

Short communication

The two faces of the Redlich–Kister equation and the limiting partial molar volume of water in 1-aminopropan-2-ol

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Abstract

The properties of the Redlich–Kister equation when expressed in power series of $x_1 - x_2$ are related to its alternative expression in terms of power series of $x_2 - x_1$, where x_1 and x_2 are mole fractions of the components 1 and 2 of a binary liquid mixture. The simple relationship between both sets of coefficients is derived and shown to conceal pitfalls while using Redlich–Kister coefficients to estimate partial molar properties of the components. The zero-powered terms, which are the same for the alternative expansions, are shown to yield four-fold the excess molar property for the equimolar mixture. Literature data for the partial molar volume of water at infinite dilution in 15 neat aminoalkanols at different temperatures are collected and tabulated. These data generally show a positive dependence of that limiting value on the temperature, the only apparent exception being in the case of 1-aminopropan-2-ol. It is demonstrated that the recently published data for this aminoalkanol [S. Mokraoui, A. Valtz, C. Coquelet, D. Richon, *Thermochim. Acta* 440 (2006) 122–128] were ill-treated and recalculated limiting values are given, which increase with increasing temperature.

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1. Introduction

The Redlich–Kister equation [1], the form of which modern authors [2] have traced to Guggenheim [3,4] and Scatchard [5], is a popular method for the algebraic representation of mixing and excess physical and thermochemical properties of binary liquid mixtures. According to Science Citation Index, by 6 October 2007 Redlich and Kister's paper [1] had been cited 2297 times.

Over the years, the actual form of the original equation has evolved and several forms, albeit mathematically equivalent, are currently in use. We believe that their inter-conversions cover a pitfall in the way to calculate excess partial molar properties at infinite dilution using Redlich–Kister coefficients, which has not yet been discussed.

On the other hand, recently Mokraoui et al. [6] reported limiting partial molar volumes of water in 1-aminopropan-2-

ol steadily decreasing from $15.3 \text{ cm}^3 \text{ mol}^{-1}$ to $14.5 \text{ cm}^3 \text{ mol}^{-1}$ when the temperature was risen from 283 K to 353 K. If correct, this would have been a remarkable finding because the expectation is a positive temperature dependence, as observed in many other aminoalkanols. Actually, it was this strange report that guided us across the subtleties of the Redlich–Kister equation and to the possible blunder underlying the miscalculated values.

2. The Redlich–Kister expansion

The excess molar volume, V_m^E , and many other thermodynamic properties of a binary liquid mixture prepared with chemical substances 1 and 2 at fixed temperature T and pressure p , are frequently described using the Redlich–Kister equation [1]. Its original form leads to

$$V_m^E = x_1 x_2 \sum_k A_k (x_1 - x_2)^k \quad (1)$$

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In the latter equation, x_1 and x_2 are mole fractions, A_k ($k=0-n$) are adjustable coefficients and n is the order of the polynomial expansion, which should be determined using a statistical significance test such as the F -test.

Following Van Ness and Abbott [7], it is instructive to perform the variable transformation $z=x_1-x_2$. Since $x_1+x_2=1$, then $x_1=(1+z)/2$ and $x_2=(1-z)/2$, and therefore:

$$z = x_1 - x_2 = 2x_1 - 1 = 1 - 2x_2 \quad (2)$$

$$\frac{1-z^2}{4} = x_1x_2 = x_1(1-x_1) = x_2(1-x_2) \quad (3)$$

In terms of variable z , which varies from -1 to $+1$, Eq. (1) becomes

$$V_m^E = \left[\frac{1-z^2}{4} \right] \sum_k A_k z^k \quad (4)$$

The latter equation condenses the original face of the Redlich–Kister equation. This face is expressed as a power series of the independent variable z which can take any of the forms given in Eq. (2), and the pre-summation factor variously written as in Eq. (3). We have chosen the running index k to vary from 0 to n . Other current choice is to use a running index j from 1 to m , in which case the summation term takes the form $\sum_j A_j z^{j-1}$.

