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Vapor–Liquid Equilibria, Density, Speed of Sound, and Viscosity of Aqueous Dipotassium Tartrate Solutions at $T = (298.15, 308.15, \text{and } 318.15)$ K

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The vapor–liquid equilibria, density, speed of sound, and viscosity of aqueous dipotassium tartrate solutions at $T = (298.15, 308.15, \text{and } 318.15)$ K have been measured. The experimental vapor–liquid equilibria data were correlated with the Pitzer equation. The experimental density and speed of sound data for dipotassium tartrate were used to obtain apparent molar volume and apparent molar compressibility values at different temperatures, and these data were satisfactorily fitted to the corresponding Pitzer equations. The limiting apparent molar volume and limiting apparent molar compressibility at each temperature were obtained from the Redlich−Mayer and an abbreviated Pitzer model. The viscosity data were fitted to an empirical equation.

Introduction

Tartrate ions in aqueous solutions are of considerable significance in many biochemical and chemical processes.[1] With respect to these solutions, conductivity studies[2,3] have been carried out on some alkali metal and ammonium tartrates in the temperature range of (278.15 to 308.15) K. Vapor pressures of saturated solutions for some tartrate salts have been measured.[4,5] However, with respect to other thermodynamic or transport properties of tartrate salts, little information can be found in the literature. There are reports on the partial molar volume of aqueous potassium tartrate,[6] in that no experimental density data have been given, and densities or apparent volumes of aqueous solutions of other tartrate salts.[7,8] Recently, we reported the refractive index, viscosity, density, and speed of sound of aqueous solutions of disodium tartrate at (298.15, 308.15, and 318.15) K.[9] This work is the continuation of our research work on the thermodynamic properties of aqueous electrolyte solutions. Here, the vapor–liquid equilibria, viscosity, density, and speed of sound of aqueous dipotassium tartrate solutions at $T = (298.15, 308.15, \text{and } 318.15)$ K are presented. As far as we know, there are no data for the aforementioned physical properties of aqueous dipotassium tartrate solutions in the literature at different temperatures. There is only one report on the osmotic coefficient and activity coefficient of aqueous dipotassium tartrate solutions at the freezing temperature.[10] The values of osmotic coefficients at other temperatures are unknown.

In this work, vapor–liquid equilibria, viscosity, density, and speed of sound measurements were performed from dilute up to near saturation at the pressure 0.1 MPa, and to calculate accurate limiting apparent volumes and compressibilities, enough data were obtained in the dilute region (below 0.1 $\text{mol}\cdot\text{kg}^{-1}$) at three temperatures. The appropriate equations were used to correlate the measured physical properties.

Experimental Section

Materials. Potassium tartrate with a purity of minimum mass fraction of 0.995 was obtained from Merck. The salt was used without further purification, and double-distilled–deionized water was used. The solutions were prepared by mass using an analytical balance (Shimatzu, 321-34553, Shimatzu Co., Japan) with an uncertainty of $\pm 1\cdot10^{-7}$ kg.

Apparatus and Procedure. In this study, the isopiestic method is used to obtain the activity of water and the osmotic coefficient in an aqueous dipotassium tartrate system at $T = (298.15, 308.15, \text{and } 318.15)$ K. The isopiestic apparatus used in this work was similar to the one used by Ochs et al.[11] This apparatus consisted of a five-leg manifold attached to round-bottom flasks. Two flasks contained the standard pure NaCl solutions; two flasks contained dipotassium tartrate solutions; and the central flask was used as a water reservoir. The apparatus was kept in a constant temperature bath for at least 120 h to achieve equilibrium. The temperature was controlled to within ± 0.005 K by a Heto temperature controller (Hetotherm PF, Heto Laboratory Equipment, Denmark). After equilibrium, the manifold assembly was removed from the bath, and each flask was weighed by an analytical balance (Shimatzu, 321-34553, Shimatzu Co., Japan) with an uncertainty of $\pm 1\cdot10^{-7}$ kg. From the weight of each flask and the initial weight of salt, the mass fraction of each solution was calculated. It was assumed that the equilibrium condition was reached when the differences between the mass fractions of each duplicate were less than 1 %. In all cases, averages of the duplicate are reported as the total isopiestic mass fraction. The water activity for the standard aqueous NaCl solutions at different concentrations and temperatures has been calculated from the correlation of Colin et al.[12] The uncertainty in the measurement of water activity was estimated to be ± 0.0002.

Densities and speed of sounds were measured with a vibrating-tube densimeter (Antoine Parr DSA-500, Austria). By this apparatus, the working temperature can be controlled within ± 0.001 K. The apparatus was calibrated at each temperature with double-distilled–deionized water and dry air. For pure water, the values (997.044, 994.028, and 990.221) $\text{kg}\cdot\text{m}^{-3}$ for density and (1497.08, 1520.08, and 1536.61) $\text{m}\cdot\text{s}^{-1}$ for ultrasonic velocity were obtained, respectively, at $T = (298.15, 308.15, \text{and } 318.15)$ K. The apparatus was also tested with the density of a known molality of aqueous NaCl using the data given by Pitzer et al.[13] The uncertainty in the measurement of

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Table 1. Isopiestic Equilibrium Molalities, Experimental Osmotic Coefficients, Activities, and Vapor Pressures for Potassium Tartrate (1) +H_2O (2) at Different Temperatures

