inner sphere water will remain. Consequently, the inner sphere two-series effect observed for the partial molal volumes (*18*) is then also observed for the transport properties at high concentrations. Even in dilute solutions the inner sphere two-series effect is reflected in the net hydration.

From conductance and other data, Spedding and Rard (*20*) deduced that water sharing is more important in the perchlorate system than in the chloride system at the same molal concentrations, and water sharing should just be starting to dominate in the chlorides when saturation occurs. If the rare earth chloride solubilities were greater, then one would expect the complete two-series effect to show up with La to Nd forming one series and Tb to Lu forming a second series and with the remaining rare earth chlorides being intermediate in behavior.

Spedding et al. (*21*) have discussed the effect of complexation on the conductance behavior of the rare earth chlorides. They conclude that outer sphere complexation gives rise to very little distinctive behavior for individual rare earth chlorides, and complexation may be affecting all of the rare earths by about equal amounts. Similar considerations hold for the relative viscosities.

Acknowledgment

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Electrical Conductances of Some Aqueous Rare Earth Electrolyte Solutions at 25°C. II. Rare Earth Chlorides

Frank H. Spedding,[1] **Joseph A. Rard, and Victor W. Saeger**
Ames Laboratory-USAEC and Department of Chemistry, Iowa State University, Ames, Iowa 50010

The electrical conductances of aqueous solutions of $LaCl_3$, $PrCl_3$, $NdCl_3$, $SmCl_3$, $EuCl_3$, $GdCl_3$, $TbCl_3$, $DyCl_3$, $HoCl_3$, $ErCl_3$, $TmCl_3$, $YbCl_3$, and $LuCl_3$ were measured over the concentration range of approximately $0.02m$ to saturation at 25°C. The equivalent conductances of the rare earth chlorides at each molality decreased smoothly from $SmCl_3$ to $LuCl_3$, and $LaCl_3$ through $NdCl_3$ had very similar conductances. By $0.16m$, the equivalent conductances of the rare earth chlorides dropped below those of the corresponding perchlorates. The differences between the chloride and perchlorate systems were discussed in terms of ionic hydration, anion-cation complex formation, the degree of cation hydrolysis, and the effect of these electrolytes on the hydrogen-bonded water structure.

The lanthanides form the most complete series of trivalent cations available for the study of aqueous solution properties. This paper is one of a series reporting accurate transport data for aqueous solutions of the rare earth perchlorates (*26*), chlorides (*23, 29*), and nitrates (*16*) up to saturation at 25°*C*.

A moderate amount of data exists concerning the nature of the rare earth ion-chloride ion interaction. Fluorescent (*18*) and visible spectra (*3*) measurements show little evidence for complex formation even at moderately high concentrations, but the formation of outer sphere complexes would probably not appreciably affect transitions within the 4f subshell. Nmr (*13*) and ultrasonic absorption (*6*) measurements also indicate the absence of strong complex formation between the rare earth and chloride ions.

Thermodynamic evidence does indicate the formation of weak, outer sphere complexes. There has been some success in determining the formation constants for the rare earth chloride complexes by thermodynamic (*4, 14*), electrochemical (*7*), and spectral (*11*) methods. Ahrland (*1*) has discussed some of the evidence for outer sphere complex formation.

Spectral (*32*) and solution viscosity (*31*) measurements have been interpreted as giving evidence for the greater disruption of the hydrogen-bonded water structure by the perchlorate ion than by the chloride ion. The disruption of the water structure should tend to enhance the

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[1] To whom correspondence should be addressed.

movement of the hydrated ions through the solution. This "structure breaking" consideration should apply up to concentrations at which the majority of the water is shared between ions.

Experimental

Most of the equipment, techniques, and procedures are the same as described in the perchlorate conductance paper (*26*). Measurements were made on solutions of the stoichiometric salts prepared by the method of Spedding et al. (*24*). Solutions prepared in this manner should have the same composition as solutions prepared directly from the anhydrous salt and conductivity water.

