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Volume Properties of Aqueous Electrolytes. 2. Application of the Pitzer Model in Estimating Apparent Molal Compressibility and Expansibility

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The Pitzer model of virial coefficients is applied to the apparent molal compressibility and expansibility of aqueous single electrolyte solutions. Pitzer coefficients for several electrolyte solutions are given in the form of temperature-dependent equations. Also, these coefficients are applied for estimating the above properties in aqueous mixed electrolytes. The Pitzer equations can predict these properties with excellent accuracy.

Introduction

The Pitzer equations are proving to be very useful in estimating various thermodynamic properties of aqueous single and mixed electrolytes (*1*, *2*). In our attempts to understand better their utility in predicting volume properties of electrolytic solutions, we (*3*) recently successfully fitted the apparent molal volumes of several single electrolytes and presented the Pitzer coefficients. We now apply these equations for estimating apparent molal compressibility ϕ_K and expansibility ϕ_E of single electrolytes. The Pitzer coefficients obtained from the single-electrolyte analysis are used for calculating these properties in mixtures.

Pitzer Equations

The details of the Pitzer equations are given elsewhere (*4*). For immediate use, the system of Pitzer equations is given below. Since the equations used for ϕ_K and ϕ_E are analogous, we give them for general property ϕ. Apparent molal property of a single electrolyte $M_{\nu_M}X_{\nu_X}$ as a function of concentration m (mol kg^{-1}) is

$$\phi = \phi^0 + \nu|Z_M Z_X|(A/2b)\ln(1 + bI^{1/2}) + 2\nu_M\nu_X RT[mB_{MX} + m^2(\nu_M\nu_X)^{1/2}C_{MX}] \quad (1)$$

where

$$B_{MX} = \beta^{(0)}_{MX} + 2\beta^{(1)}_{MX}/\alpha^2 I[1 - (1 + \alpha I^{1/2})e^{-\alpha I^{1/2}}] \quad (2)$$

$$C_{MX} = C_{MX}{}^{\phi}/2 \quad (3)$$

Note that for ϕ_K, terms $\beta^{(0)}_{MX}$ and $\beta^{(1)}_{MX}$ are $(\partial^2\beta/\partial P^2)$ and $(\partial^2\beta/\partial P^2)$, respectively. For ϕ_E, they are $(\partial^2\beta^{(0)}/\partial P\partial T)$ and $(\partial^2\beta^{(1)}/\partial P\partial T)$, respectively. Similarly, in eq 3, $(\partial^2 C^{\phi}/\partial P^2)_T$ and $(\partial^2 C^{\phi}/\partial P\partial T)$ are for ϕ_K and ϕ_E, respectively. Terms $\beta^{(0)}_{MX}$, $\beta^{(1)}_{MX}$, and C^{ϕ}_{MX} are the Pitzer coefficients. A, the Pitzer–Debye–Hückel slope, can be taken from elsewhere (*5*–*7*). ϕ^0, apparent molal compressibility or expansibility at infinite dilution, is equal to the partial molal compressibility or expansibility at infinite dilution.

Any of the Pitzer coefficients may be fitted with the temperature-dependent equation of the form

$$Y = A + BT + CT^2 \quad (4)$$

where A, B, and C are the coefficients.

Apparent molal property of an ion in mixture, ϕ_i, is given by

$$\phi_i = \phi_i{}^0 + (Z_i A/2b)\ln(1 + bI^{1/2}) + RT\sum_{i} B_{iJ} m_J + (RT/2|Z_i|^{1/2})\sum_{i} C_{iJ} m_J{}^2 |Z_J|^{1/2} + RT\sum_{k}\theta_{ik} m_k \quad (5)$$

in eq 5, the last term on the right-hand side expresses the mixing term recognizing explicitly the interactions among like-charged ions. θ_{ik} is the difference parameter and can be evaluated from the experimental ϕ and ϕ obtained from eq 5 minus last term. One notes that last term has the sum over k ions with the same charge ions and over i with the opposite charge ions. The equation for calculating θ_{ik} is

$$\theta_{ik} = \frac{\Delta\phi^*(m_1 + m_2)}{RTm_1 m_2(\nu_i + \nu_k)} \quad (6)$$

where m_1 and m_2 are molalities of the electrolytes whose cations are under consideration for interactions and ν_i and ν_k are the moles of i and k in their respective electrolytes.

The calculated ϕ_i is converted to ϕ^* by using the relation

$$\phi^* = \frac{\sum_J m_J \phi_J}{\sum_J m_J} \quad (7)$$

where $\phi_J = \sum_i \nu_i \phi_i$.

