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Abstract

From a critical evaluation of the relevant literature, Harned coefficients at 25 °C and their corresponding trace activity coefficients are reported for 72 mixtures of strong aqueous electrolytes with a common ion. The work confirms the generality of Harned's rule in its simplest form, i.e. with only one (linear) term. Unless there are good grounds to suggest that significant changes in chemical speciation are occurring, experimental measurements that do not conform to the rule should be regarded with suspicion. We find that the estimates of errors made by authors of single potentiometric investigations are often as much as an order of magnitude too optimistic. Even the work of Harned and colleagues, which was typically of high quality, is sometimes much worse than is claimed or implied in the literature. The unfortunately common practice of introducing a quadratic term (as a second adjustable parameter) to extend Harned's rule is unjustified, within the true current limits of experimental error. Harned's rule with just one coefficient provides a description of aqueous-strong-electrolyte mixing thermodynamics which is widely applicable, satisfactorily accurate and without sign of any systematic problem.

Introduction

Although most systems of applied thermodynamic interest are mixtures, for obvious reasons of both logic and pragmatism, much greater theoretical and experimental attention is given to pure substances. A common belief is that physicochemical property simulation should be more troublesome with multi-component than single-component systems, and this is certainly the case with aqueous complexing systems. However, with aqueous strong electrolytes, so-called 'linear mixing rules' of various kinds have become well-established and imply the opposite, i.e. as long as the pure binary (one salt plus water) systems have already been characterised, the thermodynamic behaviour of mixtures is comparatively simpler and hence easier to model. Of all the mixing rules available to predict the thermodynamic properties of multicomponent strong electrolyte systems, one of the most valuable is Harned's rule for activity coefficients, described first by Harned[1,2] and subsequently explored and confirmed by many others.

Harned's rule is the method of choice whenever the equation can be parameterised from experimental measurements on mixtures.[3] In contrast, Zdanovskii's rule[4,5] lacks precision when used to calculate the separate activity coefficients for each component in a mixture (although it is more universally applicable, requiring only that the pure solutions have been characterised). Unfortunately, the Harned equation parameters – or so-called 'Harned coefficients' – are widely scattered in the literature, sometimes seriously inconsistent and often presented in different algebraic forms. Since the work of Harned and Robinson in 1968,[6] many experimental studies have been reported but there has been no comprehensive review, nor corresponding analysis and evaluation, of them. The purposes of the present work are accordingly fourfold: to assemble the available values, to compare them against published activity coefficient data for each respective solution mixture, to perform an up-to-date critical assessment and to determine some recommended values for practical use at convenient ionic strengths.

Harned's rule is of great importance on account of its remarkable generality and simplicity. It describes how the logarithm of the activity coefficient of one strong electrolyte, B, in a two-component solution mixture at constant ionic strength varies in direct proportion to the concentration of the other strong electrolyte, C (ref 7, p. 438):

$$\tag{1}$$

where   is the Harned coefficient and   is the concentration of C on the molal scale. Other forms of Harned's equation appearing in the literature include:

$$\tag{2}$$

$$\tag{3}$$

and

$$\tag{4}$$

where   is the ionic strength *contribution* of component C,   is the stoichiometric ionic strength and   is the ionic strength *fraction* of component C. These equations can of course be written in terms of natural logarithms but this is done only infrequently. Since the equations (1) to (4), and those with a natural log base, all apply at a given ionic strength, they differ (at that ionic strength) just in the magnitude of their coefficients   ,   and   .

To deal with observed deviations from the above equations (1) to (4), most notably for (a) the mixed systems {NaOH + NaCl}(aq) and {KOH + KCl}(aq), (b) systems with ions of higher charge and (c) solutions at high ionic strength, equations which include a quadratic term have also often been described. Equation (5) is an example.

$$\tag{5}$$

However, references in the literature to quadratic extensions such as appear in equation (5) find that the   coefficient is almost invariably small. This raises questions about its validity, as will be considered below.

