m_i	molal concentration of electrolyte i, mol·kg^{-1}
m_T	total molality of mixed electrolyte, mol·kg^{-1}
I	total ionic strength of mixed electrolyte, mol·kg^{-1}
y_i	ionic strength fraction of electrolyte i in mixture
Φ	molal osmotic coefficient of solution
ν_i	number of ions formed by the dissociation of one molecule of electrolyte i
f^{ϕ}	Debye–Hückel term in Pitzer's equations for single electrolytes [$= -0.3290I^{1/2}/(1 + 1.2I^{1/2})$]
B^{ϕ}	ionic-strength-dependent parameter in Pitzer's equations for single electrolytes
$\beta^{(0)}$, $\beta^{(1)}$	coefficients of B^{ϕ} term in Pitzer's equations for single electrolytes
C^{ϕ}	constant parameter in Pitzer's equations for single electrolytes
Θ°, Θ', ψ	mixing parameters for Pitzer's ternary solution equations
$\sigma(\Phi)$	standard deviation of fitting equations for Φ
ϕ_i^0	osmotic coefficient of pure electrolyte i at the total ionic strength of the mixture for Scatchard's neutral electrolyte equations
b_{ij}	mixing parameters for Scatchard's neutral electrolyte equations
h_i	osmolality fraction of electrolyte i in mixture
$\gamma_{\pm}$	mean molal activity coefficient

Registry No. NaCl, 7647-14-5; $MgCl_2$, 7786-30-3.

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Osmotic and Activity Coefficients of Aqueous $La(NO_3)_3$ and Densities and Apparent Molal Volumes of Aqueous $Eu(NO_3)_3$ at 25 °C

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The osmotic coefficients of aqueous $La(NO_3)_3$ have been measured from 1.3435 to 8.4591 mol·kg^{-1} at 25 °C by using the isopiestic method. Some earlier osmotic coefficients for this salt are too high, apparently due to a stock solution concentration error, and have been normalized to the present results. These combined data and other activity data were then used to generate recommended values for the osmotic coefficients, water activities, and mean molal activity coefficients of $La(NO_3)_3$. The solubility of $La(NO_3)_3 \cdot 6H_2O$(cr) was determined to be 4.6147 ± 0.0056 mol·kg^{-1} by the isopiestic method; this is in excellent agreement with the IUPAC recommended value of 4.610 ± 0.005 mol·kg^{-1}. Density data were measured for aqueous $Eu(NO_3)_3$ from 0.03996 to 1.1014 mol·kg^{-1} at 25 °C by using pycnometry. These results are in fairly good agreement with published low-concentration densities measured with a magnetic float.

Introduction

Activity and osmotic coefficient data for aqueous electrolyte solutions have numerous applications including solubility, chemical speciation, and reaction thermodynamics calculations. Density data are required for buoyancy calculations, for the interconversion of mass and volumetric concentration scales, and for the calculation of partial molal volumes which are related to the pressure derivatives of solute and solvent activities.

We have published isopiestic data for a total of 40 aqueous rare earth chloride, perchlorate, and nitrate solutions from low concentrations to saturation or supersaturation at 25 °C (*1*–*6*). Isopiestic measurements yield water activities, osmotic coefficients, and activity coefficients as a function of molality. For the first three parts of this series (*1*–*3*), the stock solution preparations, analyses, and the isopiestic measurements were done at Ames Laboratory (Iowa State University). For the last part (*6*) all of this was done at Lawrence Livermore National Laboratory (LLNL). However, for six of the rare earth nitrates,

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published as parts 4 and 5 of this series (*4*, *5*), the stock solutions were prepared and analyzed at Ames Laboratory but were then rebottled and shipped to LLNL for isopiestic measurements. These six salts are $La(NO_3)_3$, $Pr(NO_3)_3$, $Nd(NO_3)_3$, $Dy(NO_3)_3$, $Ho(NO_3)_3$, and $Lu(NO_3)_3$.

The rare earth nitrate solutions prepared at Ames Laboratory were analyzed there for concentration by mass titration with EDTA and by conversion to the anhydrous sulfate (*3*). Four of the six stock solutions that were shipped to LLNL were reanalyzed at LLNL by the gravimetric sulfate method. For $Nd(NO_3)_3$, $Ho(NO_3)_3$, and $Lu(NO_3)_3$ the reanalyses agreed well within their experimental precision with the Ames Laboratory values. However, for $Dy(NO_3)_3$ the concentration reported by Ames Laboratory was somewhat below the concentration measured at LLNL, which indicates that some evaporation of water occurred during the handling or shipping of this stock solution to LLNL. This new concentration analysis was used for reporting data for $Dy(NO_3)_3$ (*5*). Unfortunately, for $La(NO_3)_3$ and $Pr(NO_3)_3$ the amount of stock solution shipped to LLNL was adequate for the isopiestic measurements but was insufficient for concentration analyses. Therefore, for these two salts, concentration calculations were based solely on the reported stock solution concentrations. Since we ran out of the first $La(NO_3)_3$ stock solution during the isopiestic measurements, a second one was sent to us by Ames Laboratory and was used for the three lowest concentration points. This second stock solution was reanalyzed at LLNL and the measured concentration agreed with the reported value.

We recently reported isopiestic solubility data for several rare earth nitrate solutions at 25 °C (*7*, *8*). Osmotic coefficients for saturated solutions generally agreed to within 0.1–0.3% of interpolated values from the earlier studies (*3–6*), which is very good agreement and confirms the usual precision and reproducibility of isopiestic measurements (*9*). A solubility determination for $Pr(NO_3)_3\cdot 6H_2O(cr)$ using a new stock solution analyzed at LLNL agreed with other reported solubilities but the osmotic coefficient was discrepant (*8*). Subsequent measurements for $La(NO_3)_3\cdot 6H_2O(cr)$, reported below, indicate similar problems for $La(NO_3)_3$.

