Liquid Density of Aqueous Blended Alkanolamines and *N*-Methylpyrrolidone as a Function of Concentration and Temperature

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Experimental values of the liquid density have been determined for ternary systems formed by aqueous mixtures of diethanolamine (DEA) + methyldiethanolamine (MDEA), DEA + 2-amino-2-methy-l-propanol (AMP), DEA + 2-piperidineethanol (2PE), DEA + *N*-methylpyrrolidone (NMP), MDEA + AMP, MDEA + 2PE, MDEA + NMP, and NMP + AMP. The data were obtained at (313.15, 323.15, and 333.15) K, the total concentration of the blended solvents studied was in the range 30–60 mass %, and for each concentration at least seven points were obtained with different proportions of each solvent. The estimated accuracy of the data is ±0.0004 g·cm⁻³. From the density data, molar excess volumes were derived, which were correlated using the Redlich–Kister equation; the final expression includes the functionality of both concentration and temperature.

Introduction

Because of the extensive use of both aqueous solutions of two blended alkanolamines and physical solvents blended with a chemical solvent[1] in gas sweetening applications, a complete knowledge of several thermophysical properties of such solvents is of considerable interest.

In this work density data in the range 313.15–333.15 K are reported for eight ternary systems which are formed by aqueous solutions of two blended polar solvents; the latter solvent mixtures are based on secondary diethanolamine (DEA), tertiary *N*-methyldiethanolamine (MDEA), primary and hindered 2-amino-2-methyl-1-propanol (AMP), and cyclic 2-piperidineethanol (2PE) and on the physical solvent *N*-methylpyrrolidone (NMP). The ternary systems and their concentrations studied are DEA + MDEA + H_2O at 30, 40, and 50 mass %; DEA + AMP + H_2O at 30, 40, and 50 mass % (additionally this system was also studied at 95 mass % at 333.15 K); DEA + 2PE + H_2O at 30, 40, and 50 mass % at 313.15 K; DEA + NMP + H_2O at 30, 40, and 50 mass % at 313.15 and 323.15 K (whereas at 333.15 K it was studied at 40, 50, and 60 mass %); MDEA + AMP + H_2O at 30, 40, and 50 mass %; MDEA + 2PE + H_2O and MDEA + NMP + H_2O at 30, 40, and 50 mass % at 313.15 K (whereas at 323.15 and 333.15 K both systems were studied at 40, 50, and 60 mass %); and NMP + AMP + H_2O at 30, 40, and 50 mass % at 313.15 and 323.15 K; at 333.15 K the concentrations studied were 40, 50, and 60 mass %. For a given total concentration of blended solvents at least seven experimental points were obtained varying the ratio of the polar solvents for each of the eight ternary systems considered. From the experimental density data, molar excess volumes were derived and correlated with the well-known Redlich–Kister equation, which includes adjustable parameters with concentration and temperature functionality.

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Table 1. Review of the Literature that Reports Density Data for Some of the Systems Studied in This Work

system	*T*/K	no. of data points	total mass %
DEA	292.85–361.35[8]	5	
	297.2–432.9[9]	8	
	298.15–333.15[4]	8	
	298.15–353.15[18]	7	
MDEA	288.15–333.15[10]	10	
	292.85–361.35[8]	5	
	303.15–353.15[11]	8	
	296.3–470.9[9]	8	
	298.15–353.15[5]	7	
	298.15–343.15[12]	5	
NMP	283.15–333.15[4]	11	
2PE	303.75–357.35[13]	5	
AMP	305.05–363.85[14]	6	
	303.15–353.15[15]	6	
DEA + H_2O	293.15–373.15[16]	15	10–30
	303.15–353.15[6]	12	20–30
	298.15–353.15[18]	105	5–99
MDEA + H_2O	288.15–333.15[10]	43	10–50
	303.15–353.15[11]	8	30
	303.15–353.15[15]	6	20
	333.15–373.15[16]	15	10–50
	298.15–353.15[5]	112	5–99
2PE + H_2O	298.15–356.65[13]	25	10–75
AMP + H_2O	293.15–363.85[14]	35	9–88
	303.15–353.15[15]	12	20–30
DEA + MDEA + H_2O	298.15–353.15[17]	56	25–32
	293.15–373.15[16]	20	50
	303.15–353.15[6]	42	20–30
DEA + AMP + H_2O	303.15–353.15[6]	42	20–30

Density data were also determined for the pure components DEA, MDEA, AMP, 2PE, and NMP, at the three temperatures considered and also for the binary systems DEA + H_2O, MDEA + H_2O, AMP + H_2O, 2PE + H_2O, and NMP + H_2O, at the temperatures and total concentrations mentioned above.

A review of the open literature that reports density data for DEA, MDEA, AMP, 2PE, NMP, and aqueous solutions of one and two blended alkanolamines is given in Table 1.

Experimental Section

The sample of DEA was obtained from Merck with a reported purity of 98 mol %. The MDEA, AMP, and 2PE were obtained from Aldrich with a purity of 99.0, 95.0, and 90%, respectively. The NMP was from Fluka with a purity > 99.0%. Each of the solvents was further purified in a stream of dried nitrogen under a controlled reduced pressure in an all-glass still with a 1 m packed column, and then stored over molecular sieves in order to eliminate any traces of water during handling of the samples. All the purified samples were analyzed by gas−liquid chromatography, which showed no impurities using a lower limit of detection of 0.05%, except for 2PE, whose purity was established as 96.0%.

