Densities of Univalent Cation Sulfates in Ethanol + Water Solutions

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The densities of (ammonium, sodium, and potassium) sulfates in ethanol + water solutions were measured in the temperature range of (283.15 to 298.15) K, in saline concentrations from zero to the solubility limit and at atmospheric pressure. The binary and ternary systems were correlated using polynomial expressions, with the selected parameters depending on the composition of the mixtures. Data fitting shows a good agreement with experimental values.

Introduction

The strong importance of electrolytes on aqueous−organic systems is obvious. When a electrolyte is added, strong electrostatic interactions take place, so the solution structure varies and properties of the solution change. This can be used in the engineering design of industrial processes. A lot of papers discuss the influence of electrolytes on liquid−liquid extraction,[1] solubility of nonelectrolytes in water,[2] elimination of azeotropes,[3] vapor−liquid equilibria,[4−6] extractive distillation,[7] antisolvent crytallization,[8] etc.

Considerable effort has been expended to determine and compile physical properties of aqueous electrolytes, so many authors have studied the aqueous properties of electrolytes.[9−11] There are a lot of compilations related to density in the literature.[12−14] But aqueous alcoholic solutions have not been so deeply studied, and density data are even more scarce than aqueous solution data. With regard to the ternary solutions of this paper, we have not been able to find more data in the literature on experimental values for the studied ternary systems, except for data on Na_2SO_4 mixtures related to dilution of the salt[15,16] and data on K_2SO_4 saturated solutions.[17]

This paper is a part of our work on the thermodynamics of the salt effect of electrolytes on the densities and refractive index of ethanol + water mixtures at ambient temperatures. The present paper presents experimental densities of water + ethanol + (ammonium, sodium, or potassium sulfate) mixtures that have been measured as a function of the molality of the salt and the salt-free mass fraction of the solution at atmospheric pressure and from (283.15 to 298.15) K at 5 K intervals. In the sodium sulfate system, because of the important effect of the temperature at 283.15 K, a lot of points are insoluble at this temperature, and so this temperature is rejected.

Experimental Section

Materials. Sodium sulfate (mass fraction >0.99), ammonium sulfate (mass fraction >0.995), and potassium sulfate (mass fraction >0.99) were used in sample preparation. All solids employed were supplied by Merck. The chemicals were recently acquired and stored under sun and humidity protection conditions. The water used to prepare solutions was Milli-Q quality (resistivity, 18.2 MΩ·cm) and was provided by this technical center. The ethanol employed was supplied by Merck (Lichrosolv quality) with a purity better than 0.995, degassed with ultrasound, and stored over freshly activated molecular sieves (type 4a or 3a, 1/16 in., Aldrich Catalog No. 20.860-4 or 20.858-2, respectively) for several days before use. The measured densities of ethanol and the collected densities of water[18] are presented in Table 1.

Stock solutions of concentrated sulfates were prepared by mass, using a Mettler AT261 Delta Range balance, with an uncertain better than $\pm 10^{-4}$ g. They yielded an error better than $\pm 5\cdot10^{-5}$ in the salt-free mass and molar fraction and $\pm 5\cdot10^{-4}$ mol·kg^{-1} in the molality of the salt.

Measurements. The densities of the mixtures were measured with an Anton Paar vibrating-tube DMA-60/602 densimeter with an accuracy bettter than $\pm 10^{-4}$ g·cm^{-3}. Apparatus calibration was realized periodically by testing substances with known density values. A double liquid reference was used for the calibration (Millipore quality water and air). The temperature in the cell was measured by means of a digital precision Anton Paar MKT 100 thermometer, with a precision of $\pm 10^{-3}$ K. A polyscience controller bath model 9010 with a temperature stability of ± 0.01 K was used to thermostat the samples at the desired temperature.

Results and Discussion

In the study of nonelectrolyte systems, excess molar volumes (V^E) are commonly accepted and widely used. In this work, this convention was used for the correlation of the binary ethanol + water mixtures:

$$V^E = \sum_{i=1}^{2} x_i M_i (\rho^{-1} - \rho_i^{-1}) \quad (1)$$

where M_i stands for the molar mass; ρ and ρ_i are the density of the binary mixture and the density of the pure component i, respectively; and x_i is the mole fraction of component i. Densities of the mixture are given in Table 1. The computed excess molar volumes of the binary mixtures were fitted using the following Redlich−Kister expression:[19]

$$V^E = w_i w_j \sum_{p=0}^{S} B_p (w_i - w_j)^p \quad (2)$$

where w_i and w_j are the mass fractions of components i and j, respectively; B_p are fitting parameters; and S is the degree of