All dilutions were prepared by weight from samples of the corresponding stock solutions and conductivity water, and all weights were corrected to vacuum. Saturated solutions were prepared by concentrating samples of the corresponding stock solutions followed by three-week equilibrations in a constant temperature bath. A few of the dilute solution resistance measurements were performed in commercially available Leeds and Northrup cells. The majority of the solutions were run in a single capillary cell which had also been used for some of the perchlorate solution measurements.

This capillary cell was calibrated against the standards of Jones and Prendergast (*10*). Two 0.1 and two 1.0 normal standards were used, and a cell constant of 253.50 ± 0.02 cm^{-1} was obtained. Each reported specific conductivity is the average of two to four measurements. This average is reliable to ±0.02% above 0.05m and is reliable to about ±0.1% below this concentration. No cell electrode oxidation occurred with the rare earth chloride solutions.

The concentrated stock and saturated solutions were analyzed by oxide, EDTA, or sulfate methods (*20, 24*). Secondary stock solutions were prepared from the concentrated stock solutions by weight dilution, and each of these was analyzed by one or more of the above methods and by standard gravimetric or volumetric chloride methods. The average analyses of each secondary stock solution agreed to 0.04% or better with the concentration calculated from the concentrated stock solution analysis. The concentrated stock solution concentrations were therefore estimated to be reliable to at least ±0.1%. The anion and cation analyses for each secondary stock agreed to within experimental error (0.1%) in all cases.

Except for $LaCl_3$, the measurements for each separate salt were performed on a series of dilutions prepared by weight from a single concentrated stock solution. Two stocks of $LaCl_3$ were separately prepared and analyzed, and their conductances measured to determine the reproducibility of the experimental data.

Errors and Data Treatment

The calculations were done in the same manner as for the rare earth perchlorates, and errors of similar magnitude were obtained (*26*). The density data of Spedding and coworkers (*17, 27*) were used to calculate the densities of the solutions studied. The density data were fitted to fifth-order polynomials of the form:

$$d = \sum_{i=0}^{5} A_i m^i \qquad (1)$$

where m is the molality. The density coefficients are reported in Table I.

The conductance data are reported in Table II (deposited with the ACS Microfilm Depository Service). In this table L is the specific conductivity corrected for the solvent conductivity, and Λ is the equivalent conductance. The number of experimental concentrations ranged from 18 to 33, and for seven salts at least 30 concentrations were studied. The units of Λ are (absolute ohm)$^{-1}$ cm^2 equivalent^{-1}, and those of L are (absolute ohm)$^{-1}$ cm^{-1}. The data are generally reported to five figures, although they are uncertain in the fourth figure. The extra figure was included since the internal consistency of the data is greater than its absolute accuracy.

Seventh-order polynomials of the form:

$$\Lambda = \sum_{i=0}^{7} B_i m^{i/2} \qquad (2)$$

were used to represent the equivalent conductance data from 0.1m to saturation, and these coefficients are listed in Table III. These equations represent the equivalent conductance data within experimental error. The inverse squares of the probable errors in the equivalent conductances were used as the weighting factors. These probable errors were calculated by the method of propagation of errors by use of the experimental errors in the solution concentrations, densities, and specific conductivities along with the uncertainties in the molecular weights (IUPAC, 1969). The calculated probable errors ranged from less than 0.01 to about 0.05 ohm^{-1} cm^2 equivalent^{-1}, with the smaller errors occurring for the more concentrated solutions. The molal solubilities, to be used with Equations 1 and 2, are listed in Table IV.

Relative percent differences were calculated from the equation:

$$\frac{\Lambda_{RECl_3} - \Lambda_{LuCl_3}}{\Lambda_{LuCl_3}} \times 100 \qquad (3)$$

where RE represents the rare earth of interest. These percent differences were used to represent the data in convenient graphical forms. The real differences between the conductances of the various rare earth chlorides cannot be seen on a small scale direct plot but can be seen on these ratio curves.