Finally, but of great theoretical importance, Harned's rule leads directly to the evaluation of so-called 'trace activity coefficients', which are the extrapolated values for the activity coefficient of component B in the limit of zero concentration. In particular, from equation (2) for an infinitely small concentration of B in C such that   :

$$\tag{6}$$

As with mean activity coefficients of pure salt solutions, there is little theoretical guidance for the behaviour of the trace activity coefficient at high ionic strength. However, it follows from Debye-Hückel theory that under dilute conditions the trace activity coefficient of B in a solution of C is related to the trace activity coefficient of C in a solution of B at the same ionic strength by

$$\underline{\qquad\qquad} \quad \underline{\qquad\qquad} \tag{7}$$

where   and   are the algebraic charges on the cation and anion respectively. An excellent example of a real mixture displaying this behaviour, even up to high ionic strength, is {HCl + NaCl}(aq) (ref 7, p. 433). For many systems it is further observed that the trace activity coefficients fall between the activity coefficients of the pure salts at the same ionic strength. In cases where the two Harned coefficients   and   are not available from

experiment, they may be estimated using other equations. For example, by Zdanovskii's rule (see ref 8) or by the Pitzer equations (see ref 9–10).

The forms of Harned's rule shown above apply to ternary electrolyte mixtures. A logical extension to Harned's rule for representing the activity coefficient of an electrolyte in a multicomponent system is

$$\tag{8}$$

where parameter ⬚ is specific to component B in the presence of component X.[11] As ⬚ can be derived from measurements on ternary electrolyte solutions, equation (8) is predictive for mixtures comprising more than two electrolytes without the need for further adjustable parameters. Predictions with equation (8) have been found to agree with activity coefficients and solubility data for quaternary and higher order strong electrolyte mixtures within the estimated experimental uncertainty of the data.[11–16] This means that the Harned coefficients obtained in this work from common ion mixtures can be used more generally to describe strong electrolyte systems with any number of components at 25 °C.

Methodology

An extensive search of the chemical literature located more than 1,400 Harned coefficients from 93 references. These data have been assembled and entered into the FIZ database of our JESS (Joint Expert Speciation System)[17,18] software package. The references are listed in the Supplementary Material. For compatibility of the Harned coefficients with an existing database structure designed for other physicochemical properties, the concentrations of the mixture components in this database refer to the line having constant ionic strength connecting the endpoints (i.e. of the pure, binary solutions) and a concentration transform is applied when the electrolyte component is not 1:1. Concentrations can be specified in units of either 'm' ($mol.kg^{-1}$) or 'M' ($mol.dm^{-3}$). Other forms of Harned's rule appearing in the literature are transformed automatically into equation (2), the JESS canonical form.

A computer program was then developed to generate sets of activity coefficient data from each Harned parameter in the database (with activity coefficients of the pure salts calculated from the Pitzer equations) so that values calculated by it could be analysed and compared with the corresponding activity coefficient data previously assembled from the literature.[17] Database entries were assessed and weighted in the usual way.[17] Significant inconsistencies between data *under the same conditions* were resolved in favour of the work judged to be of better standard.

Further computer programs were then used to express every individual activity coefficient value in terms of both a Harned coefficient and a trace activity coefficient for all recognised aqueous strong electrolyte mixtures at a given I and having ⬚. The internal

consistency of the resulting data sets for given mixtures at each ionic strength could thus be examined. In this way, indisputable outliers were identified and eliminated. Averages and spreads for each Harned coefficient could finally be determined, yielding a recommended value for the trace activity coefficient under each particular condition along with a rough estimate of its uncertainty where possible. These uncertainty estimates were framed as maximum deviations, cognisant of the relatively small number of calculations being averaged in most cases.

