Revision of the Osmotic Coefficients, Water Activities and Mean Activity Coefficients of the Aqueous Trivalent Rare Earth Chlorides at $T = 298.15$ K

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Abstract The analysis by Wang et al. (J. Solution Chem. 35:1137–1156, 2006) of the osmotic and activity coefficients of fourteen of the aqueous rare earth trichlorides at the temperature 298.15 K is based mainly on the isopiestic results reported by Spedding et al. (J. Chem. Eng. Data 21:341–360, 1976) and Rard and Spedding (J. Chem. Eng. Data 27:454–461, 1982), but corrected for more recent critical reviews of the osmotic coefficients of the KCl(aq) and CaCl$_2$(aq) reference solutions used in these measurements. Also included were the potential difference (emf) measurements by Malatesta et al. (Phys. Chem. Chem. Phys. 4:121–126, 2002) for LaCl$_3$(aq) using cells with a liquid ion-exchange membrane electrode and earlier results for concentration cells with transport for the rare earth trichlorides. Our reexamination of osmotic coefficients calculated from these potential differences of cells with a liquid ion-exchange electrode indicates their values are skewed relative to osmotic coefficients obtained more directly from the isopiestic measurements, being significant higher for molalities between (0.025 and 0.3) mol·kg^{-1} and significantly lower at higher molalities. As a consequence, the combined results yielded mean, molality-based, activity coefficients that are systematically too large for the light and intermediate rare earths. Although restricted to a narrower molality region of about (0.02–0.04) mol·kg^{-1}, potential difference measurements for aqueous rare earth trichlorides from concentration cells with AgCl(s) electrodes appear to yield more reliable results. The extended ion-interaction model parameters of Wang et al. have been reevaluated using the cited isopiestic results together with osmotic coefficients derived from the concentration cells with transport as reported by Spedding et al. (J. Chem. Eng. Data 21:341–360, 1976). Mean activity coefficients calculated using these new model parameters should be more accurate than those previously reported.

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1 Introduction

In 1976 Spedding et al. [1] reported the results of extensive isopiestic measurements for the aqueous rare earth trichlorides $RECl_3(aq)$, where RE = (La, Pr, Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, and Y), from $\sim (0.1$ to $0.2)$ mol·kg^{-1} to the saturated solution molalities at 298.15 K. Subsequently, Rard and Spedding [2] reported more extensive isopiestic results for $YCl_3(aq)$ including two measurements in the oversaturated (supersaturated) molality region. They [1, 2] represented the resulting molality-based osmotic coefficients with equations containing the Debye–Hückel limiting law term together with seven additional empirical terms in various fractional powers of the molality. These equations were then used to calculate the water activities and the mean, molality-based, activity coefficients of the solutes. As noted by Spedding et al. [1], earlier isopiestic measurements of Mason [3] for several of the $RECl_3(aq)$, where RE = (La, Ce, Pr, Nd, and Y), from ~ 0.06–0.07 to ~ 2.0–2.1 mol·kg^{-1} were made almost two decades before the development of ion-exchange methods to purify rare earths and therefore most of Mason's rare earth samples probably were contaminated with several percent of the neighboring rare earths.

Pitzer et al. [4] represented the osmotic coefficients of the $RECl_3(aq)$ [1] with his ion-interaction model (Pitzer equation) [5], but the standard deviations are two to six times larger than those for the empirical equations of Spedding et al. [1]. The larger standard deviations in the ion-interaction model fits are not surprising because there are between 13.5 and 15.5 water molecules per RE^{3+} in the saturated solutions (less than twice the number of waters needed to fill just the primary hydration spheres of the RE^{3+} ions [6, 7]), and extensive water sharing/ion pairing must of necessity be present in the concentrated $RECl_3(aq)$ solutions. Consequently, models based in part on essentially the same assumptions as Debye–Hückel type models, such as Pitzer's ion-interaction model, will not be able to accurately represent these thermodynamic properties at such high molalities.

Archer [8] proposed an extended version of Pitzer's ion-interaction model in which the $C_{M,X}$ coefficient is no longer a constant but is now assumed to have a dependence on the ionic strength I. Lacking any guidance from theory, Archer assumed that $C_{M,X}$ has the same type of exponential functional dependence on $I^{1/2}$ as the $B_{M,X}$ term. By including the additional $C_{M,X}^{(1)}$ adjustable parameter, Archer's model is able to accurately represent thermodynamic properties of many aqueous electrolytes to significantly higher ionic strengths. Oakes et al. [9] further extended this model with an additional $C_{M,X}^{(2)}$ term having the same exponential dependence on $I^{1/2}$ in order to represent molality-based osmotic coefficients of $Ca(NO_3)_2(aq)$ to ≈ 20 mol·kg^{-1} where the water : ion molar ratio is less than one-to-one.

Pitzer et al. [10] proposed a more general approach, where the exponential terms are a function of $I^{1/2}$ for $B_{M,X}^{\phi}$, of I for $C_{M,X}^{\phi}$, of $I^{3/2}$ for $D_{M,X}^{\phi}$, etc. By using several of these additional higher-order coefficients, molality-based osmotic and mean activity coefficients of aqueous electrolytes can be accurately modeled to extreme molalities where water sharing occurs between ions but without explicitly including ionic association in the model. Also, by using this form with the exponential terms being dependent on $I^{n/2}$ (where $n = 1$ for "B" terms, $n = 2$ for "C" terms, $n = 3$ for "D" terms, etc.), much simpler expressions

are obtained for the natural logarithm of the mean activity coefficients compared to the corresponding expressions that would be obtained if all of the higher order terms were exponential functions of $I^{1/2}$.