The dilute solution conductances of all of the rare earth chlorides studied have been reported (*19, 21, 22, 25, 30*). The resistances of six of the salts reported here were measured to low concentrations and should be reliable to about 0.1% at these low concentrations. Assuming an equal reliability for the literature values, then the two sets of data should agree to 0.2% or better in the region of overlap. Graphical comparison indicated that this was the case. Jones and Bickford (*9*) measured the equivalent conductance of $LaCl_3$ to about 1m, and the data of this research agree with their values to within 0.1%.

Results

In Figure 1 the specific conductivities of $GdCl_3$, $Gd(ClO_4)_3$, and $Gd(NO_3)_3$ are shown as a function of the molality. In Figure 2 the equivalent conductance curves are given for these salts. The perchlorate data are from the earlier paper (*26*), and the nitrate data are from Rard (*16*). At low concentrations the specific conductivities and the equivalent conductances fall in the order expected from the differences in the limiting ionic conductances. At all concentrations the rare earth chloride solutions have higher specific conductivities than the corresponding perchlorates.

For the equivalent conductances above 0.16m, the opposite is true. This reverse order for the specific conductivities results from the fact that 1 cm^3 of a rare earth perchlorate solution contains a different amount of salt and water than the corresponding chloride, at the same molal concentration. This occurs since the densities of

Table I. Density Polynomial Coefficients for Chlorides

	$A_0 \times 10^1$	$A_1 \times 10^1$		
Salt	$A_4 \times 10^4$	$A_5 \times 10^5$	$A_2 \times 10^2$	$A_3 \times 10^8$
$LaCl_3$	9.971277	2.248507	−1.535259	1.64134
	−2.97661	2.85305		
$PrCl_3$	9.971619	2.302711	−1.417505	1.25351
	−2.57603	2.57038		
$NdCl_3$	9.971757	2.343180	−1.326872	0.697387
	−1.10401	0.959772		
$SmCl_3$	9.971415	2.397484	−1.407813	1.72431
	−4.67610	4.69583		
$EuCl_3$	9.971321	2.400198	−1.339700	1.03067
	−1.76966	0.791157		
$GdCl_3$	9.971482	2.445674	−1.413651	1.317935
	−2.50683	1.76306		
$TbCl_3$	9.971433	2.459172	−1.412103	1.24437
	−2.99622	3.472853		
$DyCl_3$	9.971616	2.501525	−1.389354	0.510472
	0.562664	−1.80744		
$HoCl_3$	9.971275	2.540806	−1.514183	1.61852
	−3.54946	3.24661		
$ErCl_3$	9.971577	2.567409	−1.427477	1.09091
	−2.32193	2.37679		
$TmCl_3$	9.971597	2.596590	−1.384439	0.836655
	−1.35214	0.851877		
$YbCl_3$	9.971807	2.642649	−1.350703	0.709944
	−1.55003	1.66078		
$LuCl_3$	9.971273	2.678113	−1.488819	1.84755
	−4.96995	5.02469		

Table III. Conductance Polynomial Coefficients for Chlorides

	B_0	B_1	B_2	B_3
Salt	B_4	B_5	B_6	B_7
$LaCl_3$	123.437546	−191.286752	387.335371	−571.029774
	495.423030	−256.398930	73.213099	−8.77352118
$PrCl_3$	114.929248	−118.846661	145.388177	−152.4686419
	87.5885244	−30.7011658	7.00565173	−0.778194961
$NdCl_3$	121.326562	−171.956615	323.839653	−462.298230
	389.422071	−197.355301	55.7282392	−6.63804017
$SmCl_3$	125.908335	−215.748555	478.534918	−745.282120
	678.381643	−363.558304	106.160321	−12.9231724
$EuCl_3$	120.765284	−175.545348	342.816492	−508.717326
	443.145239	−229.200510	65.2725779	−7.79560067
$GdCl_3$	121.943758	−184.227716	358.890498	−522.151848
	445.793583	−226.311144	63.5875468	−7.54463307
$TbCl_3$	121.643402	−189.945605	386.819627	−584.164751
	516.183423	−269.487390	77.3477510	−9.33743725
$DyCl_3$	122.232251	−194.706015	396.260651	−592.513956
	516.756641	−265.515096	74.9096355	−8.88701583
$HoCl_3$	120.175836	−183.460975	369.125686	−564.523862
	507.597885	−270.265888	79.1767184	−9.75243209
$ErCl_3$	120.363538	−183.747701	365.678931	−552.193787
	489.834751	−257.640366	74.7466424	−9.13398163
$TmCl_3$	118.671733	−171.839718	328.098680	−488.173521
	426.398065	−221.325262	63.6147989	−7.71753621
$YbCl_3$	119.589566	−175.643258	330.681918	−479.115211
	406.612279	−205.139404	57.5057613	−6.83142137
$LuCl_3$	118.080032	−164.060811	294.336909	−418.869239
	348.921780	−173.354589	48.1673447	−5.69718599

