Glossary

C_i	ion concentration, kg mol/m^3
C^*	interfacial concentration of molecular chlorine, kg mol/m^3
d	jet diameter, m
D	diffusion coefficient, m^2/s
H°	Henry's law coefficient for pure water, Pa m^3/(kg mol)
H	Henry's law coefficient corrected for ions in solution, Pa m^3/(kg mol)
h, h_-, h_+, h_G	empirical parameters, m^3/(kg mol)
I	ion valence
l	jet length, m
q	liquid flow rate, m^3/s
t	contact time, s
π	3.141 59

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Relative Viscosity and Density Data for Some Alkali Halides in Aqueous 20% Sucrose Solution at 25 °C

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Relative viscosity and density data are presented for seven alkali halides in aqueous 20% sucrose solution at 25 °C. The relative viscosities were fitted to the Jones-Dole equation. Corresponding data for aqueous solutions are also presented in most instances and the coefficients of the Jones-Dole equation compared where possible with literature values. *B* coefficients are shown to be additive properties of the constituent ions in aqueous 20% sucrose solution. Individual ionic *B* coefficients are assigned.

Introduction

For concentrations below 0.2 M, the dependence of the relative viscosity, η_r, of an electrolyte solution upon the molar concentration, C, of the electrolyte is generally in accordance with the Jones-Dole (*1*, *2*) equation

$$\eta_r = 1 + AC^{1/2} + BC \qquad (1)$$

The A coefficient of this equation is an index of ionic interactions. Falkenhagen (*3*) derived an explicit expression for A in terms of limiting ionic equivalent conductances λ and solvent properties. For a 1:1 electrolyte in a solvent of viscosity η_0 and dielectric constant ϵ_0, the expression reduces to

$$A = \frac{1.45}{\eta_0(2\epsilon_0 T)^{1/2}}\left[\frac{\lambda_1 + \lambda_2}{4\lambda_1\lambda_2} - \frac{(\lambda_1 - \lambda_2)^2}{(3 + 2^{1/2})\lambda_1\lambda_2(\lambda_1 + \lambda_2)}\right] \qquad (2)$$

where the subscripts 1 and 2 designate cation and anion, respectively.

Table I. Equivalent Conductances of Cesium Chloride Solutions in Aqueous 20% Sucrose at 25 °C

C	Λ, cm^2 Ω^{-1} g equiv^{-1}
0.002 313 8	92.579
0.010 528	89.865
0.016 607	88.587
0.020 629	87.918
0.040 881	85.446
0.054 490	84.313

If the influence of incomplete dissociation (or ion association) and the consequences of an approximation made in Falkenhagen's theory are neglected, the B coefficient may be regarded as an index of ion-solvent interaction.

Experimental Section

Relative viscosities were determined by using a flared-capillary viscometer described in a previous publication (*4*). For this viscometer kinetic energy corrections were unnecessary for water and 20% sucrose solution (*4*, *5*) and hence for the systems included in the present study. Thus, relative viscosities were calculated according to

$$\eta_r = \rho t/(\rho_0 t_0) \qquad (3)$$

where ρ represents density and t flow time. The subscript 0 refers to values for the solvent. The uncertainty in the measurements of flow times was about 0.01%. For the minimum of three consecutive flow times which were averaged for each solution the reproducibility was usually much better than 0.01%.

Densities were determined by using tare pycnometry. Three single-stemmed bulb pycnometers of approximately 30-cm^3 capacity were fabricated such that their masses were equal within 0.002 g and all contained the same volume of liquid (within 0.02 cm^3) when filled to a scratch mark on the stem of each. An accompanying tare of closely similar mass and shape

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Table II. Relative Viscosities and Densities of Aqueous and Aqueous 20% Sucrose Solutions of Single Electrolytes at 25 °C[f]