Many of the earlier $La(NO_3)_3$ and $Pr(NO_3)_3$ isopiestic equilibrations were done simultaneously with $Nd(NO_3)_3$ samples present in other cups in the chamber (*4*). Several series of equilibrations were made with different samples from the same stock solutions. Since consistent results were obtained for the various equilibrations, simple weighing errors can be eliminated as the source of the problem. The $CaCl_2$ reference solution for our present redetermination of $La(NO_3)_3$ and $Pr(NO_3)_3$ osmotic coefficients is the same as used for the earlier determination (*4*), so it can be eliminated as the source of the discrepancy. Its concentration has been determined several times (*9*) and consistent results obtained. Also, ten of the earlier $Nd(NO_3)_3$ high concentration equilibrations with $CaCl_2$ standards were done simultaneously with $MgCl_2$ but without $La(NO_3)_3$ or $Pr(NO_3)_3$ samples. Separate measurements for $MgCl_2$ gave osmotic coefficients that agreed with these values within 0.2% (*9*, *10*). This provides additional evidence that the $CaCl_2$ reference concentrations reported earlier (*4*) were accurate.

The only remaining possibility is errors in some of the rare earth nitrate stock solution concentrations in that study (*4*). As noted above, $Nd(NO_3)_3$ samples were included in most of the equilibrations (*4*) and its stock solution concentration was independently analyzed both in Ames and at LLNL. Osmotic coefficients for $Nd(NO_3)_3$ have also been determined at the Technical University of Gdańsk by Libuś et al. (*11*) up to 4.8342 $mol\cdot kg^{-1}$, and their osmotic coefficients are within 0.12–0.45% of ours. Since osmotic coefficients are reproducible to about 0.2% under favorable conditions and the reference solution osmotic coefficients are uncertain by about an equal amount (*9*), agreement for the two $Nd(NO_3)_3$ studies is excellent. Thus, data for $Nd(NO_3)_3$ can be trusted and only the earlier results for $La(NO_3)_3$ and $Pr(NO_3)_3$ seem to be in error.

Discrepancies for $La(NO_3)_3$ and $Pr(NO_3)_3$ in the earlier study (*4*) relative to the new solubility values can be accounted for if their actual stock solution concentrations were higher than determined at Ames Laboratory. This is in the same direction as the difference noted for $Dy(NO_3)_3$, and the same explanation seems appropriate. That is, some evaporation probably occurred for $La(NO_3)_3$ and $Pr(NO_3)_3$ stock solutions prior to their arrival at LLNL. Thus, isopiestic data have been redetermined for $La(NO_3)_3$ using the second stock solution supplied by Ames Laboratory and are reported here. Although the three lowest concentration points in the earlier study were measured with samples from this second stock solution, no discrepancies were obvious at that time since molality concentration errors are greatly reduced at low concentrations.

Since similar discrepancies occur for $Pr(NO_3)_3$, isopiestic measurements are in progress at LLNL and will be reported elsewhere.

Spedding et al. (*12*, *13*) have reported densities for solutions of 13 aqueous rare earth nitrates from about 0.0015 to 0.09–0.27 $mol\cdot kg^{-1}$ at 25 °C by using a magnetically controlled float, and they have extended the data for 11 of these salts to saturation by using pycnometry (*14*). However, density data for $Eu(NO_3)_3$ extend only from 0.001 38 to 0.169 94 $mol\cdot kg^{-1}$ (*13*). Jeżowska-Trzebiatowska et al. (*15*) have reported five density points for $Eu(NO_3)_3$ from 0.40894 to 0.67400 $mol\cdot kg^{-1}$ using pycnometry. These two density studies for $Eu(NO_3)_3$ do not overlap in concentration.

A convenient deviation function for comparing density data is the density of the solution minus that of the solvent, all divided by the molality. For the majority of aqueous electrolytes, this deviation function is a monotonic decreasing function of concentration. The $Eu(NO_3)_3$ density data of Spedding et al. (*13*) show this typical behavior, whereas the values of Jeżowska-Trzebiatowska et al. (*15*) for this function are not only discrepant but increase rather than decrease with concentration. Thus, new density data are needed for $Eu(NO_3)_3$, and they are reported in this paper.

Experimental Section

The isopiestic experiments were performed at 25.00 ± 0.005 °C (IPTS-68) using stainless steel isopiestic chambers that have been described in detail elsewhere (*8*). Isopiestic equilibration times were generally 4–6 days over most of the concentration range, but were increased to 6–12 days for the lower concentrations. All sample weights were converted to masses by using their appropriate densities.

Our $Eu(NO_3)_3$ stock solution was prepared at LLNL from Analytical Reagent HNO_3 and Eu_2O_3 (supplied by Ames Laboratory), and it was adjusted to its equivalence pH by using dilute HNO_3. This equivalence pH was determined by titration of samples of unadjusted stock solution with the same dilute HNO_3 solution. The $La(NO_3)_3$ stock solution was prepared by the same method at Ames Laboratory, and it was supplied to us as an analyzed stock solution.