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Table 1. Density and Excess Molar Volumes for the Binary Mixtures of Ethanol (1) + Water (2) in the Temperature Range from (283.15 to 298.15) K

		ρ/g·cm^{-3} at t/K				V^E/cm^3·mol^{-1} at t/K			
w_1	x_1	298.15	293.15	288.15	283.15	298.15	293.15	288.15	283.15
0.00000	0.00000	0.99704	0.99820	0.99910	0.99970	0.0000	0.0000	0.0000	0.0000
0.02665	0.01059	0.99213	0.99329	0.99416	0.99482	−0.0410	−0.0385	−0.0353	−0.0335
0.05108	0.02062	0.98796	0.98919	0.99010	0.99075	−0.0855	−0.0820	−0.0770	−0.0722
0.07349	0.03009	0.98440	0.98570	0.98669	0.98745	−0.1320	−0.1277	−0.1217	−0.1166
0.09629	0.04000	0.98101	0.98240	0.98351	0.98442	−0.1848	−0.1801	−0.1739	−0.1689
0.12148	0.05130	0.97736	0.97892	0.98020	0.98130	−0.2465	−0.2426	−0.2369	−0.2330
0.14660	0.06295	0.97384	0.97559	0.97708	0.97839	−0.3118	−0.3095	−0.3052	−0.3026
0.16910	0.07372	0.97072	0.97267	0.97436	0.97590	−0.3727	−0.3721	−0.3696	−0.3691
0.20244	0.09030	0.96602	0.96829	0.97032	0.97221	−0.4636	−0.4667	−0.4678	−0.4710
0.21890	0.09876	0.96364	0.96606	0.96826	0.97036	−0.5082	−0.5134	−0.5164	−0.5223
0.24359	0.11184	0.95993	0.96258	0.96505	0.96743	−0.5739	−0.5817	−0.5880	−0.5976
0.27188	0.12741	0.95544	0.95836	0.96109	0.96379	−0.6452	−0.6568	−0.6663	−0.6802
0.29597	0.14118	0.95135	0.95446	0.95742	0.96035	−0.7011	−0.7152	−0.7278	−0.7444
0.31857	0.15456	0.94734	0.95063	0.95375	0.95687	−0.7504	−0.7669	−0.7811	−0.8003
0.34572	0.17124	0.94226	0.94574	0.94903	0.95238	−0.8038	−0.8231	−0.8391	−0.8613
0.37231	0.18828	0.93713	0.94071	0.94418	0.94766	−0.8529	−0.8729	−0.8912	−0.9144
0.39397	0.20269	0.93269	0.93637	0.93992	0.94356	−0.8862	−0.9074	−0.9259	−0.9514
0.42546	0.22455	0.92616	0.92993	0.93359	0.93739	−0.9325	−0.9540	−0.9728	−1.0001
0.44853	0.24131	0.92117	0.92503	0.92878	0.93258	−0.9602	−0.9828	−1.0023	−1.0276
0.47353	0.26020	0.91575	0.91965	0.92346	0.92736	−0.9893	−1.0115	−1.0307	−1.0569
0.49980	0.28096	0.90990	0.91390	0.91778	0.92170	−1.0143	−1.0378	−1.0570	−1.0819
0.52386	0.30082	0.90453	0.90852	0.91242	0.91648	−1.0357	−1.0575	−1.0755	−1.1025
0.54772	0.32138	0.89905	0.90314	0.90708	0.91112	−1.0507	−1.0744	−1.0917	−1.1164
0.57086	0.34218	0.89376	0.89788	0.90183	0.90593	−1.0643	−1.0877	−1.1035	−1.1284
0.60109	0.37077	0.88675	0.89089	0.89493	0.89906	−1.0765	−1.0991	−1.1152	−1.1389
0.62033	0.38984	0.88224	0.88645	0.89041	0.89464	−1.0804	−1.1045	−1.1163	−1.1421
0.65167	0.42250	0.87482	0.87904	0.88312	0.88734	−1.0809	−1.1035	−1.1171	−1.1403
0.66942	0.44193	0.87068	0.87493	0.87904	0.88322	−1.0813	−1.1043	−1.1176	−1.1379
0.70032	0.47749	0.86328	0.86754	0.87168	0.87594	−1.0701	−1.0918	−1.1041	−1.1251
0.71869	0.49976	0.85898	0.86322	0.86740	0.87165	−1.0647	−1.0847	−1.0973	−1.1162
0.75361	0.54464	0.85055	0.85484	0.85902	0.86332	−1.0369	−1.0575	−1.0671	−1.0858
0.76719	0.56305	0.84723	0.85152	0.85568	0.86006	−1.0216	−1.0416	−1.0491	−1.0700
0.80225	0.61337	0.83856	0.84288	0.84710	0.85145	−0.9687	−0.9885	−0.9956	−1.0129
0.81874	0.63851	0.83445	0.83878	0.84305	0.84744	−0.9375	−0.9568	−0.9658	−0.9833
0.84994	0.68895	0.82663	0.83097	0.83522	0.83959	−0.8662	−0.8853	−0.8903	−0.9050
0.86696	0.71817	0.82235	0.82669	0.83094	0.83536	−0.8202	−0.8389	−0.8426	−0.8589
0.90050	0.77970	0.81364	0.81801	0.82224	0.82669	−0.7006	−0.7198	−0.7197	−0.7357
0.92349	0.82518	0.80739	0.81173	0.81598	0.82040	−0.5870	−0.6039	−0.6036	−0.6168
0.94663	0.87398	0.80098	0.80530	0.80954	0.81394	−0.4513	−0.4665	−0.4640	−0.4748
0.97516	0.93885	0.79282	0.79705	0.80131	0.80569	−0.2434	−0.2523	−0.2495	−0.2570
1.00000	1.00000	0.78522	0.78933	0.79364	0.79793	0.0000	0.0000	0.0000	0.0000