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Table I. Pitzer Coefficients for ϕ_K of Aqueous Single Electrolytes at 25 °C

electrolytes	$(\partial^2\beta^{(0)}/\partial P_2) \times 10^8$	$(\partial^2\beta^{(0)}/\partial P^2) \times 10^8$	$(\partial^2C^\phi/\partial P^2) \times 10^9$	$\sigma(\phi_K) \times 10^4$, cm^3 mol^{-1} bar^{-1}
KCl (*20*)[a]	4.5	1.34	−1.93	0.05
KBr (*21*)	4.0	0.825	−0.968	0.04
NaBr (*20*)	8.4	0.930	−0.894	0.08
RbCl (*20*)	6.1	1.30	−1.79	0.10
CsCl (*20*)	9.3	1.08	−1.12	0.10
CsBr (*20*)	4.7	1.45	−2.99	0.09
CsI (*20*)	2.2	1.19	−4.02	0.05
$MnSO_4$ (*22*)	1.1	15.17	−133.7	0.18
$CoSO_4$ (*22*)	1.0	17.07	−164.7	0.10
$NiSO_4$ (*22*)	0.9	17.82	−181.0	0.10
$ZnSO_4$ (*22*)	1.0	16.98	−166.2	0.09
$CdSO_4$ (*22*)	0.8	17.19	−194.1	0.09
$LaCl_3$ (*23*)	0.8	7.51	−46.36	0.16

[a] Number in parentheses indicates the reference source of data.

General Equations

Experimental sound velocities u from the literature can be converted into adiabatic compressibility β as

$$\beta = 1/u^2d \tag{8}$$

β is then used to estimate the apparent molal compressibility ϕ_K as

$$\phi_K = \frac{1000(\beta d_0 - \beta_0 d)}{mdd_0} + \frac{\beta M}{d} \tag{9}$$

where β_0 and d_0 are the adiabatic compressibility and the density of pure water, taken from elsewhere (*8*, *9*).

Mean apparent molal compressibility of the mixture $\phi_K{}^*$ can be calculated from

$$\phi_K{}^* = \frac{1000(\beta^* d_0 - \beta_0 d^*)}{\sum_J m_J d^* d_0} + \frac{\sum_J \beta_J M_J}{\sum_J m_J d^*} \tag{10}$$

The experimental ϕ_E and $\phi_E{}^*$ can be obtained from the slope of ϕ_V or $\phi_V{}^*$, the apparent molal volume of a single or mixed aqueous electrolytes. This is discussed in detail in our earlier work (*4*).

Results and Discussion

Single Electrolytes. We used the literature data on apparent molal compressibility and expansibility of single-electrolyte solutions in order to generate the Pitzer coefficients using eq 1 to 3. Following our earlier work (*3*) and that of Rogers and Pitzer (*6*), we fitted the entire array of the data without $\beta^{(1)}_{MX}$ term for both the properties with good degree of accuracy. Table I lists the Pitzer coefficients for ϕ_K of the single electrolytes at 25 °C together with the standard deviation (σ). Similarly, Table II lists the temperature dependence of the Pitzer coefficients as calculated by eq 4 for ϕ_K of those electrolyte solutions for which ϕ_K were known at more than three temperatures. Both the tables include the molality range used in the fits and source of experimental data. Figure 1 shows the difference (Δ) between the experimental and the predicted ϕ_K as a function of molality. The fits were random throughout and the average standard deviation of the is 0.9 × 10^{-4} cm^3 mol^{-1} bar^{-1} with slightly higher σ for sulfates. Carpio et al. (*10*) have recently measured the adiabatic compressibilities of aqueous

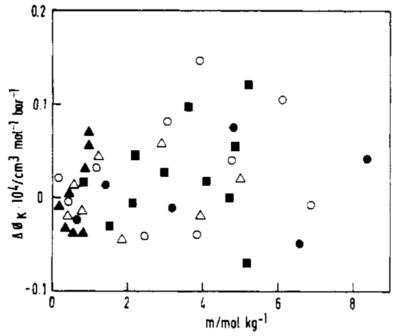

Figure 1. $\Delta\phi_K$ (experimental ϕ_K − calculated ϕ_K) against m. ●, NaBr, 25 °C, ref *20*; ○, CsCl, 25 °C, ref *20*; △, RbCl, 25 °C, ref *20*; ▲, Na_2CO_3, 45 °C, ref *26*; ■, $MgCl_2$, 35 °C, ref *31*.