Results and Discussion

In line with the majority of experimental investigations, we chose to examine the available data at 25 °C and at rounded ionic strengths of (0.1, 0.5, 1.0, 2.0 and 3.0) mol.kg^{-1}. No consideration has been given, and hence no conclusions can properly be drawn, for ionic strengths above 3.0 mol.kg^{-1}. A total of 72 ternary common ion mixtures were found with sufficient data to permit evaluation of the Harned coefficient for at least one rounded ionic strength. Of these 72 mixtures, there were 23 'complementary salt pairs', i.e. in which the activity coefficient of electrolyte 1 had been characterised (as B) in the presence of electrolyte 2 (as C) and *vice versa*. Over 500 Harned coefficients and trace activity coefficients were obtained in this way. The results are summarised in Table 1, which for simplicity and the avoidance of mistakes employs JESS notation to identify the systems. The Harned coefficients,   , are given for the JESS canonical form, equation (2). Electrolyte mixtures with a common ion are represented as 'ionB|electrolyteC' where ionB is the distinctive ion of the target electrolyte (i.e. to which the activity coefficient refers) and electrolyteC is the other (diluting) electrolyte in the mixture. For example, 'H+1|Na+1/Cl-1' refers to the activity coefficient of HCl in a mixture with NaCl and its complementary partner 'Na+1|H+1/Cl-1' refers to the activity coefficient of NaCl in a mixture with HCl. Note also that in the specification of a non-1:1 electrolyteC charge balance is assumed implicitly.

<Insert Table 1 near here>

Inspection of Table 1 shows that Harned's rule provides a highly satisfactory description of the activity coefficients for most common ion mixtures: in about 85% of cases the estimated standard deviation of the trace activity coefficient is less than 0.01 (estimated maximum deviation ≤ 0.02), which is a realistic uncertainty for activity coefficients of many independently-measured pure electrolyte solutions. Thus, only one adjustable parameter is necessary to achieve good agreement with experimental data. Remarkably good results are found in particular at $I = 1.0$ mol.kg^{-1} (Figure 1).
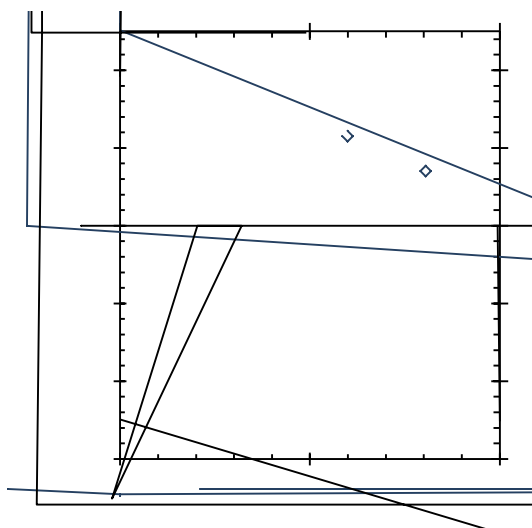
Figure 1. at $I = 1.0$ mol.kg^{-1} and $t = 25$ °C with M = {Mg,Ca,Sr,Ba,Mn,Ni} (from top to bottom). Each series is displaced downward 0.01 for clarity. Data sources: Mg[19]; Ca[20,21]; Sr[22–24]; Ba[22,25–28]; Mn[29,30]; Ni[31–32]. Slopes of the straight lines correspond to Harned coefficients.

The most accurate data involve mixtures of HCl(aq) with a large variety of salts, providing a backbone of dependable measurements which is attributable to the effectiveness below 4 mol.kg^{-1} of the so-called 'Harned cell' – Ag(s)/AgCl(s) | HCl(aq) | H$_2$(g) | Pt(s). Fewer than half a dozen instances had a spread of calculated trace activity coefficients > 0.02. Given that trace activity coefficients are obtained from extrapolations of the target ion to zero concentration, the outcome summarised in Table 1 constitutes a robust validation of the generality of Harned's rule.