Table 2. Fitting Parameters of Equation 2 and Root Mean Square Deviation (σ) of Equation 6 for the Ethanol (1) + Water (2) System

	298.15 K	293.15 K	288.15 K	283.15 K
B_0	−4.0589	−4.1513	−4.2257	−4.3309
B_1	−1.7716	−1.7839	−1.7278	−1.7057
B_2	−1.2604	−1.2668	−1.1796	−1.1248
B_3	−2.3129	−2.4869	−2.8688	−3.1537
B_4	0.9677	1.3243	1.4270	1.7115
B_5	−0.64715	−0.7367	−0.3972	−0.3459
B_6	−1.8482	−2.3253	−2.2972	−2.6758
σ	0.0015	0.0015	0.0014	0.0017

the polynomial expansion. Table 2 shows adjusted parameters with eq 2. Generally, the Redlich−Kister variable is most frequently the volume fraction or mole fraction. The low solubility of sulfates in ethanol + water mixtures, due to the insolubility of sulfates in ethanol, lead to the use of the ethanol mass fraction rather than the mole fraction. Later in ternary density calculus, both the mass fraction and the mole fraction of ethanol of the mass-free solution are low, but the mass fraction is higher than the mole fraction. So the mass fraction is chosen for the density solvent correlation. Another reason is the extended use of the salt-free mass fraction and concentration of salt to express solubility of a solute in binary solvents.[20−22]

Densities for the salt + water binary mixtures are tabulated in Table 3. These binary data were fitted to polynomials of the form:[23]

$$\rho_{SW} = \rho_W + \sum_{i=1}^{N} A_i m^{(i+1)/2} \qquad (3)$$

Table 3. Density for the Binary Mixtures of Salt + Water as a Function of Molality in the Temperature Range from (283.15 to 298.15) K

m	ρ/g·cm^{-3} at t/K			
mol·kg^{-1}	298.15	293.15	288.15	283.15
	$(NH_4)_2SO_4$ + Water			
0.3027	1.01968	1.02104	1.02214	1.02303
0.6334	1.04200	1.04351	1.04487	1.04595
1.2913	1.08131	1.08303	1.08461	1.08601
1.9176	1.11370	1.11556	1.11726	1.11886
2.6017	1.14448	1.14637	1.14826	1.15000
3.2290	1.16922	1.17111	1.17304	1.17483
3.8967	1.19244	1.19434	1.19629	1.19813
4.5345	1.21212	1.21398	1.21596	1.21784
5.1716	1.22967	1.23152	1.23344	1.23525
5.8906	1.24740			
	K_2SO_4 + Water			
0.0427	1.00300	1.00423	1.00516	1.00583
0.0943	1.01002	1.01129	1.01231	1.01304
0.1740	1.02065	1.02203	1.02314	1.02399
0.2657	1.03269	1.03413	1.03536	1.03631
0.3482	1.04327	1.04477	1.04608	1.04712
0.4416	1.05507	1.05666	1.05806	1.05919
0.5235	1.06520	1.06688	1.06833	1.06952
0.6093	1.07568	1.07738	1.07891	
0.7110	1.08790			
	Na_2SO_4 + Water			
0.1002	1.00975	1.01104	1.01205	1.01281
0.2329	1.02593	1.02737	1.02862	1.02963
0.4700	1.05383	1.05551	1.05702	1.05830
0.6914	1.07890	1.08075	1.08248	1.08401
0.9264	1.10452	1.10662	1.10856	
1.1554	1.12871	1.13086		
1.4004	1.15369			
1.6147	1.17482			
1.8476	1.19710			