However, the Redlich–Kister equation presents many times a new face, which is algebraically and numerically equivalent to the original face. In fact, exchanging the component subscripts in Eq. (1) leads to

$$V_m^E = x_1x_2 \sum_k B_k (x_2 - x_1)^k \quad (5)$$

We introduce the convenient independent variable $s=x_2-x_1$, which also varies from -1 to $+1$ and is merely equal to $-z$. Hence $x_1=(1-s)/2$ and $x_2=(1+s)/2$. As a result:

$$s = x_2 - x_1 = 2x_2 - 1 = 1 - 2x_1 \quad (6)$$

$$\frac{1-s^2}{4} = x_1x_2 = x_2(1-x_2) = x_1(1-x_1) \quad (7)$$

$$V_m^E = \left[\frac{1-s^2}{4} \right] \sum_k B_k s^k \quad (8)$$

Because $s=-z$, $s^2=z^2$ and the pre-summation factors are identical in Eqs. (4) and (8). However, the power terms are related by

$$s^k = (-1)^k z^k \quad (9)$$

And rewriting Eq. (8) in terms of variable z yields:

$$V_m^E = \left[\frac{1-z^2}{4} \right] \sum_k B_k (-1)^k z^k \quad (10)$$

Comparison of Eqs. (4) and (10) gives the following important result:

$$A_k = B_k (-1)^k \quad (11)$$

In other words, for a given set of experimental data the fitting coefficients A_k obtained using $z=x_1-x_2$ as independent variable are related by Eq. (11) to the parameters B_k when $s=x_2-x_1$ is chosen as independent variable. Thus $A_k=B_k$ for the coefficients of even-powered terms and $A_k=-B_k$ for the coefficients of odd-powered terms. As stated, this alternate correspondence does not depend on the starting number for the running index. Eq. (11) entails an interesting consequence. If, inadvertently, at an experimental mole fraction x'_2 we calculate $x'_1 x'_2 \sum_k B_k (1-2x'_2)^k$, we obtain V_m^E for the composition $x''_2 = 1 - x'_2$. In the event x''_2 is also an experimental mole fraction, this procedure may lead to a false check of calculations.

The physical meaning of A_0 , which equals B_0 , is useful in practice. Indeed, the equimolar mixture implies $x_1x_2=0.25$ and $x_1-x_2=0$, and consequently at this composition V_m^E is given by $0.25 \times A_0$. That is, the correctness of computer-fitted A_0 values can be appreciated at a glance by comparing $0.25A_0$ with table- or graph-read excess molar values for the equimolar mixture.

3. Excess partial molar volumes

Excess partial molar volumes of the components, V_i^E , have many times been estimated using Redlich–Kister coefficients. From chemical thermodynamics [7]:

$$V_i^E = V_m^E + (1-x_i) \frac{dV_m^E}{dx_i} \quad (12)$$

Here V_m^E is calculated using the Redlich–Kister equation in the form of Eq. (1), (4), (5) or (8). For the differential dV_m^E/dx_i , which rigorously should be written as the partial derivative $(\partial V_m^E/\partial x_i)_{T,p}$, we note that $dz=dx_1-dx_2=2dx_1=-2dx_2$ [7] and, similarly, that: $ds=dx_2-dx_1=2dx_2=-2dx_1$.

Of particular interest are the limiting excess partial molar volumes, $V_i^{E,\infty}$, from which the corresponding partial molar volumes at infinite dilution, V_i^∞ , are easily calculated using the following relationship, where V_i^* is the molar volume of pure substance i at the same T and p :

$$V_i^\infty = V_i^* + V_i^{E,\infty} \quad (13)$$

Frequently the expressions relating limiting $V_i^{E,\infty}$ properties to Redlich–Kister coefficients are obtained from the full, involved expressions for V_i^E . All the same, Lampreia and co-workers [8] devised a much simpler derivation, which has been repeated for different properties [9,10]. Thus, making use of the thermodynamic relationship between the excess molar volume of a binary mixture and the excess apparent molar volume of component i , $\phi(V_i)^E$:

$$\phi(V_i)^E = \frac{V_m^E}{x_i} \quad (14)$$

in terms of Eq. (4) we obtain

$$\phi(V_1)^E = \left[\frac{1-z}{2} \right] \sum_k A_k z^k \quad (15)$$

$$\phi(V_2)^E = \left[\frac{1+z}{2} \right] \sum_k A_k z^k \quad (16)$$

Another thermodynamic result is

$$\lim(x_i = 0) \phi(V_i)^E = V_i^{E,\infty} \quad (17)$$

Since $x_1 = 0$ for $z = -1$ and $x_2 = 0$ for $z = 1$, from combining Eqs. (15)–(17) follow that

$$V_1^{E,\infty} = \sum_k A_k (-1)^k \quad (18)$$

$$V_2^{E,\infty} = \sum_k A_k \quad (19)$$

Both Eqs. (18) and (19) are well-known results.