Wang et al. [11][1] reanalyzed the molality-based osmotic coefficients ϕ and mean activity coefficients $\gamma_{\pm}$ of RECl$_3$(aq) solutions at the temperature 298.15 K. The source isopiestic results [1, 2] are based on use of the molality-based osmotic coefficients of KCl(aq) and CaCl$_2$(aq) as isopiestic (equal water activity at equilibrium) reference solutions. Some years after the isopiestic results were reported [1, 2], the osmotic coefficients of both of these reference solutions were reviewed and critically assessed [12, 13] based on much larger sets of thermodynamic measurements. Based on these more recent assessments, the recommended osmotic coefficients of KCl(aq) [12] and those of CaCl$_2$(aq) [13] over most of the molality range were changed by about 0.1 % to 0.3 %, except for CaCl$_2$(aq) solutions between $0.7 \leq m/\text{mol·kg}^{-1} \leq 1.9$ where larger differences of up to twice this size occur. Wang et al. [11] recalculated the molality-based osmotic coefficients ϕ of the RECl$_3$(aq) based on these revised results for the KCl(aq) and CaCl$_2$(aq) reference solutions, and also included values of ϕ from the cell potential difference (emf) measurements for concentration cells with transport for various RECl$_3$(aq) as analyzed by Spedding et al. [1] and from a cell with a liquid ion-exchange membrane electrode of Maletesta et al. [14] for LaCl$_3$(aq) solutions, as revised in a later publication [15]. They then represented these osmotic coefficients using an extended form of Pitzer's ion-interaction model of the form described by Pitzer et al. [10].

Potential difference measurements using liquid ion-exchange membrane cells have the advantage that they can be used for measurements to much lower molalities than traditional concentration cells with Ag/AgCl electrodes, where chloride ions from electrode dissolution can make a significant contribution to the total chloride molality in the dilute solutions. However, they have several disadvantages including slow drifts of the emfs with time at low and high molalities and systematic offsets when new measurements were made, although Malatesta et al. [14] reported that the changes of emf with molality were consistent for the various series of measurements. They also noted that the activity coefficients for LaCl$_3$(aq) from their measurements are not consistent with those from isopiestic measurements or emfs of the concentration cells with transport since the activity coefficients from emfs for the cell with a liquid ion-exchange membrane electrode refer to LaCl$_3 \cdot n$H$_2$O where n is the number of moles of water molecules transported into the liquid ion-exchange membrane electrode per mole of LaCl$_3$. The value of n was not measured directly but was estimated to be $n \approx 24$ by comparison with the isopiestic results [1].

The mean molality-based activity coefficients from the equation and parameters of Wang et al. [11] for the light and intermediate RECl$_3$(aq), RE = (La, Pr, Nd, Sm, Eu, Gd, Tb), are significantly higher than the corresponding values reported by Spedding et al. [1], as are the osmotic coefficients calculated from their model parameters from (0.025 to 0.2) mol·kg^{-1} where they are affected most strongly by the potential difference results of Malatesta et al. [15]. Matatesta et al. [14, 15] represented relative mean activity coefficients from their measurements using the standard form of Pitzer's ion-interaction model [5]. We calculated osmotic coefficients from both the original set of ion-interaction model parameters that pertain to LaCl$_3 \cdot n$H$_2$O being transported into the liquid ion-exchange membrane electrode [14] and their revised parameters that were corrected for the water transport across the membrane [15], and in Table 1 compare them with the smoothed osmotic coefficients reported by Spedding et al. [1] from their isopiestic results and also

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[1] Equation 2 of reference [11] is missing a factor of $+\{2(v_{\text{M}}v_{\text{X}})/v\}m\beta_{\text{M,X}}^{(1)}\exp(-\alpha_{B1}I^{1/2})$.

Table 1 Comparison of molality-based osmotic coefficients of LaCl$_3$(aq) at 298.15 K from the original model of Malatesta et al. [14], the revised model of Malatesta et al. [15], from emfs (cell potential differences) for concentration cells with transport as analyzed by Spedding et al. [1] and their smoothed results from isopiestic measurements

m/mol·kg^{-1}	ϕ Malatesta et al. [14]	ϕ Malatesta et al. [15]	ϕ Spedding, emf [1]	ϕ Spedding, isopiestic [1]
0.005	0.8640	0.8646	0.8712	
0.010	0.8349	0.8362$_5$	0.8428	
0.015	0.8190	0.8212	0.8257	
0.020	0.8089	0.8118	0.8134	
0.025	0.8018	0.8052	0.8037	
0.030	0.7968	0.8012	0.7957	
0.1	0.7753	0.7920		0.7760
0.2	0.7704	0.8052		0.7953
0.3	0.7732	0.8280		0.8267
0.4	0.7817	0.8592		0.8637
0.5	0.7943	0.8976		0.9046
0.6	0.8092$_5$	0.9416		0.9487
0.7	0.8253	0.9901		0.9958
0.8	0.8414	1.0420		1.0454

those derived by them from the potential differences for concentration cells with transport. Malatesta et al. [14, 15] did not report the value of the Debye–Hückel constant used in their models; we assumed the value $A_\phi = 0.3915$ kg$^{1/2}$·mol$^{-1/2}$, which is intermediate between 0.391 and 0.392 kg$^{1/2}$·mol$^{-1/2}$ in common usage at that time. As can be seen from Table 1, the molality dependence of the osmotic coefficients from the results of Malatesta et al., whether corrected for water transport or not, are inconsistent with both the isopiestic results and the concentration cell with transport results.