the solutions are different, and because the perchlorate and chloride salts have appreciably different molecular weights. The rare earth nitrates complex more than the perchlorates and chlorides; this complex formation is reflected in the lower nitrate salt conductances as seen in Figure 2.

In Figure 3 the percent difference between the experimental conductances and the values calculated from Equation 2 are shown for two of the salts studied. The two sets of data for $LaCl_3$, measured on solutions prepared from two separate stock solutions, fall within about 0.07% of each other over the entire concentration range. The solid curves represent the percent probable errors. The two $LaCl_3$ runs fall well within experimental error of each other.

Figures 4 and 5 are plots of the relative percent differences as a function of the molality. The Lu data on this type of plot fall along the molality axis.

Relative percent differences and relative viscosities (*23, 29*) are shown for the rare earth chloride salts at 2.5 and 3.5*m* in Figure 6. In the perchlorate conductance paper (*26*), a plot of the relative percent differences for both the chloride and perchlorate salts is given at 1.0 and 2.0*m*.

The bulk of thermodynamic (*1, 4, 14*) and spectral (*3, 11, 13*) measurements indicates that the rare earth and chloride ions form weak, outer sphere complexes. X-ray diffraction data (*2, 12*) indicate that a sheath of water remains between the chloride and rare earth ions, even near saturation. The main difference in complex formation between the rare earth perchlorate and chloride systems is that the rare earth chlorides form appreciable amounts of outer sphere complexes at relatively low concentrations, whereas the rare earth perchlorates presumably do not form appreciable amounts of outer sphere complexes until forced to do so by the mutual hydration needs of the ions.

Table IV. Aqueous Rare Earth Chloride Molal Solubilities at 25°C

Rare earth	Molality	Rare earth	Molality
La	3.8959	Dy	3.6310
Pr	3.8940	Ho	3.6965
Nd	3.9292	Er	3.7821
Sm	3.6401	Tm	3.8794
Eu	3.5864	Yb	4.0028
Gd	3.5906	Lu	4.1202
Tb	3.5727	...	...

There have been some equilibrium constant measurements performed on the rare earth chloride systems at an ionic strength of one (*4, 7, 14*). These measurements indicate that the light rare earth chlorides have the same numerical value for the first apparent formation constants within experimental error, and the heavy rare earths probably have smaller first formation constants. The equilibrium constants are near unity, and the various measurements are in qualitative agreement. Measurements of the first apparent formation constants at an ionic strength of three (*11*) indicate a weak maximum in the middle of the series.

The greater amount of complex formation in the rare earth chloride solutions should be a major factor in causing the conductances of the chloride salts to drop below those of the corresponding perchlorate salts, since fewer ions will remain in the chloride system to conduct the electrical current. The qualitative features of the relative percent differences, across the rare earth chloride series, change little with concentration (Figure 6). The same general shape also occurs at infinite dilution (*22*). Rare earth chloride viscosity data (*23, 29*) and the activity of water in rare earth chloride solutions (*28*) show related behavior over most of the concentration range.