In a similar fashion, from Eqs. (8) and (14) we write

$$\phi(V_1)^E = \left[\frac{1+s}{2} \right] \sum_k B_k s^k \quad (20)$$

$$\phi(V_2)^E = \left[\frac{1-s}{2} \right] \sum_k B_k s^k \quad (21)$$

Furthermore, because now $x_1 = 0$ for $s = 1$ and $x_2 = 0$ for $s = -1$, we obtain

$$V_1^{E,\infty} = \sum_k B_k \quad (22)$$

$$V_2^{E,\infty} = \sum_k B_k (-1)^k \quad (23)$$

As before, Eqs. (22) and (23) are not new. However, in view of Eq. (11), they may constitute an unsuspected pitfall for the less-experienced researcher if A_k coefficients are mistaken as B_k coefficients or the other way round.

4. The dependence on temperature of the limiting partial molar volume of water in aminoalkanols

The partial molar volume of water at infinite dilution in organic solvents and its isobaric variation with temperature are valuable probes for understanding the nature of aqueous liquid mixtures. In this field, most authors use subscript 1 to identify component water and subscript 2 for the organic component. Following this convention, we will be primarily interested in V_1^∞ and $(\partial V_1^\infty / \partial T)_p$ when the organic solvent is an aminoalkanol, for which there is a wealth of published thermodynamic data.

The volumetric properties of 15 aqueous aminoalkanol systems over the entire composition range and at different temperatures have been investigated in recent years by research groups led by Mather [11–15], Palepu [16,17], Henni [18–21], Lampreia [9] and Richon [6]. The names and formulae of these 15 aminoalkanols are given in Table 1.

The collection of aminoalkanols in Table 1 shows a large variety of structures and includes primary and secondary alcohols, mono-ols, diols and one triol, and primary, secondary and tertiary amines, and also one diamine. On the basis of published V_1^∞ values and of V_1^∞ values calculated by us using published Redlich–Kister coefficients and density data for water taken from Ref. [22], together with Eq. (13), we constructed Table 2.

Analysis of Table 2 shows that, with very few irregularities, V_1^∞ increases with increasing temperature, except for the original values reported for water in 1-aminopropan-2-ol. This aminoalkanol contains a secondary alcohol and primary amine functional groups and the noted apparent exceptional dependence of V_1^∞ on temperature seems puzzling. Although the thermodynamic relationships used in Ref. [6] are faultless, on closer inspection we found that the Redlich–Kister coefficients in Table 4 in Ref. [6] only reproduced experimental V_m^E values while using Eq. (8) in spite of the fact that the Redlich–Kister expansion and derived equations in Ref. [6] are in terms of Eq. (4). In short, in Table 5 in Ref. [6] the subscripts for $V_i^{E,\infty}$ should be interchanged. Furthermore, back calculations performed by us demonstrated that reported V_i^∞ values for both components [6] were miscalculated in terms of Eq. (13) by combining $V_i^{E,\infty}$

Table 1
Acronym, nomenclature and chemical formula of aminoalkanols of interest

Acronym	Common name	IUPAC name	Structural formula
MEA	Ethanolamine	2-Aminoethanol	NH ₂ CH ₂ CH ₂ OH
IPA	Isopropanolamine	1-Aminopropan-2-ol	CH ₃ CH(OH)CH ₂ NH ₂
MMEA	<i>N</i> -Methylethanolamine	2-(Methylamino)ethanol	CH ₃ NHCH ₂ CH ₂ OH
AMP	–	2-Amino-2-methylpropan-1-ol	(CH ₃) ₂ C(NH ₂)CH ₂ OH
MEEA	<i>N</i> -Ethylethanolamine	2-(Ethylamino)ethanol	CH ₃ CH ₂ NHCH ₂ CH ₂ OH
DMEA	<i>N,N</i> -Dimethylethanolamine	2-(Dimethylamino)ethanol	(CH ₃) ₂ NCH ₂ CH ₂ OH
DEA	Diethanolamine	2,2'-Iminobisethanol	NH(CH ₂ CH ₂ OH) ₂
AEEA	–	2-((2-Aminoethyl)amino)ethanol	NH ₂ CH ₂ CH ₂ NHCH ₂ CH ₂ OH
<i>n</i> -PEA	<i>N</i> -Propylethanolamine	2-(Propylamino)ethanol	CH ₃ CH ₂ CH ₂ NHCH ₂ CH ₂ OH
MDEA	<i>N</i> -Methyldiethanolamine	2,2'-(Methylimino)bisethanol	CH ₃ N(CH ₂ CH ₂ OH) ₂
DEEA	<i>N,N</i> -Diethylethanolamine	2-(Diethylamino)ethanol	(CH ₃ CH ₂) ₂ NCH ₂ CH ₂ OH
DIPA	Diisopropanolamine	1,1'-Iminobispropan-2-ol	NH(CH ₂ CH(OH)CH ₃) ₂
EDEA	<i>N</i> -Ethyl-diethanolamine	2,2'-(Ethylimino)bisethanol	CH ₃ CH ₂ N(CH ₂ CH ₂ OH) ₂
TEA	Triethanolamine	2,2',2''-Nitrilotrisethanol	N(CH ₂ CH ₂ OH) ₃
<i>n</i> -BDEA	<i>N</i> -Butyldiethanolamine	2,2'-(Butylimino)bisethanol	CH ₃ CH ₂ CH ₂ CH ₂ N(CH ₂ CH ₂ OH) ₂