Table 4. Fitting Parameters Equation 3 and Root Mean Square Deviation (σ) of Equation 6 for the Salt + Water System

	298.15 K	293.15 K	288.15 K	283.15 K
		$(NH_4)_2SO_4$ + Water		
A_1	$8.14197\cdot10^{-2}$	$8.21206\cdot10^{-2}$	$8.33502\cdot10^{-2}$	$8.46635\cdot10^{-2}$
A_2	$-9.40181\cdot10^{-3}$	$-9.41902\cdot10^{-3}$	$-1.03509\cdot10^{-2}$	$-1.11807\cdot10^{-2}$
A_3	$-5.56910\cdot10^{-3}$	$-5.83563\cdot10^{-3}$	$-5.49766\cdot10^{-3}$	$-5.27245\cdot10^{-3}$
A_4	$1.16846\cdot10^{-3}$	$1.24096\cdot10^{-3}$	$1.18482\cdot10^{-3}$	$1.15473\cdot10^{-3}$
σ	$1\cdot10^{-5}$	$2\cdot10^{-5}$	$2\cdot10^{-5}$	$3\cdot10^{-5}$
		K_2SO_4 + Water		
A_1	$1.41973\cdot10^{-1}$	$1.43347\cdot10^{-1}$	$1.44528\cdot10^{-1}$	$1.46213\cdot10^{-1}$
A_2	$-1.27795\cdot10^{-2}$	$-1.32207\cdot10^{-2}$	$-1.26126\cdot10^{-2}$	$-1.31995\cdot10^{-2}$
A_3	$-4.81059\cdot10^{-3}$	$-5.02539\cdot10^{-3}$	$-6.05459\cdot10^{-3}$	$-6.27254\cdot10^{-3}$
σ	$1\cdot10^{-5}$	$2\cdot10^{-5}$	$2\cdot10^{-5}$	$2\cdot10^{-5}$
		Na_2SO_4 + Water		
	298.15	293.15	288.15	283.15
A_1	$1.30595\cdot10^{-1}$	$1.32047\cdot10^{-1}$	$1.34219\cdot10^{-1}$	$1.37314\cdot10^{-1}$
A_2	$-1.19856\cdot10^{-2}$	$-1.24300\cdot10^{-2}$	$-1.43461\cdot10^{-2}$	$-1.84690\cdot10^{-2}$
A_3	$-3.26071\cdot10^{-3}$	$-3.33995\cdot10^{-3}$	$-2.44148\cdot10^{-3}$	
σ	$2\cdot10^{-5}$	$3\cdot10^{-5}$	$1\cdot10^{-5}$	$4\cdot10^{-5}$

where ρ_W and ρ_{SW} are the density of water and of the salt + water mixture, respectively; m is the molality of the salt in the solution; A_i are the fitting parameters; and N is the number of parameters. These parameters are presented in Table 4.

For the ternary systems of the salt + water + ethanol solutions (Tables 5 to 7), a polynomial expansion[24] similar to that used for the salt + water solutions was used to represent ternary densities:

$$\rho = \rho_{EW} + \sum_{i=1}^{3} C_i m^{(i+1)/2} \quad (4)$$

where ρ and ρ_{EW} are the density of the ternary solution and of ethanol + water solution density and C_i are the fitting parameters. In these cases, these parameters are dependent on the mass fraction of the salt-free system:

$$C_i = A_i + \sum_{j=1}^{3} C_{ij} w_E^j \quad (5)$$

where w_E is the ethanol mass fraction of the salt-free solution; C_{ij} are the adjustable parameters; and A_i are the parameters of the salt + water solutions, corresponding to $w_E = 0$. These parameters are collected from Table 4. In this way, binary systems (salt + water, ethanol + water) are perfectly represented in the equation of the ternary solution. Fitting parameters of ternary solutions are presented in Table 8.

All parameters were obtained using an unweighted least-squares method applying a fitting algorithm due to Marquard.[25] The root mean square deviation at each correlation is enclosed