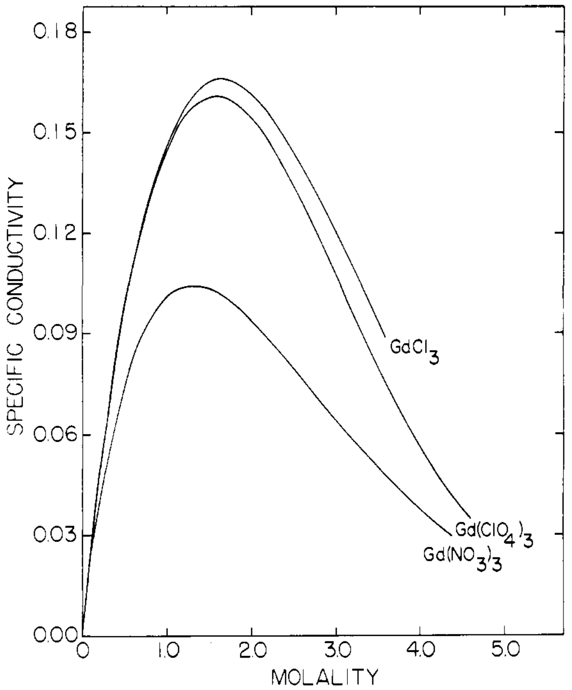

Figure 1. Specific conductivities of three aqueous gadolinium electrolytes

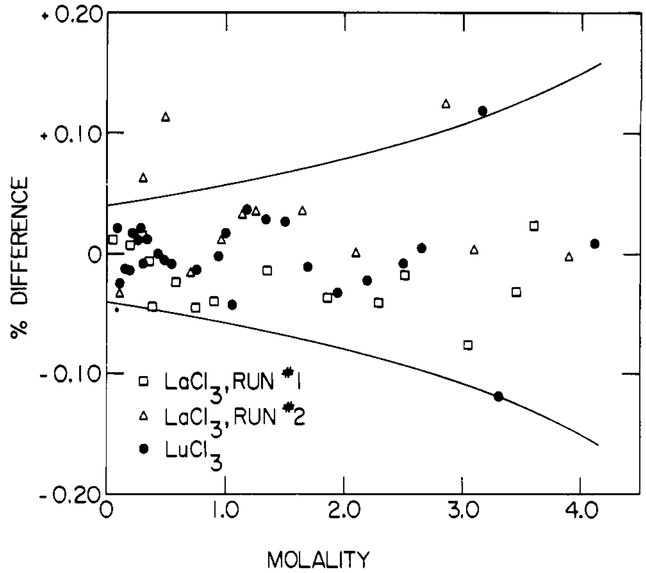

Figure 3. Percent differences between calculated and experimental equivalent conductances. Solid curves represent total probable errors