C	ρ, g cm^{-3}	η_r	$10^4\Delta$	C	ρ, g cm^{-3}	η_r	$10^4\Delta$	C	ρ, g cm^{-3}	η_r	$10^4\Delta$
Aqueous Solutions											
NaCl[a]											
0.000991	0.99709	1.0002	0.5	0.024915	0.99808	1.0031	−1.5	0.124358	1.00217	1.0123	−0.6
0.001975	0.99713	1.0003	1.2	0.047287	0.99901	1.0051	0.8	0.149168	1.00319	1.0145	−0.2
0.004949	0.99725	1.0007	1.0	0.049810	0.99911	1.0056	−2.1	0.173950	1.00420	1.0166	0.7
0.009940	0.99746	1.0012	2.0	0.074684	1.00014	1.0077	0.2	0.198722	1.00520	1.0188	0.2
0.019841	0.99787	1.0024	0.6	0.099132	1.00114	1.0100	−0.3				
KCl[b]											
0.001954	0.99713	1.0001	1.4	0.049030	0.99936	1.0005	−0.8	0.147700	1.00397	1.0000	0.0
0.004887	0.99727	1.0002	0.8	0.074994	1.00057	1.0003	0.7	0.171721	1.00508	0.9998	−0.3
0.009754	0.99750	1.0004	−0.5	0.097851	1.00163	1.0003	−0.2	0.197426	1.00630	0.9996	0.1
0.025038	0.99822	1.0005	−0.5	0.120943	1.00272	1.0001	0.3				
CsCl[c]											
0.004792	0.99765	1.0000	0.8	0.072175	1.00638	0.9978	0.2	0.148668	1.01612	0.9945	0.2
0.012457	0.99867	1.0001	−1.0	0.074560	1.00666	0.9978	−1.0	0.173241	1.01929	0.9935	−0.2
0.024904	1.00027	0.9997	−1.3	0.098295	1.00969	0.9965	1.7	0.197446	1.02239	0.9922	2.4
0.048639	1.00331	0.9985	1.6	0.123886	1.01297	0.9957	−1.1	0.197801	1.02240	0.9926	−2.3
NaI											
0.009840	0.99800[d]	1.0007	−0.3	0.074399	1.00557	1.0021	0.8	0.146557	1.01381	1.0035	−0.1
0.024754	0.99989	1.0012	−0.9	0.097721	1.00822	1.0026	0.3	0.172133	1.01673	1.0038	0.9
0.049423	1.00271	1.0017	−0.5	0.122280	1.01104	1.0030	0.6	0.198000	1.01969	1.0044	−1.2
KI											
0.009905	0.99824	0.9997	−0.1	0.073362	1.00588	0.9952	0.4	0.147173	1.01475	0.9897	−0.6
0.024367	0.99999	0.9988	−0.8	0.096931	1.00872	0.9935	−0.1	0.171596	1.01768	0.9876	1.5
0.048594	1.00291	0.9970	0.6	0.122133	1.01174	0.9916	−0.3	0.197602	1.02081	0.9858	−0.8
CsI											
0.001945	0.99744	1.0000	−0.3	0.073959	1.01198	0.9924	1.0	0.146975	1.02669	0.9843	0.5
0.004886	0.99803	0.9999	−2.3	0.096730	1.01657	0.9898	1.6	0.171234	1.03158	0.9817	−0.3
0.024489	1.00198	0.9980	−2.5	0.120467	1.02136	0.9873	0.3	0.197121	1.03682	0.9789	−1.3
0.049117	1.00697	0.9950	1.0								
KBr											
0.001968	0.99722	1.0001	0.0	0.073741	1.00330	0.9982	−1.2	0.148023	1.00957	0.9951	1.1
0.004910	0.99747	1.0002	−0.1	0.098188	1.00537	0.9973	−1.6	0.172543	1.01163	0.9944	−1.9
0.024673	0.99915	0.9996	0.8	0.123768	1.00753	0.9960	1.4	0.198011	1.01378	0.9931	0.9
0.049719	1.00127	0.9988	0.8								
Aqueous 20% Sucrose Solutions											
NaCl[e]											
0.001098	1.07940	1.0003	0.8	0.028244	1.08044	1.0042	−1.7	0.142622	1.08483	1.0164	0.6
0.002183	1.07945	1.0005	1.3	0.056022	1.08155	1.0072	0.0	0.168645	1.08582	1.0190	1.0
0.005381	1.07957	1.0011	−0.4	0.056653	1.08156	1.0071	1.2	0.190787	1.08669	1.0213	0.8
0.011090	1.07979	1.0020	−1.4	0.080237	1.08247	1.0099	−1.1	0.202452	1.08714	1.0228	−1.7
0.020722	1.08015	1.0030	1.5	0.109123	1.08357	1.0129	−0.1				
KCl[c]											
0.010269	1.07982	1.0002	2.9	0.083648	1.08365	1.0000	−1.0	0.166360	1.08659	0.9986	0.2
0.026963	1.08058	1.0003	2.1	0.107788	1.08412	0.9998	−2.4	0.189794	1.08760	0.9981	0.8
0.054727	1.08180	1.0004	−0.5	0.143116	1.08561	0.9992	−1.3	0.213978	1.08855	0.9976	1.4
CsCl											
0.005254	1.08001[d]	1.0001	−0.5	0.081225	1.08951	0.9961	0.2	0.161167	1.09935	0.9913	0.6
0.013093	1.08101[d]	0.9998	−0.8	0.106814	1.09268	0.9946	0.1	0.188191	1.10268	0.9897	−0.2
0.026506	1.08271	0.9990	1.2	0.135579	1.09618	0.9927	1.7	0.213814	1.10586	0.9882	−1.1
0.052805	1.08599	0.9979	−1.9								
NaI											
0.010594	1.08059	1.0010	−2.6	0.079457	1.08820	1.0030	−1.0	0.161416	1.09721	1.0049	1.4
0.026769	1.08239	1.0014	0.3	0.106864	1.09124	1.0037	−0.8	0.183685	1.09965	1.0054	0.9
0.053917	1.08538	1.0021	1.0	0.134563	1.09425	1.0041	2.1	0.213845	1.10305	1.0065	−2.4
KI											
0.005288	1.07988	0.9998	−0.3	0.080586	1.08857	0.9930	0.8	0.159232	1.09761	0.9856	0.5
0.026686	1.08246	0.9982	−2.4	0.106546	1.09156	0.9906	0.2	0.186423	1.10075	0.9829	1.6
0.053250	1.08541	0.9955	0.3	0.132621	1.09456	0.9881	0.8	0.213635	1.10394	0.9807	−2.4
CsI											
0.002075	1.07975	0.9999	−0.4	0.054010	1.08991	0.9930	−0.1	0.131690	1.10513	0.9824	−0.4
0.005375	1.08040	0.9996	−1.5	0.082249	1.09521	0.9892	−0.5	0.157792	1.11050	0.9785	2.1
0.026588	1.08453	0.9966	1.0	0.105272	1.09995	0.9860	0.3	0.184922	1.11552	0.9751	−1.5
KBr											
0.002120	1.07963	1.0004	−1.7	0.080904	1.08595	0.9965	2.9	0.160959	1.09237	0.9922	2.5
0.005503	1.07990	1.0003	−1.1	0.106649	1.08808	0.9956	−2.1	0.191439	1.09481	0.9906	0.6
0.026659	1.08160	0.9997	−2.2	0.129178	1.08982	0.9939	2.9	0.214234	1.09668	0.9897	−3.6
0.051672	1.08361	0.9984	−1.1								