Table 5. Density for the Ternary Mixtures of Ammonium Sulfate for Various Mass Fractions w_e in Ethanol (w_e) + Water ($1 - w_e$) in the Temperature Range from (283.15 to 298.15) K

	m	ρ/g·cm^{-3} at t/K					m	ρ/g·cm^{-3} at t/K			
w_e	mol·kg^{-1}	298.15	293.15	288.15	283.15	w_e	mol·kg^{-1}	298.15	293.15	288.15	283.15
0.02733	0.6345	1.03688	1.03840	1.03966	1.04074	0.18156	1.4655	1.05806	1.06025	1.06236	1.06436
0.02724	1.2789	1.07535	1.07702	1.07848	1.07988	0.17997	2.1993	1.09288	1.09514	1.09721	1.09921
0.02978	1.9048	1.10724	1.10908	1.11078	1.11233	0.20472	0.4422	0.99594	0.99825	1.00041	1.00245
0.02869	2.5697	1.13756	1.13936	1.14106	1.14271	0.20649	0.8895	1.02255	1.02491	1.02716	1.02928
0.02460	3.1918	1.16316	1.16494	1.16673	1.16843	0.20523	1.3150	1.04589	1.04827	1.05054	1.05269
0.02712	3.7967	1.18415	1.18595	1.18773	1.18947	0.20585	1.7562	1.06749	1.06986	1.07212	1.07439
0.02672	4.4512	1.20500	1.20667	1.20854	1.21029	0.20674	2.1961	1.08701	1.08949	1.09174	1.09396
0.03125	5.0531	1.22119	1.22291	1.22470	1.22643	0.23105	0.6074	1.00171	1.00420	1.00661	1.00894
0.05407	0.6542	1.03336	1.03485	1.03613	1.03735	0.22991	1.2698	1.03878	1.04127	1.04372	1.04608
0.05851	1.3077	1.07113	1.07285	1.07431	1.07577	0.23161	1.7958	1.06384	1.06638	1.06883	1.07121
0.05445	1.9977	1.10708	1.10880	1.11047	1.11206	0.25487	0.3470	0.98155	0.98428	0.98689	0.98941
0.05404	2.6303	1.13541	1.13719	1.13887	1.14056	0.25171	0.6956	1.00328	1.00595	1.00857	1.01105
0.05104	3.2960	1.16213	1.16393	1.16567	1.16744	0.25489	1.0404	1.02175	1.02444	1.02706	1.02964
0.05322	3.9449	1.18431	1.18611	1.18786	1.18966	0.25438	1.3797	1.03926	1.04194	1.04455	1.04710
0.05716	4.5892	1.20366	1.20541	1.20717	1.20893	0.25219	1.7189	1.05583	1.05845	1.06106	1.06351
0.07731	0.5779	1.02453	1.02609	1.02743	1.02865	0.27730	0.4368	0.98313	0.98603	0.98881	0.99154
0.07699	1.1560	1.05949	1.06119	1.06271	1.06415	0.28127	0.8738	1.00753	1.01040	1.01319	1.01592
0.07828	1.7269	1.08958	1.09134	1.09294	1.09458	0.27688	1.3172	1.03132	1.03414	1.03692	1.03961
0.07995	2.3068	1.11649	1.11832	1.12003	1.12172	0.30079	0.3300	0.97219	0.97528	0.97824	0.98114
0.08045	2.8672	1.14014	1.14198	1.14362	1.14546	0.30069	0.6535	0.99130	0.99430	0.99725	1.00013
0.08285	3.4279	1.16095	1.16283	1.16458	1.16641	0.30229	0.9691	1.00822	1.01118	1.01410	1.01696
0.08079	3.9689	1.17998	1.18179	1.18354	1.18545	0.30270	1.2865	1.02412	1.02700	1.02989	1.03271
0.10431	0.5885	1.02085	1.02251	1.02400	1.02534	0.32729	0.3957	0.97094	0.97415	0.97728	0.98038
0.10291	1.1936	1.05711	1.05889	1.06051	1.06208	0.33052	0.8013	0.99331	0.99640	0.99945	1.00249
0.10310	1.7823	1.08789	1.08975	1.09152	1.09317	0.35411	0.2149	0.95460	0.95795	0.96127	0.96454
0.10712	2.3800	1.11484	1.11675	1.11853	1.12036	0.35842	0.4184	0.96596	0.96928	0.97250	0.97576
0.10811	2.9591	1.13854	1.14044	1.14227	1.14410	0.35250	0.6309	0.97924	0.98251	0.98568	0.98884
0.10762	3.5509	1.16054	1.16247	1.16435	1.16620	0.35276	0.8466	0.99075	0.99392	0.99707	1.00022
0.12940	0.7214	1.02528	1.02711	1.02876	1.03024	0.37575	0.2878	0.95448	0.95795	0.96134	0.96471
0.12965	1.4508	1.06647	1.06838	1.07021	1.07187	0.37420	0.3988	0.96150	0.96488	0.96820	0.97150
0.13296	2.1670	1.10062	1.10259	1.10454	1.10628	0.37529	0.5851	0.97178	0.97515	0.97843	0.98171
0.13440	2.6738	1.12210	1.12409	1.12606	1.12787	0.39756	0.1576	0.94215	0.94568	0.94920	0.95270
0.15946	0.4472	1.00329	1.00527	1.00707	1.00868	0.40058	0.3250	0.95148	0.95500	0.95849	0.96194
0.15728	0.8987	1.03137	1.03342	1.03531	1.03703	0.40202	0.4934	0.96066	0.96414	0.96758	0.97101
0.15356	1.3535	1.05717	1.05922	1.06120	1.06298	0.42911	0.1826	0.93674	0.94040	0.94400	0.94762
0.15317	1.7968	1.07957	1.08166	1.08368	1.08553	0.42485	0.3693	0.94872	0.95227	0.95583	0.95939
0.15595	2.2440	1.09961	1.10177	1.10378	1.10569	0.44971	0.0982	0.92719	0.93096	0.93465	0.93837
0.15630	2.6905	1.11837	1.12048	1.12247	1.12441	0.45212	0.2002	0.93272	0.93646	0.94013	0.94382
0.18018	0.7306	1.01774	1.01988	1.02191	1.02378	0.44962	0.2973	0.93891	0.94259	0.94623	0.94985