m_{NaCl} / $(mol \cdot kg^{-1})$	m_1 / $(mol \cdot kg^{-1})$	φ_{exptl}	a_s	P_{exptl} / (kPa)
		$T = 298.15$ K		
0.0000	0.0000	1.000	1.0000	3.173
0.4209	0.3635	0.713	0.9861	3.129
0.6665	0.6040	0.683	0.9779	3.103
0.8168	0.7532	0.675	0.9729	3.087
1.3639	1.2852	0.677	0.954	3.027
1.7990	1.7020	0.691	0.9384	2.977
1.9497	1.8606	0.691	0.9329	2.960
2.3109	2.2119	0.704	0.9193	2.917
2.4832	2.3787	0.711	0.9127	2.896
2.5428	2.4657	0.705	0.9104	2.888
2.9170	2.8375	0.719	0.8956	2.841
3.2915	3.2361	0.728	0.8804	2.793
4.1768	4.1683	0.759	0.8429	2.674
4.3912	4.3793	0.77	0.8335	2.644
4.5934	4.6313	0.771	0.8245	2.615
4.7731	4.8447	0.775	0.8164	2.590
6.2815	6.7200	0.805	0.7464	2.368
6.3753	6.8692	0.804	0.7419	2.353
		$T = 308.15$ K		
0.0000	0.0000	1.000	1.0000	5.575
0.3858	0.3282	0.721	0.9873	5.504
0.8142	0.7152	0.706	0.9731	5.425
1.0316	0.9198	0.702	0.9657	5.383
1.2426	1.1349	0.692	0.9584	5.343
1.4434	1.3275	0.694	0.9514	5.303
1.9852	1.8536	0.705	0.9318	5.194
2.3452	2.2152	0.712	0.9183	5.119
2.9504	2.8422	0.725	0.8947	4.987
3.6820	3.5849	0.751	0.8646	4.819
4.2194	4.1767	0.764	0.8415	4.690
4.9654	5.0470	0.780	0.8084	4.505
5.6089	5.8361	0.793	0.7788	4.340
6.1067	6.4352	0.806	0.7554	4.209
		$T = 318.15$ K		
0.0000	0.0000	1.000	1.0000	9.508
0.2979	0.2468	0.741	0.9902	9.414
0.4175	0.3446	0.745	0.9862	9.377
0.6412	0.5504	0.721	0.9788	9.306
0.7094	0.6181	0.712	0.9765	9.284
1.0933	0.9764	0.707	0.9634	9.159
1.3958	1.2757	0.702	0.9527	9.057
1.6759	1.5360	0.712	0.9426	8.961
2.1494	1.9954	0.723	0.9250	8.793
2.7913	2.6463	0.737	0.9000	8.555
3.0947	2.9341	0.751	0.8877	8.438
4.2241	4.1535	0.777	0.8399	7.982
4.8070	4.8126	0.791	0.8140	7.735
5.5732	5.7492	0.804	0.7789	7.401

Table 2. Pitzer Model Parameters and Standard Deviationes, σ (φ) and σ (P), for Potassium Tartrate (1) + H_2O (2) at Different Temperatures

T / K	$\beta^{(0)}$	$\beta^{(1)}$	C_φ	σ (φ)	σ (P) / (kPa)
298.15	0.07202	0.64313	−0.00344	0.004	0.002
308.15	0.07777	1.07280	−0.00400	0.006	0.005
318.15	0.09016	1.17597	−0.00532	0.005	0.061

density and speed of sound was estimated to be ± 0.003 $kg \cdot m^{-3}$ for density and 0.1 $m \cdot s^{-1}$ for ultrasonic velocity.

The viscosities of dipotassium tartrate solutions were determined with a suspended Ubbelohde-type viscometer (Julabo, MD-18V, Germany) at $T = (288.15 \text{ to } 328.15)$ K in which the temperature was controlled with a precision of 0.01 K. The flow times were measured using a stopwatch. The precision of the used stopwatch was ± 0.01 s. The dynamic viscosity, η, was calculated by the following relation

$$\eta = dK(t - \theta) \tag{1}$$

where t is the flow time; K is the viscometer constant; θ is the Hagenbach correction factor; and d is the density. The viscometer constant, K, is determined by calibrating at working temperatures with distilled water using viscosity values from the literature.[14] The uncertainty for the dynamic viscosity determination was estimated to be ± 0.5 %. Each measurement was repeated five times.

Results and Discussion

Isopiestic Results. Isopiestic equilibrium molalities with reference standard solutions of NaCl in water as reported in Table 1 enabled the calculation of the osmotic coefficient, Φ, of the solutions of dipotassium tartrate in water from

$$\varphi = \frac{\nu^* \varphi^* m^*}{\nu m_1} \tag{2}$$

where ν^* and ν are the sum of stoichiometric numbers of the anion and cation in the reference solution and the solution of dipotassium tartrate, respectively; m_1 is the molality of the dipotassium tartrate solution; m^* is the molality of the reference standard in isopiestic equilibrium with this solution; and φ^* is the osmotic coefficient of the isopiestic reference standard, calculated at m^*. The necessary φ^* values at any m^* at different temperatures were calculated from the correlation given by Colin et al.[12] From the calculated osmotic coefficient data, the activity of water in dipotassium solution and the vapor pressure of this solution were determined at isopiestic equilibrium molalities, with the help of the following relations

$$\varphi = -\frac{\ln a_s}{\nu m_1 M_2} \tag{3}$$

$$\ln a_s = \ln\left(\frac{p}{p^*}\right) + \frac{(B - V_s^*)(p - p^*)}{RT} \tag{4}$$

where a_s is the activity of water; M_2 is the molar mass of the water; and B is the second virial coefficient of water vapor. V_s* is the molar volume, and p* is the vapor pressure of pure water. The second virial coefficients of water vapor at each

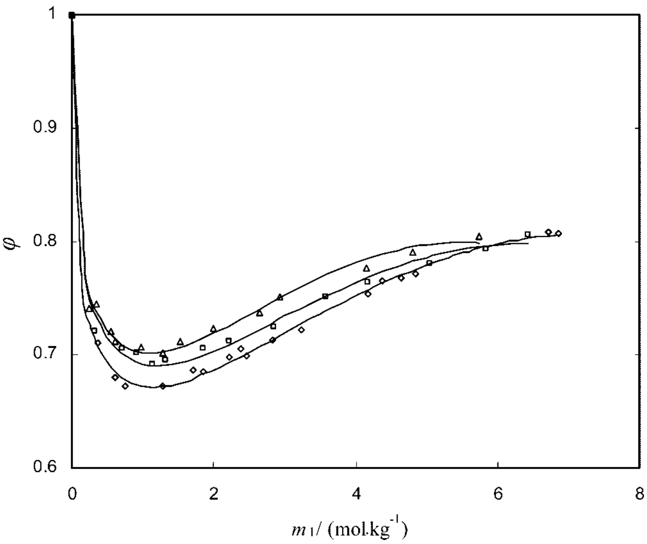

Figure 1. Experimental and calculated osmotic coefficient, φ, plotted against molality of salt, m_1, for the potassium tartrate (1) + H_2O (2) system studied with the Pitzer model: ◇, 298.15 K; □, 308.15 K; △, 318.15 K; −, Pitzer model.