[a] Densities calculated by using the Root equation presented by Jones and Christian (*6*). [b] Densities obtained by graphical interpolation of the data of Jones and Talley (*7*), using successive approximations to the molarity of the solutions. [c] Densities obtained by graphical interpolation from a set of five measured values. [d] Obtained by graphical interpolation. [e] Densities obtained by graphical interpolation and extrapolation from a set of six measured values. [f] $\rho_{H_2O} = 0.99704_7$ g cm^{-3}, $\rho_{20\%\ \text{sucrose}} = 1.07937$ g cm^{-3}.

was sealed after introducing a mass of water equal to that contained by a pycnometer when filled to the scratch mark at 25 °C.

The volume V of each pycnometer was obtained by weighing the water contained at 25 °C. Also the difference in mass m_1 between the pycnometer filled with water and the sealed tare was obtained in each case. In practice it was found most convenient not to adjust the meniscus level in the stem to coincide with the scratch mark. Instead, after thermal equilibration, the small separation between meniscus and scratch mark was measured with a cathetometer. Knowing the area of cross section of the capillary used in the stem, one can calculate a correction to the volume.

When measuring the density of a solution, we weighed the filled pycnometer and tare and the difference in mass m_2 was obtained. The Δd, the difference in density between the solution and water, is given by

$$\Delta d = [(m_2 - m_1)/V](1 - d_a/d_m) \qquad (4)$$

where d_a is the density of air and d_m the density of the balance weights. With this procedure it is not necessary to know the density of air to the high accuracy which is required if a tare is not used.

For solutions of sodium or potassium chloride in water, densities were generaly obtained by interpolation of literature data (*6*, *7*). However, a few check determinations were made.

The temperature of the water bath used in the viscosity and density measurements was controlled at 25 ± 0.002 °C.