Densities were measured with an Anton Paar DMA 45 density meter, which was calibrated at each temperature with distilled water and ethylene glycol, making use of accurate values of density reported in the literature.[2,3] The temperature was controlled within ±0.002 K as measured with a Systemteknik platinum resistance thermometer. A Sartorius 2006 MP analytical balance with a precision of ±0.0001 g was used for mass measurements of the aqueous solutions. A complete description of the experimental methodology has been described in previous work from our laboratory.[4]

Results and Discussion

We determined the precision of our results through the repetition of experimental measurements during independent runs of freshly prepared samples for both pure components and several mixtures of known concentration, at two different temperatures within the range studied. The results indicated that the precision of our density measurements is ±0.0002 g·cm^{-3}. To establish the accuracy of our measurements, we carried out a comparison with literature data for pure DEA and MDEA, since there are more data reported for these alkanolamines than for any of the other solvents considered in this work, in the temperature range studied. The mean absolute deviation between our values and those reported in the literature for DEA,[4,9,10] at the three temperatures considered, is 0.000 54 g.cm^{-3}, whereas for MDEA[4,7,8,10−12] it is 0.000 56 g·cm^{-3}. Table 2 shows the results of our measurements and literature data not only for DEA and MDEA but also for the other three pure solvents studied in this work. Further, the accuracy of our measurements was also established through the comparison of our density values with literature data[4] for the system H_2O + MDEA in the whole concentration range, at 313.15 K. This comparison gives a mean absolute deviation of 0.0002 g·cm^{-3}. Hence, we deem the precision, ±0.0002 g·cm^{-3}, and accuracy, ±0.00043 g·cm^{-3}, of our results to be adequate.

Although there are experimental density data reported in the literature[6] for the aqueous blends of DEA + MDEA and DEA + AMP, it is not possible to carry out a direct comparison with our data, since both sets of data do not cover the same concentration range. However, if the correlation reported in the literature[6] for the aqueous mixtures of DEA + MDEA and DEA + AMP is used to extrapolate values to cover the values of 30, 40, and 50 mass % of total concentration of the alkanolamines, the mean absolute difference obtained is 0.0008 g·cm^{-3} for DEA + MDEA and 0.0011 g·cm^{-3} for DEA−AMP. These differences are clearly outside the uncertainty of our data, but they are within the overall mean absolute percentage deviation of the correlation,[6] which is 0.031%.

Table 2. Comparison of Density Values from This Work with Literature for Pure Components

system	T/K	ρ/g·cm^{-3} this work	literature
DEA	313.15	1.0847	1.0838,[4] 1.0843,[8] 1.0846,[9] 1.08401[18]
	323.15	1.0774	1.0771,[4] 1.0778,[8] 1.0781,[9] 1.07732[18]
	333.15	1.0703	1.0700,[4] 1.0714,[8] 1.0716,[9] 1.07074[18]
MDEA	313.15	1.0250	1.02445,[5] 1.0246,[8] 1.0247,[9] 1.0267,[10] 1.0249,[11] 1.02519[12]
	323.15	1.0174	1.01666,[5] 1.0170,[8] 1.0173,[9] 1.0194,[10] 1.0174,[11] 1.01699[12]
	333.15	1.0096	1.00900,[5] 1.0093,[8] 1.0099,[9] 1.0123,[10] 1.0098,[11] 1.00960[12]
NMP	313.15	1.0157	1.0120[4]
	323.15	1.0054	1.0030[4]
	333.15	0.9974	0.9934[4]
2PE	313.15	0.9843	0.9849[13]
	323.15	0.9762	0.9772[13]
	333.15	0.9687	0.9695[13]
AMP	313.15	0.9172	0.9211,[14] 0.9179[15]
	323.15	0.9092	0.9134,[14] 0.9096[15]
	333.15	0.9007	0.9055,[14] 0.9011[15]

The experimental density data obtained in this work for the eight ternary systems are given in Tables 3−10. It is observed that the order in the magnitude of the density values, at a given temperature and concentration, is the following: DEA + MDEA + H_2O > DEA + NMP + H_2O > DEA + 2PE + H_2O > DEA + AMP + H_2O > MDEA + NMP + H_2O > MDEA + AMP + H_2O > NMP + AMP + H_2O. This order is consistent with that for density values for the pure solvents, at a given temperature, which is DEA > MDEA > NMP > 2PE > AMP. The functionality observed for the density versus concentration for each of the eight aqueous blended solvents was linear in the range of temperature studied.

From the experimental density data, molar excess volumes (V^E) were derived. The procedure has been described in ref 6.

All the molar excess volumes determined in this work are negative, which is common for this type of (water + polar organic) mixtures; the V^E values become less negative with increasing temperature.

Molar excess volumes and their dependence on both concentration and temperature are used together with models of the liquid state to interpret the type and strength of molecular interactions; also, excess volumes are used to back-calculate density values of mixtures. In this work we make use of the well-known Redlich−Kister equation to correlate the excess volume of the eight ternary systems studied here in order to have general parameters that will allow us to interpolate and even extrapolate reliable density values.

Correlation of the Experimental Data

The molar excess volume for a multicomponent mixture can be calculated from the molar volume of the mixture and those corresponding to the pure components, according to the following equation:

$$V^E = V_m - \sum x_i V_i^\circ \qquad (1)$$

For a ternary system, eq 1 takes the following form:

$$V^E = V_m - (x_1 V_1^\circ + x_2 V_2^\circ + x_3 V_3^\circ) \qquad (2)$$

The molar excess volume for a ternary system can be considered as the result of the contributions from the three pairs of components present in the ternary:

Table 3. Experimental Density and Molar Excess Volume of DEA (1) + MDEA (2) + H$_2$O (3)