The relation for calculation of mean activity coefficients from the osmotic coefficients is:

$$\ln\gamma_\pm = \phi - 1 + \int_0^m \{(\phi - 1)/m\}\mathrm{d}m \tag{1}$$

from which it can be seen that too high values of the osmotic coefficients at low molalities, as calculated from Malatesta's ion-interaction model parameters [15], will shift the integral term in the positive direction and yield mean activity coefficients that are systematically too large. Because of this herein we reevaluate the parameters of an extended ion-interaction model [10, 11] for the RECl$_3$(aq) at 298.15 K using the published isopiestic results and concentration cell with transport results [1], but not including the liquid ion-exchange membrane cell results.

2 The Extended Pitzer Model

The extended ion-interaction model of Pitzer et al. [10] for the osmotic coefficient, which was accepted by Wang et al. [11] for the RECl$_3$(aq), can be written in the following general form:

$$\phi = 1 - |z_M z_X| A_\phi I^{1/2} \big/ (1 + bI^{1/2}) + (2\nu_M \nu_X / \nu) m \left\{ \beta_{M,X}^{(0)} + \beta_{M,X}^{(1)} \exp(-\alpha_{B1} I^{1/2}) \right\}$$
$$+ \left(4(\nu_M \nu_X)^{3/2} / \nu \right) m^2 \left\{ C_{M,X}^{(0)} + C_{M,X}^{(1)} \exp(-\alpha_{C1} I) + C_{M,X}^{(2)} \exp(-\alpha_{C2} I) + C_{M,X}^{(3)} \exp(-\alpha_{C3} I) \right\}$$

(2a)

where M denotes the cation (in this case a RE^{3+}) and subscript X the anion (Cl^-), z_i the valence (with sign) of ion i ($i = M$ or X), $b = 1.2$ $kg^{1/2} \cdot mol^{1/2}$, ν_i denotes the number of ions of type i formed by the stoichiometric dissociation of one formula unit of the electrolyte, and $\nu = \nu_M + \nu_X$ is the total (stoichiometric) ionization number of an electrolyte of general formula $M_{\nu_M} X_{\nu_X}$. For the $RECl_3(aq)$, $\nu_{RE} = 1$, $\nu_{Cl} = 3$, $\nu = 4$, $z_{RE} = +3$, and $z_{Cl} = -1$ so this equation becomes:

$$\phi = 1 - 3A_\phi I^{1/2} \big/ (1 + bI^{1/2}) + (3/2)m \left\{ \beta_{M,X}^{(0)} + \beta_{M,X}^{(1)} \exp(-\alpha_{B1} I^{1/2}) \right\}$$
$$+ \left(3^{3/2} \right) m^2 \left\{ C_{M,X}^{(0)} + C_{M,X}^{(1)} \exp(-\alpha_{C1} I) + C_{M,X}^{(2)} \exp(-\alpha_{C2} I) + C_{M,X}^{(3)} \exp(-\alpha_{C3} I) \right\}$$

(2b)

The corresponding general expression for the mean activity coefficient is:

$$\ln \gamma_\pm = - |z_M z_X| A_\phi \{ I^{1/2} / (1 + bI^{1/2}) + (2/b)\ln(1 + bI^{1/2}) \}$$
$$+ \{ 2(\nu_M \nu_X)/\nu \} m [2\beta_{M,X}^{(0)} + (\beta_{M,X}^{(1)}) \{ g(\alpha_{B1} I^{1/2}) + \exp(-\alpha_{B1} I^{1/2}) \}]$$
$$+ \{ 2(\nu_M \nu_X)^{3/2}/\nu \} m^2 [3C_{M,X}^{(0)} + C_{M,X}^{(1)} \{ g(\alpha_{C1} I) + 2\exp(-\alpha_{C1} I) \}$$
$$+ C_{M,X}^{(2)} \{ g(\alpha_{C2} I) + 2\exp(-\alpha_{C2} I) \} + C_{M,X}^{(3)} \{ g(\alpha_{C3} I) + 2\exp(-\alpha_{C3} I) \}]$$

(3a)

which for the $RECl_3(aq)$ becomes:

$$\ln \gamma_\pm = - 3A_\phi \{ I^{1/2} / (1 + bI^{1/2}) + (2/b)\ln(1 + bI^{1/2}) \}$$
$$+ (3/2)m [2\beta_{M,X}^{(0)} + (\beta_{M,X}^{(1)}) \{ g(\alpha_{B1} I^{1/2}) + \exp(-\alpha_{B1} I^{1/2}) \}]$$
$$+ (3^{3/2}/2)m^2 [3C_{M,X}^{(0)} + C_{M,X}^{(1)} \{ g(\alpha_{C1} I) + 2\exp(-\alpha_{C1} I) \}$$
$$+ C_{M,X}^{(2)} \{ g(\alpha_{C2} I) + 2\exp(-\alpha_{C2} I) \} + C_{M,X}^{(3)} \{ g(\alpha_{C3} I) + 2\exp(-\alpha_{C3} I) \}]$$

(3b)

where

$$g(x) = (2/x^2)\{ 1 - (1 + x)\exp(-x) \}$$

(4)

and $x = (\alpha_{B1} I^{1/2}, \alpha_{C1} I, \alpha_{C2} I,$ or $\alpha_{C3} I)$.