A sample of the $La(NO_3)_3$ stock solution was evaporated to dryness and then thermally decomposed to La_2O_3 at 800 °C. Both this La_2O_3 and some of the original Eu_2O_3 were analyzed by direct current arc optical emission spectroscopy (DCAOES), and the Eu_2O_3 by X-ray fluorescence spectroscopy (XRFS), at LLNL to quantitatively determine impurities present. The La_2O_3 contained (in weight percent) 0.001% Mg, $\leq$0.001% Ca, and $\leq$0.0001% Sr, B, and Cu; other rare earth elements were below their detection limits. DCAOES indicated the Eu_2O_3 contained 0.06% Gd, $\leq$0.001% Cu, 0.0005% each of B and Fe, 0.0004% Mg, and $\leq$0.0001% Si; XRFS gave 0.04% Gd

Table I. Isopiestic Molalities of $La(NO_3)_3$ Solutions with $CaCl_2$ Reference Solutions at 25 °C

$[CaCl_2]$, mol·kg^{-1}	$[La(NO_3)_3]$, mol·kg^{-1}	$\Phi(La(NO_3)_3)$
1.4051	1.3435	0.9156
1.6016	1.5554	0.9524
1.7004	1.6644	0.9712
1.7279	1.6945	0.9768
1.8696	1.8529	1.0049
2.0163	2.0191	1.0348
2.1230	2.1417	1.0569
2.2419	2.2796	1.0819
2.3324	2.3867	1.1005
2.4947	2.5783	1.1356
2.5918	2.6978	1.1552
2.7166	2.8489	1.1822
2.8281	2.9845	1.2067
2.9387[a]	3.1225	1.2301
3.0717[a]	3.2879	1.2592
3.1782[a]	3.4205	1.2828
3.2710[a]	3.5379	1.3030
3.3848	3.6833	1.3275
3.4890	3.8172	1.3500
3.5743	3.9265	1.3686
3.7035	4.0911	1.3974
3.7724	4.1802	1.4124
3.8353	4.2645	1.4251
3.8877	4.3319	1.4367
3.9472	4.4117	1.4487
4.0781	4.5809	1.4774
4.1049	4.6140[b]	1.4838
4.1049	4.6150[c]	1.4834
4.1045	4.6150[d]	1.4832
4.1839	4.7184	1.5003
4.2508	4.8066	1.5143
4.3896	4.9898	1.5433
4.4662	5.0885	1.5598
4.5947	5.2595	1.5857
4.6658	5.3515	1.6007
4.7605	5.4767	1.6196
4.8586	5.6062	1.6389
4.9602	5.7368	1.6596
5.0288	5.8292	1.6722
5.1102	5.9347	1.6879
5.1753	6.0176	1.7007
5.3014	6.1752	1.7258
5.3850	6.2823	1.7411
5.4729	6.3941	1.7570
5.5958	6.5477	1.7792
5.7715	6.7598	1.8111
5.8497	6.8529	1.8248
5.9036	6.9178	1.8338
6.0485	7.0814	1.8598
6.1948	7.2487[e]	1.8835
6.3340	7.4036[e]	1.9055
6.4778	7.5540	1.9288
6.6571	7.7414	1.9551
6.7583	7.8529	1.9672
6.9453	8.0342[e]	1.9934
7.1354	8.2163	2.0174
7.2482	8.3233	2.0304
7.3986	8.4591	2.0480

[a] Quadruplicate $CaCl_2$ samples used for these determinations. [b] First solubility determination, with 4-day equilibration. The $La(NO_3)_3$ saturation molality is 4.6140 ± 0.0008 and the $CaCl_2$ molality 4.1049 ± 0.0007. [c] Second solubility determination, with 6-day equilibration. The $La(NO_3)_3$ saturation molality is 4.6150 ± 0.0008 and the $CaCl_2$ molality 4.1049 ± 0.0011. [d] Third solubility determination, with 13-day equilibration. The $La(NO_3)_3$ saturation molality is 4.6150 ± 0.0008 and the $CaCl_2$ molality 4.1045 ± 0.0011. [e] Single $La(NO_3)_3$ sample; see text for details.

and 0.006% Dy. Other rare earth elements were below their detection limits.

Stock solution concentration analyses were performed by conversion of triplicate samples of the stock solution to the anhydrous sulfate at 500 °C. To avoid coprecipitation of nitrate ions in the rare earth sulfate, nitrate ions were decomposed prior to the H_2SO_4 addition. This decomposition was done either by doing two evaporations of stock solution samples with excess HCl, or by thermally decomposing the nitrate followed by another evaporation with HCl. The resulting rare earth chlorides/oxychlorides are readily converted to the sulfates by evaporation with excess H_2SO_4.

Table II. Densities and Apparent Molal Volumes of Aqueous $Eu(NO_3)_3$ at 25 °C

m, mol·kg^{-1}	c, mol·dm^{-3}	d, g·cm^{-3}	ϕ_v, cm^3·mol^{-1}
0.039 961	0.039 763	1.008 489	50.32
0.062 242	0.061 860	1.014 756	51.82
0.121 97	0.120 82	1.031 451	53.37
0.202 01	0.199 21	1.053 479	54.85
0.359 49	0.351 28	1.095 89	56.76
0.561 24	0.541 82	1.148 52	58.58
0.721 76	0.689 92	1.189 07	59.82
0.901 84	0.852 34	1.233 18	61.11
1.101 4	1.027 7	1.280 40	62.44

As noted above, we used the second $La(NO_3)_3$ stock solution that had been sent by Ames Laboratory; it had been analyzed there for concentration both by mass titration with EDTA and by oxalate precipitation followed by thermal decomposition to the oxide. Our sulfate analysis agreed well with these other analyses, and the mean value of 4.4551 ± 0.0040 mol·kg^{-1} was used in calculations. Here and elsewhere the uncertainty limits are one standard deviation. Two separate sulfate analyses at LLNL for the $Eu(NO_3)_3$ stock concentration gave 1.1013 ± 0.0001 and 1.1015 ± 0.0003 mol·kg^{-1}. Assumed molecular masses are 324.92 for $La(NO_3)_3$, 565.98 for $La_2(SO_4)_3$, 337.975 for $Eu(NO_3)_3$, and 592.09 g·mol^{-1} for $Eu_2(SO_4)_3$.

