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Densities and Excess Molar Volumes of Binary Mixtures of Bis(2-hydroxyethyl)ammonium Acetate + Water and Monoethanolamine + Bis(2-hydroxyethyl)ammonium Acetate at Temperatures from (303.15 to 353.15) K

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In the present work, binary mixtures of bis(2-hydroxyethyl)ammonium acetate + water and monoethanolamine + bis(2-hydroxyethyl)ammonium acetate were prepared with different compositions, and their densities were measured over the temperature range from (303.15 to 353.15) K. Excess molar volumes were calculated using the measured density values and correlated using a Redlich−Kister-type polynomial equation.

Introduction

The development of new hybrid solvents for the absorption of carbon dioxide from natural gas have drawn great attention because of the ability of CO_2 as a greenhouse gas that can bring harm especially to humans and the environment. Currently, common solvents such as aqueous methyldiethanolamine (MDEA), monoethanolamine (MEA), and diethanolamine (DEA) are used for CO_2 removal from natural gas, although they cause serious environmental concerns because of their volatility and corrosiveness.[1] In the United States, 95% of natural gas “sweetening”operations are mainly amine-based absorption, where 1 mol of CO_2 reacts with 2 equiv of MEA to produce carbamate.[2] This process, however, seems to be energy-intensive, although its effectiveness in capturing CO_2 cannot be denied. Therefore, more studies are being carried out in order to improve the energy efficiency of CO_2 absorption using nonvolatile and noncorrosive hybrid solvents. Recently, some researchers reported that ionic liquids possess an effective and high capability for CO_2 capture and have several advantages over other molecular solvents used for this purpose.[3−6] These ionic liquids have negligible vapor pressure and hence are better suited for recycling operations such as distillation, absorption, etc. Among the available ionic liquids, imidazolium-based ionic liquids have been found to be more ideal for CO_2 removal.[3−6] Anthony et al.[7] found that CO_2 has the highest solubilty in 1-butyl-3-methylimidazolium hexafluorophosphate, [bmim]-[PF_6], among other gases (ethane, ethylene, methane, argon, oxygen, carbon monoxide, hydrogen, and nitrogen), whereas Kumelan et al.[8] reported the solubility of CO_2 in 1-hexyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide, [hmim]-[Tf_2N]. On the other hand, Yuan et al.[9] found that the solubility of CO_2 in hydroxylammonium ionic liquids increased with an increase in pressure, and the opposite trend was found in the case of temperature. A similar study involving other hydroxylammonium ionic liquids was made by our group, and a similar trend was observed.[10]

Most of the previous studies involving carbon dioxide absorption have mainly been focused on either ionic liquids or amines individually, but literature reports pertaining to the studies on the combinations of amines with ionic liquids as either binary mixtures or ternary mixtures with water for CO_2 removal are not readily available. Iglesias et al.[11] studied the physical properties (density and ultrasonic velocity) of 2-hydroxyethylammonium formate (2-HEAF) and its solutions in water and found that these solutions show total miscibility over the temperature range (288.15 to 323.15) K at atmospheric pressure. The present synthesized bis(2-hydroxyethyl)ammonium acetate (BHEAA) ionic liquids have been found to be miscible with water in all proportions. Our experiments have also shown that MEA, which is a commercially inexpensive amine, is also miscible with BHEAA. Hence, the present work was aimed at preparing binary mixtures of the ionic liquid with cost-effective amines to form hybrid solvents for their potential application in the effective capture of CO_2. The binary mixtures used in the present study involve the combinations BHEAA + water and BHEAA + MEA. Since the basic physical properties are essential for the design, scale-up, and sizing of the equipment for commercial applications, the densities of the binary mixtures at temperatures from (303.15 to 353.15) K have been measured, and on the basis of these measured densities, the excess molar volumes have also been estimated.