Table 6. Density for the Ternary Mixtures of Potassium Sulfate for Various Mass Fractions w_e in Ethanol (w_e) + Water ($1 - w_e$) in the Temperature Range from (283.15 to 298.15) K

	m	ρ/g·cm^{-3} at t/K					m	ρ/g·cm^{-3} at t/K			
w_e	mol·kg^{-1}	298.15	293.15	288.15	283.15	w_e	mol·kg^{-1}	298.15	293.15	288.15	283.15
0.02598	0.0887	1.00439	1.00570	1.00670	1.00744	0.10244	0.2003	1.00653	1.00818	1.00955	1.01074
0.02505	0.1728	1.01575	1.01713	1.01819	1.01904	0.10757	0.2563	1.01287	1.01461		
0.02565	0.2663	1.02778	1.02924	1.03038	1.03135	0.13046	0.0477	0.98256	0.98427	0.98567	0.98690
0.02551	0.3489	1.03826	1.03979	1.04102	1.04207	0.12949	0.1008	0.98963	0.99140	0.99283	0.99412
0.03168	0.4328	1.04767	1.04933	1.05064	1.05176	0.12887	0.1515	0.99633	0.99815	0.99962	1.00095
0.02785	0.5226	1.05948	1.06111			0.13398	0.1962	1.00129	1.00314		
0.05178	0.0715	0.99762	0.99893	0.99993	1.00076	0.14489	0.0423	0.97973	0.98156	0.98305	0.98440
0.04956	0.1396	1.00711	1.00848	1.00954	1.01041	0.14714	0.0790	0.98426	0.98616	0.98770	0.98913
0.05183	0.2111	1.01604	1.01751	1.01865	1.01958	0.14723	0.1138	0.98873	0.99069	0.99226	0.99371
0.04940	0.2778	1.02501	1.02657	1.02774	1.02877	0.14742	0.1557	0.99414	0.99607		
0.05076	0.3464	1.03345	1.03504	1.03629	1.03738	0.17343	0.0421	0.97572	0.97781	0.97955	0.98118
0.05210	0.4263	1.04317	1.04478			0.17224	0.0795	0.98075	0.98284	0.98462	0.98628
0.07248	0.0649	0.99341	0.99483	0.99589	0.99681	0.17296	0.1203	0.98589	0.98805	0.98986	
0.07285	0.1289	1.00185	1.00331	1.00441	1.00538	0.19875	0.0322	0.97085	0.97317	0.97516	0.97707
0.07173	0.1936	1.01048	1.01199	1.01318	1.01419	0.19407	0.0670	0.97601	0.97834	0.98029	0.98219
0.07422	0.2611	1.01881	1.02041	1.02166	1.02276	0.19762	0.0942	0.97905	0.98132		
0.08031	0.3230	1.02557	1.02720			0.22287	0.0494	0.96950	0.97205	0.97430	0.97650
0.10493	0.0516	0.98675	0.98831	0.98950	0.99055	0.24572	0.0294	0.96337	0.96615	0.96862	0.97104
0.10430	0.0995	0.99320	0.99483	0.99606	0.99709	0.24996	0.0580	0.96650	0.96922		
0.10577	0.1520	0.99982	1.00148	1.00280	1.00396						