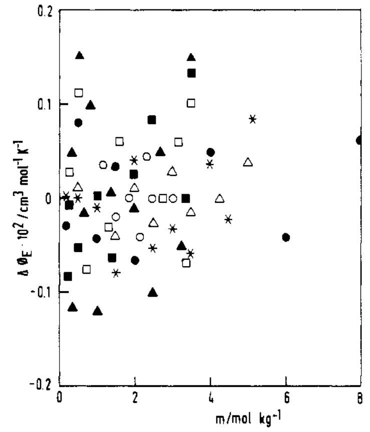

Figure 2. $\Delta\phi_E$ (experimental ϕ_E − calculated ϕ_E) against m. ●, NaBr, 50 °C, ref *29*; ○, HCl, 40 °C, ref *27*; △, KCl, 35 °C, ref *11*; ▲, $PrCl_3$, 75 °C, ref *34*; □, $NdCl_3$, 25 °C, ref 34; ■, $LaCl_3$, 50 °C, ref 34; *, $MgCl_2$, 25 °C, ref 31.

Table II. Temperature Dependence of Pitzer Coefficients for ϕ_K of Aqueous Single Electrolytes ($F = (\partial^2\beta^{(0)}/\partial P^2)$, $S = (\partial^2C^\phi/\partial P^2)$)

electrolyte, temp range, °C, m_{max}, mol kg^{-1}	term	10^8A	$10^{10}B$	$10^{12}C$	$10^4\sigma(\phi_K)$, cm^3 mol^{-1} K^{-1}
HCl (*24*),[a] 0–45, 1.0	F	9.29	−3.37	1.22	0.03
	S	−9.91	6.87	−1.29	
NaOH (*24*), 0–45, 1.0	F	−105.1	754	−13.0	0.03
	S	73.33	−51.1	8.75	
Na_2SO_4 (*25*), 0–50, 1.5	F	29.3	−7.77		0.18
	S	−8.39	2.16		
$MgSO_4$ (*25*), 0–50, 2.2	F	225.1	−135.1	21.6	0.19
	S	−128.2	82.3	−13.5	
$MgCl_2$ (*25*), 0–50, 5.2	F	10.96	3.08		0.09
	S	−1.434	0.410		
$NaHCO_3$ (*26*), 0–50, 1.0	F	14.4	−3.15		0.09
	S	73.4	−53.7	9.39	
Na_2CO_3 (*26*), 0–50, 1.0	F	230.4	−140.0	22.0	0.18
	S	−113.1	68.9	−10.9	

[a] Number in parentheses indicates the reference source of data.

Table III. Temperature Dependence of Pitzer Coefficients for ϕ_E of Aqueous Single Electrolytes ($F = (\partial^2\beta^{(0)}/\partial P\partial T)$, $S = (\partial^2C^\phi/\partial P\partial T)$)

electrolyte, temp range, °C, m_{max}, mol kg^{-1}	term	10^5A	10^8B	10^9C	$10^2\partial(\phi_E)$, cm^3 mol^{-1} K^{-1}
HCl (*24*, *27*, *28*),[a] 0–75,	F	−4.79	29.49	−0.453	0.01
3.0	S	1.190	−7.34	0.113	
HBr (*27*), 25–75	S	0.443	−2.77	0.043	
3.0	S	0.443	−2.77	0.043	
HI (*27*), 25–75, 3.0	F	−1.41	8.69	−0.1.35	0.02
	S	0.9330	−1.99	0.031	
$HClO_4$ (*27*), 25–75, 3.0	F	−14.46	89.52	−1.39	0.03
	S	14.06	−90.80	1.47	
NaBr (*29*), 15–55, 8.0	F	−2.05	12.23	−0.184	0.07
	S	0.151	−0.867	0.012	
	S	0.151	−0.867	−0.012	
$NaNO_3$ (*29*), 15–55, 8.0	F	−3.99	24.17	−0.367	0.10
	S	0.585	−35.59	0.054	
NaOH (*24*, *30*), 0–70,	F	5.23	−10.34	1.35	0.08
25.0	S	−10.25	6.35		
Na_2SO_4 (*31*), 0–50, 1.5	F	−0.368	3.44	−0.080	0.01
	S	−0.024	0.378	−0.011	
KBr (*29*), 15–55, 4.0	F	−6.19	38.75	−0.608	0.07
	S	1.76	−11.24	0.18	
KNO_3 (*29*), 15–55 4.0	F	−8.12	51.99	−0.835	0.05
	S	3.87	−25.10	0.409	
$MgCl_2$ (*11*, *29*, *31*),	F	−0.144	−0.605	−0.033	0.15
0–55, 5.2	S	−0.049	0.126		
$SrCl_2$ (*29*), 15–55, 2.5	F	−13.7	87.2	−0.014	0.17
	S	3.88	−24.8	0.397	
$BaCl_2$ (*29*), 15–55, 1.5	F	−16.1	101.0	−1.59	0.15
	S	8.059	−50.16	0.784	
$MgSO_4$ (*31*), 0–508, 2.2	F	−1.51	9.11	−0.141	0.08
	S	−0.481	1.92	−0.001	
$Cd(NO_3)_2$ (*29*, *32*),	F	−6.63	41.4	−0.649	0.08
15–85, 13.0	S	1.02	−6.36	0.099	
$SmCl_3$ (*33*), 0–80, 3.5	F	−2.18	12.8	−0.190	0.15
	S	0.327	−1.95	0.029	
$GdCl_3$ (*33*), 0–80, 3.5	F	−2.25	13.9	−0.208	0.19
	S	0.285	−1.66	0.025	
$DyCl_3$ (*33*), 0–80, 3.5	F	−2.35	13.7	−0.202	0.15
	S	0.404	−2.39	0.036	
$ErCl_3$ (*33*), 0–80, 3.5	F	−2.80	16.5	−0.245	0.17
		S	0.287	−1.68	0.025
$YbCl_3$ (*33*), 0–80, 3.5	F	−2.47	14.4	−0.212	0.15
	S	0.349	−2.05	0.036	
$LaCl_3$ (*34*), 15–80, 3.5	F	−1.87	10.8	−0.157	0.15
	S	0.421	−2.52	0.038	
$PrCl_3$ (*34*), 0–80, 3.9	F	−2.28	13.3	−0.197	0.19
	S	0.308	−1.84	0.028	
$NdCl_3$ (*34*), 0–80, 3.9	F	−2.24	13.5	−0.199	0.15
	S	0.674	−4.06	0.061	