Several aspects of the results shown in Table 1 are noteworthy, the initial three having fundamental implications. First, Harned's rule works for strong electrolytes regardless of their charge type, i.e. it is not confined to 1:1 electrolytes. Second, the trace activity coefficients change systematically with ionic strength and they tend to change only slowly above $I = 1.0$ mol.kg^{-1}. Third, in more than half of the complementary salt pairs, the trace activity coefficients of electrolyte 1 and electrolyte 2 (in mixtures with the other electrolyte) are in close agreement with equation (7) . Fourth, the high percentages shown in Table 1 for some mixtures, e.g. Mg+2|K+1/Cl-1 and K+1|Mg+2/Cl-1, are no cause for concern: they arise simply because (unusually) the activity coefficient curves of the two neat, binary electrolytes intersect; this happens at about the ionic strength in question (so, in calculating the percentage, the divisor tends to zero, making any change whatsoever large in fractional terms).

The results in Table 1 provide a useful set of parameters for the prediction of activity coefficients in mixtures of aqueous strong electrolytes. The overall sense of orderly behaviour, despite the diversity of systems and sources, supports the notion that Harned's rule is widely applicable. The random nature of the worst-fitting results suggests that experimental error is the main cause of deviations from linear behaviour.

A number of systems where the data were poorly fitting were examined in detail. With the system {NaBr + LiBr}(aq) at various ionic strengths, the activity coefficients of NaBr from Yan[33] do not accord with Harned's rule: the corresponding parameter values vary systematically at given ionic strength. This may be due to the type of ion selective electrode used. Similar issues were found with the data for {NaF + NaClO$_4$}(aq) at lower ionic strengths from Hernandez-Luis et al.[34] who also used an ion selective electrode but in this case we attribute the waywardness to anomalies which these authors themselves identified.

Linear or quadratic dependence?

Following what appears to be the mistaken lead by Harned and Cook, many reports in the literature concerned with the thermodynamics of mixing aqueous strong electrolytes reach a categorical conclusion in favour of the existence of the quadratic / term in equation (5). A list of such statements is given in the Supplementary Material. However, it is found in almost all of such cases, and often acknowledged, that the observed curvature is minor: for example, < 2 % in the study by Harned and Harris;[35] too small for quantitative evaluation by Robinson and Lim;[36] "even if and terms are required to represent the experimental results, it is found that they are extremely small" by Robinson and Stokes;[7] "reliable estimates of activity coefficients for the salt could still be obtained … by omitting the quadratic term" by Khoo et al.;[37] *etc*. It is against this background that the results of the following case studies are most pertinent. They suggest not only that terms are unjustified but also that quadratic terms are being used to over-fit the experimental data, sometimes blatantly.