313.15 K				323.15 K				333.15 K			
m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}
30.03	0	1.0260	−0.174	30.16	0	1.0213	−0.184	30.21	0	1.0144	−0.163
25.11	4.81	1.0249	−0.213	25.13	5.17	1.0197	−0.213	24.95	4.96	1.0123	−0.187
19.91	10.02	1.0233	−0.244	19.92	10.15	1.0179	−0.241	19.97	10.19	1.0108	−0.218
15.10	15.02	1.0220	−0.275	14.82	15.48	1.0164	−0.273	14.84	15.15	1.0092	−0.250
9.95	20.03	1.0203	−0.304	9.78	20.17	1.0146	−0.299	9.79	20.21	1.0073	−0.274
5.02	24.91	1.0188	−0.334	4.76	25.09	1.0130	−0.329	5.38	24.97	1.0056	−0.292
0	29.97	1.0171	−0.359	0	30.03	1.0115	−0.356	0	30.16	1.0041	−0.331
39.84	0	1.0384	−0.282	39.93	0	1.0326	−0.274	39.80	0	1.0263	−0.275
35.01	4.96	1.0369	−0.314	34.86	5.34	1.0311	−0.310	34.92	4.99	1.0245	−0.302
30.17	10.04	1.0355	−0.348	29.99	10.30	1.0295	−0.342	29.98	10.15	1.0226	−0.328
25.11	15.43	1.0340	−0.383	24.94	14.97	1.0276	−0.370	24.88	15.26	1.0209	−0.362
20.07	20.02	1.0322	−0.413	20.07	19.82	1.0259	−0.399	19.98	20.01	1.0195	−0.401
14.99	25.26	1.0305	−0.444	14.79	25.03	1.0240	−0.429	15.03	25.09	1.0178	−0.432
9.99	30.46	1.0291	−0.482	10.23	29.75	1.0226	−0.461	10.17	29.89	1.0159	−0.457
5.19	35.15	1.0275	−0.512	4.75	35.23	1.0207	−0.494	4.98	35.11	1.0138	−0.482
0	40.08	1.0255	−0.537	0	40.09	1.0192	−0.526	0	39.80	1.0121	−0.514
49.77	0	1.0494	−0.370	50.26	0	1.0442	−0.379	50.34	0	1.0381	−0.386
44.90	5.11	1.0483	−0.419	45.18	5.11	1.0425	−0.417	45.10	5.06	1.0357	−0.409
39.86	10.17	1.0468	−0.461	40.22	10.17	1.0408	−0.453	39.96	9.97	1.0337	−0.442
34.93	14.93	1.0450	−0.493	35.11	15.35	1.0391	−0.491	34.87	15.01	1.0318	−0.476
29.85	19.94	1.0433	−0.530	30.06	19.92	1.0370	−0.519	30.20	19.96	1.0302	−0.511
24.97	24.93	1.0417	−0.567	24.75	25.20	1.0350	−0.552	25.01	25.16	1.0282	−0.544
20.06	29.97	1.0400	−0.601	19.97	29.90	1.0332	−0.582	19.87	30.07	1.0260	−0.570
15.30	34.74	1.0384	−0.635	15.09	34.99	1.0315	−0.616	15.03	34.91	1.0241	−0.599
10.06	40.02	1.0365	−0.669	10.16	39.90	1.0296	−0.646	9.93	39.97	1.0223	−0.636
4.98	44.66	1.0344	−0.694	4.66	45.24	1.0275	−0.679	4.77	44.94	1.0202	−0.664
0	49.83	1.0326	−0.726	0	49.97	1.0258	−0.710	0	49.96	1.0184	−0.695

Table 4. Experimental Density and Molar Excess Volume of DEA (1) + NMP (2) + H$_2$O (3)

313.15 K				323.15 K				333.15 K			
m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}
30.03	0	1.0260	−0.174	30.16	0	1.0213	−0.184	39.80	0	1.0263	−0.275
25.07	5.03	1.0237	−0.195	25.03	4.96	1.0182	−0.195	35.16	4.85	1.0234	−0.285
20.27	9.86	1.0213	−0.212	20.08	10.31	1.0157	−0.215	30.34	9.74	1.0204	−0.296
14.96	15.00	1.0184	−0.226	14.97	15.02	1.0126	−0.224	25.12	14.60	1.0170	−0.304
9.88	19.95	1.0158	−0.242	9.81	19.91	1.0095	−0.235	20.11	19.90	1.0144	−0.327
4.94	25.06	1.0134	−0.259	4.90	25.32	1.0071	−0.255	14.87	24.93	1.0112	−0.339
0	30.03	1.0107	−0.270	0	30.30	1.0037	−0.252	10.05	29.78	1.0082	−0.349
39.84	0	1.0384	−0.282	39.93	0	1.0326	−0.274	4.96	35.15	1.0053	−0.365
35.04	5.07	1.0358	−0.296	34.83	5.34	1.0300	−0.299	0	39.83	1.0021	−0.370
29.96	9.91	1.0331	−0.315	30.29	9.80	1.0271	−0.306	50.34	0	1.0381	−0.386
24.82	15.11	1.0308	−0.343	24.96	15.25	1.0241	−0.324	44.99	5.52	1.0344	−0.392
19.94	20.04	1.0279	−0.351	20.14	20.08	1.0215	−0.344	40.06	9.83	1.0311	−0.400
15.13	25.04	1.0257	−0.375	15.00	25.14	1.0188	−0.365	35.32	14.76	1.0284	−0.420
9.94	30.11	1.0231	−0.396	9.88	29.84	1.0159	−0.382	30.11	19.68	1.0251	−0.433
4.95	35.00	1.0208	−0.421	5.03	35.05	1.0132	−0.398	25.21	24.82	1.0222	−0.450
0	39.89	1.0181	−0.434	0	39.76	1.0100	−0.403	20.20	29.89	1.0192	−0.467
49.77	0	1.0494	−0.370	50.26	0	1.0442	−0.379	14.92	34.87	1.0161	−0.484
45.02	5.37	1.0477	−0.412	44.82	5.21	1.0412	−0.404	10.02	40.01	1.0134	−0.507
40.24	10.07	1.0452	−0.434	40.09	9.69	1.0384	−0.421	5.06	45.23	1.0106	−0.527
34.96	14.84	1.0422	−0.451	34.90	15.37	1.0353	−0.439	0	49.84	1.0075	−0.538
29.80	20.16	1.0397	−0.479	30.00	20.34	1.0326	−0.461	60.17	0	1.0481	−0.484
24.95	25.34	1.0377	−0.514	25.26	25.09	1.0301	−0.485	49.90	10.21	1.0414	−0.514
19.93	30.29	1.0352	−0.539	19.84	29.97	1.0269	−0.501	39.89	20.17	1.0352	−0.552
14.90	35.03	1.0323	−0.552	14.94	34.78	1.0242	−0.522	30.29	29.92	1.0294	−0.591
10.34	40.18	1.0306	−0.590	9.78	39.85	1.0214	−0.544	20.17	39.96	1.0233	−0.629
4.75	45.02	1.0274	−0.604	5.01	45.00	1.0193	−0.579	10.00	49.88	1.0174	−0.672
0	50.05	1.0253	−0.634	0	50.1	1.0163	−0.593	0	59.65	1.0114	−0.704