The parameters of Eq. 2b were evaluated using osmotic coefficients from the available isopiestic studies [1, 2], recalculated using more recent critically assessed osmotic coefficients for the reference standards KCl(aq) [12] and $CaCl_2$(aq) [13] as described by Wang et al. [11], and osmotic coefficients for concentration cells with transport as analyzed by Spedding et al. [1]. Following Pitzer's recommendation for electrolytes when at least one ion is univalent [5], α_{B1} was fixed at $\alpha_{B1} = 2.0$ $kg^{1/2} \cdot mol^{-1/2}$. Few systems have been modeled by this generalized ion-interaction model, mainly because Prof. Pitzer passed away shortly after Ref. [10] was submitted for publication. Pitzer et al. [10] used $\alpha_{C1} = 0.15$ $kg \cdot mol^{-1}$ and $\alpha_{C2} = 0.25$ $kg \cdot mol^{-1}$ for modeling the $CaCl_2$(aq) system and for $MgCl_2$(aq) for the full molality range, and these values were accepted by Wang et al.

[11], along with $\alpha_{C3} = 0.35$ kg·mol^{-1}, for modeling the osmotic coefficients of the RECl$_3$(aq), RE(ClO$_4$)$_3$(aq) and RE(NO$_3$)$_3$(aq). We also accepted these values. Thus there are six adjustable parameters of Eqs. 2b and 3b: $\beta_{M,X}^{(0)}$, $\beta_{M,X}^{(1)}$, $C_{M,X}^{(0)}$, $C_{M,X}^{(1)}$, $C_{M,X}^{(2)}$, and $C_{M,X}^{(3)}$ for each electrolyte.

Wang et al. [11] used the same basic model as given by Eqs. 2a, 2b and 3a, 3b to represent the osmotic coefficients of the RECl$_3$(aq), RE(ClO$_4$)$_3$(aq), and RE(NO$_3$)$_3$(aq) at 298.15 K to the highest experimental ionic strengths. They found that the standard uncertainties u(ϕ) showed sharp decreases with increasing number of model parameters np up to $np = 7$ for the RE(NO$_3$)$_3$(aq) and to $np = 6$ for the RECl$_3$(aq) and RE(ClO$_4$)$_3$(aq), with no significant further improvement resulting from including additional higher order terms with their corresponding parameters. Based on this information, $np = 6$ models were accepted for all of the RECl$_3$(aq).

The evaluated parameters of Eq. 2b are reported in Table 2, and calculated values of ϕ, a_w (water activity) and $\gamma_\pm$ using these parameters are reported in Tables 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, and 16 at rounded values of the molalities and at the saturated solution molalities. The solubilities of the RECl$_3$·7H$_2$O(cr), RE = (La, Pr), and RECl$_3$· 6H$_2$O(cr), RE = (Nd, Sm, Eu, Gd, Tb, Dy, Ho, Er, Tm, Yb, Lu, Y), were taken from the IUPAC–NIST critical assessments [16–18] when reported to four significant figures or are our assessment from their tabulated solubilities when fewer significant figures were recommended by them.

Table 2 Parameters and standard uncertainties u(ϕ) for the new extended ion-interaction (Pitzer) model based on literature values of osmotic coefficients ϕ of aqueous rare earth chlorides from dilute solution to the maximum ionic strength I at $T = 298.15$ K

Rare earth chloride	$(3/2)\beta_{M,X}^{(0)}$	$(3/2)\beta_{M,X}^{(1)}$	$(3^{3/2})C_{M,X}^{(0)}$	$(3^{3/2})C_{M,X}^{(1)}$	$(3^{3/2})C_{M,X}^{(2)}$	$(3^{3/2})C_{M,X}^{(3)}$	u(ϕ)	Maximum I (mol·kg^{-1})
LaCl$_3$	0.5616	8.63	−0.00946	1.2108	−2.7704	3.2556	0.0024	23.4
PrCl$_3$	0.6260	8.52	−0.01460	1.0725	−2.7293	3.2002	0.0018	23.4
NdCl$_3$	0.7175	8.23	−0.02658	0.7719	−2.1103	2.4514	0.0021	23.6
SmCl$_3$	0.7659	8.10	−0.03652	0.7180	−2.0854	2.3792	0.0022	21.8
EuCl$_3$	0.8001	8.18	−0.04201	0.6648	−1.9780	2.2348	0.0017	21.5
GdCl$_3$	0.7576	8.28	−0.03119	0.8117	−2.1763	2.4557	0.0023	21.5
TbCl$_3$	0.6423	8.78	−0.00776	1.2024	−2.8692	3.2386	0.0023	21.4
DyCl$_3$	0.4623	9.42	0.02879	1.7440	−3.8773	4.4478	0.0023	21.8
HoCl$_3$	0.7282	8.97	−0.01622	0.9787	−2.5418	2.8281	0.0019	22.2
ErCl$_3$	0.6258	9.06	0.00686	1.2410	−2.9878	3.4068	0.0022	22.7
TmCl$_3$	0.5418	9.24	0.02551	1.4673	−3.3887	3.9240	0.0019	23.3
YbCl$_3$	0.6279	8.89	0.01211	1.1928	−2.8607	3.3011	0.0025	24.0
LuCl$_3$	0.5547	9.31	0.02706	1.3803	−3.1675	3.6831	0.0023	24.7
YCl$_3$	0.6737	8.78	0.00122	1.0851	−2.6842	3.0775	0.0020	24.5