Table 3. Experimental Density, d, and Speed of Sound, u, Data for Potassium Tartrate (1) + H_2O (2) at Different Temperatures

m_1 (mol·kg^{-1})	$10^{-3}\cdot d$ (kg·m^{-3})	u (m·s^{-1})	$10^6\cdot V_\varphi$ (m^3·mol^{-1})	$10^{10}\cdot\kappa_s$ Pa^{-1}	$10^{14}\cdot\kappa_\varphi$ (m^3·mol·Pa^{-1})
		T = 298.15 K			
0.0398	1.002883	1502.55	79.09	4.42	−11.20
0.0498	1.004290	1503.74	79.97	4.40	−10.89
0.0615	1.005993	1505.42	79.85	4.39	−10.99
0.0697	1.007177	1506.52	79.80	4.37	−10.99
0.0797	1.008558	1508.75	80.74	4.36	−11.49
0.0897	1.010006	1509.47	80.56	4.35	−10.99
0.0996	1.011422	1510.30	80.52	4.33	−10.66
0.3000	1.039015	1535.65	82.73	4.08	−9.79
0.4985	1.064812	1559.54	84.44	3.86	−9.09
0.6922	1.088799	1582.17	85.71	3.67	−8.54
0.8934	1.112364	1605.24	87.03	3.49	−8.04
1.2845	1.155123	1648.29	89.03	3.19	−7.22
1.5788	1.185123	1680.48	90.11	2.99	−6.75
1.9646	1.221196	1718.34	91.58	2.77	−6.15
2.4427	1.262588	1763.99	92.86	2.55	−5.56
2.9234	1.299599	1805.25	94.24	2.36	−5.03
3.3913	1.331798	1841.49	95.56	2.21	−4.57
3.8581	1.362454	1876.19	96.36	2.09	−4.20
4.3238	1.389867	1907.02	97.24	1.98	−3.87
4.7850	1.414660	1935.37	98.07	1.89	−3.57
5.2381	1.437729	1960.58	98.69	1.81	−3.32
5.6922	1.459068	1984.76	99.29	1.74	−3.09
		T = 308.15 K			
0.0398	0.999780	1541.43	81.08	4.30	−10.21
0.0498	1.001178	1542.57	81.72	4.29	−9.94
0.0615	1.002846	1543.94	81.79	4.27	−9.94
0.0697	1.004013	1544.95	81.74	4.26	−9.94
0.0797	1.005386	1546.71	82.51	4.25	−10.21
0.0897	1.006800	1547.66	82.49	4.23	−9.99
0.0996	1.008210	1548.32	82.31	4.22	−9.67
0.3000	1.035425	1570.49	84.46	3.99	−8.88
0.4985	1.060927	1591.56	86.02	3.78	−8.25
0.6922	1.084647	1611.60	87.19	3.60	−7.77
0.8934	1.107996	1631.93	88.39	3.43	−7.32
1.2845	1.150412	1670.11	90.22	3.15	−6.60
1.5788	1.180157	1698.69	91.23	2.96	−6.18
1.9646	1.216053	1732.59	92.58	2.76	−5.63
2.4427	1.257213	1773.43	93.76	2.54	−5.10
2.9234	1.294090	1810.80	95.06	2.36	−4.62
3.3913	1.326153	1843.62	96.33	2.22	−4.20
3.8581	1.356741	1875.29	97.07	2.09	−3.87
4.3238	1.384056	1903.71	97.92	1.99	−3.56
4.7850	1.408825	1929.52	98.71	1.90	−3.29
5.2381	1.431813	1953.35	99.31	1.82	−3.05
5.6922	1.453123	1975.52	99.88	1.75	−2.84
		T = 318.15 K			
0.0398	1541.43	0.995909	82.47	4.23	−9.34
0.0498	1542.57	0.997282	83.29	4.21	−9.34
0.0615	1543.94	0.998942	83.15	4.20	−9.23
0.0697	1544.95	1.000092	83.16	4.19	−9.34
0.0797	1546.71	1.001442	84.02	4.17	−9.54
0.0897	1547.66	1.002857	83.79	4.16	−9.34
0.0996	1548.32	1.004238	83.75	4.15	−9.02
0.3000	1570.49	1.031194	85.68	3.93	−8.25
0.4985	1591.56	1.056452	87.17	3.74	−7.69
0.6922	1611.60	1.079979	88.26	3.57	−7.24
0.8934	1631.93	1.103168	89.38	3.40	−6.83
1.2845	1670.11	1.145296	91.11	3.13	−6.16
1.5788	1698.69	1.174959	92.01	2.95	−5.78
1.9646	1732.59	1.210586	93.34	2.75	−5.27
2.4427	1773.43	1.251594	94.45	2.54	−4.78
2.9234	1810.80	1.288337	95.70	2.37	−4.33
3.3913	1843.62	1.320321	96.93	2.23	−3.94
3.8581	1875.29	1.350826	97.63	2.11	−3.63
4.3238	1903.71	1.378097	98.45	2.00	−3.34
4.7850	1929.52	1.402829	99.22	1.91	−3.09
5.2381	1953.35	1.425776	99.81	1.84	−2.87
5.6922	1975.52	1.447055	100.36	1.77	−2.67

working temperature were calculated using the equation provided by Rard and Platford.[15] Molar volumes of liquid water were calculated using the density of water at different temperatures.[16] The vapor pressures of pure water were calculated using the equation of state of Saul and Wagner.[17] After the establishment of isopiestic equilibrium, water activities were calculated using eq 3. The results are also collected in Table 1. The values of p for aqueous dipotassium tartrate calculated using eq 4 are also given in Table 1.