$$V^E_{123} = V^E_{12} + V^E_{13} + V^E_{23} \qquad (3)$$

For a binary system, the polynomial Redlich−Kister equation used to represent the excess volume has the following form:

$$V^E_{12} = x_1 x_2 \sum_{i=0}^{n} A_i (x_1 - x_2)^i \qquad (4)$$

where A_i corresponds to adjustable parameters and x is the mole fraction.

Substituting eq 4 in eq 3, for each pair of components in the ternary system, the polynomial Redlich−Kister equation for a ternary system is then obtained:

$$V^E_{123} = x_1 x_2 \sum_{i=0}^{n} A_i (x_1 - x_2)^i + x_1 x_3 \sum_{i=0}^{n} B_i (x_1 - x_3)^i +$$
$$x_2 x_3 \sum_{i=0}^{n} C_i (x_2 - x_3)^i \quad (5)$$

Equation 5 allows the calculation of the molar excess volume for a ternary system using two different ap-

Table 5. Experimental Density and Molar Excess Volume of DEA (1) + 2PE (2) + H₂O (3) at 313.15 K

m_1/%	m_2/%	ρ_m/g·cm^{-3}	V^E/cm^3·mol^{-1}
30.03	0	1.0260	−0.174
24.97	5.12	1.0226	−0.212
19.88	10.31	1.0187	−0.239
14.50	14.86	1.0146	−0.262
9.83	19.87	1.0108	−0.281
5.13	24.84	1.0068	−0.296
0	30.00	1.0025	−0.311
39.84	0	1.0384	−0.282
34.96	4.99	1.0347	−0.315
30.27	9.94	1.0312	−0.348
24.82	14.90	1.0266	−0.368
19.93	19.95	1.0224	−0.387
15.03	25.06	1.0181	−0.403
10.07	29.62	1.0135	−0.407
5.03	34.68	1.0088	−0.413
0	39.98	1.0042	−0.422
49.77	0	1.0494	−0.370
45.26	4.98	1.0463	−0.419
39.98	9.73	1.0420	−0.448
34.98	14.97	1.0380	−0.483
30.05	19.98	1.0337	−0.506
24.82	25.08	1.0289	−0.519
19.92	29.92	1.0245	−0.534
15.01	35.27	1.0200	−0.551
9.97	39.99	1.0152	−0.553
4.98	45.16	1.0102	−0.551
0	50.22	1.0057	−0.563

proximations: (a) using the adjustable parameters derived for each of the binary systems to "predict" the excess volume of the ternary system at a given temperature[6] and (b) using this equation to correlate experimental excess volume data for the ternary system; under the latter, experimental excess volume data from the binaries can also

be included. In both cases, the number of adjustable parameters used to take into account the functionality of excess volume with concentration is established through the standard deviation obtained during the fitting procedure at a given temperature.

From a previous work,[7] it has been established that for binary systems three adjustable parameters are enough to represent with good accuracy the excess volume as a function of concentration, at a given temperature, using the Redlich−Kister equation. Assuming that at least three adjustable parameters are needed to represent each of the binary systems from a ternary, at a given temperature, then it would be necessary to have a total of nine parameters to represent the excess volume of a given ternary system, that is, three for each A_i, B_i, and C_i, respectively.

Considering that most of the experimental V^E data derived in this work are available at three different temperatures, the temperature functionality was also incorporated into the adjustable parameters of eq 5. Assuming a linear dependence with temperature, the adjustable parameters can be optimized for a given ternary system; in which case, A_i in eq 5 takes the following form:

$$A_0 = A_{00} + A_{01}T$$

$$A_1 = A_{10} + A_{11}T$$

$$A_2 = A_{20} + A_{21}T \qquad (6)$$

The same functionality is applied to the adjustable parameters B_i and C_i of eq 5.

With the scheme mentioned above, the adjustable parameters needed to represent the excess volume of a

Table 6. Experimental Density and Molar Excess Volumes of DEA (1) + AMP (2) + H₂O (3)