The α_{Ci} exponential coefficients were fixed at the values recommended by Wang et al. [11] for aqueous rare earth chloride and rare earth perchlorate solutions: $\alpha_{B1} = 2.0$ kg$^{1/2}$·mol$^{-1/2}$, $\alpha_{C1} = 0.15$ kg·mol^{-1}, $\alpha_{C2} = 0.25$ kg·mol^{-1}, and $\alpha_{C3} = 0.35$ kg·mol^{-1}. The units of $\beta_{M,X}^{(0)}$ and $\beta_{M,X}^{(1)}$ are kg·mol^{-1}, whereas those of $C_{M,X}^{(0)}$, $C_{M,X}^{(1)}$, $C_{M,X}^{(2)}$, and $C_{M,X}^{(3)}$ are kg^2·mol^{-2}. The maximum ionic strength for nearly all of the rare earth chlorides is the solubility limit but for YCl$_3$(aq) the range of experimental osmotic coefficients extends slightly into the oversaturation (supersaturated) region

Table 3 Molality-based osmotic coefficients, water activities, and mean activity coefficients of $LaCl_3(aq)$ at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7815	0.9944	0.3289	1.8	1.6350	0.8089	0.7162
0.2	0.7930	0.9886	0.2866	2.0	1.7574	0.7762	0.8710
0.3	0.8211	0.9824	0.2724	2.2	1.8767	0.7427	1.0607
0.4	0.8592	0.9755	0.2701	2.4	1.9910	0.7087	1.2897
0.5	0.9033	0.9680	0.2749	2.5	2.0459	0.6917	1.4204
0.6	0.9509	0.9597	0.2844	2.6	2.0993	0.6748	1.5626
0.7	1.0006	0.9508	0.2977	2.8	2.2010	0.6414	1.8838
0.8	1.0520	0.9412	0.3145	3.0	2.2961	0.6087	2.2581
0.9	1.1048	0.9309	0.3346	3.2	2.3850	0.5770	2.6910
1.0	1.1590	0.9199	0.3581	3.4	2.4682	0.5462	3.1886
1.2	1.2717	0.8959	0.4168	3.5	2.5079	0.5313	3.4640
1.4	1.3898	0.8692	0.4934	3.6	2.5464	0.5165	3.7585
1.5	1.4504	0.8549	0.5396	3.8	2.6207	0.4879	4.4099
1.6	1.5116	0.8401	0.5918	3.896	2.6551	0.4745	4.7548

Table 4 Molality-based osmotic coefficients, water activities, and mean activity coefficients of $PrCl_3(aq)$ at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7842	0.9944	0.3303	1.8	1.6502	0.8073	0.7410
0.2	0.7983	0.9886	0.2894	2.0	1.7789	0.7739	0.9085
0.3	0.8279	0.9823	0.2761	2.2	1.9056	0.7393	1.1174
0.4	0.8665	0.9753	0.2745	2.4	2.0283	0.7041	1.3742
0.5	0.9105	0.9677	0.2797	2.5	2.0878	0.6865	1.5227
0.6	0.9576	0.9594	0.2897	2.6	2.1458	0.6690	1.6859
0.7	1.0066	0.9505	0.3034	2.8	2.2573	0.6342	2.0602
0.8	1.0573	0.9409	0.3205	3.0	2.3625	0.6001	2.5053
0.9	1.1096	0.9306	0.3410	3.2	2.4616	0.5669	3.0303
1.0	1.1636	0.9196	0.3651	3.4	2.5551	0.5347	3.6456
1.2	1.2767	0.8955	0.4254	3.5	2.5999	0.5191	3.9910
1.4	1.3966	0.8686	0.5049	3.6	2.6435	0.5037	4.3638
1.5	1.4587	0.8541	0.5534	3.8	2.7275	0.4738	5.1993
1.6	1.5219	0.8391	0.6084	3.894	2.7657	0.4602	5.6374

As expected, the values of $\gamma_\pm$ of the $RECl_3(aq)$ reported in Tables 3, 4, 5, 6, 7, 8, and 9 are smaller than those calculated using the model parameters of Wang et al. [11], and are in somewhat better agreement with the values reported in the source isopiestic studies [1, 2] but with differences commensurate with changes in the osmotic coefficients of the KCl(aq) and $CaCl_2(aq)$ reference solutions.

Table 5 Molality-based osmotic coefficients, water activities, and mean activity coefficients of NdCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7836	0.9944	0.3288	1.8	1.6620	0.8061	0.7484
0.2	0.7983	0.9886	0.2882	2.0	1.7930	0.7723	0.9210
0.3	0.8273	0.9823	0.2748	2.2	1.9224	0.7373	1.1374
0.4	0.8651	0.9754	0.2729	2.4	2.0484	0.7017	1.4056
0.5	0.9084	0.9678	0.2778	2.5	2.1098	0.6838	1.5618
0.6	0.9553	0.9595	0.2875	2.6	2.1699	0.6659	1.7344
0.7	1.0048	0.9506	0.3011	2.8	2.2861	0.6305	2.1337
0.8	1.0563	0.9409	0.3183	3.0	2.3967	0.5956	2.6144
0.9	1.1098	0.9306	0.3391	3.2	2.5017	0.5616	3.1885
1.0	1.1651	0.9195	0.3636	3.4	2.6014	0.5287	3.8701
1.2	1.2812	0.8951	0.4251	3.5	2.6493	0.5126	4.2561
1.4	1.4039	0.8679	0.5065	3.6	2.6961	0.4969	4.6751
1.5	1.4672	0.8533	0.5560	3.8	2.7864	0.4663	5.6217
1.6	1.5315	0.8381	0.6124	3.930	2.8429	0.4470	6.3230