In this work, for the correlation of osmotic coefficient data of aqueous dipotassium tartrate solutions, we used the ion-interaction model of Pitzer.[18] The Pitzer model has the following form for a binary 1:2 electrolyte solution[18]

$$\varphi - 1 = -2\left(\frac{A_\varphi I^{0.5}}{1 + bI^{0.5}}\right) + \frac{4}{3}m_1 B^\varphi + 2\frac{(2)^{\frac{3}{2}}}{3}m_1^2 C^\varphi \qquad (5a)$$

where

$$B^\varphi = \beta^{(0)} + \beta^{(1)}\exp(-\alpha_1\sqrt{I}) \qquad (5b)$$

$$A_\Phi = \frac{1}{3}\sqrt{2\pi N_A d_0}\left(\frac{e^2}{4\pi\epsilon_0 DkT}\right) \qquad (5c)$$

In these equations, $\beta^{(0)}$, $\beta^{(1)}$, and C^φ are Pitzer's ion-interaction parameters obtained by fitting of experimental data; α and b are adjustable parameters; and d_0, N_A, e, k, and ϵ_0 are the density of water, the Avogadro number, the electronic charge, Boltzmann's constant, and the permittivity of the vacuum, respectively. I is the ionic strength on the molality basis. For adjustable parameters, the values $\alpha_1 = 2.0$ kg$^{1/2}$·mol$^{-1/2}$ and $b = 1.2$ kg$^{1/2}$·mol$^{-1/2}$ were used. Table 2 summarizes the Pitzer ion-interaction parameters obtained from fitting of the experimental osmotic coefficient data to the Pitzer model. In Figure 1, the experimental and calculated osmotic coefficients calculated from the Pitzer model are plotted against dipotassium molalities at different temperatures. On the basis of standard deviations reported in Table 2 and Figure 1, we conclude that the Pitzer model can be satisfactorily used for the correlation of the osmotic coefficient of disodium tartarte solutions from dilute to near saturation.

Volumetric and Isentropic Compressibility Results. The experimental density d and speed of sound u values as a function of dipotassium tartrate concentration m_1 at three temperatures are reported in Table 3. The apparent molar volumes V_φ of aqueous dipotassium tartrate solutions were calculated from the densities of the solutions d using the following equation

$$V_\varphi = \frac{M_1}{d} - \frac{(d - d_0)}{m_1 d d_0} \qquad (6)$$

where M_1 is the molar mass of dipotassium salt. The calculated V_φ values are also given in Table 3.

Using the speed of sound and density values reported in Table 3, the isentropic compressibilities, k_s, were calculated from the Laplace-Newton equation:

$$\kappa_s = (du^2)^{-1} \qquad (7)$$

The apparent molar isentropic compressibility, κ_φ, is another thermodynamic property of the solution that can be regarded as the extent to which water of hydration around the solute molecule can be compressed. Solutions become less compressible as κ_φ values become more negative. The apparent molar isentropic compressibility is computed from the relation

$$\kappa_\varphi = \frac{(\kappa_s d_0 - \kappa_{s0} d)}{m_1 d d_0} + \frac{\kappa_s M_1}{d} \qquad (8)$$

where κ_{s0} and κ_s are the isentropic compressibility of pure water and the mixture, respectively. The calculated isentropic compressibility, κ_s, and apparent molar isentropic compressibility, κ_φ, values for the aqueous dipotassium tartrate solutions at different temperatures are also given in Table 3.

For evaluation of the limiting apparent molar volume, $V_\varphi{}^0$ (equal to the partial molar volume at infinite dilution) of potassium tartrate in water solutions, the V_φ values in the dilute

Table 4. Coefficients of the Redlich−Mayer (Equation 9) Equation and an Abbreviated Form of the Pitzer (Equation 15) Equations and Standard Deviations, σ (V_φ), for Potassium Tartrate (1) + H_2O (2) at Different Temperatures

T	$10^6 \cdot V_\varphi^{0\,a}$			$10^6 \cdot \sigma\ (V_\varphi^0)^a$	$10^6 \cdot V_\varphi^{0\,b}$	$10^9 \cdot \beta_v^0$	$10^6 \cdot \sigma\ (V_\varphi^0)^b$
K	($m^3 \cdot mol^{-1}$)	$10^6 \cdot S_v$	$10^6 \cdot b_v$	($m^3 \cdot mol^{-1}$)	($m^3 \cdot mol^{-1}$)	($kg \cdot mol^{-1} \cdot Pa^{-1}$)	($m^3 \cdot mol^{-1}$)
298.15	77.24	9.74	4.13	0.27	77.29	−1.078	0.27(7)[c]
308.15	79.10	10.68	0.84	0.21	79.16	−0.785	0.21(7)[c]
318.15	80.44	11.74	−2.03	0.25	80.52	−0.547	0.25(7)[c]

[a] Obtained from the Redlich−Mayer equation. [b] Obtained from an abbreviated form of the Pitzer model (eq 15). [c] The number of experimental data used in the calculation of parameters of the Redlich−Mayer and an abbreviated form of the Pitzer model.

Table 5. Coefficients of the Redlich−Mayer (Equation 12) Equation and the Abbreviated Form of the Pitzer (Equation 15) Equation and Standard Deviations, σ (κ_φ) for Potassium Tartrate (1) + H_2O (2) at Different Temperatures

T	$10^{14} \cdot \kappa_\varphi^{0\,a}$			$10^{14} \cdot \sigma(\kappa_\varphi)^a$	$10^{14} \cdot \kappa_\varphi^{0\,b}$	$10^{17} \cdot \beta_\kappa^0$	$10^{14} \cdot \sigma(\kappa_\varphi)^b$
K	($m^3 \cdot mol^{-1} \cdot Pa^{-1}$)	$10^{14} \cdot S_\kappa$	$10^{14} \cdot b_\kappa$	($m^3 \cdot mol^{-1} \cdot Pa^{-1}$)	($m^3 \cdot mol^{-1} \cdot Pa^{-1}$)	($m^3 \cdot mol^{-1} \cdot Pa^{-2}$)	($m^3 \cdot mol^{-1} \cdot Pa^{-1}$)
298.15	−11.05	−1.96	7.44	0.57	−11.52	0.114	0.24(7)[c]
308.15	−10.00	−2.37	9.18	0.64	−10.53	0.090	0.16(7)[c]
318.15	−9.20	−2.83	10.10	0.76	−9.87	−0.034	0.15(7)[c]

[a] Obtained from the Redlich−Mayer equation. [b] Obtained from an abbreviated form of the Pitzer model (eq 15). [c] The number of experimental data used in the calculation of parameters of the Redlich−Mayer and an abbreviated form of the Pitzer model.