$CaCl_2$ solutions were used as isopiestic reference standards. The concentration of this stock solution was determined by conversion of samples to $CaSO_4$ and by dehydration; the analyses results are given elsewhere (*9*). Assumed molecular masses are 110.986 for $CaCl_2$ and 136.138 g·mol^{-1} for $CaSO_4$.

Table I contains the experimental molalities of $La(NO_3)_3$ in isopiestic equilibrium with a $CaCl_2$ reference solution. Duplicate samples were used in the majority of these equilibrations; the average molalities for each electrolyte are reported and they are reliable to better than ±0.087%, with most molalities being precise to at least ±0.05%. For a few equilibrations, four $CaCl_2$ samples were used.

Crystals of $La(NO_3)_3 \cdot nH_2O$ were grown by seeding a slightly supersaturated solution followed by slow evaporation of water in a desiccator. An extra isopiestic cup containing saturated solution and crystals was added to the chamber for solubility determinations. See ref *8* for details. Measured solubilities using 4-, 6-, and 13-day equilibrations were in excellent agreement, so true saturation had been reached. The experimental solubility is 4.6147 ± 0.0010 mol·kg^{-1}, where the uncertainty limits are statistical only. Including the stock solution concentration uncertainty gives a total uncertainty of ±0.0056 mol·kg^{-1}. Chemical analysis of the solid phase by thermal decomposition at 825 °C gave $n = 6.06 \pm 0.1$; i.e., the hexahydrate was present. Our solubility value is in excellent agreement with the IUPAC recommended value (*16*) of 4.610 ± 0.005 mol·kg^{-1}.

Our isopiestic measurements for $La(NO_3)_3$ extend 3.844 mol·kg^{-1} into the supersaturated concentration region. A total of 29 supersaturated solution experiments were performed which involved 4$^1/_2$ months of experimentation. Supersaturated $La(NO_3)_3$ solutions show very little tendency to spontaneously crystallize, and problems occurred in only two of these attempts. In both cases only one $La(NO_3)_3$ sample went dry while the other sample remained in solution. Since the reference solution molalities were in agreement, these two runs should be reliable and are reported in Table I. In a third experiment, only one $La(NO_3)_3$ sample was used since the other was left out to be dissolved and reconcentrated.

Table III. Coefficients for $Eu(NO_3)_3$ Density Polynomials

B_i	molality	molarity
B_2	0.290556	0.290284
B_3	-0.0279792	-0.0197875
B_4	0.0182804	0.0117012
B_5	-0.0307829	-0.0062726
B_6	0.0094975	
max concn	1.1014	1.0277
σ(d)	0.000020	0.000019

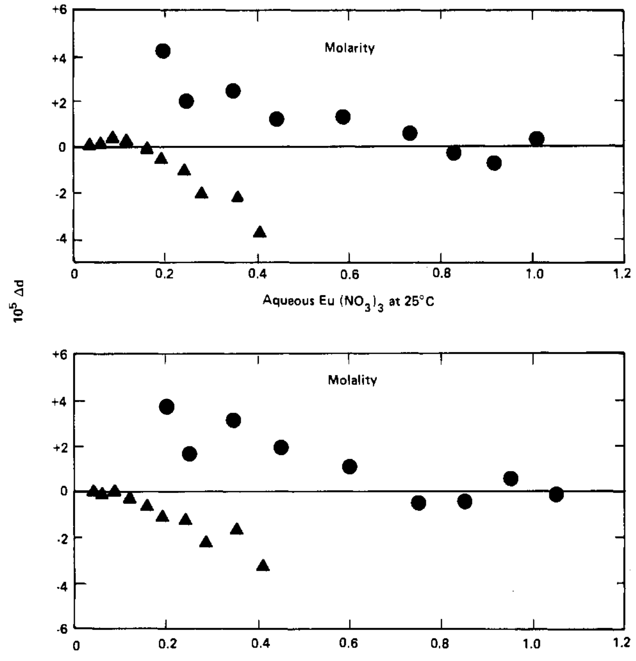

Figure 1. Differences between experimental densities and least-squares equations for $Eu(NO_3)_3$ solutions in $g\cdot cm^{-3} \times 10^5$: (●) this research; (▲) Spedding et al. (*13*).

Densities of $Eu(NO_3)_3$ solutions were measured in duplicate at 25.00 ± 0.005 °C by using a matched pair of 30.87-cm^3 single-stem pycnometers. The pycnometer volumes had been calibrated 7–9 times each by using purified water and assuming a water density of 0.997 045 $g\cdot cm^{-3}$ (*17*). Table II contains the $Eu(NO_3)_3$ densities which are precise to about (2–3) × 10^{-5} $g\cdot cm^{-3}$. These measurements extend only to 1.1014 $mol\cdot kg^{-1}$ since the amount of stock solution was limited.

Density Results for Aqueous $Eu(NO_3)_3$

Density data for $Eu(NO_3)_3$ solutions were represented by the least-squares equations

$$d = d^0 + \sum_{i=2}^{n} B_i X^{i/2} \tag{1}$$

where $X = m$ in $mol\cdot kg^{-1}$ or c in $mol\cdot dm^{-3}$, and d^0 = 0.997 045 $g\cdot cm^{-3}$ (*17*). Table II contains our experimental densities, and Table III contains the least-squares B_i for eq 1. These coefficients were calculated by giving equal weight to our densities and to those of Spedding et al. (*13*).