For calculation of the A coefficients, data were available for solvent dielectric constants (*8*, *9*) and viscosities (*10*, *11*) and for all the limiting ionic equivalent conductances (*12*, *13*) except that for cesium ion in 20% sucrose. We therefore measured the conductances of some solutions of cesium chloride in 20% sucrose using conventional techniques. The temperture of the oil bath used in the conductance measurements was controlled at 25 ± 0.002 °C.

Materials

Water. Demineralized water from a bulk supply was once distilled from glass, yielding water of average specific conductivity $1.3 \times 10^{-6}\ \Omega^{-1}\ \text{cm}^{-1}$. This water was used for all recrystallizations and the preparation of all solutions except those used for conductance measurements. For this purpose a second distillation yielding water of specific conductivity $1.1 \times 10^{-6}\ \Omega^{-1}\ \text{cm}^{-1}$ was made.

Sucrose. BDH microanalytical reagent sucrose was dried at 30 °C under vacuum.

Sodium and Potassium Chlorides. The analytical reagent grade chloride was recrystallized once from water and the dried product was fused in a platinum crucible.

Cesium Chloride. BDH laboratory reagent grade cesium chloride was 3 times recrystallized from water. Flame photometric analysis (*14*) indicated the presence of impurities in the following insignificant amounts (ppm): Li, <1; Na, 18; K, 1; Rb, 40.

Sodium and Potassium Iodides. The corresponding anhydrous carbonate (BDH laboratory reagent, once recrystallized) was neutralized with freshly distilled BDH hydriodic acid. The products were twice recrystallized from water to which a few drops of hydriodic acid were added.

Cesium Iodide. Stoichiometric quantities of solutions of BDH laboratory reagent barium iodide and cesium sulfate were mixed and the resultant barium sulfate was removed by centrifugation and filtration. The cesium iodide was 3 times recrystallized from water.

Potassium Bromide. Univar (Ajax Chemical Co. Sydney) grade potassium bromide was 3 times recrystallized.

Table III. Coefficients k and l of the Root Equation $\rho = \rho_0 + kC - lC^{3/2}$ for Electrolytes in Aqueous Solution and Aqueous 20% Sucrose Solution at 25 °C

	data points	k	SE(k)[a]	l	SE(l)[a]	$10^5\Delta_{av}$
			Aqueous Solutions[b]			
CsCl	5	0.1285	±0.0003	0.0003	±0.0007	0.6
NaI	8	0.1146	±0.0001	0.0007	±0.0003	0.5
KI	9	0.1207	±0.0001	0.0011	±0.0003	0.4
CsI	10	0.2019	±0.0002	0.0005	±0.0005	0.8
KBr	10	0.08526	±0.00007	0.0018	±0.0002	0.3
			Aqueous 20% Sucrose Solutions[c]			
NaCl	6	0.0393	±0.0004	0.0027	±0.0012	1
KCl	5	0.0457	±0.0002	0.0056	±0.0005	1
CsCl	8	0.1262	±0.0004	0.0053	±0.0010	2
NaI	9	0.1119	±0.0007	0.0031	±0.0016	3
KI	7	0.1126	±0.0004	−0.0051	±0.0009	1
CsI	7	0.1926	±0.0004	−0.0087	±0.0011	1
KBr	9	0.0830	±0.0007	0.0052	±0.0018	3

[a] SE = standard error. [b] $\rho_0 = 0.99704_7$ g cm^{-3}. [c] $\rho_0 = 1.07937$ g cm^{-3}.

Prior to use all salts were dried by heating to above 100 °C under vacuum and were stored in vacuum desiccators. All solutions were prepared by weight and vacuum corrections were applied to all masses.

Results and Discussion

Conductance. In Table I we present the values of the measured equivalent conductance, Λ, for cesium chloride at various concentrations, C, in 20% aqueous sucrose solution. The limiting equivalent conductance Λ^0 was estimated by using the relation (*15*)

$$\Lambda^0_{\text{calcd}} = \Lambda + \frac{(B_1\Lambda + B_2)C^{1/2}}{1 + (B\mathring{a} - B_1)C^{1/2}} \qquad (5)$$

in which B, B_1, and B_2 are constants of Debye–Hückel–Onsager theory. The resulting values of Λ^0_{calcd} were extrapolated against C to $C = 0$ to yield a final value for Λ^0. Three extrapolations were made by using å = 2.5, 3.0, and 4.0 Å. All resulted in the value for $\Lambda^0 = 95.2_1$ cm^2 Ω^{-1} g equiv^{-1}. Since $\lambda^0_{\text{Cl}^-}$ in this solvent is known to be equal to 48.2, $\lambda^0_{\text{Cs}^+}$ may be taken as 47.0 cm^2 Ω^{-1} g equiv^{-1}.