Case Study 1: {NaOH + NaCl}(aq)
The system {NaOH + NaCl}(aq) is rare amongst strong electrolyte mixtures in the sense that the activity coefficients of both substances can be measured potentiometrically and that at least three independent reputable investigators have examined the system under similar conditions. This system is of special interest for various reasons but particularly because it deviates without doubt from Zdanovskii's rule[5,38] and it shows a high uncertainty in the trace activity coefficient (0.05) at $I = 3.0$ mol.kg^{-1} in Table 1. Comparing the results of the three independent studies reveals discrepancies between the reported activity coefficients (Figure 2) that become glaring when the data are plotted so as to reveal the Harned slopes (Figure 3).
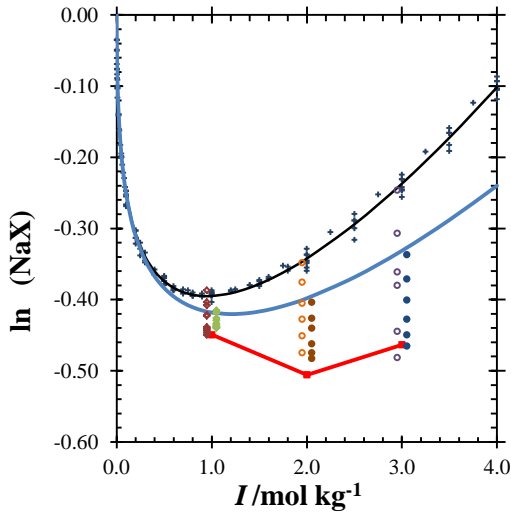
Figure 2. Results from Case Study 1 - natural logarithms of the activity coefficients for the system {NaOH + NaCl}(aq), showing activity coefficient curves for pure NaOH(aq), , as the upper black line, for pure NaCl(aq), , as the middle blue line and for the trace activity coefficient of NaCl(aq),), as determined by Falciola et al.[39] as the lower two red straight line segments. Values for (open symbols) and (filled symbols) at $I = 1.0$ mol.kg^{-1} [40] and $I = (2.0$ and $3.0)$ mol.kg^{-1} [41] are displaced to either side by 0.05 mol.kg^{-1} for clarity of representation.
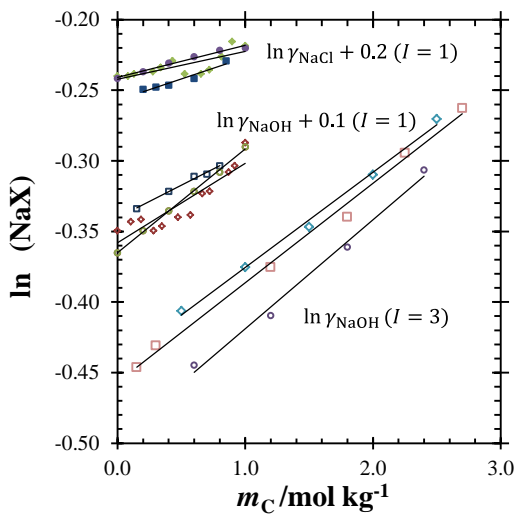
Figure 3. Additional results from Case Study 1 - natural logarithms of the activity coefficients for the system {NaOH + NaCl}(aq), showing linear fits to three independent sets of experimental data, together with the scatter of measured points. Data at $I = 3.0$ mol.kg^{-1} come from Harned and Harris[35] (diamonds), Falciola et al.[39] (squares) and Mironov and Sokolova[41] (circles). Data in the top two sets come from Harned and Cook[40] (diamonds), Falciola et al.[39] (squares) and Mironov and Sokolova[41] (circles).

Taken individually there would be little reason to discard any of these studies: the authors are all of good reputation and, within the limits discussed below, their results appear to be

internally consistent. However, while it seems clear that Harned's rule is obeyed at all ionic strengths within the limits of experimental error (Figure 3), the slopes of the trend-lines fitted by linear regression – corresponding to the Harned coefficients – differ significantly; this systematic effect is clearly the main problem of practical importance but the scatter of the data points around the lines even casts doubt the precisions claimed by the authors (0.2 mV Mironov and Sokolova,[41] 0.5 mV Harned and Cook[40] and 0.34 Falciola et al.[39]). The frequently-referred-to curvature of Harned and Cook's data is seen in Figure 3 to be in sharp contrast to the other results and one can therefore conclude beyond reasonable doubt that it is artefactual. Falciola et al. [39] on the other hand seem too conveniently ambivalent when they opine "for both components the behaviour can, with a reasonable approximation, be represented by Harned's rule (linear dependence of lg γ on m), although a quadratic-type dependence is more appropriate". In our view, the Falciola et al. [39] data are linear within their experimental error.

From an overall perspective, the three studies all conclusively agree on one key point: that both trace activity coefficients fall well below the activity coefficients of the pure systems at the same ionic strength, making this system anomalous among strong electrolyte mixtures. Exact quantification of the trace activity coefficients is made difficult because the measured activity coefficients (in terms of natural logs) are discrepant by as much as 0.01 at $I = 1$ mol.kg^{-1} and 0.05 at $I = 3$ mol.kg^{-1}. This belies the implied precision of activity coefficients in the literature, which is typically reported as three or four decimal places – see ref. 42 for instance. In fact these discrepancies in the activity coefficients of ternary mixtures between independent investigators are similar in size to the differences between critically-assessed values in JESS for the binary salts from independent workers, as can be seen in Figure 2 scattered around the (topmost) curve for pure NaOH(aq).