Table 6 Molality-based osmotic coefficients, water activities, and mean activity coefficients of SmCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7847	0.9944	0.3290	1.8	1.6821	0.8040	0.7806
0.2	0.8017	0.9885	0.2894	2.0	1.8148	0.7699	0.9643
0.3	0.8325	0.9822	0.2770	2.2	1.9455	0.7346	1.1951
0.4	0.8717	0.9752	0.2759	2.4	2.0726	0.6988	1.4814
0.5	0.9161	0.9675	0.2816	2.5	2.1343	0.6808	1.6483
0.6	0.9640	0.9592	0.2922	2.6	2.1947	0.6629	1.8327
0.7	1.0143	0.9501	0.3067	2.8	2.3109	0.6273	2.2590
0.8	1.0668	0.9404	0.3249	3.0	2.4211	0.5925	2.7712
0.9	1.1212	0.9299	0.3469	3.2	2.5250	0.5586	3.3813
1.0	1.1774	0.9187	0.3727	3.4	2.6229	0.5259	4.1024
1.2	1.2955	0.8940	0.4377	3.5	2.6697	0.5100	4.5090
1.4	1.4201	0.8665	0.5238	3.6	2.7151	0.4944	4.9490
1.5	1.4845	0.8518	0.5763	3.641	2.7334	0.4881	5.1395
1.6	1.5498	0.8364	0.6361				

The standard uncertainties u(ϕ) of the fits reported in Table 2 are the same or very close to those reported previously by Wang et al. [11] for the heavier rare earth trichlorides, RE = (Ho, Er, Tm, Yb, Lu), and for YCl$_3$(aq) whose thermodynamic properties are close to those of YbCl$_3$(aq) and TmCl$_3$(aq). However, for the lighter and intermediate rare earths

Table 7 Molality-based osmotic coefficients, water activities, and mean activity coefficients of EuCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7890	0.9943	0.3324	1.6	1.5705	0.8344	0.6704
0.2	0.8077	0.9884	0.2940	1.8	1.7047	0.8016	0.8263
0.3	0.8395	0.9820	0.2823	2.0	1.8389	0.7672	1.0249
0.4	0.8794	0.9750	0.2821	2.2	1.9710	0.7316	1.2749
0.5	0.9247	0.9672	0.2887	2.4	2.0993	0.6955	1.5859
0.6	0.9734	0.9588	0.3002	2.5	2.1616	0.6775	1.7675
0.7	1.0248	0.9496	0.3160	2.6	2.2224	0.6594	1.9683
0.8	1.0783	0.9397	0.3356	2.8	2.3396	0.6237	2.4334
0.9	1.1339	0.9291	0.3592	3.0	2.4505	0.5888	2.9934
1.0	1.1914	0.9177	0.3870	3.2	2.5549	0.5548	3.6614
1.2	1.3119	0.8928	0.4569	3.4	2.6531	0.5220	4.4517
1.4	1.4388	0.8649	0.5495	3.5	2.7000	0.5061	4.8976
1.5	1.5042	0.8499	0.6060	3.587	2.7396	0.4926	5.3151

Table 8 Molality-based osmotic coefficients, water activities, and mean activity coefficients of GdCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7884	0.9943	0.3324	1.6	1.5933	0.8322	0.6941
0.2	0.8067	0.9884	0.2937	1.8	1.7309	0.7989	0.8610
0.3	0.8391	0.9820	0.2821	2.0	1.8683	0.7639	1.0745
0.4	0.8802	0.9749	0.2822	2.2	2.0034	0.7279	1.3446
0.5	0.9270	0.9672	0.2893	2.4	2.1344	0.6913	1.6821
0.6	0.9775	0.9586	0.3016	2.5	2.1979	0.6730	1.8799
0.7	1.0307	0.9493	0.3182	2.6	2.2600	0.6548	2.0991
0.8	1.0861	0.9393	0.3390	2.8	2.3797	0.6187	2.6091
0.9	1.1436	0.9285	0.3639	3.0	2.4931	0.5834	3.2267
1.0	1.2030	0.9170	0.3932	3.2	2.6002	0.5490	3.9686
1.2	1.3272	0.8916	0.4671	3.4	2.7014	0.5159	4.8534
1.4	1.4579	0.8632	0.5653	3.5	2.7499	0.4998	5.3559
1.5	1.5251	0.8480	0.6255	3.590	2.7925	0.4856	5.8457

the standard uncertainties are slightly larger than for earlier models [11], $\Delta\{u(\phi)\} = (0.0002$ to $0.0007)$, which is of marginal significance. Some of this difference may have arisen from having fewer values of ϕ to constrain the model fits at molalities below the range of the isopiestic measurements.