[a] Number in parentheses indicates the reference source of data.

$Ca(NO_3)_2$, $ZnCl_2$, and $Zn(NO_3)_2$ solutions. On analyzing their data with the Pitzer equations, we obtained large systematic deviations. This may apparently be due to the structural changes occurring in the solutions. The similar behavior was observed by us (*3*) during the ϕ_V fit. We do not give the Pitzer coefficients for these solutions as they may not have physical significance. The Pitzer coefficients of aqueous $CaCl_2$ and problems associated with the fit are discussed elsewhere (*4*).

We list the temperature dependence of the Pitzer coefficients for ϕ_E in Table III along with the standard deviation of the fit. Figure 2 depicts the difference (Δ) between experimental and predicted ϕ_E as a function of molality. The average standard deviation of the fit for ϕ_E is 0.10×10^{-2} cm^3 mol^{-1} K^{-1}. ϕ_E of aqueous KCl (*11*) did not vary with temperature (5–45 °C). For KCl, the Pitzer coefficients $(\partial^2\beta/\partial P\partial T)$ and $(\partial^2C^\phi/\partial P\partial T)$ are 1.34×10^{-8} and -1.93×10^{-9}, respectively. All the three tables do not include the work on aqueous NaCl as Rogers and Pitzer (*6*) have discussed them in detail. During the single-electrolyte fitting, first the attempts were made to treat ϕ^0 unknown and evaluate it through the least-squares method as other Pitzer coefficients. Since ϕ^0 thus obtained was in excellent agreement with the literature data (given in the original papers of the experimental data), we directly used those values and evaluated the Pitzer coefficients for each electrolyte solution.

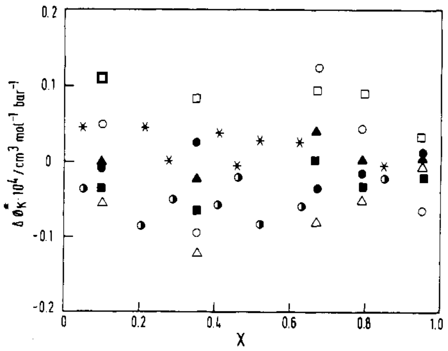

Figure 3. $\Delta\phi_K^*$ (experimental − calculated) against X (mole fraction of second component) without and with θ_{ik}. NaCl-$MgCl_2$: ○ without, ● with, ref *35*. Na_2SO_4-$MgCl_2$: △ without, ▲ with, ref *35*. $MgSO_4$-$MgCl_2$: □ without, ■ with, ref *35*. KCl-KBr: ◑ without, * with, ref *36*.