This is not the first time Harned's results have been questioned. Pitzer and Kim (ref 43, p. 5707) commented that Harned and Gancy's[44] results for {HCl + KCl}(aq) may be subject to larger error than they assumed. This conclusion was corroborated by citing several sources: Lietzke and O'Brien[45] found that both activity coefficients in the mixture obeyed Harned's rule in linear form; mixing in the common ion system {HBr + KBr}(aq)[46] was more consistent with the careful work of Guntelberg;[47] and Bates et al.[48] have shown that the Ag-AgCl electrode of the kind used by Harned and Gancy[44] is more erratic than had been thought at the time. All these factors lead us to reject Harned and Cook's[40] data from our analysis. We have also favoured the results of Falciola et al.[39] over those of Mironov and Sokolova[41] since, in the same study by the latter authors, the data for the corresponding K$^+$ system show some worrying inconsistencies.

Case study 2: {KBr + K$_2$SO$_4$}(aq)
Careful consideration of the recent work by Zhang et al.,[49] depicted in Figure 4, provides a graphic illustration of the over-fitting to be found in this area of research.
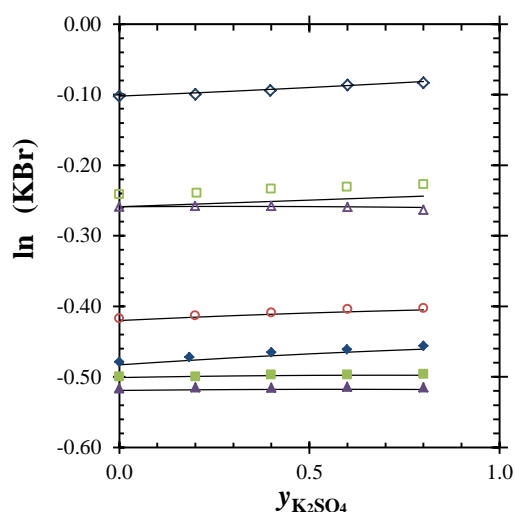
Figure 4. Results from Case Study 2 - activity coefficient change in mixtures of {KBr + K$_2$SO$_4$}(aq) as reported by Zhang et al.[49] for ionic strengths (top to bottom) I = (0.01, 0.1, 0.2, 0.5, 1.0, 2.0, 3.0) mol.kg^{-1}, with lines indicating fits calculated from parameters reported by Zhang et al.[49]

The authors used a 'battery cell without liquid junction', designated as K-ISE|KBr(m$_1$),K$_2$SO$_4$(m$_2$)|Br-ISE, to measure the activity coefficients in mixtures of KBr(aq) and K$_2$SO$_4$(aq) with a precision of 0.1 mV, typical of such ion-selective electrodes. The data at each ionic strength were fitted to the equation:

i.e. a function with two adjustable parameters, and , describing the change in away from the activity coefficient of the neat solution, , as a function of the ionic strength fraction of K$_2$SO$_4$, . These fits show no sign whatsoever of a systematic effect that would warrant a second adjustable parameter. By eye, the lines look as straight as could be expected within the scatter of small random effects. There is some indication, one might conclude, of a positive slope at lower ionic strengths but even this seems doubtful given that at 0.2 the slope is negative. Accordingly, Harned's rule with one (linear) parameter describes these measurements more than adequately. Moreover, the effect of ionic strength on mixing behaviour is negligible or close to it, in line with much evidence that at higher concentrations Harned coefficients become less dependent on concentration.[50] We also note, incidentally, that the experimental data at 0.1 are not matched using the author's tabulated parameters and that their value of (in their Table 1) is 0.934 at 0.0052 mol.kg^{-1} compared to literature values of 0.926 or 0.927.[51–54]