Table 9 Molality-based osmotic coefficients, water activities, and mean activity coefficients of TbCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7917	0.9943	0.3367	1.6	1.6161	0.8300	0.7301
0.2	0.8089	0.9884	0.2978	1.8	1.7580	0.7961	0.9123
0.3	0.8414	0.9820	0.2863	2.0	1.8997	0.7605	1.1469
0.4	0.8837	0.9748	0.2870	2.2	2.0386	0.7238	1.4452
0.5	0.9320	0.9670	0.2949	2.4	2.1729	0.6867	1.8198
0.6	0.9840	0.9583	0.3083	2.5	2.2379	0.6682	2.0399
0.7	1.0387	0.9490	0.3261	2.6	2.3013	0.6498	2.2843
0.8	1.0954	0.9388	0.3482	2.8	2.4231	0.6133	2.8544
0.9	1.1542	0.9279	0.3747	3.0	2.5384	0.5777	3.5476
1.0	1.2149	0.9162	0.4060	3.2	2.6473	0.5431	4.3843
1.2	1.3422	0.8904	0.4849	3.4	2.7506	0.5097	5.3886
1.4	1.4766	0.8616	0.5905	3.5	2.8003	0.4935	5.9624
1.5	1.5458	0.8461	0.6556	3.572	2.8354	0.4820	6.4083

Table 10 Molality-based osmotic coefficients, water activities, and mean activity coefficients of DyCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7937	0.9943	0.3408	1.8	1.7712	0.7947	0.9408
0.2	0.8087	0.9884	0.3008	2.0	1.9170	0.7586	1.1895
0.3	0.8412	0.9820	0.2892	2.2	2.0601	0.7214	1.5080
0.4	0.8846	0.9748	0.2902	2.4	2.1984	0.6837	1.9103
0.5	0.9342	0.9669	0.2988	2.5	2.2652	0.6649	2.1477
0.6	0.9873	0.9582	0.3128	2.6	2.3305	0.6462	2.4122
0.7	1.0427	0.9488	0.3313	2.8	2.4558	0.6093	3.0316
0.8	1.0999	0.9386	0.3541	3.0	2.5745	0.5732	3.7898
0.9	1.1588	0.9276	0.3813	3.2	2.6872	0.5381	4.7128
1.0	1.2196	0.9159	0.4134	3.4	2.7949	0.5042	5.8331
1.2	1.3475	0.8900	0.4944	3.5	2.8473	0.4877	6.4798
1.4	1.4834	0.8610	0.6037	3.6	2.8988	0.4714	7.1922
1.5	1.5539	0.8454	0.6713	3.631	2.9147	0.4664	7.4275
1.6	1.6256	0.8291	0.7492				

Table 11 Molality-based osmotic coefficients, water activities, and mean activity coefficients of HoCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.8017	0.9942	0.3451	1.8	1.7832	0.7935	0.9835
0.2	0.8214	0.9882	0.3083	2.0	1.9307	0.7571	1.2473
0.3	0.8541	0.9817	0.2980	2.2	2.0763	0.7195	1.5875
0.4	0.8956	0.9745	0.2995	2.4	2.2182	0.6814	2.0214
0.5	0.9429	0.9666	0.3084	2.5	2.2872	0.6623	2.2796
0.6	0.9941	0.9579	0.3226	2.6	2.3549	0.6433	2.5689
0.7	1.0484	0.9485	0.3417	2.8	2.4858	0.6056	3.2532
0.8	1.1051	0.9383	0.3653	3.0	2.6107	0.5687	4.1013
0.9	1.1642	0.9273	0.3937	3.2	2.7296	0.5329	5.1448
1.0	1.2256	0.9155	0.4273	3.4	2.8430	0.4983	6.4215
1.2	1.3552	0.8894	0.5126	3.5	2.8978	0.4815	7.1610
1.4	1.4928	0.8602	0.6277	3.6	2.9514	0.4650	7.9765
1.5	1.5639	0.8445	0.6991	3.696	3.0019	0.4495	8.8374
1.6	1.6363	0.8281	0.7812				

Table 12 Molality-based osmotic coefficients, water activities, and mean activity coefficients of ErCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7968	0.9943	0.3417	1.8	1.7878	0.7930	0.9727
0.2	0.8146	0.9883	0.3035	2.0	1.9377	0.7563	1.2374
0.3	0.8475	0.9818	0.2926	2.2	2.0861	0.7184	1.5806
0.4	0.8900	0.9747	0.2939	2.4	2.2311	0.6799	2.0208
0.5	0.9386	0.9667	0.3026	2.5	2.3018	0.6606	2.2839
0.6	0.9911	0.9581	0.3168	2.6	2.3712	0.6413	2.5798
0.7	1.0462	0.9486	0.3356	2.8	2.5058	0.6031	3.2835
0.8	1.1036	0.9384	0.3590	3.0	2.6349	0.5657	4.1632
0.9	1.1632	0.9273	0.3870	3.2	2.7586	0.5293	5.2568
1.0	1.2250	0.9155	0.4202	3.4	2.8777	0.4941	6.6114
1.2	1.3552	0.8894	0.5043	3.5	2.9357	0.4769	7.4044
1.4	1.4938	0.8601	0.6182	3.6	2.9928	0.4601	8.2857
1.5	1.5656	0.8443	0.6890	3.783	3.0955	0.4301	10.161
1.6	1.6388	0.8278	0.7707				