Figure 1 shows the differences between the experimental densities and the least-squares fit to eq 1. The data of Spedding et al. (*13*) are about 4 × 10^{-5} $g\cdot cm^{-3}$ below ours, which only slightly exceeds our experimental precision of (2–3) × 10^{-5} $g\cdot cm^{-3}$. Spedding et al. used a magnetically controlled float, and their densities are slightly more precise than ours. However, the major source of error was their stock solution concentration uncertainty of ±0.1% which exceeds our concentration uncertainty. Thus, these two studies probably agree within experimental error. Densities of Jeżowska-Trzebiatowska et al. (*15*) are too high by 0.7–1.8% and were rejected.

Table IV. $La(NO_3)_3$ Osmotic Coefficients from Freezing Point Depression Data[a]

m, $mol\cdot kg^{-1}$	Φ_f[b]	Φ[c]	m, $mol\cdot kg^{-1}$	Φ_f[b]	Φ[c]
0.022 311	0.8002	0.7952	0.051 483	0.7670	0.7639
0.023 699	0.7964	0.7915	0.087 282	0.7514	0.7511
0.043 599	0.7734	0.7697	0.177 14	0.7363	0.7420[d]

[a] Based on the data of Hall and Harkins (*20*). [b] Osmotic coefficient at the freezing temperature. [c] Osmotic coefficient at 25 °C. [d] This point given zero weight in the least-squares fits.

Apparent molal volumes for $Eu(NO_3)_3$ were calculated by use of the equation

$$\phi_V = \frac{10^3(d^0 - d)}{m d^0 d} + \frac{M_2}{d} \tag{2}$$

where M_2 is the molecular mass of $Eu(NO_3)_3$. These values are also given in Table II. The density differences of 4 × 10^{-5} $g\cdot cm^{-3}$ between us and Spedding et al. (*13*) affect ϕ_V by 1.2 $cm^3\cdot mol^{-1}$ at 0.04 $mol\cdot kg^{-1}$, but change ϕ_V by only 0.03 $cm^3\cdot mol^{-1}$ at 1.1 $mol\cdot kg^{-1}$.

Activity Results for Aqueous $La(NO_3)_3$

Osmotic coefficients, Φ, of $La(NO_3)_3$ were calculated by using the equation for isopiestic equilibrium

$$\Phi = \frac{\nu^* m^* \Phi^*}{\nu m} \tag{3}$$

where m is the molality of $La(NO_3)_3$, $\nu = 4$ is the number of ions formed by the dissociation of one molecule of $La(NO_3)_3$, and the corresponding quantities for $CaCl_2$ reference solutions in equilibrium with the $La(NO_3)_3$ solutions are denoted with asterisks ($\nu^* = 3$). $CaCl_2$ Φ^* values were taken from a published equation (*18*). Table I contains the isopiestic equilibrium molalities and $La(NO_3)_3$ Φ values.

Other activity data have been reported for aqueous $La(NO_3)_3$. Two freezing point depression studies were made for $La(NO_3)_3$ early this century (*19*, *20*), and they can be converted to osmotic coefficients by using available heat capacity (*21*) and enthalpy of dilution data (*22*). The four points of Noyes and Johnston (*19*) are of low precision and are scattered, so they were rejected. Freezing point data of Hall and Harkins (20) were converted to osmotic coefficients at 25 °C, and these derived values are given in Table IV. Yakimov and Guzhavina's eight vapor pressure points (*23*) were rejected because of considerable scatter; osmotic coefficients calculated from them are about right at 1.17 $mol\cdot kg^{-1}$ but are 3–10% high at other concentrations.

Kirgintsev and Luk'yanov (*24*) reported isopiestic data for seven $La(NO_3)_3$ compositions from 1.450 to 2.922 $mol\cdot kg^{-1}$ using NaCl as the isopiestic reference solution. Osmotic coefficients were recalculated from these data by using eq 3 and Hamer and Wu's (*25*) NaCl Φ^* equation. Their point at 1.978 $mol\cdot kg^{-1}$ is discrepant from their other values, but their other six points are in fair agreement with the values in Table I (≃0.5% lower). It was concluded previously (*4*) that their data are unreliable since the sulfate method of analysis is unreliable without precautions to destory nitrate ions. In fact, they actually used oxalate precipitation followed by ignition to La_2O_3. This method is less prone to coprecipitation problems, so our earlier criticism (*4*) of that part of their study (*24*) was unwarranted. However, they also used evaporation of samples followed by thermal decomposition, which has been shown to give high apparent concentrations (and low Φ values) owing to incomplete decomposition of the oxynitrate (*6*).

In addition to these other studies, there is our earlier isopiestic data relative to KCl and $CaCl_2$ reference solutions (*4*).

Table V. Isopiestic Molalities from Rard, Miller, and Spedding (*4*) Normalized to Table I Data