Case Study 3: {CaBr$_2$ + CaCl$_2$}(aq)
The system {CaBr$_2$ + CaCl$_2$}(aq) has been studied by Tialowska-Mocharla and Atkinson[42] with Ca^{2+} and Br$^-$ ion-selective electrodes. The work appears to have been carefully performed but, regrettably, several aspects of it are problematic. First, the potential of the cell

was reported to be stable to within 0.5 mV, corresponding to an uncertainty of +/- 0.005 in the activity coefficients even though Tialowska-Mocharla and Atkinson[42] give 4 significant figures in the logarithms of their activity coefficients. Second, analysis of their data with the equations of Pitzer[43] leads to ternary interaction parameters that vary randomly with ionic strength, and with the (Cl^-, Br^-) interaction parameters tending to be large and positive, in contrast to literature reports that they should be independent of ionic strength and zero-valued[43,55] or small and negative.[56,57] Finally, regarding the significance of the   parameters reported by Tialowska-Mocharla and Atkinson,[42] consider Figure 5.
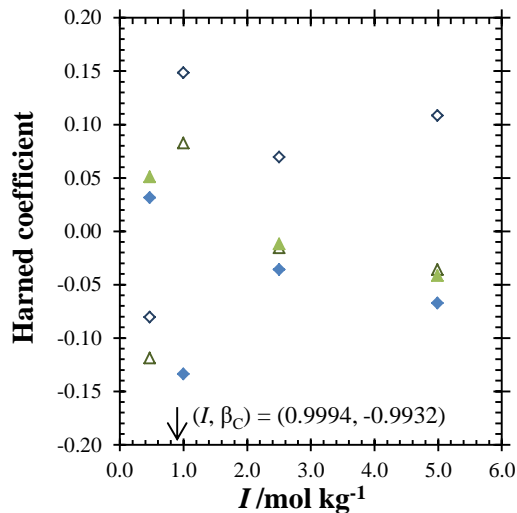
Figure 5. Harned coefficients (, open diamond;   , filled diamond;   , open triangle;   , filled triangle) for the system $\{CaBr_2 + CaCl_2\}(aq)$ reported by Tialowska-Mocharla and Atkinson[42] according with

and

.

The   values change systematically but the   values are evidently scattered. It would be difficult to interpolate or extrapolate these   values to other ionic strengths and be confident of achieving anything meaningful.

Case Study 4: $\{CsCl + MgCl_2\}(aq)$
As a last example, the system $\{CsCl + MgCl_2\}(aq)$ has been studied by Hu et al.[58] with Cs^+ ion-selective electrode. They concluded that Harned's rule with quadratic term fitted the experimental activity coefficients very well. Their Harned coefficients are shown in Figure 6.
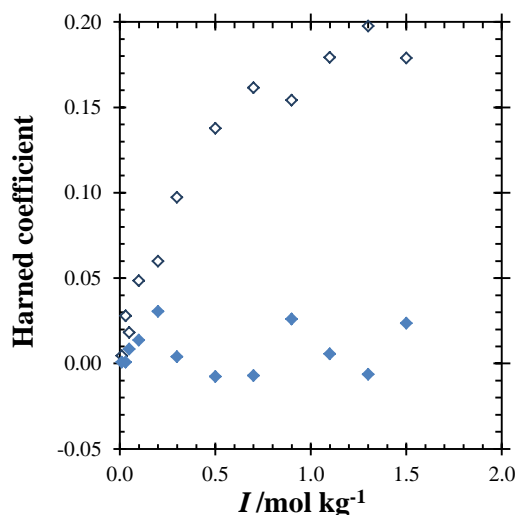
Figure 6. Harned coefficients (, open diamond;   , filled diamond) for the system {CsCl + MgCl$_2$}(aq) reported by Hu et al.[58] according with

.