Table 1. Comparison of Measured Densities (ρ/g·cm^{-3}) for Pure MEA and Pure BHEAA at Temperatures from (303.15 to 353.15) K

	MEA		BHEAA	
T/K	this work	lit.	this work	lit.[a]
303.15	1.01067	1.0081[b] 1.00828[c] 1.00817[d] 1.0082[e] 1.0091[f]	1.16862	1.16639
313.15	1.00277	1.0004[b] 1.00034[c] 1.00021[d] 1.0010[e] 1.0013[f]	1.16229	1.16012
323.15	0.99480	0.9927[b] 0.99219[d] 0.9940[e] 0.9934[f]	1.15584	1.15396
333.15	0.98675	0.9850[b] 0.98410[d]	1.14928	1.14764
343.15	0.97862	0.9773[b] 0.97594[d]	1.14276	1.14122
353.15	0.97040	0.9696[b] 0.96768[d]	1.13612	1.13466

[a] Data from ref 1. [b] Data from ref 12. [c] Data from ref 13. [d] Data from ref 14. [e] Data from ref 15. [f] Data from ref 16.

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Table 2. Experimental Densities ρ and Excess Molar Volumes V^E for BHEAA (1) + Water (2) Mixtures

x_1	$\rho/\mathrm{g \cdot cm^{-3}}$	$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	x_1	$\rho/\mathrm{g \cdot cm^{-3}}$	$V^E/\mathrm{cm^3 \cdot mol^{-1}}$
	T/K = 303.15			T/K = 333.15	
0.0000	0.99570	0.0000	0.0000	0.98320	0.0000
0.0678	1.07890	−0.5051	0.0678	1.06330	−0.4992
0.0995	1.10040	−0.6787	0.0995	1.08390	−0.6698
0.1432	1.11980	−0.8367	0.1432	1.10260	−0.8290
0.2005	1.13360	−0.8847	0.2005	1.11610	−0.8875
0.3159	1.15160	−1.0164	0.3159	1.13370	−1.0401
0.4659	1.16060	−0.9186	0.4659	1.14270	−0.9804
0.6045	1.16254	−0.5806	0.6045	1.14328	−0.5589
0.7465	1.16500	−0.3394	0.7465	1.14550	−0.3043
0.8700	1.16660	−0.1322	0.8700	1.14740	−0.1361
1.0000	1.16862	0.0000	1.0000	1.14928	0.0000
	T/K = 313.15			T/K = 343.15	
0.0000	0.99220	0.0000	0.0000	0.97780	0.0000
0.0678	1.07410	−0.4996	0.0678	1.05720	−0.4972
0.0995	1.09520	−0.6706	0.0995	1.07800	−0.6776
0.1432	1.11420	−0.8250	0.1432	1.09660	−0.8391
0.2005	1.12790	−0.8771	0.2005	1.10990	−0.8948
0.3159	1.14570	−1.0138	0.3159	1.12750	−1.0553
0.4659	1.15470	−0.9293	0.4659	1.13640	−0.9962
0.6045	1.15611	−0.5583	0.6045	1.13720	−0.5939
0.7465	1.15860	−0.3232	0.7465	1.13950	−0.3539
0.8700	1.16050	−0.1515	0.8700	1.14100	−0.1483
1.0000	1.16229	0.0000	1.0000	1.14276	0.0000
	T/K = 323.15			T/K = 353.15	
0.0000	0.98810	0.0000	0.0000	0.97180	0.0000
0.0678	1.06880	−0.4933	0.0678	1.05000	−0.4790
0.0995	1.08960	−0.6632	0.0995	1.07180	−0.6888
0.1432	1.10850	−0.8218	0.1432	1.09040	−0.8548
0.2005	1.12210	−0.8778	0.2005	1.10370	−0.9146
0.3159	1.13990	−1.0288	0.3159	1.12120	−1.0785
0.4659	1.14880	−0.9535	0.4659	1.13000	−1.0190
0.6045	1.14967	−0.5495	0.6045	1.13026	−0.6338
0.7465	1.15250	−0.3501	0.7465	1.13300	−0.3695
0.8700	1.15400	−0.1426	0.8700	1.13440	−0.1532
1.0000	1.15584	0.0000	1.0000	1.13612	0.0000