313.15 K				323.15 K				333.15 K			
m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}
30.03	0	1.0260	−0.174	30.16	0	1.0213	−0.184	30.21	0	1.0144	−0.163
24.93	4.98	1.0198	−0.235	24.86	5.26	1.0142	−0.236	25.08	5.05	1.0074	−0.213
20.15	9.96	1.0136	−0.288	19.99	10.10	1.0078	−0.286	20.08	9.88	1.0003	−0.250
15.16	15.16	1.0072	−0.343	15.09	15.19	1.0014	−0.339	15.03	14.83	0.9942	−0.313
9.99	20.13	1.0006	−0.389	10.08	20.12	0.9948	−0.384	10.01	19.90	0.9879	−0.370
5.03	25.07	0.9943	−0.435	4.69	25.08	0.9878	−0.422	4.86	25.20	0.9813	−0.427
0	29.83	0.9880	−0.476	0	29.88	0.9816	−0.464	0	29.96	0.9733	−0.428
39.84	0	1.0384	−0.282	39.93	0	1.0326	−0.274	39.80	0	1.0263	−0.275
34.87	4.96	1.0317	−0.337	34.83	5.19	1.0256	−0.334	34.74	5.09	1.0193	−0.334
30.03	9.95	1.0253	−0.397	30.04	10.38	1.0191	−0.398	30.13	10.16	1.0129	−0.398
25.12	14.89	1.0186	−0.447	25.06	14.88	1.0125	−0.441	24.92	14.96	1.0057	−0.441
19.94	19.84	1.0120	−0.501	20.10	20.05	1.0057	−0.496	19.87	20.01	0.9987	−0.490
14.96	24.99	1.0054	−0.555	15.10	24.83	0.9989	−0.537	15.06	25.08	0.9919	−0.541
10.20	29.95	0.9989	−0.601	10.34	29.77	0.9923	−0.581	9.87	30.14	0.9846	−0.579
5.07	34.98	0.9921	−0.643	5.39	34.66	0.9855	−0.619	5.14	35.09	0.9778	−0.618
0	39.89	0.9853	−0.679	0	40.27	0.9779	−0.660	0	39.83	0.9707	−0.646
49.77	0	1.0494	−0.370	50.26	0	1.0442	−0.379	50.34	0	1.0381	−0.386
44.99	5.14	1.0435	−0.461	44.83	5.27	1.0367	−0.446	45.14	5.10	1.0305	−0.444
40.17	10.02	1.0368	−0.521	40.26	10.15	1.0302	−0.509	40.00	9.84	1.0232	−0.494
34.94	14.93	1.0296	−0.572	34.79	15.25	1.0227	−0.565	34.88	15.33	1.0157	−0.560
29.96	20.05	1.0227	−0.631	30.01	19.68	1.0161	−0.608	30.02	19.87	1.0089	−0.605
24.94	24.87	1.0158	−0.678	24.99	24.66	1.0089	−0.653	25.06	25.19	1.0015	−0.660
19.92	29.82	1.0089	−0.725	19.91	30.25	1.0014	−0.709	20.01	30.20	0.9941	−0.701
15.04	34.85	1.0019	−0.768	15.16	34.95	0.9946	−0.746	14.93	34.94	0.9869	−0.735
9.92	39.84	0.9947	−0.803	9.79	39.66	0.9871	−0.767	10.02	39.94	0.9795	−0.766
5.07	44.96	0.9875	−0.835	5.38	44.99	0.9801	−0.809	5.08	44.84	0.9723	−0.796
0	50.26	0.9803	−0.875	0	49.88	0.9726	−0.830	0	49.88	0.9647	−0.818
								95.05	0	1.0700	−0.329
								74.93	20.10	1.0355	−0.668
								60.24	34.78	1.0104	−0.804
								44.88	50.51	0.9841	−0.870
								30.02	64.94	0.9602	−0.875
								15.09	79.98	0.9353	−0.759
								0	94.96	0.9111	−0.596

Table 7. Experimental Density and Molar Excess Volume of MDEA (1) + NMP (2) + H$_2$O (3)

	313.15 K				323.15 K				333.15 K		
m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}
29.97	0	1.0171	−0.359	40.09	0	1.0192	−0.526	39.80	0	1.0121	−0.514
25.18	5.00	1.0162	−0.347	35.02	4.99	1.0179	−0.507	34.89	4.91	1.0106	−0.489
19.78	10.02	1.0148	−0.326	29.71	10.30	1.0165	−0.486	29.79	9.92	1.0093	−0.470
14.91	15.12	1.0139	−0.315	24.97	15.17	1.0156	−0.476	25.08	15.18	1.0082	−0.456
9.82	20.24	1.0128	−0.299	19.92	19.97	1.0144	−0.459	20.16	19.91	1.0069	−0.436
4.91	24.81	1.0116	−0.282	15.11	24.65	1.0133	−0.445	14.86	25.13	1.0057	−0.421
0	30.03	1.0107	−0.270	9.88	30.10	1.0122	−0.431	9.91	29.77	1.0044	−0.401
40.08	0	1.0255	−0.537	5.03	34.85	1.0113	−0.422	5.00	35.02	1.0035	−0.392
35.16	5.06	1.0247	−0.527	0	39.76	1.0100	−0.403	0	39.83	1.0021	−0.370
30.01	10.21	1.0236	−0.510	49.97	0	1.0258	−0.710	49.96	0	1.0184	−0.695
25.06	15.17	1.0229	−0.503	44.73	4.99	1.0244	−0.684	45.34	4.99	1.0172	−0.677
19.80	20.09	1.0217	−0.483	40.01	10.19	1.0234	−0.672	40.24	10.28	1.0159	−0.656
15.13	25.01	1.0211	−0.478	35.19	14.97	1.0223	−0.655	35.19	15.10	1.0147	−0.636
10.23	29.96	1.0199	−0.457	30.20	20.09	1.0215	−0.648	30.12	20.15	1.0135	−0.617
4.92	35.16	1.0190	−0.446	25.07	25.25	1.0205	−0.635	25.00	25.34	1.0125	−0.604
0	39.89	1.0181	−0.434	20.10	29.95	1.0197	−0.627	20.00	30.34	1.0114	−0.588
49.83	0	1.0326	−0.726	14.86	35.00	1.0187	−0.614	14.90	35.03	1.0102	−0.568
44.99	5.25	1.0321	−0.724	9.88	39.95	1.0177	−0.601	10.07	40.23	1.0095	−0.565
39.78	10.11	1.0309	−0.701	5.10	44.91	1.0170	−0.596	5.11	44.86	1.0085	−0.551
35.14	14.91	1.0304	−0.698	0	50.10	1.0163	−0.593	0	49.84	1.0075	−0.538
30.27	20.01	1.0295	−0.684	60.28	0	1.0310	−0.907	59.99	0	1.0229	−0.874
25.18	24.98	1.0289	−0.678	50.32	9.98	1.0289	−0.872	50.34	9.65	1.0205	−0.828
19.97	29.95	1.0280	−0.664	39.64	20.36	1.0267	−0.833	40.30	20.11	1.0184	−0.796
15.03	34.61	1.0272	−0.652	29.93	30.34	1.0253	−0.824	30.14	30.04	1.0164	−0.764
10.06	39.76	1.0265	−0.644	20.01	40.37	1.0240	−0.817	20.33	40.07	1.0148	−0.748
4.91	45.30	1.0261	−0.645	10.02	50.19	1.0228	−0.812	10.10	50.24	1.0132	−0.732
0	50.05	1.0253	−0.634	0	60.21	1.0215	−0.804	0	59.65	1.0114	−0.704