Table 6. Pitzer Model (Equation 14) Parameters and Standard Deviations, σ (V_φ) and σ (d), for Potassium Tartrate (1) + H_2O (2) at Different Temperatures

T	$10^8 \cdot \beta_v^{(0)}$	$10^8 \cdot \beta_v^{(1)}$	$10^8 \cdot C_v$	$10^6 \cdot V_\varphi^0$	$10^6 \cdot \sigma\ (V_\varphi^0)$	$\sigma\ (d)$
K	($kg \cdot mol^{-1} \cdot Pa^{-1}$)	($kg \cdot mol^{-1} \cdot Pa^{-1}$)	($kg^2 \cdot mol^{-2} \cdot Pa^{-1}$)	($m^3 \cdot mol^{-1}$)	($m^3 \cdot mol^{-1}$)	($kg \cdot m^{-3}$)
298.15	0.038	0.174	−0.004	77.19[a]	0.20	0.32
	0.038	0.164	−0.004	77.24[b]	0.20	0.32
308.15	0.031	0.104	−0.003	79.13[a]	0.16	0.24
	0.031	0.110	−0.003	79.10[b]	0.16	0.24
318.15	0.026	0.026	−0.003	80.63[a]	0.17	0.19
	0.025	0.060	−0.002	80.44[b]	0.18	0.18

[a] Obtained from the Pitzer model with unrestrictive volumetric ion interaction parameters. [b] Fixed to the value obtained from the Redlich−Mayer equation (eq 9).

Table 7. Fitting Parameters of the Pitzer Model (Equation 12) with Four Parameters and Standard Deviations, σ (κ_φ) and σ (u), for Potassium Tartrate (1) + H_2O (2) at Different Temperatures

T	$10^{17} \cdot \beta_k^{(0)}$	$10^{17} \cdot \beta_k^{(1)}$	$10^{17} \cdot C_k$	$10^{14} \cdot \kappa^0_\varphi$	$10^{14} \cdot \sigma\ (\kappa_\varphi)$	$\sigma\ (u)$
K	($kg \cdot mol^{-1} \cdot Pa^{-2}$)	($kg \cdot mol^{-1} \cdot Pa^{-2}$)	($kg^2 \cdot mol^{-2} \cdot Pa^{-2}$)	($m^3 \cdot mol^{-1} \cdot Pa^{-1}$)	($m^3 \cdot mol^{-1} \cdot Pa^{-1}$)	($m \cdot s^{-1}$)
298.15	−0.194	−1.620	0.022	−12.10[a]	0.16	6.69
	−0.245	0.475	0.033	−11.05[b]	0.34	19.26
308.15	−0.157	−1.177	0.018	−10.98[a]	0.11	5.05
	−0.202	0.643	0.027	10.00[b]	0.29	15.83
318.15	−0.130	−0.914	0.014	−10.32[a]	0.11	3.89
	−0.184	1.269	0.026	−9.20[b]	0.35	15.90

[a] Obtained from the Pitzer model with unrestrictive volumetric ion interaction parameters. [b] Fixed to the value obtained from the Redlich−Mayer equation (eq 12).

region ($m < 0.1\ mol \cdot kg^{-1}$) calculated from eq 5 were fitted to the Redlich−Mayer equation[19] (eq 9)

$$V_\varphi = V_\varphi^0 + S_v m_1^{0.5} + b_v m_1 \tag{9}$$

where b_v is an empirical parameter, and S_v is defined as

$$S_v = A_v \left[\frac{1}{2} \left(\sum_i \nu_i z_i^2 \right) \right]^{\frac{3}{2}} \tag{10}$$

In eq 10, ν_i and z_i are the stoichiometric number and absolute charge of ion i, respectively. The obtained $V_\varphi{}^0$ and b_v values are given in Table 4. For the limiting theoretical slope, A_v, the values (1.8743, 2.0547, and 2.2601) $cm^3 \cdot kg^{0.5} \cdot mol^{-1.5}$ given by Anathaswamy and Atkinson[20] were used at T = (298.15, 308.15, and 318.15) K, respectively. Using the additivity rule,[21] the partial molar volume at infinite dilution of the tartrate ion can be calculated from the limiting apparent molar volume of potassium tartrate. According to the additivity rule, the partial molar volume of a salt at infinite dilution is the sum of partial molar volume at infinite dilution of its consisting ions, $V_\varphi^0 = \sum_{i=\text{ions of the salt}} \nu_i V_i^0$, in which $V_i{}^0$ is the partial molar volume at infinite dilution of ith ion. Therefore

$$V_\varphi^0 = 2V_{K^+}^0 + V_{C_4H_4O_6{}^{2-}}^0 \tag{11}$$

For partial molar volume at infinite dilution of the potassium ion $V^0{}_{K^+}$, Millero[21] has given the values ($9.02 \cdot 10^{-6}$, $9.57 \cdot 10^{-6}$, and $10.3 \cdot 10^{-6}$) $m^3 \cdot mol^{-1}$ at (298.15, 323.15, and 348.15) K, respectively. From fitting of these data to a straight line, the necessary values for $V^0{}_{K^+}$ may be estimated as ($8.916 \cdot 10^{-6}$ and $9.313 \cdot 10^{-6}$) $m^3 \cdot mol^{-1}$ at (308.15 and 318.15) K, respectively. The standard deviation of the fit is $0.27 \cdot 10^{-6}\ m^3 \cdot mol^{-1}$. Using the $V_\varphi{}^0$ values obtained from the Redlich−Mayer equation (eq 9) which are given in the left-hand side of Table 4 and the above $V^0{}_{K^+}$ values, the partial molar volume of the tartrate ion, $V^0{}_{C_4H_4O_6{}^{2-}}$, can be calculated by the help of eq 11. The $V^0{}_{C_4H_4O_6{}^{2-}}$ values ($59.20 \cdot 10^{-6}$, $61.27 \cdot 10^{-6}$, and $61.81 \cdot 10^{-6}$) $m^3 \cdot mol^{-1}$ are obtained at T = (298.15, 308.15, and 318.15) K, respectively. Our value, $V^0{}_{C_4H_4O_6{}^{2-}} = 59.20$ at 298.15 K can

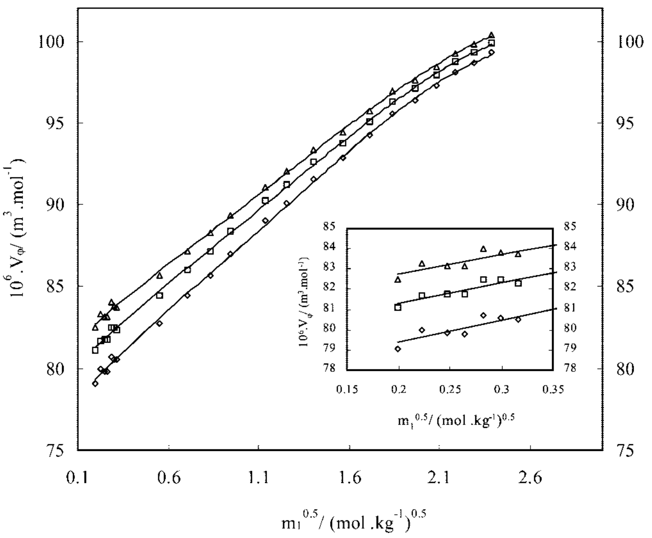

Figure 2. Experimental and calculated apparent molar volume, V_φ, plotted against square root of molality of salt, $m_1^{0.5}$, for the potassium tartrate (1) + H_2O (2) system studied with the Pitzer model: ◇, 298.15 K; □, 308.15 K; △, 318.15 K; −, the Pitzer model.