Table 7. Density for the Ternary Mixtures of Sodium Sulfate for Various Mass Fractions w_e in Ethanol (w_e) + Water ($1 - w_e$) in the Temperature Range from (283.15 to 298.15) K

	m	ρ/g·cm^{-3} at t/K				m	ρ/g·cm^{-3} at t/K			
w_e	mol·kg^{-1}	298.15	293.15	288.15	w_e	mol·kg^{-1}	298.15	293.15	288.15	
0.02555	0.2493	1.02305	1.02447	1.02570	0.15327	0.1224	0.98771	0.98967	0.99137	
0.02703	0.5028	1.05234	1.05397	1.05547	0.15049	0.2428	1.00209	1.00414	1.00597	
0.02705	0.7564	1.08055	1.08240	1.08410	0.15435	0.3672	1.01551	1.01769		
0.02830	1.0114	1.10764	1.10960		0.15209	0.5157	1.03220			
0.02695	1.2438	1.13200	1.13411		0.15080	0.7002	1.05214			
0.02554	1.5062	1.15847			0.17898	0.1245	0.98417	0.98632	0.98832	
0.02632	1.7515	1.18190			0.17982	0.2430	0.99760	0.99989	1.00202	
0.05236	0.2045	1.01293	1.01441	1.01562	0.16732	0.3405	1.01050	1.01273		
0.04767	0.4108	1.03796	1.03963	1.04106	0.17384	0.3651	1.01224	1.01450		
0.04850	0.6149	1.06086	1.06264	1.06424	0.18029	0.4872	1.02429			
0.05378	0.8174	1.08204	1.08391	1.08571	0.20441	0.1018	0.97777	0.98011	0.98237	
0.05024	1.0219	1.10433	1.10633		0.20810	0.2033	0.98875	0.99128	0.99360	
0.05194	1.2239	1.12474			0.20267	0.3049	1.00084	1.00336		
0.05281	1.5582	1.15767			0.20345	0.4008	1.01113			
0.07798	0.2109	1.00950	1.01103	1.01236	0.20007	0.5049	1.02280			
0.07508	0.4216	1.03443	1.03614	1.03766	0.22939	0.1020	0.97397	0.97655	0.97903	
0.07786	0.6329	1.05755	1.05941	1.06113	0.23033	0.2031	0.98513	0.98783	0.99038	
0.07739	0.8459	1.08063	1.08265		0.22720	0.3002	0.99620			
0.08006	1.0530	1.10175			0.25375	0.0831	0.96790	0.97075	0.97342	
0.07649	1.3006	1.12746			0.25291	0.1594	0.97656	0.97943		
0.10377	0.1646	1.00000	1.00166	1.00306	0.25349	0.2321	0.98432			
0.10359	0.3298	1.01930	1.02108	1.02265	0.24891	0.3163	0.99423			
0.10339	0.5403	1.04302	1.04497	1.04665	0.27835	0.0621	0.96140	0.96442	0.96735	
0.10260	0.6616	1.05638	1.05840		0.27174	0.0672	0.96317	0.96612	0.96885	
0.10322	0.8258	1.07379			0.27647	0.1262	0.96880	0.97184	0.97477	
0.10326	1.0632	1.09830			0.27934	0.1923	0.97545			
0.12713	0.1665	0.99660	0.99851	1.00006	0.30093	0.0555	0.95659	0.95977	0.96287	
0.12691	0.3319	1.01584	1.01783	1.01954	0.30281	0.1061	0.96186	0.96508	0.96821	
0.12591	0.5016	1.03502	1.03706		0.30316	0.1587	0.96743			
0.12905	0.6626	1.05197			0.29802	0.2236	0.97526			
0.12519	0.8711	1.07460								

as a measurement of the validity of the gathered fitting parameters, and the equation applied is expressed by

$$\sigma = \sqrt{\frac{\sum_{i}^{n} (z_{\text{experimental}} - z_{\text{predicted}})^2}{n - p}} \qquad (6)$$

In this equation the value of the property, the number of experimental data, and the number of parameters are represented by z, n, and p, respectively.

For the binary and ternary salt systems measured, the values of the density increased with concentration of the salt at constant salt-free mass fraction and temperature. At constant composition, the density decreased with an increase in temperature. At constant molality of the salt and temperature, the increase in the salt-free mass fraction of ethanol led to a decrease in the densities.