[reference], mol·kg^{-1}	[La(NO$_3$)$_3$], mol·kg^{-1}	Φ(La(NO$_3$)$_3$)
	KCl Reference	
0.20504	0.12481[a]	0.7491
0.21249	0.12990[a]	0.7454
0.50176	0.29628[a]	0.7614
0.67018	0.39041	0.7701
0.68341	0.39762	0.7710
0.81030	0.46634	0.7791
1.0147	0.57326	0.7942
1.1959	0.66600	0.8070
1.3538	0.74444	0.8189
1.5736	0.85124	0.8353
1.7837	0.95067	0.8511
1.9755	1.0401	0.8650
2.2806	1.1789	0.8872
2.3641	1.2152	0.8941
	CaCl$_2$ Reference	
1.2564	1.1886	0.8876
1.2733	1.2052	0.8914
1.2947	1.2282	0.8947
1.3186	1.2548	0.8979
1.4587	1.4031	0.9239
1.5903	1.5467	0.9480
1.6822	1.6489	0.9650
1.7515	1.7267	0.9780
1.8250	1.8090	0.9925
1.9094	1.9040	1.0096
1.9724	1.9773	1.0215
2.0519	2.0671	1.0385
2.1395	2.1653	1.0581
2.2266	2.2681	1.0756
2.2962	2.3531	1.0887
2.3751	2.4490	1.1042
2.4518	2.5374	1.1219
2.5367	2.6353	1.1417
2.6053	2.7215	1.1550
2.7061	2.8409	1.1780
2.8198	2.9802	1.2025
2.8982	3.0733	1.2210
2.9329	3.1194	1.2273
2.9706	3.1625	1.2369
3.0413	3.2511	1.2521
3.1164	3.3419	1.2697
3.1885	3.4331	1.2852
3.2634	3.5326	1.2997
3.3399	3.6276	1.3172
3.4201	3.7281	1.3353
3.5365	3.8787	1.3601
3.5962	3.9549	1.3733
3.6563	4.0337	1.3859
3.7271	4.1252	1.4013
3.7693	4.1745	1.4123
3.8288	4.2526	1.4248
3.8359	4.2634	1.4258
3.9218	4.3700	1.4461
4.0050	4.4816	1.4630
4.0743	4.5710	1.4782
4.1572	4.6797	1.4958
4.2724	4.8281	1.5211
4.3374	4.9181	1.5333
4.3962	4.9935	1.5462
4.5083	5.1414	1.5693
4.5367	5.1792	1.5750
4.6688	5.3560	1.6011
4.7452	5.4558	1.6168
4.8233	5.5595	1.6320
4.9152	5.6802	1.6502
5.0238	5.8156	1.6732
5.0703	5.8780	1.6817
5.1577	5.9954	1.6972
5.2457	6.1044	1.7152
5.3125	6.1876	1.7283
5.3978	6.2910	1.7456
5.4884	6.4116	1.7604
5.5404	6.4779	1.7695
5.5888	6.5356	1.7789

[a] These three points only were not adjusted; see text for details.

Table VI. Coefficients and Powers for La(NO$_3$)$_3$ Osmotic Coefficient Polynomial[a]

i	r_i	A_i
1	0.75	0.568 8551
2	1.00	22.395 64
3	1.25	−53.576 35
4	1.50	57.930 19
5	1.75	−33.226 22
6	2.00	9.830 907
7	2.25	−1.184 351
$\sigma(\Phi)$	0.00141	

[a] These parameters apply to 8.4591 mol·kg^{-1}.

We concluded above that these reference molalities were reliable; only the La(NO$_3$)$_3$ stock solution molality was incorrect. It should therefore be possible to "normalize" the earlier concentration scale to our present data in their overlap region, to generate a corrected set of isopiestic molalities from that earlier study (*4*).

The isopiestic reference solution used for the isopiestic measurements in Table I was CaCl$_2$, and samples from the very same CaCl$_2$ stock solution were used in the earlier study (*4*) for La(NO$_3$)$_3$ nominal concentrations of 1.1836–6.4739 mol·kg^{-1}. Thus, the new La(NO$_3$)$_3$ to CaCl$_2$ isopiestic molality ratio *R* can be used to adjust the earlier La(NO$_3$)$_3$ molalities from their isopiestic CaCl$_2$ concentrations.

A large expanded-scale plot of the isopiestic molality ratio $R = m/m^*$ was made for all of the data in Table I. For certain selected molalities of CaCl$_2$ obtained in the earlier study (*4*), the *R* values were graphically interpolated and then used to calculate what the La(NO$_3$)$_3$ molality should have been for that equilibration. From this adjusted La(NO$_3$)$_3$ molality, the final weights of La(NO$_3$)$_3$ solution for the two samples used for that particular equilibration, and the weights of stock solution added initially to these isopiestic cups, we back-calculated what the correct number of moles of La(NO$_3$)$_3$ and the weight of water were per gram of stock solution for the earlier study. This was done for seven approximately equally spaced CaCl$_2$ concentrations from the earlier study: 2.5367 (1.0036), 3.0413 (1.0061), 3.5365 (1.0068), 4.0050 (1.0084), 4.5367 (1.0087), 5.0238 (1.0099), and 5.5888 (1.0108) mol·kg^{-1}. The numbers in parentheses are the ratios of the corrected to the uncorrected La(NO$_3$)$_3$ molality for that particular CaCl$_2$ reference molality. These La(NO$_3$)$_3$ molality ratios are uncertain between about 0.0005 and 0.0023 based on uncertainties in the *R* values (which have been graphically smoothed at one stage during the calculations). This normalization was done at relatively high concentrations since the larger differences there give greater precision to the final results. The back-calculated La(NO$_3$)$_3$ stock solution concentration was nearly independent of the selected equilibrium CaCl$_2$ molalities and highly consistent, and the average value was used to recalculate the La(NO$_3$)$_3$ molalities. This shows that the earlier study has nearly the same concentration dependence for Φ as the present study.

Table V contains the normalized isopiestic results from the earlier study (*4*). Only the three lowest La(NO$_3$)$_3$ molalities in this table were not adjusted, since they were done with samples from the same stock solution used in the current study. Molality changes for La(NO$_3$)$_3$ are 1% at about 6 mol·kg^{-1} but decrease to about 0.3% at low concentrations; this variation is just that due to changes in molality with changes in the weight fraction of water present. The uncertainty in the adjusted molalities due to the normalization is fairly small. For example, this uncertainty in the highest La(NO$_3$)$_3$ molality in Table V is only 0.19% and it is 0.08% at 1.1886 mol·kg^{-1}.