Table IV. Analysis of Mixture Data on Apparent Molal Compressibilities by Pitzer Equation

system	temp, °C	std dev $\sigma \times 10^4$, cm^3 mol^{-1} bar^{-1} without θ_{ik}	std dev $\sigma \times 10^4$, cm^3 mol^{-1} bar^{-1} with θ_{ik}
NaCl-$MgCl_2$ (*35*)[a]	30	0.60	0.19
Na_2SO_4-$MgCl_2$ (*35*)	30	0.58	0.24
$MgSO_4$-$MgCl_2$ (*35*)	30	0.64	0.21
KCl-KBr (*36*)	25	0.25	0.09

[a] Number in parentheses indicates the reference source of data.

Table V. Values of θ_{ik} Used in Mixture Calculations in Eq 5 (Treated Independent of Ionic Strength)

θ_{ik}	values
Na^+-Mg^{2+}	-1.33×10^{-9}
SO_4^{2-}-Cl^-	4.32×10^{-9}
Cl^--Br^-	1.35×10^{-9}

Mixed Electrolytes. The experimental data on mixtures for compressibility are not too many in the literature. A few data (*12–16*) are found in Russian literature, but they could not be analyzed due to the want of an easy access and lack of full information on experimental data. We have therefore tested the Pitzer equations only for accurate and accessible data. Equations 5–7 were used to calculate ϕ_K^* without and with the mixing parameters θ_{ik}. Table IV lists these systems together with the standard deviations of the fit. Also listed are the standard deviations of the fit with the use of θ_{ik}. Figure 3 shows the effect of θ_{ik} on the fit for ϕ_K^* for few systems. As is apparent, the use of θ_{ik} brings a remarkable improvement in the fit of ϕ_K^* and removes the systematic deviation. Table V lists the values of θ_{ik}. The magnitude of ternary interaction term similar to that derived for ϕ_V (*17*) was very small and hence was neglected. The Pitzer analysis for ϕ_K^* of aqueous NaCl-$CaCl_2$ system from 278.15 to 308.15 K and I = 0.3 to 20 mol kg^{-1} is given in our earlier work (*4*, *18*).

We are not aware of any experimental data on ϕ_E^* except those of Kumar and Atkinson (*4*) and Krumgalz and Millero (*19*). The Pitzer analysis of Kumar and Atkinson data is given elsewhere (*4*). We only computed the ϕ_E^* data on aqueous brines, i.e., NaCl-KCl-$CaCl_2$-$MgCl_2$, using the density of Krumgalz and Millero (*19*) in the ionic strength range 8.3–9.6 mol kg^{-1} and temperature range 20–40 °C. The Pitzer coefficients from Table III, when used in eq 5–7 yield the prediction of ϕ_E^* with a standard deviation of 0.48×10^{-2} cm^3 mol^{-1} K^{-1}. No mixing parameters were needed for the fit.

In part 1 and this study, we attempted to demonstrate the utility of the Pitzer model in estimating the volume properties of aqueous single and mixed electrolyte solutions. The Pitzer equations can fit the volume properties of 1–1 electrolytes up to 9.3 *m* (for NaOH 24 *m*), 2–2 electrolytes up to 2.2 *m*, 2–1 electrolytes up to 7.4 *m* and 1–3 electrolytes up to 3.9 *m* with excellent accuracy. The results prove that this model is a powerful tool for predicting these properties. A preliminary study of these results indicates the possibilities of correlations between the structure of electrolyte solutions and the Pitzer coefficients. The discussion on such correlation is beyond the scope of this paper and will be dealt in a seperate article. Also, modified sets of Pitzer equations accounting structural changes in electrolyte solutions like $ZnCl_2$ and $Zn(NO_3)_2$ will be presented.

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Glossary

A	Pitzer–Debye–Hückel slope
b	1.2, $(kg/mol)^{1/2}$
d	density in solution, g cm^{-3}
I	ionic strength = $^1/_2\sum_i m_i Z_i^2$
M	molecular weight of an electrolyte
m	molality, mol kg^{-1}
R	gas constant = 83.1441 cm^3 mol^{-1} bar^{-1} K^{-1}
T	temperature in kelvin
u	sound velocity
Z	charge on ion

Greek Letters

β_{MX}, β_{MX}, C^{ϕ}_{MX}	Pitzer coefficients
ϕ	apparent molal property at finite concentration
ν	stoichiometric factor in an electrolyte, $\nu = \nu_M + \nu_X$
α	2.0, $(kg/mol)^{1/2}$
β	adiabatic compressibility, bar^{-1}

Superscripts

0	property at infinite dilution
*	mean property in mixture

Subscripts

E	expansibility
i	ion
J	an electrolyte
K	compressibility
M	cation
X	anion
0	property in pure water

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