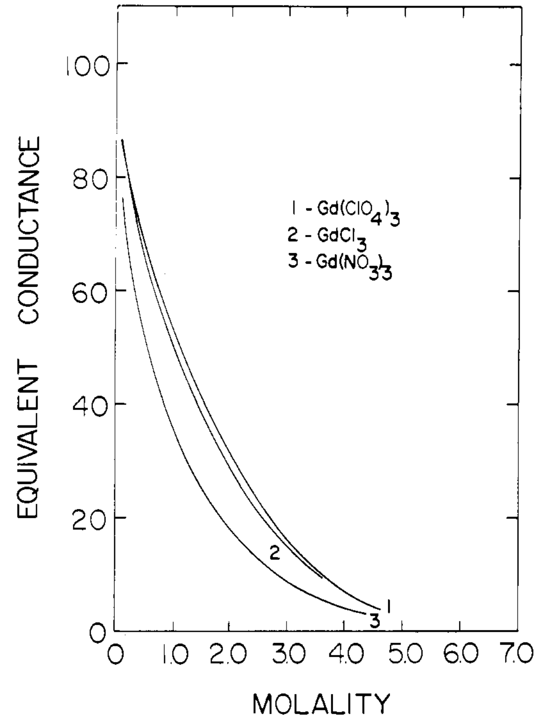

Figure 2. Equivalent conductances of three aqueous gadolinium electrolytes

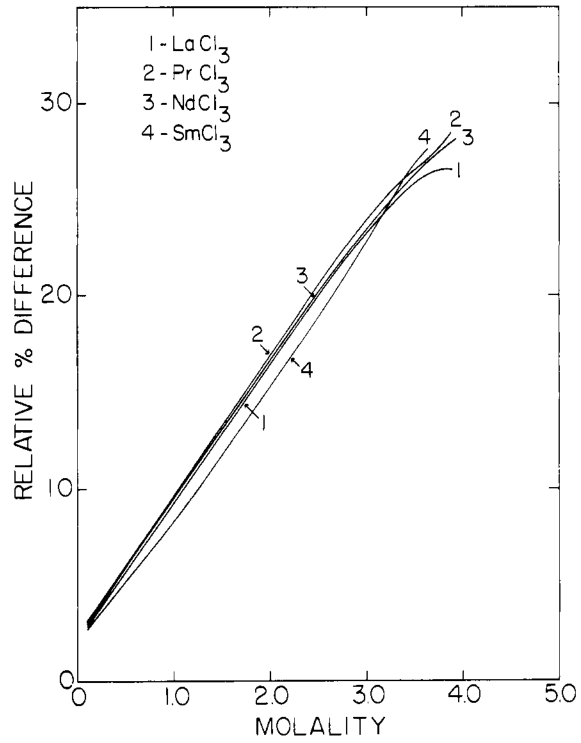

Figure 4. Percent differences in equivalent conductances relative to $LuCl_3$ for some light rare earth chlorides

No major new trends appear with concentration in the chloride solution conductances across the rare earth series. This implies that the effect of complex formation on the equivalent conductances is similar for all of the rare earth chloride solutions, although the equilibrium constants at an ionic strength of one (*4, 14*) do parallel the conductance series trend.

The viscosity B-coefficients (*31*) indicate that the large perchlorate ion disrupts the hydrogen-bonded water structure more than the chloride ion, causing a greater reduction in the resistance to flow encountered by the ions in the perchlorate solutions. This water structure disruption should increase with salt concentration until water sharing between the ions becomes important. At high concentrations water sharing will be extensive, and little or no free solvent will remain. Under this condition, the above considerations will not strictly apply, but the trends established at moderate concentrations, i.e., perchlorate solutions more conducting and less viscous, persist even to the concentrations at which the chlorides saturate out.

The reason for the continuation of this trend at high

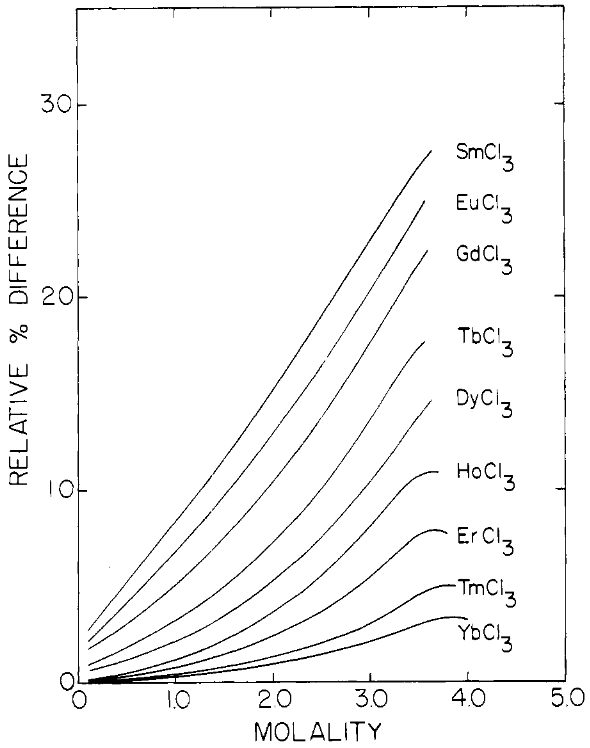

Figure 5. Percent differences in equivalent conductances relative to $LuCl_3$ for some middle and heavy rare earth chlorides