Osmotic coefficients for La(NO$_3$)$_3$ were then represented by the least-squares equation

$$\Phi = 1 - (A/3)m^{1/2} + \sum_i A_i m^{r_i} \qquad (4)$$

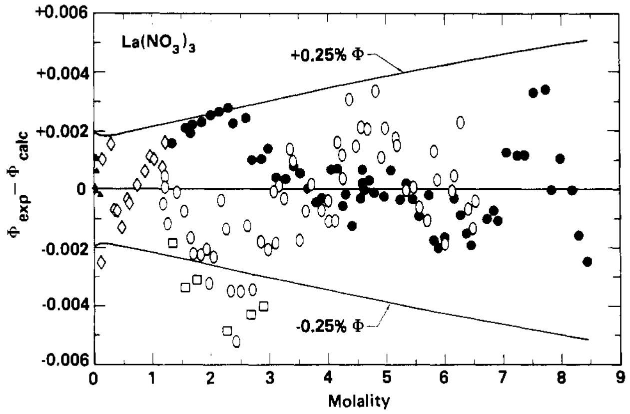

Figure 2. Differences between experimental osmotic coefficients and the least-squares eq 4 for La(NO$_3$)$_3$ solutions: (●) this research; corrected data of Rard et al. (*4*) from Table V: (◇) KCl standard and (○) CaCl$_2$ standard; (▲) freezing point depression data (*20*) from Table IV; (□) isopiestic data of Kirgintsev and Luk'yanov (*24*).

where A = 8.6430 is the Debye-Hückel limiting slope for a 3-1 electrolyte. Unit weights were given to our isopiestic data in Table I and to the freezing point depression results in Table IV, and weights of 0.5 were given to the normalized isopiestic data in Table V and to six of Kirgintsev and Luk'yanov's (*24*) points. Other data (*19*, *23*) were rejected as inaccurate.

Least-squares coefficients for the best fit to eq 4 and the standard deviation of the fit $\sigma(\Phi)$ are given in Table VI. Table VII contains calculated values of Φ, a_1, and $\gamma_\pm$. Here a_1 is the water activity of the solution, and $\gamma_\pm$ is the mean molal activity coefficient calculated from

$$\ln \gamma_\pm = -Am^{1/2} + \sum_i A_i\left(\frac{r_i + 1}{r_i}\right)m^{r_i} \tag{5}$$

Figure 2 shows the difference between the various experimental Φ values for La(NO$_3$)$_3$ and their corresponding values calculated by using the least-squares parameters in Table VI. These various sets of data are obviously now in excellent agreement.

The normalized data of Rard et al. (*4*), Table V, agree with our new Φ values within 0.0–0.2% over most of their overlapping concentration range, but slightly systematic differences of 0.3–0.5% do occur from 1.65 to 2.9 mol·kg^{-1} for the earlier study using CaCl$_2$ reference standard data. Our new Φ values are more precise and are probably more accurate than the earlier data in this concentration region. However, their KCl standard data connect up smoothly with our redetermined data of Table I.

The experimental Φ data up to 1.5 mol·kg^{-1} were also represented by Pitzer's equation (*26*) using A^ϕ = 0.3920. Following Pitzer et al. (*26*), we set $3\beta^{(1)}/2$ = 7.700 and used their recommended weighting scheme. Least-squares calculations then give $3\beta^{(0)}/2$ = 0.7374, $(3^{3/2}/2)C^\phi$ = −0.1989 and $\sigma(\Phi)$ = 0.0069. These parameters have changed slightly from those reported earlier (*4*). Higher concentration results were not included because of excessive cycling of this equation around the Φ data.

The isopiestic molality ratio of La(NO$_3$)$_3$ to CaCl$_2$, R, is greater than unity above 1.995 mol·kg^{-1}, whereas we expect it to be less than unity for dissociated salts of these valence types. Since CaCl$_2$ is a strong electrolyte, this implies that La(NO$_3$)$_3$ has considerable amounts of complex formation present at moderate and high concentrations.

Table VII. Osmotic Coefficients, Water Activities, and Activity Coefficients of La(NO$_3$)$_3$ at Even Molalities

m, mol·kg^{-1}	Φ	a_1	$\gamma_\pm$
0.01	0.8328	0.999400	0.5500
0.05	0.7642	0.997250	0.3699
0.1	0.7496	0.994613	0.3077
0.2	0.7507	0.98924	0.2588
0.3	0.7602	0.98370	0.2366
0.4	0.7720	0.97799	0.2238
0.5	0.7848	0.97212	0.2157
0.6	0.7983	0.96608	0.2105
0.7	0.8123	0.95985	0.2071
0.8	0.8270	0.95344	0.2052
0.9	0.8421	0.94685	0.2043
1.0	0.8577	0.94007	0.2042
1.2	0.8900	0.92593	0.2061
1.4	0.9235	0.91104	0.2101
1.5	0.9407	0.90332	0.2127
1.6	0.9580	0.8954	0.2157
1.8	0.9932	0.8791	0.2228
2.0	1.0288	0.8622	0.2312
2.2	1.0647	0.8447	0.2407
2.4	1.1007	0.8267	0.2513
2.5	1.1187	0.8175	0.2570
2.6	1.1366	0.8082	0.2630
2.8	1.1725	0.7893	0.2757
3.0	1.2081	0.7702	0.2894
3.2	1.2434	0.7507	0.3043
3.4	1.2785	0.7311	0.3201
3.5	1.2958	0.7212	0.3285
3.6	1.3131	0.7113	0.3371
3.8	1.3475	0.6914	0.3551
4.0	1.3814	0.6715	0.3743
4.2	1.4149	0.6517	0.3947
4.4	1.4481	0.6318	0.4162
4.5	1.4645	0.6219	0.4275
4.6	1.4808	0.6121	0.4391
4.8	1.5132	0.5925	0.4632
5.0	1.5452	0.5731	0.4887
5.2	1.5768	0.5539	0.5156
5.4	1.6080	0.5349	0.5440
5.5	1.6235	0.5255	0.5588
5.6	1.6389	0.5161	0.5739
5.8	1.6695	0.4977	0.6055
6.0	1.6998	0.4795	0.6388
6.2	1.7297	0.4617	0.6738
6.4	1.7594	0.4442	0.7107
6.5	1.7741	0.4356	0.7299
6.6	1.7888	0.4271	0.7495
6.8	1.8179	0.4103	0.7904
7.0	1.8468	0.3939	0.8334
7.2	1.8754	0.3779	0.8787
7.4	1.9038	0.3623	0.9263
7.5	1.9179	0.3547	0.9510
7.6	1.9320	0.3471	0.9764
7.8	1.9599	0.3323	1.0290
8.0	1.9877	0.3179	1.0844
8.2	2.0152	0.3040	1.1425
8.4	2.0425	0.2904	1.2036
8.4591	2.0505	0.2865	1.2223