Table 8. Fitting Parameters of Equations 4 and 5 and Standard Deviation (σ) of Equation 6 for the Salt + Ethanol + Water Ternary Systems

	298.15 K	293.15 K	288.15 K	283.15 K
		$(NH_4)_2SO_4$ + Ethanol + Water		
C_{11}	$-7.02782\cdot10^{-3}$	$-7.28346\cdot10^{-3}$	$-4.59073\cdot10^{-3}$	$-2.90400\cdot10^{-3}$
C_{12}	$-1.45176\cdot10^{-1}$	$-1.66084\cdot10^{-1}$	$-2.10859\cdot10^{-1}$	$-2.44355\cdot10^{-1}$
C_{13}	$1.64513\cdot10^{-1}$	$1.86849\cdot10^{-1}$	$2.49136\cdot10^{-1}$	$2.73642\cdot10^{-1}$
C_{21}	$-1.71686\cdot10^{-3}$	$-2.70710\cdot10^{-3}$	$-7.85266\cdot10^{-3}$	$-9.84420\cdot10^{-3}$
C_{22}	$6.48585\cdot10^{-2}$	$7.35381\cdot10^{-2}$	$1.25605\cdot10^{-1}$	$1.36207\cdot10^{-1}$
C_{23}	$-3.26390\cdot10^{-2}$	$-1.63876\cdot10^{-2}$	$-7.83337\cdot10^{-2}$	$-3.31991\cdot10^{-2}$
C_{31}	$2.93081\cdot10^{-3}$	$3.04168\cdot10^{-3}$	$4.59132\cdot10^{-3}$	$4.88330\cdot10^{-3}$
C_{32}	$-6.18570\cdot10^{-3}$	$-4.66433\cdot10^{-4}$	$-1.30977\cdot10^{-2}$	$-7.74154\cdot10^{-3}$
C_{33}	$-7.12989\cdot10^{-2}$	$-1.01204\cdot10^{-1}$	$-9.79081\cdot10^{-2}$	$-1.38782\cdot10^{-1}$
σ	$5\cdot10^{-5}$	$5\cdot10^{-5}$	$6\cdot10^{-5}$	$7\cdot10^{-5}$
		K_2SO_4 + Ethanol + Water		
C_{11}	$7.51021\cdot10^{-2}$	$5.87778\cdot10^{-2}$	$1.24097\cdot10^{-1}$	$1.55135\cdot10^{-1}$
C_{12}	-1.39824	-1.09597	-1.52326	-1.97275
C_{13}	2.97421	2.65165	3.19682	4.16579
C_{21}	$-3.84825\cdot10^{-1}$	$-3.44654\cdot10^{-1}$	$-6.37317\cdot10^{-1}$	$-8.02542\cdot10^{-1}$
C_{22}	4.65418	4.43843	5.56203	7.98676
C_{23}	-4.94995	-8.60780	-8.48082	$-1.16727\cdot10^{1}$
C_{31}	$2.72618\cdot10^{-1}$	$2.47801\cdot10^{-1}$	$4.91226\cdot10^{-1}$	$6.32578\cdot10^{-1}$
C_{32}	-1.83175	-2.21683	-1.33032	-2.60003
C_{33}	$-1.33187\cdot10^{1}$	-7.52937	$-1.53395\cdot10^{1}$	$-2.54223\cdot10^{1}$
σ	$4\cdot10^{-5}$	$4\cdot10^{-5}$	$4\cdot10^{-5}$	$4\cdot10^{-5}$
		Na_2SO_4 + Ethanol + Water		
C_{11}	$-6.25687\cdot10^{-3}$	$1.87055\cdot10^{-2}$	$1.32234\cdot10^{-2}$	
C_{12}	$-2.26232\cdot10^{-1}$	$-4.81838\cdot10^{-1}$	$-4.26611\cdot10^{-1}$	
C_{13}	$2.20135\cdot10^{-1}$	$6.12006\cdot10^{-1}$	$3.49005\cdot10^{-1}$	
C_{21}	$-3.16479\cdot10^{-2}$	$-1.12801\cdot10^{-1}$	$-9.70512\cdot10^{-2}$	
C_{22}	$2.40758\cdot10^{-1}$	1.03596	$8.39135\cdot10^{-1}$	
C_{23}	$-2.37571\cdot10^{-1}$	$-9.52032\ 10^{-1}$	$3.87111\cdot10^{-1}$	
C_{31}	$1.18132\cdot10^{-2}$	$5.58340\ 10^{-2}$	$3.88144\cdot10^{-2}$	
C_{32}	$-3.55686\cdot10^{-2}$	$-3.06821\ 10^{-1}$	$-2.96637\cdot10^{-2}$	
C_{33}	$-4.01771\cdot10^{-1}$	-1.72036	-3.90283	
σ	$4\cdot10^{-5}$	$4\cdot10^{-5}$	$4\cdot10^{-5}$	

Conclusions

This paper reports experimental data for the density of ammonium, sodium, and potassium sulfate in ethanol + water mixtures from (283.15 to 298.15) K. These data were correlated by the method of least-squares to polynomials. For all systems, the calculated values are in good agreement with the experimental values presented in the tables, at all temperatures and range of salt concentrations. The density solutions are described adequately by these equations, and these polynomials can be used for the density prediction to salt saturation.

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