Table 3. Experimental Densities ρ and Excess Molar Volumes V^E for BHEAA (1) + MEA (2) Mixtures

x_1	$\rho/\mathrm{g \cdot cm^{-3}}$	$V^E/\mathrm{cm^3 \cdot mol^{-1}}$	x_1	$\rho/\mathrm{g \cdot cm^{-3}}$	$V^E/\mathrm{cm^3 \cdot mol^{-1}}$
	T/K = 303.15			T/K = 333.15	
0.0000	1.01067	0.0000	0.0000	0.98675	0.0000
0.1000	1.04310	0.0100	0.1000	1.02190	−0.1244
0.2028	1.06717	0.1750	0.2028	1.04692	0.0146
0.3000	1.08483	0.3833	0.3000	1.06558	0.1817
0.4000	1.09884	0.6805	0.4000	1.07989	0.4907
0.5000	1.10844	1.1713	0.5000	1.08975	1.0001
0.5998	1.11624	1.6918	0.5998	1.09853	1.4621
0.7001	1.12506	1.9888	0.7001	1.10723	1.8020
0.8004	1.13464	2.0692	0.8004	1.11723	1.8569
0.9000	1.14910	1.4330	0.9000	1.13150	1.2412
1.0000	1.16862	0.0000	1.0000	1.14928	0.0000
	T/K = 313.15			T/K = 343.15	
0.0000	1.00277	0.0000	0.0000	0.97862	0.0000
0.1000	1.03600	−0.0259	0.1000	1.01473	−0.1749
0.2028	1.06048	0.1253	0.2028	1.04005	−0.0426
0.3000	1.07848	0.3204	0.3000	1.05904	0.1111
0.4000	1.09258	0.6227	0.4000	1.07344	0.4259
0.5000	1.10228	1.1189	0.5000	1.08338	0.9437
0.5998	1.11045	1.6167	0.5998	1.09248	1.3877
0.7001	1.11921	1.9298	0.7001	1.10114	1.7432
0.8004	1.12886	2.0103	0.8004	1.11129	1.7893
0.9000	1.14313	1.3964	0.9000	1.12550	1.1814
1.0000	1.16229	0.0000	1.0000	1.14276	0.0000
	T/K = 323.15			T/K = 353.15	
0.0000	0.99480	0.0000	0.0000	0.97040	0.0000
0.1000	1.02900	−0.0749	0.1000	1.00750	−0.2295
0.2028	1.05373	0.0721	0.2028	1.03309	−0.1033
0.3000	1.07206	0.2530	0.3000	1.05241	0.0365
0.4000	1.08627	0.5587	0.4000	1.06693	0.3547
0.5000	1.09605	1.0620	0.5000	1.07694	0.8814
0.5998	1.10452	1.5422	0.5998	1.08635	1.3076
0.7001	1.11325	1.8688	0.7001	1.09496	1.6799
0.8004	1.12309	1.9351	0.8004	1.10524	1.7185
0.9000	1.13711	1.3503	0.9000	1.11940	1.1177
1.0000	1.15584	0.0000	1.0000	1.13612	0.0000

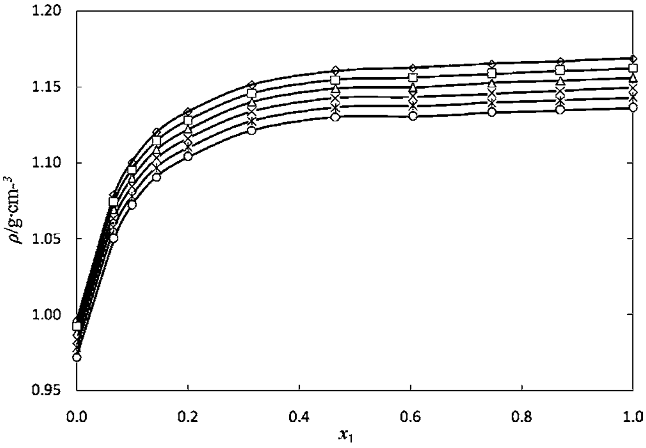

Figure 1. Density ρ vs BHEAA mole fraction x_1 for BHEAA (1) + water (2) mixtures: ◇, 303.15 K; □, 313.15 K; △, 323.15 K; ×, 333.15 K; ∗, 343.15 K; ○, 353.15 K.