Table 2
 Partial molar volume of water ($\text{cm}^3 \text{mol}^{-1}$) at infinite dilution in aminoalkanols at different temperatures

T (K)	MEA		IPA		MMEA		AMP		
278.15	15.8 [13]	–	–	–	15.2 [13] ^a	–	–	–	–
283.15	–	–	(15.3) [6]	14.7 ^b	–	–	–	–	–
288.15	15.9 [13]	–	(15.2) [6]	14.8 ^b	15.3 [13] ^a	15.3 [21]	–	–	–
293.15	–	–	(15.1) [6]	14.8 ^b	–	–	–	–	–
298.15	16.1 [11]	16.0 [16]	(15.0) [6]	14.9 ^b	15.3 [13] ^a	–	–	–	–
303.15	16.2 [11]	–	(15.0) [6]	14.9 ^b	16.0 [13] ^a	15.3 [21]	–	–	–
308.15	–	16.9 [16]	(14.9) [6]	15.0 ^b	–	–	15.1 [17] ^a	15.1 [15]	–
313.15	16.3 [11]	–	(14.8) [6]	15.1 ^b	15.2 [13] ^a	15.4 [21]	15.1 [17] ^a	–	14.5 [18]
318.15	–	17.3 [16]	(14.8) [6]	15.1 ^b	–	–	15.2 [17] ^a	–	–
323.15	–	–	(14.7) [6]	15.1 ^b	–	15.5 [21]	15.2 [17] ^a	15.2 [15]	15.2 [18]
328.15	–	–	(14.7) [6]	15.2 ^b	–	–	15.2 [17] ^a	–	–
333.15	16.4 [11]	–	(14.6) [6]	15.3 ^b	15.4 [13] ^a	15.8 [21]	15.2 [17] ^a	–	15.4 [18]
338.15	–	–	(14.6) [6]	15.3 ^b	–	–	15.3 [17] ^a	–	–
343.15	–	–	(14.6) [6]	15.3 ^b	–	15.9 [21]	15.3 [17] ^a	15.4 [15]	15.1 [18]
348.15	–	–	(14.6) [6]	15.4 ^b	–	–	15.4 [17] ^a	–	–
353.15	16.4 [11]	–	(14.5) [6]	15.5 ^b	15.2 [13] ^a	–	15.5 [17] ^a	15.8 [15]	–
T (K)	MEEA		DMEA		DEA		AEEA	<i>n</i> -PEA	MDEA
278.15	14.9 [14] ^a	–	–	12.8 [13] ^a	–	–	–	–	–
283.15	–	13.3 [9] ^c	–	–	–	–	–	–	–
288.15	14.5 [14] ^a	11.6 [9] ^c	–	12.4 [13] ^a	–	–	–	–	–
293.15	–	12.3 [9] ^c	–	–	–	–	–	–	–
298.15	14.8 [14] ^a	14.0 [9] ^c	11.3 [16]	13.6 [13] ^a	16.9 [11]	16.9 [16]	15.0 [20]	15.0 [15]	14.2 [12]
303.15	14.4 [14] ^a	14.1 [9] ^c	–	13.0 [13] ^a	16.5 [11]	–	15.0 [20]	15.2 [15]	14.0 [12]
308.15	–	–	13.3 [16]	–	–	16.5 [16]	–	–	–
313.15	14.5 [14] ^a	–	–	13.7 [13] ^a	16.6 [11]	–	15.0 [20]	15.2 [15]	14.4 [12]
318.15	–	–	14.6 [16]	–	–	17.3 [16]	–	–	–
323.15	–	–	–	–	17.0 [11]	–	15.1 [20]	–	14.3 [12]
333.15	14.8 [14] ^a	–	–	13.3 [13] ^a	17.6 [11]	–	15.3 [20]	15.9 [15]	14.6 [12]
343.15	–	–	–	–	18.3 [11]	–	15.5 [20]	–	14.8 [12]
353.15	15.2 [14] ^a	–	–	14.5 [13] ^a	17.9 [11]	–	–	16.3 [15]	15.5 [12]
T (K)	DEEA		DIPA		EDEA		TEA		<i>n</i> -BDEA
278.15	–	16.4 [14] ^a	–	–	–	–	–	–	–
288.15	–	14.3 [14] ^a	–	–	–	–	–	–	–
298.15	8.7 [16]	13.7 [14] ^a	–	–	14.4 [12]	13.6 [16]	16.0 [11]	16.0 [16]	15.0 [15]
303.15	–	11.5 [14] ^a	–	–	14.4 [12]	–	16.0 [11]	–	15.0 [15]
308.15	9.5 [16]	–	–	–	–	14.9 [16]	–	16.4 [16]	–
313.15	–	10.6 [14] ^a	–	–	14.4 [12]	–	16.0 [11]	–	15.1 [15]
318.15	11.7 [16]	–	12.4 ^d [19] ^a	–	–	15.4 [16]	–	16.9 [16]	–
323.15	–	–	12.9 [19] ^a	–	–	–	–	–	–
333.15	–	18.9 [14] ^a	13.7 [19] ^a	–	14.7 [12]	–	16.3 [11]	–	–
343.15	–	–	16.1 [19] ^a	–	–	–	–	–	–
353.15	–	19.0 [14] ^a	–	–	15.3 [12]	–	16.8 [11]	–	15.9 [15]