Although the   values are subject to some scatter, they at least display a plausible trend with ionic strength. However, the   values are small and flip randomly between positive and negative values. This strongly suggests that the   values are not significant and that the curvature in the activity coefficients described by Hu et al.[58] is more likely due to a systematic error, probably of experimental origin. These results again illustrate the widespread over-fitting in the literature that we have identified in general.

Conclusion

In spite of frequent usage of the quadratic extension to Harned's rule in the literature (see examples in the Supplementary Material), the resulting   parameters often display unrealistic characteristics. In particular, the   values do not tend to differ from zero by statistically-significant amounts nor do they vary smoothly with the ionic strength of the system. We consider the second point to be particularly damning. Scattered   values are unquestionably contrary to what one would expect if the underlying data possessed significant / real quadratic dependence. Accordingly, in such cases the reported curvature of the activity coefficients with respect to concentration has to be rejected.

There is a strong tendency in the literature, within the area of solution chemistry at least, to pick out the data of highest quality (as critically assessed) and then to treat such data as if they were error-free. However, later independent investigations of similar or better quality often yield results that differ significantly from the earlier selected values, as illustrated in Figure 2. We have reached similar conclusions in other contexts, even for properties like density which can nowadays be measured routinely to high precision.[59] The problem is that no matter how carefully experimental work is performed it is impossible to avoid systematic errors. Such errors cannot be quantified by internal scrutiny. In other words, experts tend to

over-emphasise the accuracy of individual work (often, but not always, their own). As a common consequence it becomes difficult for them, and others following them, to 'see the forest for the trees'. A broader perspective in the context of Harned's rule makes it clear that using the β parameters in equation (5) to fit small observed effects is not justified and has been a major distraction in the literature. Apart from enticing subsequent investigators to over-fit their experimental data, theoretical frameworks with an abundance of empirical parameters (such as occur in the Pitzer equations) can accommodate these spurious effects and, hence, suffer from various deleterious consequences associated with too many degrees of freedom.

Once it is accepted that aqueous strong electrolytes obey Harned's rule (with one parameter), **it becomes much easier to identify and to reject anomalous experimental measurements.** This is the great advantage of having a simple theoretical equation with a minimum of flexibility. If they do not conform to Harned's rule, experimental measurements of electrolyte solution mixing should be regarded with suspicion unless there are good grounds to suggest significant changes in chemical speciation. It may one day become possible to characterise more subtle real effects but considerably improved experimental techniques, much superior to those available today, will be needed. Until then, it is safer to conclude that fits involving an extended Harned equation with a quadratic term are merely following systematic errors. We have found that the estimates of errors made by authors of single potentiometric investigations are typically as much as an order of magnitude too optimistic. Although the experimental work of Harned and colleagues was usually of high quality, particularly given the technological limitations of their time, the accuracy often claimed in the literature for their results is worse, and sometimes much worse, than stated.

We are not at all suggesting that hypothetical models should take precedence over observation but rather depending on the axiom that scientific knowledge comes from agreement between experiment and theory. When, and only when, such agreement is reached is it sensible to assess the overall result in terms of the simplicity of the model's premises, the patterns evident in the data and the likely magnitude of experimental error. Judged by these criteria, this work shows that Harned's rule with only one coefficient provides a description of aqueous-strong-electrolyte mixing thermodynamics which is widely applicable, satisfactorily accurate and without sign of any systematic problem.

On the other hand, there is an undeniable weakness in using the Harned coefficients reported here which arises because they each refer only to a particular ionic strength and to only one complementary salt pair. This is especially worrying because these values are, of course, obtained by extrapolation and often by thermodynamic calculation rather than direct measurement. The key issue in understanding the physicochemical behaviour of strong electrolytes remains: there is currently no sound fundamental model for their specific ion interactions.[60] Such a model would accurately predict and smooth Harned coefficients as a thermodynamically-consistent function of ionic strength. Until then, application of Harned's rule with one linear parameter to endorse experimentally measured data will be the outcome from this work of most benefit.

Acknowledgement

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