Table 8. Experimental Density and Molar Excess Volume of MDEA (1) + 2PE (2) + H$_2$O (3)

	313.15 K				323.15 K				333.15 K		
m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}
29.97	0	1.0171	−0.359	40.09	0	1.0192	−0.526	39.80	0	1.0121	−0.514
25.18	4.71	1.0154	−0.366	34.84	4.70	1.0167	−0.515	34.95	5.09	1.0096	−0.506
19.93	9.95	1.0128	−0.356	30.10	9.84	1.0145	−0.514	30.08	9.97	1.0070	−0.492
14.97	15.03	1.0106	−0.354	25.16	14.79	1.0119	−0.502	24.90	14.94	1.0045	−0.482
9.97	19.63	1.0079	−0.336	20.07	19.74	1.0090	−0.481	19.97	19.89	1.0017	−0.463
5.23	24.95	1.0055	−0.329	15.09	24.98	1.0063	−0.467	15.12	24.98	0.9989	−0.445
0	30.00	1.0025	−0.307	9.96	29.81	1.0034	−0.445	9.97	30.26	0.9960	−0.425
40.08	0	1.0255	−0.537	4.96	34.93	1.0005	−0.424	5.01	34.85	0.9930	−0.396
35.07	4.69	1.0233	−0.533	0	40.00	0.9975	−0.399	0	39.80	0.9900	−0.369
30.07	9.94	1.0212	−0.536	49.97	0	1.0258	−0.710	49.96	0	1.0184	−0.695
25.04	15.02	1.0186	−0.524	44.87	4.86	1.0234	−0.701	44.93	4.72	1.0159	−0.682
20.02	20.14	1.0157	−0.504	39.75	10.33	1.0208	−0.696	39.98	10.18	1.0133	−0.676
14.89	25.20	1.0131	−0.491	34.85	15.28	1.0181	−0.679	34.91	15.16	1.0103	−0.650
10.11	29.89	1.0102	−0.466	29.87	19.91	1.0153	−0.655	30.06	19.79	1.0077	−0.630
4.98	34.80	1.0072	−0.441	25.00	24.88	1.0126	−0.638	24.92	25.17	1.0048	−0.612
0	39.98	1.0042	−0.416	20.24	29.98	1.0096	−0.613	20.33	29.92	1.0021	−0.591
49.83	0	1.0326	−0.726	14.95	34.85	1.0067	−0.588	15.30	35.11	0.9992	−0.569
44.81	4.89	1.0305	−0.727	10.05	39.86	1.0040	−0.569	10.05	40.04	0.9963	−0.542
40.01	9.83	1.0282	−0.722	4.97	44.83	1.0011	−0.544	4.96	45.22	0.9934	−0.519
35.18	14.80	1.0256	−0.709	0	50.03	0.9983	−0.524	0	49.95	0.9906	−0.492
30.11	20.18	1.0229	−0.697	60.28	0	1.0310	−0.907	59.99	0	1.0229	−0.874
25.10	25.23	1.0200	−0.674	49.73	9.77	1.0257	−0.870	50.07	10.21	1.0178	−0.854
20.02	30.13	1.0172	−0.652	40.03	20.05	1.0204	−0.845	39.98	20.21	1.0122	−0.810
14.80	35.03	1.0142	−0.623	29.90	30.23	1.0149	−0.807	29.95	30.10	1.0069	−0.771
9.95	40.09	1.0114	−0.602	19.75	39.97	1.0092	−0.752	20.14	39.89	1.0015	−0.727
5.06	44.83	1.0086	−0.576	9.94	50.24	1.0038	−0.715	10.02	49.80	0.9959	−0.673
0	50.22	1.0057	−0.555	0	59.99	0.9984	−0.666	0	59.95	0.9903	−0.622

ternary system, as a function of both concentration and temperature, are 18. The corresponding adjustable parameter values for the different ternary systems studied here were obtained using the least squares technique together with the Gauss−Newton method to satisfy the following objective function:

$$FO = \min \sum_{i=1}^{N} (V_{iexp}^E - V_{ical}^E)^2 \qquad (7)$$

The quality of the fitting procedure was established through the standard deviation, which in turn was evalu-

ated for each ternary system, according to the following relation:

$$\sigma_{VE} = \sqrt{\frac{\sum_{i=1}^{N}(V_{iexp}^E - V_{ical}^E)^2}{N - m}}$$

with N the number of experimental points and m the number of adjustable parameters.