Table 13 Molality-based osmotic coefficients, water activities, and mean activity coefficients of TmCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7953	0.9943	0.3412	1.8	1.7935	0.7924	0.9773
0.2	0.8119	0.9884	0.3022	2.0	1.9455	0.7555	1.2467
0.3	0.8450	0.9819	0.2911	2.2	2.0962	0.7173	1.5975
0.4	0.8884	0.9747	0.2925	2.4	2.2436	0.6784	2.0494
0.5	0.9381	0.9668	0.3014	2.5	2.3157	0.6589	2.3206
0.6	0.9914	0.9580	0.3158	2.6	2.3864	0.6395	2.6263
0.7	1.0472	0.9485	0.3348	2.8	2.5239	0.6009	3.3565
0.8	1.1051	0.9383	0.3584	3.0	2.6562	0.5631	4.2752
0.9	1.1649	0.9272	0.3865	3.2	2.7836	0.5263	5.4260
1.0	1.2268	0.9154	0.4198	3.4	2.9069	0.4906	6.8646
1.2	1.3574	0.8892	0.5042	3.5	2.9673	0.4731	7.7133
1.4	1.4967	0.8599	0.6188	3.6	3.0271	0.4560	8.6621
1.5	1.5691	0.8440	0.6901	3.8	3.1452	0.4226	10.911
1.6	1.6429	0.8274	0.7726	3.879	3.1914	0.4098	11.950

Table 14 Molality-based osmotic coefficients, water activities, and mean activity coefficients of YbCl$_3$(aq) at rounded values of the molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7933	0.9943	0.3383	2.0	1.9497	0.7550	1.2421
0.2	0.8108	0.9884	0.2996	2.2	2.1014	0.7167	1.5938
0.3	0.8440	0.9819	0.2885	2.4	2.2502	0.6776	2.0487
0.4	0.8871	0.9748	0.2897	2.5	2.3231	0.6580	2.3225
0.5	0.9364	0.9668	0.2983	2.6	2.3950	0.6384	2.6320
0.6	0.9896	0.9581	0.3124	2.8	2.5350	0.5996	3.3747
0.7	1.0456	0.9486	0.3313	3.0	2.6701	0.5614	4.3147
0.8	1.1038	0.9383	0.3546	3.2	2.8007	0.5242	5.4991
0.9	1.1643	0.9273	0.3827	3.4	2.9273	0.4881	6.9878
1.0	1.2269	0.9154	0.4158	3.5	2.9893	0.4705	7.8692
1.2	1.3588	0.8891	0.5002	3.6	3.0506	0.4532	8.8565
1.4	1.4990	0.8597	0.6147	3.8	3.1715	0.4196	11.203
1.5	1.5718	0.8438	0.6859	4.0	3.2908	0.3873	14.151
1.6	1.6460	0.8271	0.7683	4.003	3.2925	0.3868	14.201
1.8	1.7971	0.7921	0.9727				

Table 15 Molality-based osmotic coefficients, water activities, and mean activity coefficients of LuCl$_3$(aq) at rounded molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7972	0.9943	0.3431	2.0	1.9497	0.7550	1.2587
0.2	0.8134	0.9883	0.3041	2.2	2.1021	0.7166	1.6162
0.3	0.8456	0.9819	0.2928	2.4	2.2516	0.6775	2.0793
0.4	0.8882	0.9747	0.2939	2.5	2.3250	0.6578	2.3583
0.5	0.9372	0.9668	0.3027	2.6	2.3972	0.6382	2.6738
0.6	0.9902	0.9581	0.3170	2.8	2.5381	0.5992	3.4321
0.7	1.0459	0.9486	0.3360	3.0	2.6744	0.5609	4.3940
0.8	1.1039	0.9383	0.3596	3.2	2.8064	0.5235	5.6101
0.9	1.1640	0.9273	0.3879	3.4	2.9349	0.4872	7.1454
1.0	1.2263	0.9154	0.4214	3.5	2.9981	0.4695	8.0581
1.2	1.3577	0.8892	0.5066	3.6	3.0608	0.4520	9.0839
1.4	1.4978	0.8598	0.6223	3.8	3.1850	0.4181	11.536
1.5	1.5706	0.8439	0.6944	4.0	3.3084	0.3853	14.645
1.6	1.6449	0.8272	0.7778	4.120	3.3825	0.3663	16.904
1.8	1.7965	0.7921	0.9851				

Table 16 Molality-based osmotic coefficients, water activities, and mean activity coefficients of YCl$_3$(aq) at rounded molalities and saturated solution at $T = 298.15$ K

m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$	m (mol·kg^{-1})	ϕ	a_w	$\gamma_\pm$
0.1	0.7941	0.9943	0.3385	1.8	1.7968	0.7921	0.9777
0.2	0.8129	0.9884	0.3004	2.0	1.9480	0.7552	1.2467
0.3	0.8465	0.9819	0.2897	2.2	2.0981	0.7170	1.5968
0.4	0.8897	0.9747	0.2911	2.4	2.2451	0.6782	2.0481
0.5	0.9390	0.9667	0.2999	2.5	2.3171	0.6587	2.3190
0.6	0.9921	0.9580	0.3143	2.6	2.3879	0.6393	2.6247
0.7	1.0480	0.9485	0.3333	2.8	2.5256	0.6007	3.3557
0.8	1.1062	0.9382	0.3569	3.0	2.6582	0.5629	4.2762
0.9	1.1666	0.9271	0.3852	3.2	2.7859	0.5260	5.4295
1.0	1.2292	0.9152	0.4187	3.4	2.9091	0.4903	6.8690
1.2	1.3609	0.8890	0.5038	3.5	2.9692	0.4729	7.7164
1.4	1.5006	0.8595	0.6189	3.6	3.0285	0.4558	8.6618
1.5	1.5731	0.8436	0.6905	3.8	3.1448	0.4227	10.892
1.6	1.6468	0.8271	0.7731	3.942	3.2259	0.4000	12.798

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