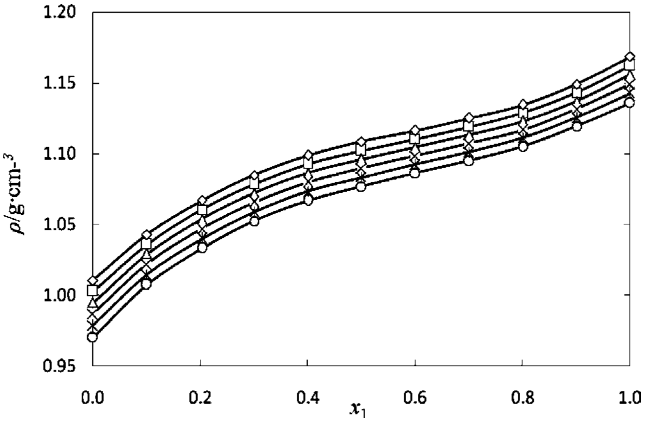

Figure 2. Density ρ vs BHEAA mole fraction x_1 for BHEAA (1) + MEA (2) mixtures: ◇, 303.15 K; □, 313.15 K; △, 323.15 K; ×, 333.15 K; ∗, 343.15 K; ○, 353.15 K.

Experimental Section

Chemicals. The ionic liquid BHEAA used in this study was synthesized in our laboratory, and its structure and physical properties were established previously.[1,10] The water content was determined using a coulometer Karl Fischer titrator (DL 39, Mettler Toledo) and the HYDRANAL-Coulomat AG reagent (Riedel-de Haen). It was found that the mass fraction of water in the BHEAA used in this study was $166 \cdot 10^{-6}$. AR-grade MEA (Aldrich, 99 % purity) was used, while the water used for the preparation of the samples was doubly distilled and deionized.

All of the samples were freshly prepared and retained at room temperature for 24 h to ensure total solubility. All of the samples were kept in airtight sample bottles and closed with caps to avoid the effects of atmospheric humidity on the samples. The samples were prepared using an analytical balance (model AS120S, Mettler Toledo) with a precision of ± 0.0001 g and later converted to mole fraction.

Density Measurements. The densities of all of the binary mixtures as well as pure MEA and pure BHEAA ionic liquid were measured using an oscillating U-tube density meter (model DMA-5000, Anton Paar) at temperatures from (303.15 to 353.15) K with an uncertainty of ± 0.01 K. The density meter included an automatic correction for the viscosity of the sample and was calibrated frequently using Millipore-quality water and dry air as instructed by the supplier. The apparatus is precise to within $1 \cdot 10^{-5}\ \mathrm{g \cdot cm^{-3}}$, and the uncertainty of the measurements was better than $3 \cdot 10^{-5}\ \mathrm{g \cdot cm^{-3}}$. Table 1 compares the measured

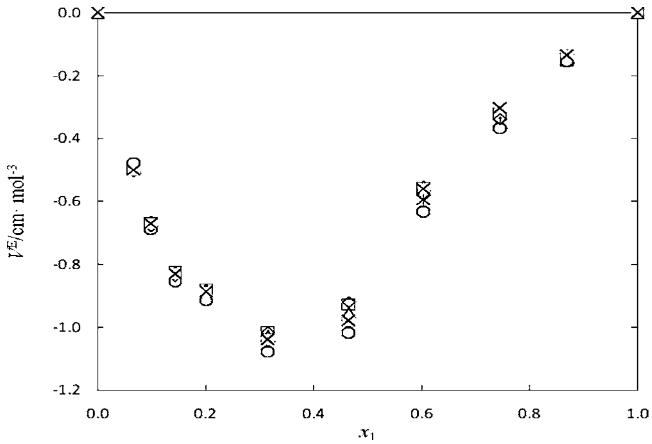

Figure 3. Excess molar volume V^E vs mole fraction x_1 for BHEAA (1) + water (2) binary mixtures: ◇, 303.15 K; □, 313.15 K; △, 323.15 K; ×, 333.15 K; ∗, 343.15 K; ○, 353.15 K.