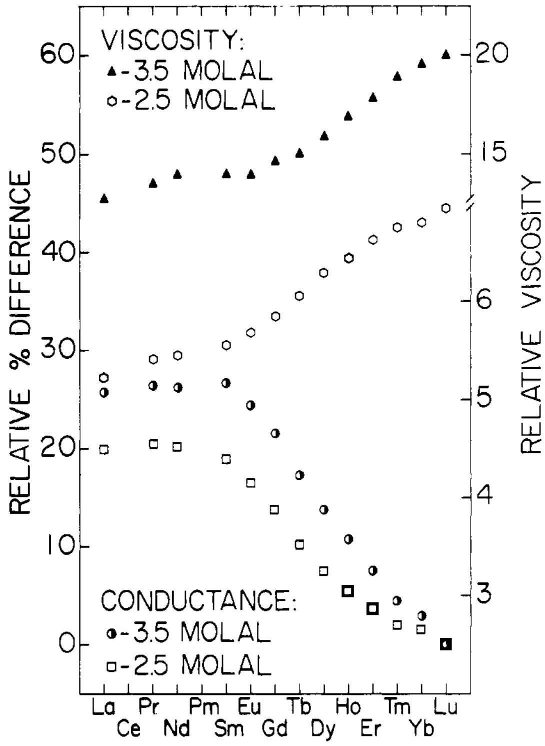

Figure 6. Relative percent differences in equivalent conductances and relative viscosities at constant molality for rare earth chlorides

concentrations may be due to the strength of the cation–water–anion interactions in the various systems. Ion-pairing occurs to a greater extent in the chloride systems (*3*); therefore, one might expect chloride–water–rare earth bonds to be stronger than the same type of bonds in the perchlorate systems at equal molalities. Since the conductance process involves the breaking and reforming of these bonds as the ions migrate in opposite directions, the relative strength of the ion-pairs will contribute to the larger perchlorate conductances in these concentrated solutions.

The pH values of the rare earth chloride solutions are given in Figure 7. These pH values indicate that hydrolysis probably occurs to a greater degree in the perchlorate systems (*26*) than for the corresponding chloride systems. Although the hydrogen ion contribution to conductance will contribute to the larger perchlorate solution conductance, it should not be the major reason for the qualitative difference in behavior between the two anion series.

All of the crossovers between the corresponding rare earth chloride and perchlorate equivalent conductances occur between 0.11 and 0.16*m*. The crossovers tend to occur at higher concentrations for the heavy rare earths than for the light ones. This is to be expected from the dilute solution equilibrium constant data, but the effect seems to be quite small. That the crossovers occur at low concentrations seems to indicate that the weak ion-pairing cannot be the only major cause for the crossovers, and the water structure disruption, as mentioned above, also contributes to the differences between the chloride and perchlorate systems. The viscosities of the rare earth chloride solutions are about 1.5% greater than for the corresponding perchlorate solutions at the crossover concentrations (*23, 29*), and this gives a rough measure of the importance of structural modifications. At high concentrations, however, ion-pairing effects probably dominate.

The surface charge density of the rare earth ions increases across the rare earth series, as the lanthanide contraction occurs, and this leads to an increase in the total number of water molecules associated with the cations, until concentrations at which water sharing between ions becomes important. The isomolal conductance should decrease down the rare earth series since the heavy rare earth ions will drag more water molecules than the light rare earth ions. Complex formation seems to be affecting all of the rare earth chlorides about the same amount and therefore should not alter this trend.

Spedding et al. (*24*) have suggested, from apparent molal volume data, that a decrease in the inner sphere hydration occurs across the rare earth series with the light rare earth ions having a higher inner sphere coordination number than the heavy ones. This change in primary hydration causes the isomolal conductance to change more rapidly with rare earth in the middle of the rare earth series and gives rise to the approximate S shape in the conductance across the rare earth series.