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Glossary

d	density of solution, g·cm^{-3}
d^0	density of pure water, g·cm^{-3}
ϕ_V	apparent molal volume of solute, cm^3·mol^{-1}
Φ	molal osmotic coefficient
ν	number of ions formed by the dissociation of one molecule of solute
m	molal concentration of solute, mol·kg^{-1}
c	molar concentration of solute, mol·dm^{-3}

*	symbols with asterisks refer to isopiestic reference solution
B_i	least-squares coefficients of eq 1
A_i	least-squares coefficients of eq 4 and 5
r_i	powers of eq 4 and 5
A	Debye–Hückel constant for 3–1 electrolytes
M_1	molecular mass of water, g·mol^{-1}
M_2	molecular mass of solute, g·mol^{-1}
$\beta^{(0)}$, $\beta^{(1)}$, C^{ϕ}	parameters for Pitzer's equations
A^{ϕ}	Debye–Hückel constant (1–1 charge type) for Pitzer's Φ equation
σ	standard deviation of fitting equations
$\gamma_{\pm}$	mean molal activity coefficient
a_1	water activity
Φ_f	osmotic coefficient at the freezing temperature of the solution
R	isopiestic molality ratio of $La(NO_3)_3$ to $CaCl_2$

Registry No. $La(NO_3)_3$, 10099-59-9; $Eu(NO_3)_3$, 10138-01-9.

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Viscosities of Gaseous R13B1, R142b, and R152a

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The viscosities of gaseous bromitrifluoromethane (R13B1), 1-chloro-1,1-difluoroethane (R142b), and 1,1-difluoroethane (R152a) were measured with an oscillating disk viscometer of the Maxwell type at 273.15–448.15 K up to 10 MPa for R13B1 and at 298.15–423.15 K up to 5 MPa for R142b and R152a. Two empirical equations were obtained; one is for the viscosity at atmospheric pressure as a function of temperature, and the other is for the viscosity in the whole range of the present measurement as a function of temperature and density. The intermolecular force constants of Stockmayer 12-6-3 potential were determined from the temperature dependence of the viscosity at atmospheric pressure as follows: ϵ/k = 235 K, σ = 0.506 nm, and δ = 0.058 for R13B1; ϵ/k = 362 K, σ = 0.499 nm, and δ = 0.36 for R142b; ϵ/k = 312 K, σ = 0.463 nm, and δ = 0.62 for R152a.

Many halogenated hydrocarbons are commonly used as refrigerants and expected to be used as working fluids of turbines. However, the experimental data of gas viscosity needed for the design of related equipments are scarce and the reliability of the literature data is uncertain because of the large discrepancies among them. Therefore, the measurement of the gas viscosities of halogenated hydrocarbons under pressure is being continued by the present authors, and the viscosity data for chlorodifluoromethane (R22), dichlorodifluoromethane (R12), chlorotrifluoromethane (R13), 1,2,2-trichloro-1,1,2-trifluoroethane (R113), 1,2-dichloro-1,1,2,2-tetrafluoroethane (R114), and chloropentafluoroethane (R115) were reported previously (*1–4*). The viscosities of gaseous bromotrifluoromethane (R13B1), 1-chloro-1,1-difluoroethane (R142b), and 1,1-difluoroethane (R152a) are described in the present paper.

Table I. Physical Properties of Refrigerants

	R13B1	R142b	R152a
chem formula	$CBrF_3$	$CClF_2CH_3$	CHF_2CH_3
mol wt[a]	148.910	100.496	66.050
T_b,[a] K	215.4	263.4	248.2
T_c,[a] K	340.2	410.2	386.6
P_c,[b] MPa	3.97	4.12	4.50
ρ_c,[b] kg·m^{-3}	760	435	365
dipole moment,[c] D	0.7	2.1	2.3

[a] T_b, boiling point at atmospheric pressure; T_c, critical temperature; P_c, critical pressure; ρ_c, critical density. [b] Makita, T. *Viscosity and Thermal Conductivity*; Baifukan: Tokyo, 1975; p 224 and 225. [c] Reid, R. C.; Prausnitz, J. M.; Sherwood, T. K. *The Properties of Gases and Liquids*; McGraw Hill: New York, 1977; p 630 and 636.

The viscosity of gaseous R13B1 has been measured by Tsui (*5*), Reed et al. (*6*), Wilbers (*7*), Karbanov et al. (*8*), and Kletskii et al. (*9*). Figure 1 shows the temperatures and pressures at which the literature data were obtained. The present measurement covers the area with oblique lines. The point C.P. denotes the critical point. No experimental data for the viscosities of R142b and R152a could be found in the literature.

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