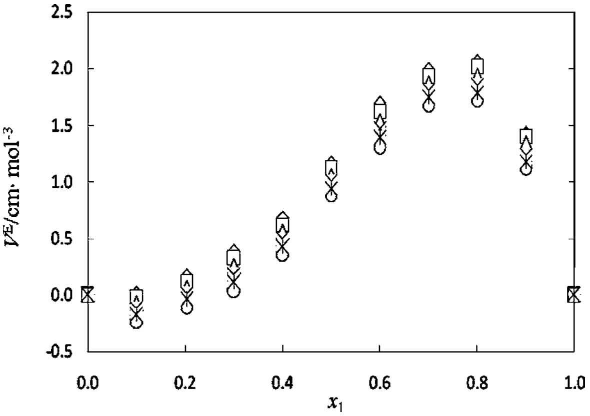

Figure 4. Excess molar volume V^E vs mole fraction x_1 for BHEAA (1) + MEA (2) binary mixtures: ◇, 303.15 K; □, 313.15 K; △, 323.15 K; ×, 333.15 K; ∗, 343.15 K; ○, 353.15 K.

density values of pure BHEAA and MEA with the available literature values[1,12−16] for validation of the results. As can be seen from Table 1, the densities of pure MEA are in good agreement with the values reported in the literature. However, for pure BHEAA, the density values were found to be slightly different than the previously reported data. This small difference may be due to the presence of small amounts of impurities or differences in water content. All of the measurements for each sample were performed in triplicate, and the average values are reported and considered for further analysis.

Result and Discussion

In this present work, binary mixtures, namely, BHEAA + water and BHEAA + MEA, were prepared with different concentrations, and the densities were measured at temperatures from (303.15 to 353.15) K at atmospheric pressure. The objective of this study was to provide information regarding the physical properties of different hybrid solutions at various temperatures and compositions. The measured density values of the pure MEA and BHEAA were compared with the available literature values (see Table 1), and satisfactory agreement was found.

The measured experimental density values for the BHEAA + water and BHEAA + MEA systems over the temperature range (303.15 to 353.15) K are presented in Tables 2 and 3, respectively. The variations of the densities with concentration at the different temperatures studied are shown in Figures 1 and 2. From analysis of the data, it was found that the densities depend more strongly on the mole fraction of BHEAA or MEA in the solution than on temperature. However, the trend for both systems with respect to temperature was the same, as the density decreased with increasing temperature. On the other hand, the density for the BHEAA + water system increased with increasing BHEAA composition up to $x_{\mathrm{BHEAA}} \approx 0.5$, after which the increment was too small and became nearly constant (Figure 1). In contrast to the BHEAA + water system, the density values for the BHEAA + MEA system increased with BHEAA composition.

The excess molar volumes for the two binary systems were calculated from the experimental density values at each temperature using the following equation:

$$V^E = \frac{x_1M_1 + x_2M_2}{\rho} - \frac{x_1M_1}{\rho_1} - \frac{x_2M_2}{\rho_2} \qquad (1)$$

where ρ is the density of the mixture, x_1 and x_2 are the mole fractions of pure components 1 and 2, respectively, and M_1 and M_2 are the molar masses of pure components 1 and 2, respectively.