A prominent high-concentration two-series effect occurred in the rare earth perchlorate conductances (*26*) above 3.5*m*. At these high concentrations most of the water should be shared between ions. The smaller hydrated rare earth ions will have a greater surface charge density than the larger hydrated ions; therefore, the strength of the outer sphere ion-pairs should increase across the rare earth series at these high concentrations. This gives rise to a decrease in conductance and an increase in viscosity across the rare earth series at high concentrations. When the inner sphere hydration decrease occurs, the "release" of this tightly bound water should help to reduce both the viscosity and the amount of ion-pairing present. This gives rise to the increase in

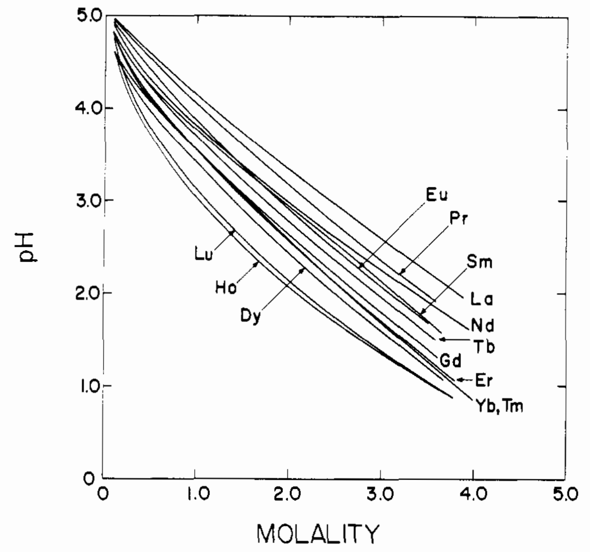

Figure 7. pH values of rare earth chlorides as function of molality

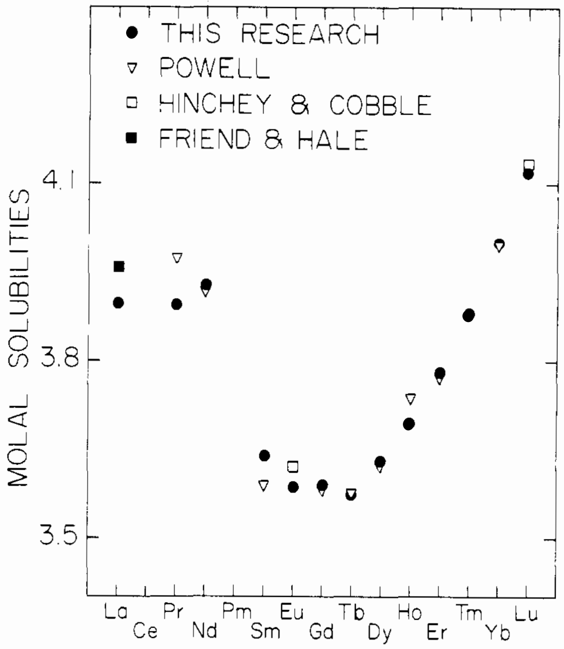

Figure 8. Solubilities of aqueous rare earth chlorides at 25°C

the isomolal perchlorate conductance between Nd and Tb.

At high concentrations this two-series effect is beginning to appear in the chloride viscosities, but since the chlorides begin to saturate out by $3.57m$, the trend never becomes well established. The appearance of the complete high-concentration two-series effect in the perchlorate solutions and its near absence in the chloride solutions at this concentration were attributed to more extensive water sharing in the concentrated perchlorate solutions (*26*).

In Figure 8 the molal solubilities of the aqueous rare earth chlorides are given at 25°C. Values reported in the literature (*5, 8, 15*) are also given on this graph, and in some cases, the agreement is not as good as might be desired.

If the conductances are plotted against the molar concentration instead of the molality, certain qualitative differences are observed in the curves. The equivalent conductance curve for each rare earth perchlorate at intermediate molarities recrosses the corresponding rare earth chloride curve, but this does not happen on the molal scale. It should be recalled that in comparing equal molarities, the ratio of the moles of salt to solute varies, since the densities change, whereas in comparing equal molalities, this ratio is constant. To compare the same amount of salt and solute in all cases, we have interpreted our data on the molal concentration scale.

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