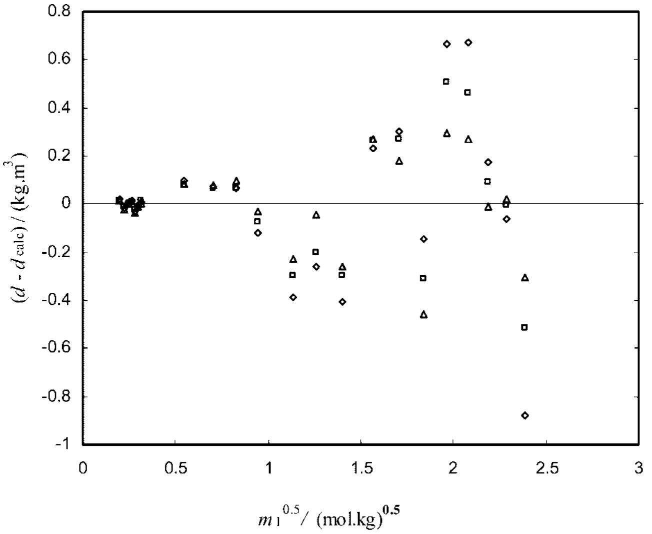

Figure 3. Plot of the difference between the experimental and calculated density data, d, against square root of molality using the Pitzer model (eq 14) for the potassium tartrate (1) + H_2O (2) system: ◇, 298.15 K; □, 308.15 K; △, 318.15 K.

be compared with the average values $V^0_{C_4H_4O_6}{}^{2-} = (60 \pm 0.4) \cdot 10^{-6}$ $m^3 \cdot mol^{-1}$, obtained from a volumetric study of aqueous disodium tartrate, diammonium tartrate, and sodium potassium tartrate by Apelblat et al.,[7] and $V^0_{C_4H_4O_6}{}^{2-} = 59.77 \cdot 10^{-6}$ $m^3 \cdot mol^{-1}$ obtained previously from a volumetric study of disodium tartrate,[9] and they are in good agreement. However, Halasey[6] has reported the value $V_\varphi{}^0 = 83.2 \cdot 10^{-6}$ $m^3 \cdot mol^{-1}$ for dipotassium tartrate in water at 298.15 K. This gives $V^0_{C_4H_4O_6}{}^{2-} = 65.16 \cdot 10^{-6}$ $m^3 \cdot mol^{-1}$, which is higher than the above values. Since no densities but only a $V_\varphi{}^0$ value is given,[6] it is difficult to see the reason for the discrepancy between our results and those of Halasey.[6] Apelblat et al.[7] observed that a rather higher value has also been obtained by Halasey[6] for partial molar volume at infinite dilution of potassium citrate.

An equation of the form[19]

$$\kappa_\varphi = \kappa^0_\varphi + S_\kappa m_1^{0.5} + b_\kappa m_1 \tag{12}$$

was used for correlating the experimental apparent molar isentropic compressibility data in the dilute region ($m < 0.1$).

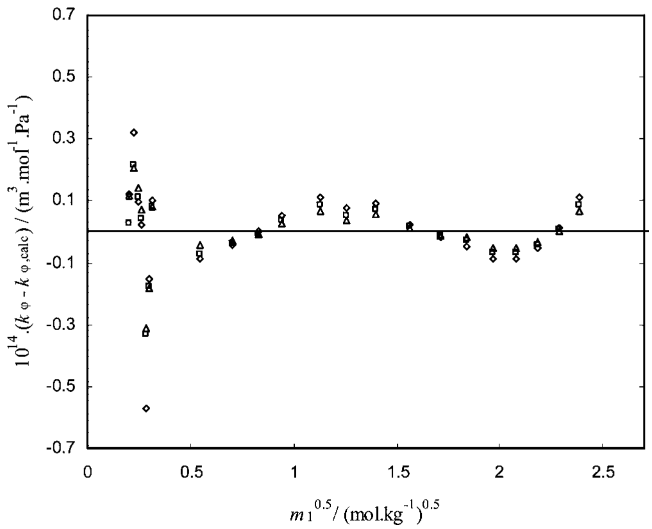

Figure 4. Plot of difference between the experimental and calculated apparent molar isentropic compressibility, κ_φ, against square root of molality using the Pitzer model (eq 14) for the potassium tartrate (1) + H_2O (2) system: ◇, 298.15 K; □, 308.15 K; △, 318.15 K.

Here κ^0_φ is the limiting apparent molar isentropic compressibility; b_κ is an empirical parameter; and S_κ is defined as

$$S_\kappa = -A_\kappa\left[\frac{1}{2}\left(\sum_i \nu_i z_i^2\right)\right]^{3/2} \tag{13}$$

