H₂O, DEA + MDEA + H₂O, DEA + AMP + H₂O, and MEA + 2-PE + H₂O. The pair-interaction parameters for MEA-MDEA, MEA-AMP, DEA-MDEA, DEA-AMP, and MEA-2-PE are determined from these ternary systems and are presented in Table 9. In Table 9, the AAD % of density calculations for each ternary system are also presented. Except for the system 5 mass % MEA + 15 mass % 2-PE + H₂O (AAD % = 0.1%), the density calculations for all the ternary systems are less than 0.1%. The overall AAD % is about 0.031% for a total of 312 data points. In Figure 5, the results of density

Figure 3. Density of *N*-methyldiethanolamine aqueous solutions for concentrations up to 50 mass % MDEA: (points) experimental data; (solid lines) values calculated using eq 1.

t/°C



Figure 4. Density of 2-amino-2-methyl-1-propanol aqueous solutions for various concentrations: (points) experimental data; (solid lines) values calculated using eq 1.

calculations for MEA + MDEA + H_2O is shown for six concentrations (a total of 30 mass %) obtained from two data sources (Li and Shen, 1992; Hagewiesche et al., 1995).



Figure 5. Density of monoethanolamine + *N*-methyldiethanolamine aqueous solutions for seven concentrations: (points) experimental data; (solid lines) values calculated using eq 3.



Figure 6. Density of diethanolamine + *N*-methyldiethanolamine aqueous solutions for eight concentrations: (points) experimental data; (solid lines) calculated values using eq 3.

As shown in Figure 5, the density calculations using the Redlich-Kister equation for the ternary system MEA + $MDEA + H_2O$ are quite satisfactory. The results of density calculations for six DEA + MDEA + H₂O systems are shown in Figure 6. The DEA + MDEA + H₂O systems cover a wide concentration range, the total amine concentration varies from 30 to 50 mass %. As shown in Figure 6, the density calculations are quite satisfactory. Figure 7 shows the density calculations for $DEA + AMP + H_2O$ for temperature 25 to 80 °C. The AAD % for the density calculations for DEA + AMP + H₂O is 0.03% for 42 data points. From the results presented in Table 8, it can be said that the density of ternary aqueous solutions of blended amine tested in this stydy can be well-correlated by the Redlich-Kister equation. A summary of the results of density calculations for pure fluids, single-amine aqueous solutions, and ternary aqueous solutions of blended amines are presented in Table 10. The overall AAD % for 686 data



Figure 7. Density of diethanolamine + 2-amino-2-methyl-1-propanol aqueous solutions for various concentrations: (points) experimental data; (solid lines) calculated values using eq 3.

Table 10.Summary of the Results of DensityCalculations for Aqueous Alkanolamine Solutions Usingthe Redlich-Kister Equation

system	no. of data points	AAD %
H ₂ O	38	0.034
MEA	21	0.038
DEA	13	0.033
MDEA	26	0.054
AMP	12	0.033
2-PE	5	0.012
$MEA + H_2O$	14	0.054
$DEA + H_2O$	27	0.042
$MDEA + H_2O$	78	0.059
$AMP + H_2O$	47	0.068
$2 - PE + H_2O$	25	0.063
$MEA + MDEA + H_2O$	68	0.037
$MEA + AMP + H_2O$	42	0.045
$DEA + MDEA + H_2O$	118	0.021
$DEA + AMP + H_2O$	42	0.030
$MEA + 2-PE + H_2O$	42	0.041
overall	686	0.041

points is 0.041%, which is satisfactory for the design calculations.

Conclusion

The densities of aqueous mixtures of diethanolamine + N-methyldiethanolamine + water, DEA + 2-amino-2methyl-1-propanol + water, and monoethanolamine + 2-piperidineethanol + water were measured from 30 °C to 80 °C. A Redlich-Kister equation of the excess volume was applied to represent the available density data of five aqueous solutions of blended alkanolamines, MEA + $MDEA + H_2O$, $MEA + AMP + H_2O$, $MEA + 2-PE + H_2O$, $DEA + MDEA + H_2O$, and $DEA + AMP + H_2O$. Based on the available density data in the literature, a generalized set of parameters in the Redlich-Kister density equation was determined. The overall AAD % for the density calculations is 0.041% for 686 density data points including pure fluids, single-amine aqueous solutions, and ternary aqueous solutions of blended amines. The results are satisfactory. The present correlation can be used for density calculations of blended amine aqueous solutions for industrial design calculations.

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