^a Calculated in terms of Eq. (13) using the Redlich–Kister coefficients reported in this reference and density data for water from Ref. [22].

^b Recalculated in terms of Eq. (13) using the Redlich–Kister coefficients reported in Ref. [6] and density data for water from Ref. [22].

^c Calculated in terms of Eq. (13) using the $V_1^{E,\infty}$ values reported in Ref. [9] and density data for water from Ref. [22].

^d At 318.65 K.

for a given component with V_i^* for the other component. Using the same Redlich–Kister coefficients, in Table 2 we present recalculated values of limiting partial molar volumes of water in 1-aminopropan-2-ol that nicely show the expected positive dependence on temperature.

5. Conclusion

We have condensed the various, equivalent forms found in the literature to express the Redlich–Kister expansion into Eqs. (4) and (8), which typify faces A and B of this flexible equation for describing excess molar thermodynamic data of binary liquid mixtures. Using a single mole fraction as variable, Eq. (4) for face A unfolds into:

$$V_m^E = x_1(1 - x_1) \sum_k A_k (2x_1 - 1)^k \quad (24)$$

$$V_m^E = x_2(1 - x_2) \sum_k A_k (1 - 2x_2)^k \quad (25)$$

Similarly, from Eq. (8) for face B:

$$V_m^E = x_1(1 - x_1) \sum_k B_k (1 - 2x_1)^k \quad (26)$$

$$V_m^E = x_2(1 - x_2) \sum_k B_k (2x_2 - 1)^k \quad (27)$$

The relationship $A_k = B_k (-1)^k$ obtained in Eq. (11) allows an easy comparison between coefficients reported in terms of one face and the corresponding coefficients calculated by software prepared for use in terms of the other face. Similar care should be taken when evaluating $V_1^{E,\infty}$ and $V_2^{E,\infty}$ in terms of fitted coefficients.

We demonstrate that $A_0 = B_0 = 4V_m^E$ (at $x_1 = x_2 = 0.5$).

We compiled literature data for the effect of temperature on the partial molar volume of water at infinite dilution in 15 aminoalkanols. These data (Table 2) strongly suggest a positive dependence on temperature, the only apparent exception being in 1-aminopropan-2-ol [6]. We show that the aforementioned dual aspect of the Redlich–Kister equation is most probably at the origin of these reported strange values, which after proper calculation yielded the expected positive dependence on temperature.

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