The values of κ^0_φ and b_κ obtained at each temperature are listed in Table 5. The parameters κ^0_φ, S_κ, and b_κ have a meaning similar to that in eq 9 for apparent molar volume. A_κ is the Debye–Huckel slope for the apparent molar compressibility of water for which the values $(-3.7784 \cdot 10^{-4}$, $-4.5589 \cdot 10^{-4}$, and $-5.4505 \cdot 10^{-4})$ $cm^3 \cdot kg^{1/2} \cdot mol^{-3/2} \cdot bar^{-1}$ taken from Anathaswamy and Atkinson[20] were used at $T = (298.15, 308.15,$ and $318.15)$ K, respectively. The specific interaction theory of Pitzer has been used for correlating the volumetric and compressibility properties of the aqueous and nonaqueous electrolyte solutions with good accuracy. The details of the Pitzer equations are given elsewhere.[20] This model[20] has the following form for 1:2 electrolyte solutions

$$X_\varphi = X^0_\varphi - 3\frac{A_x}{b}\ln(1 + bm_1^{0.5}) - 4RTm_1(B_x + 4m_1C_x) \tag{14a}$$

where B_x is the binary interaction parameter which is defined as

$$B_x = \beta_x^{(0)} + \beta_x^{(1)}\frac{2}{\alpha_1 m_1}[1 - (1 + \alpha_1 m_1^{0.5})\exp(-\alpha_1 m_1^{0.5})] \tag{14b}$$

In eq 14, X_φ represents V_φ or k_φ. A_x is the limiting theoretical slope A_v or A_κ. The second and third terms on the right-hand side of eq 14a indicate the Debye–Huckel and virial coefficient expressions, respectively.

Pinheiro et al.[22] have suggested that the abbreviated form of the Pitzer model may also be used to obtain $V_\varphi{}^0$ values. This abbreviated form of the Pitzer model has the following form for 1:2 electrolyte solutions

$$X_\varphi = X^0_\varphi - 3\frac{A_x}{b}\ln(1 + bm_1^{0.5}) - 4RTm_1\beta_z^0 \tag{15}$$

We decided to use eq 15 to obtain $V_\varphi{}^0$ and $\kappa_\varphi{}^0$ values. The results of fitting for V_φ and $\boldsymbol{\kappa_\varphi}$data in the dilute region are

Table 8. Fitting Parameters of the Pitzer Model (Equation 14) with Five Parameters and Standard Deviations, σ (κ_φ) and σ (u), for Potassium Tartrate (1) + H_2O (2) at Different Temperatures

T / K	$10^{17}\cdot\beta_k^{(0)}$ / ($\mathrm{kg\cdot mol^{-1}\cdot Pa^{-2}}$)	$10^{17}\cdot\beta_k^{(1)}$ / ($\mathrm{kg\cdot mol^{-1}\cdot Pa^{-2}}$)	$10^{17}\cdot\beta_k^{(2)}$ / ($\mathrm{kg\cdot mol^{-1}\cdot Pa^{-2}}$)	$10^{17}\cdot C_k$ / ($\mathrm{kg^2\cdot mol^{-2}\cdot Pa^{-2}}$)	$10^{14}\cdot\kappa^0_\varphi$ / ($\mathrm{m^3\cdot mol^{-1}\cdot Pa^{-1}}$)	$10^{14}\cdot\sigma$ (κ_φ) / ($\mathrm{m^3\cdot mol^{-1}\cdot Pa^{-1}}$)	σ (u) / ($\mathrm{m\cdot s^{-1}}$)
298.15	−0.170	−4.483	3.899	0.016	−11.59[a]	0.14	1.15
	−0.158	−8.018	8.502	0.012	−11.05[b]	0.15	3.45
308.15	−0.141	−3.088	2.625	0.013	−10.61[a]	0.09	1.09
	−0.118	−7.988	8.735	0.007	10.00[b]	0.13	5.83
318.15	−0.118	−2.347	1.975	0.011	−10.03[a]	0.09	0.91
	−0.085	−8.788	10.009	0.002	−9.20[b]	0.15	8.18

[a] Obtained from the Pitzer model with unrestrictive volumetric ion interaction parameters. [b] Fixed to the value obtained from the Redlich−Mayer equation (eq 12).

Table 9. Experimental Viscosity, η, and Density, d, for Potassium Tartrate (1) + H_2O (2) at Different Temperatures

m_1 / ($\mathrm{mol\cdot kg^{-1}}$)	$10^{-3}\cdot d$ / ($\mathrm{kg\cdot m^{-3}}$)	η / ($\mathrm{m\cdot Pa\cdot s}$)
	T = 298.15 K	
0.0222	1.000227	0.879
0.0312	1.001272	0.892
0.0516	1.004265	0.902
0.0730	1.007196	0.911
0.0909	1.009682	0.917
0.1822	1.022149	0.948
0.2842	1.035396	0.985
0.3843	1.048001	1.012
0.4639	1.057858	1.040
0.6883	1.084655	1.125
1.0976	1.129431	1.294
1.4682	1.166427	1.474
1.8781	1.204158	1.710
2.6016	1.262747	2.243
3.4047	1.317793	3.301
4.0775	1.357871	4.296
5.1880	1.414292	6.378
5.3782	1.422909	7.066
	T = 308.15 K	
0.0222	0.997160	0.718
0.0312	0.998223	0.723
0.0516	1.001145	0.731
0.0730	1.004035	0.737
0.0909	1.006486	0.750
0.1822	1.018786	0.773
0.2842	1.031860	0.799
0.3843	1.044309	0.821
0.4639	1.054049	0.847
0.6883	1.080554	0.916
1.0976	1.124908	1.054
1.4682	1.161613	1.206
1.8781	1.199098	1.389
2.6016	1.257381	1.801
3.4047	1.312208	2.613
4.0775	1.352159	3.367
5.1880	1.408438	4.841
5.3782	1.417036	5.339
	T = 318.15 K	
0.0222	0.993306	0.595
0.0312	0.994475	0.601
0.0516	0.997256	0.609
0.0730	1.000113	0.612
0.0909	1.002537	0.623
0.1822	1.014707	0.639
0.2842	1.027648	0.665
0.3843	1.039979	0.693
0.4639	1.049634	0.705
0.6883	1.075926	0.764
1.0976	1.119972	0.879
1.4682	1.156458	1.006
1.8781	1.193744	1.157
2.6016	1.251764	1.483
3.4047	1.306403	2.120
4.0775	1.346258	2.700
5.1880	1.402441	3.829
5.3782	1.411025	4.176

Table 10. Parameters of Equation 18 along with Standard Deviations, σ (η), for the Potassium Tartrate (1) + H_2O (2) System at Different Temperatures

T / K	ln a_0	a_0	b_0	C_0	σ ln(η)	σ (η)
298.15	−0.1259	0.8817	0.3522	0.0066	0.018	0.067
308.15	−0.3336	0.7164	0.3579	0.0030	0.018	0.052
318.15	−0.5173	0.5961	0.3619	0.0001	0.016	0.035

presented in Tables 4 and 5. From these tables, we conclude that the performance of the Pitzer model in the form of eq 15 is very similar to that of the Redlich−Mayer equation in predicting $V_\varphi{}^0$ and $\kappa_\varphi{}^0$ values for the studied system at different temperatures.