The calculated excess molar volumes for the binary systems are presented in Tables 2 and 3 and shown graphically in Figures 3 and 4. Excess molar volumes of mixtures are commonly related to the differences in and changes of structure undergone by the pure components. From the results it can be seen that the excess molar volume is negative for the BHEAA + water system, with the most negative value occurring at x_{BHEAA}= 0.3159. The results shown are consistent with previous reported V^E values for similar systems.[12,17−20] The negative values for

Table 4. Parameter Values and Standard Deviations of the Redlich−Kister Equation for Excess Molar Volume V^E at Temperatures from (303.15 to 353.15) K

T/K	A_0	A_1	A_2	A_3	A_4	A_5	A_6	σ/cm^3·mol^{-1}
				BHEAA (1) + Water (2)				
303.15	−3.466	4.786	4.785	−9.972	−25.35	15.33	26.65	0.083
313.15	−3.476	5.167	5.661	−11.63	−28.20	16.52	28.73	0.072
323.15	−3.530	5.686	6.023	−15.57	−31.61	22.08	34.45	0.067
333.15	−3.638	5.703	7.115	−13.59	−32.16	18.69	32.43	0.063
343.15	−3.730	5.548	7.032	−14.25	−33.67	20.36	35.46	0.075
353.15	−3.879	5.346	8.057	−13.11	−38.20	19.18	40.63	0.103
				BHEAA (1) + MEA (2)				
303.15	4.717	9.972	4.727	−0.917	9.389	1.243	−13.61	0.086
313.15	4.481	9.923	4.147	−0.886	10.66	1.296	−14.83	0.066
323.15	4.228	10.00	3.764	−1.878	10.63	2.687	−14.90	0.046
333.15	3.960	9.957	3.091	−1.439	12.79	1.106	−18.98	0.039
343.15	3.710	10.00	2.538	−1.726	13.93	1.281	−20.78	0.038
353.15	3.435	10.04	1.998	−2.023	15.14	1.473	−22.82	0.050

the BHEAA + water system could be attributed to a possible efficient packing of water molecules with BHEAA and/or a possible attractive interaction during the mixing of BHEAA with water. This interaction, however, strongly depends on the composition as well as the temperature. In contrast to the BHEAA + water system, the BHEAA + MEA system exhibits mostly positive values of the excess molar volume, except at higher temperatures, where V^E is slightly negative for dilute ionic liquid solutions and positive for concentrated ionic liquid solutions. This change in excess molar volume may be attributed to the strong interactions with amine solutions at low ionic liquid concentrations, and it becomes weaker with increasing ionic liquid concentration. Figure 4 also indicates the change in the interaction between BHEAA and MEA with concentration as well as with temperature.

The calculated V^E values were correlated using the following Redlich−Kister-type polynomial equation:

$$V^E = x_1 x_2 \sum_{i=0}^{6} A_i (x_1 - x_2)^i \qquad (2)$$

where the A_i are the parameters of the Redlich−Kister polynomial equation, which were obtained using the method of least squares. The parameters of the Redlich−Kister equation representing the excess molar volumes for the BHEAA + water and BHEAA + MEA systems are presented in Table 4 together with the standard deviation, which was estimated using the following equation:

$$\sigma = \left[\frac{\sum (V^E_{\text{exptl}} - V^E_{\text{calcd}})^2}{n} \right]^{1/2} \qquad (3)$$

The small values of the standard deviation for the entire set of data proved that the present form of the Redlich−Kister-type polynomial equation with the constants given in Table 4 is suitable and sufficient to represent the present estimated excess molar volumes at temperatures from (303.15 to 353.15) K for the binary mixtures studied in the present work.

Conclusion

Densities for the binary systems bis(2-hydroxyethyl)ammonium acetate + water and bis(2-hydroxyethyl)ammonium acetate + monoethanolamine were measured over the temperature range (303.15 to 353.15) K. The experimental density values were then used for the calculation of excess molar volumes, which were then correlated using a Redlich−Kister-type polynomial equation. The densities of both binary systems decreased with increasing temperature, whereas the excess molar volumes were totally negative for BHEAA + water system and mostly positive for the BHEAA + MEA system except at higher temperatures, indicating the nature of the interactions between them. Since the binary mixtures could be used as a hybrid solvent for the capture of CO_2 from natural gases, the present data on density and excess molar volumes will certainly be useful for the efficient design of gas−liquid contacting equipment for commercial applications.

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