The adjusted parameters corresponding to each system studied along with the standard deviation of the fit are

Table 9. Experimental Density and Molar Excess Volume of MDEA (1) + AMP (2) + H₂O (3)

313.15 K				323.15 K				333.15 K			
m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}
29.97	0	1.0171	−0.359	30.03	0	1.0115	−0.356	30.16	0	1.0041	−0.331
24.92	5.20	1.0121	−0.384	25.15	4.93	1.0065	−0.375	25.12	4.76	0.9992	−0.351
19.96	9.94	1.0073	−0.401	20.08	9.84	1.0015	−0.394	20.21	9.86	0.9938	−0.365
15.00	15.00	1.0024	−0.422	14.99	14.94	0.9965	−0.415	14.84	15.15	0.9889	−0.396
10.05	20.07	0.9976	−0.445	10.25	19.88	0.9916	−0.433	10.14	19.77	0.9849	−0.428
5.05	25.11	0.9928	−0.465	5.14	24.81	0.9866	−0.447	5.30	24.89	0.9788	−0.422
0	29.83	0.9880	−0.476	0	29.88	0.9816	−0.464	0	29.96	0.9733	−0.428
40.08	0	1.0255	−0.537	40.09	0	1.0192	−0.526	39.80	0	1.0121	−0.514
34.92	5.15	1.0203	−0.561	34.81	4.85	1.0139	−0.539	34.79	5.23	1.0067	−0.538
29.99	10.05	1.0153	−0.580	29.99	10.17	1.0088	−0.570	29.97	10.18	1.0019	−0.568
24.88	14.92	1.0102	−0.594	25.26	14.93	1.0039	−0.588	24.96	15.13	0.9964	−0.577
19.71	20.28	1.0050	−0.620	20.11	19.94	0.9986	−0.601	20.03	20.04	0.9914	−0.597
14.82	24.87	1.0002	−0.631	15.22	25.02	0.9935	−0.620	15.12	25.03	0.9863	−0.615
9.96	29.99	0.9953	−0.654	10.12	29.72	0.9885	−0.628	10.08	29.77	0.9812	−0.623
5.14	34.82	0.9904	−0.666	4.95	35.12	0.9830	−0.644	4.78	35.23	0.9756	−0.639
0	39.89	0.9853	−0.679	0	40.27	0.9779	−0.660	0	39.83	0.9707	−0.646
49.83	0	1.0326	−0.726	49.97	0	1.0258	−0.710	49.96	0	1.0184	−0.695
44.84	5.16	1.0274	−0.754	44.90	5.60	1.0202	−0.744	44.82	4.99	1.0130	−0.715
39.89	10.04	1.0222	−0.771	39.91	9.81	1.0150	−0.737	39.96	10.13	1.0076	−0.739
35.13	14.97	1.0173	−0.798	35.22	14.95	1.0101	−0.773	35.13	14.87	1.0024	−0.753
29.89	19.82	1.0119	−0.803	30.15	20.17	1.0046	−0.791	29.97	20.17	0.9967	−0.769
25.03	24.95	1.0067	−0.824	25.25	25.42	0.9991	−0.808	24.97	24.86	0.9915	−0.776
20.06	29.98	1.0015	−0.839	19.85	30.02	0.9939	−0.808	20.06	29.64	0.9864	−0.788
14.90	34.80	0.9962	−0.842	14.71	34.82	0.9885	−0.807	15.22	35.07	0.9808	−0.807
9.83	39.93	0.9908	−0.850	10.02	39.48	0.9835	−0.813	10.05	40.10	0.9753	−0.812
5.19	44.90	0.9858	−0.865	5.00	44.78	0.9780	−0.825	4.95	44.78	0.9701	−0.811
0	50.26	0.9803	−0.875	0	49.88	0.9726	−0.830	0	49.88	0.9647	−0.818

Table 10. Experimental Density and Molar Excess Volume of NMP (1) + AMP (2) + H₂O (3)

313.15 K				323.15 K				333.15 K			
m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}	m_1/%	m_2/%	ρ/g·cm^{-3}	V^E/cm^3·mol^{-1}
30.03	0	1.0107	−0.270	30.30	0	1.0037	−0.252	39.83	0	1.0021	−0.370
25.11	5.24	1.0075	−0.326	24.93	4.99	1.0000	−0.289	35.08	4.81	0.9987	−0.417
20.21	9.78	1.0038	−0.353	19.88	10.03	0.9968	−0.339	29.76	10.10	0.9945	−0.456
15.11	14.73	0.9998	−0.383	15.30	14.79	0.9935	−0.378	24.99	14.98	0.9906	−0.490
9.98	20.08	0.9957	−0.419	10.13	20.05	0.9892	−0.405	20.05	20.14	0.9865	−0.526
5.43	24.83	0.9921	−0.450	5.03	24.99	0.9857	−0.443	14.84	24.98	0.9825	−0.552
0	29.83	0.9880	−0.476	0	29.88	0.9816	−0.464	10.03	29.93	0.9786	−0.586
39.89	0	1.0181	−0.434	39.76	0	1.0100	−0.403	4.94	34.87	0.9746	−0.614
34.98	5.07	1.0146	−0.485	35.03	4.77	1.0066	−0.447	0	39.83	0.9707	−0.646
30.22	9.80	1.0108	−0.519	29.95	10.01	1.0022	−0.477	49.84	0	1.0075	−0.538
24.97	15.01	1.0064	−0.550	24.95	14.73	0.9982	−0.502	45.02	4.80	1.0036	−0.577
19.98	19.89	1.0024	−0.581	19.98	20.22	0.9940	−0.543	40.09	10.03	0.9992	−0.615
15.41	24.79	0.9984	−0.612	15.08	24.92	0.9902	−0.571	35.27	14.86	0.9950	−0.643
9.86	30.11	0.9938	−0.637	10.14	30.09	0.9865	−0.614	29.87	20.16	0.9903	−0.670
4.97	35.03	0.9896	−0.661	5.06	34.98	0.9825	−0.640	24.84	24.88	0.9861	−0.691
0	39.89	0.9853	−0.679	0	40.27	0.9779	−0.660	20.05	30.03	0.9817	−0.722
50.05	0	1.0253	−0.634	50.10	0	1.0163	−0.593	14.95	34.94	0.9774	−0.743
44.76	5.26	1.0211	−0.680	44.91	5.13	1.0119	−0.625	10.02	40.27	0.9729	−0.775
39.82	9.76	1.0169	−0.699	40.08	10.24	1.0076	−0.661	5.01	45.25	0.9687	−0.800
35.03	14.81	1.0125	−0.731	34.90	15.09	1.0031	−0.679	0	49.88	0.9647	−0.818
30.11	19.99	1.0080	−0.763	29.99	19.69	0.9990	−0.698	59.65	0	1.0114	−0.704
25.05	24.79	1.0035	−0.779	24.91	24.78	0.9945	−0.722	49.85	10.35	1.0023	−0.781
19.96	30.03	0.9986	−0.798	20.08	29.95	0.9900	−0.748	39.76	19.91	0.9932	−0.814
15.27	35.10	0.9942	−0.826	14.80	35.25	0.9854	−0.773	29.72	30.02	0.9838	−0.856
9.67	39.94	0.9896	−0.835	10.05	39.85	0.9814	−0.793	19.93	39.91	0.9747	−0.894
5.12	45.59	0.9848	−0.870	5.02	45.07	0.9768	−0.813	10.09	50.31	0.9652	−0.933
0	50.26	0.9803	−0.875	0	49.88	0.9726	−0.830	0	59.99	0.9566	−0.964