We applied eq 14 for analyzing V_φ and κ_φ data for the entire concentration range. First, we treated $V_\varphi{}^0$ or $\kappa_\varphi{}^0$ as a parameter. In this case, from the fitting of V_φ and κ_φ data, the unrestrictive sets of volumetric and acoustic ion interaction parameters are obtained which are presented as a first row in Tables 6 and 7, respectively. The experimental and calculated apparent molar volume, V_φ, plotted against square root of molality of salt, $m_1{}^{0.5}$, using the Pitzer model with unrestrictive parameters of Table 6 is shown in Figure 2. In the second procedure, in the fitting of the V_φ and κ_φ data to the Pitzer model (eq 14), at each working temperature $V_\varphi{}^0$ and $\kappa_\varphi{}^0$ values are fixed to the limiting apparent molar volume and limiting apparent molar isentropic compressibility values obtained, respectively, from eqs 9 and 12. In this case, the restrictive sets of volumetric and acoustic ion interaction parameters are obtained which are summarized, respectively, in Tables 6 and 7 as a second row at each temperature.

From the obtained standard deviations reported in Tables 6 and 7, it can be seen that the experimental V_φ and κ_φ data can be satisfactorily represented by the Pitzer equations at different temperatures. To show the differences between the experimental and calculated densities and apparent isentropic compressibilities in a better manner, the values for $d - d_{\mathrm{calcd}}$ and $\kappa_\varphi - \kappa_{\varphi\mathrm{calcd}}$ are plotted against the square root of molality as shown, respectively, in Figures 3 and 4. In preparing these figures, the unrestrictive parameters of Tables 6 and 7 were used. As can be seen from Tables 6 and 7, the $V_\varphi{}^0$ and $\kappa_\varphi{}^0$ values predicted by the Pitzer model (eq 14) are in good agreement with those obtained from the Redlich−Mayer equation. However, the obtained standard deviations for speed of sound reported in Table 7 are rather large. To improve the quality of fitting κ_φ to the Pitzer model (eq 14), we decided to add the extra term, $\beta_x{}^{(2)}(2/\alpha_2{}^2 m_1)[1 -(1 = \alpha_2 m_1{}^{0.5}) \exp(-\alpha_2 m_1{}^{0.5})]$, to the right-hand side of eq 14b. In this case, the values $\alpha_1 = 1.4\ \mathrm{kg^{1/2}\cdot mol^{-1/2}}$ and $\alpha_2 = 2\ \mathrm{kg^{1/2}\cdot mol^{-1/2}}$ were used. The results of fitting are given in Table 8. As can be seen from the Table 8, by adding the extra term to the Pitzer model, we obtain smaller standard deviations for speed of sound u.

Several empirical as well as theoretical equations are available in the literature for describing the concentration dependence of

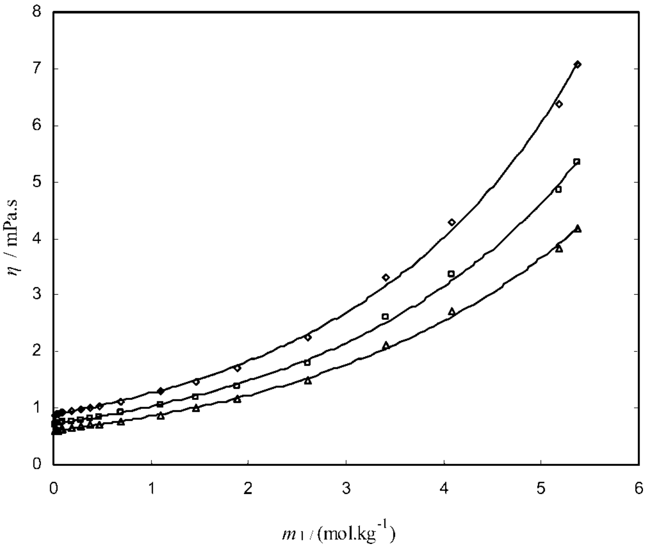

Figure 5. Experimental and calculated viscosity data, η, plotted against molality of salt, m_1, for the potassium tartrate (1) + H_2O (2) system: ◇, 298.15 K; □, 308.15 K; △, 318.15 K; −, eq 18.

viscosity. However, most of these equations fail to be applicable when the concentration becomes high. Horvath[23] has reviewed these equations, and it was shown that[23–25] a semiempirical equation

$$\eta = a_0 \exp(b_0 m_1 + c_0 m_1^2) \tag{16}$$

is useful over wide concentration ranges. In eq 16, a_0, b_0, and c_0 are adjustable temperature-dependent parameters. The results of fitting experimental viscosity data of Table 9 to this equation are presented in Table 10. Figure 5 shows the concentration dependence of viscosities obtained from the experiment and those generated using eq 16 with the parameters taken from Table 10. From Figure 5 and the standard deviations given in Table 10, we conclude that eq 16 adequately fits the viscosity data of aqueous solutions of dipotassium tartrate from dilute to high concentration. As pointed out by Wahab et al.,[26] the value a_0 corresponds to the viscosity at infinite dilution. From Table 10, we also note that the obtained value for a_0 at each temperature is in very good agreement with that of pure water at the corresponding temperature. The maximum error corresponds to 298.15 K which is less than 1 %, and at (308.15 and 318.15) K, the error is (0.4 and 0.01) %, respectively.

Conclusions

The accurate osmotic coefficient data measured at $T = (298.15, 308.15, \text{and } 318.15)$ K for aqueous dipotassium tartrate solutions were fitted satisfactorily to the Pitzer model. The apparent molar volumes and apparent molar compressibilities were calculated from the measured density and speed of sound data at $T = (298.15, 308.15, \text{and } 318.15)$ K, and these data were also fitted to the corresponding Pitzer equation with good accuracy. It was found that both the Redlich−Mayer and the abbreviated form of the Pitzer equations are suitable for the dilute region ($m < 0.1$), and the agreement between the limiting values for apparent molar volumes and compressibilities obtained by these equations is excellent. The viscosity data were adequately fitted to an empirical equation.

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