given in Table 11. The Redlich−Kister equation reproduces the experimentally derived excess volume of the eight ternary systems considered in this study within a mean standard deviation of ±0.007 cm^3·mol^{-1}, which is satisfactory considering that both the concentration and temperature functionality of the data are included in such a correlation.

Conclusions

Density data of aqueous blended polar solvents have been measured for eight systems; all the ternary systems studied have industrial interest, since they are being used or could be used soon for the sweetening of hydrocarbon-rich gas streams. Reliable density data are now available in a large range of temperature and concentration.

The experimentally derived molar excess volume was correlated satisfactorily by the Redlich−Kister equation; the correlation procedure included the functionality with both temperature and concentration which represents a further contribution to results from works reported previously in the literature.

The behavior observed for the excess volume of the systems studied in this work is the one that traditionally has been found for aqueous organic systems.

Table 11. Parameters of the Redlich−Kister Equation Obtained from the Correlation of the Molar Excess Volume for Aqueous Blended Solvents as a Function of Concentration and Temperature

parameters		$DEA + MDEA + H_2O$	$DEA + NMP + H_2O$	$DEA + 2PE + H_2O$	$DEA + AMP + H_2O$
A_0	A_{00}	−13.0467	−87.1251	−13.7657	−3.1884
	A_{01}	0.0292	0.2612		−0.0075
A_1	A_{10}	−29.8877	47.0257	−1.2945	−564.7317
	A_{11}	0.1246	−0.1245		1.7806
A_2	A_{20}	−7090.0316	−2281.6405	144.2067	−3783.1254
	A_{21}	22.3027	6.8864		11.7650
B_0	B_{00}	−6.0581	168.0799	13.6796	−163.0005
	B_{01}	0.0460	−0.5115		0.5499
B_1	B_{10}	−31.4697	455.8372	43.6896	−437.8592
	B_{11}	0.1955	−1.3634		1.5009
B_2	B_{20}	−36.1600	303.6494	28.4957	−297.3488
	B_{21}	0.1775	−0.9086		1.0169
C_0	C_{00}	−511.8139	−104.9756	−38.0842	−49.0584
	C_{01}	1.5878	0.3103		0.1499
C_1	C_{10}	−1265.0168	−208.3619	−86.3117	−95.8773
	C_{11}	3.9721	0.6514		0.3346
C_2	C_{20}	−796.0145	−114.9714	−56.5399	−61.1762
	C_{21}	2.4985	0.3667		0.2089
$\sigma_{V^E}/cm^3 \cdot mol^{-1}$		0.0069	0.0063	0.0062	0.0087

parameters		$MDEA + NMP + H_2O$	$MDEA + 2PE + H_2O$	$MDEA + AMP + H_2O$	$NMP + AMP + H_2O$
A_0	A_{00}	−103.4105	−83.9041	17.6422	−103.1095
	A_{01}	0.3212	0.2564	−0.0573	0.3114
A_1	A_{10}	31.3360	−369.0140	−181.2139	−146.3929
	A_{11}	−0.0838	1.1040	0.5618	0.4310
A_2	A_{20}	1736.0202	722.1554	−719.4952	−1441.0648
	A_{21}	−5.2490	−2.2575	2.5301	4.3090
B_0	B_{00}	115.4842	77.8437	−357.4139	20.0808
	B_{01}	−0.3611	−0.2487	1.1521	−0.0555
B_1	B_{10}	346.8323	246.7665	−872.9198	99.3376
	B_{11}	−1.0381	−0.7398	2.8653	−0.2428
B_2	B_{20}	237.1200	171.4058	−547.8907	70.6008
	B_{21}	−0.7147	−0.5192	1.7978	−0.1682
C_0	C_{00}	−22.1629	−81.9179	0.0623	−14.5274
	C_{01}	0.0599	0.2189	−0.0003	0.0290
C_1	C_{10}	17.2495	−189.6134	27.1497	10.9783
	C_{11}	−0.0307	0.5246	−0.0397	−0.0357
C_2	C_{20}	36.6420	−131.3114	13.3931	22.9983
	C_{21}	−0.0916	0.3586	−0.0165	−0.0796
$\sigma_{V^E}/cm^3 \cdot mol^{-1}$		0.0061	0.0057	0.0083	0.0078

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