Densities. Measured and/or interpolated densities are included in Table II and Root equation coefficients for a least-squares fit of the measured densities are presented in Table III. The precision of the density measurements was a few parts in 10^5.

Viscosities. Values for the relative viscosities of the solutions studied are presented in Table II. These data were fitted to the Jones–Dole equation using the method of least squares. Table II includes values of Δ, the difference between the experimental value of η_r and that calculated from the Jones–Dole equation using the values of A and B obtained for the corresponding system by least-squares fit of the data. The resultant values of the A and B coefficients together with the standard errors of the estimate of each are sumarized in Table IV. In each instance the expected value of the A coefficient, calculated by using eq 2, is presented for comparison. No explanation is offered for the significant discrepancy observed, in several of the aqueous sucrose systems, between the calculated value of the A coefficient and that obtained experimentally. If the theoretical value of A is substituted in the Jones–Dole equation, then $\eta_r - 1 - AC^{1/2}$ should be linear in C with slope B. However, for the systems in question, we find the fit of our data using this function to be significantly worse than for the case where both A and B are treated as adjustable parameters.

Table IV. Coefficients *A* and *B* of the Jones-Dole Equation (Eq 1) for Electrolytes in Aqueous Solution and Aqueous 20% Sucrose Solution at 25 °C

	A	SE(*A*)[a]	*A*(theory)	*B*	SE(*B*)[a]	*B*(lit.)
			Aqueous Solution			
NaCl	0.0060	±0.0005	0.0060	0.081	±0.001	0.079 (*6, 16*)
KCl	0.0050	±0.0003	0.0050	−0.013	±0.001	−0.014 (*7, 18*)
CsCl	0.0052	±0.0006	0.0049	−0.050	±0.002	
NaI	0.0053	±0.0004	0.0060	0.010	±0.001	
KI	0.0052	±0.0004	0.0050	−0.084	±0.001	−0.076 (*19*) −0.088 (*20*)
CsI	0.0038	±0.0008	0.0049	−0.116	±0.002	−0.118 (*21*)
KBr	0.0054	±0.0006	0.0050	−0.046	±0.002	−0.049 (*22*)
			Aqueous 20% Sucrose Solutions			
NaCl	0.0082	±0.0005	0.0052	0.093	±0.001	
KCl	0.0075	±0.0009	0.0043	−0.027	±0.002	
CsCl	0.0056	±0.0005	0.0043	−0.068	±0.001	
NaI	0.0057	±0.0009	0.0053	0.017	±0.002	
KI	0.0035	±0.0007	0.0044	−0.099	±0.002	
CsI	0.0033	±0.0007	0.0044	−0.143	±0.002	
KBr	0.0077	±0.0012	0.0043	−0.066	±0.003	

[a] SE = standard error.

Table V. Differences between *B* Coefficients

Alkali Halides in Aqueous 20% Sucrose Solution at 25 °C

$B_{\mathrm{NaCl}} - B_{\mathrm{NaI}} = 0.076 \pm 0.003$

$B_{\mathrm{KCl}} - B_{\mathrm{KI}} = 0.072 \pm 0.004$

$B_{\mathrm{CsCl}} - B_{\mathrm{CsI}} = 0.075 \pm 0.003$

Alkali Halides in Aqueous Solution at 25 °C

$B_{\mathrm{NaCl}} - B_{\mathrm{NaI}} = 0.071 \pm 0.002$

$B_{\mathrm{KCl}} - B_{\mathrm{KI}} = 0.071 \pm 0.002$

$B_{\mathrm{CsCl}} - B_{\mathrm{CsI}} = 0.066 \pm 0.004$

$B_{\mathrm{NaCl}} - B_{\mathrm{KCl}} = 0.094 \pm 0.002$

Literature

$B_{\mathrm{NaCl}} - B_{\mathrm{KCl}} = 0.093$ (*17*)

Table VI. Ionic *B* Coefficients

ion	water		20% aq sucrose (this work)
	this work	lit.	
Na^+	0.088	0.086 (*17*)	0.107
K^+	(−0.0065)	(−0.007) (*17*)	(−0.014)
Cs^+	−0.044	−0.045 (*23*)	−0.055
Cl^-	(−0.0065)	(−0.007) (*17*)	(−0.014)
Br^-	−0.040	−0.042 (*17*) −0.032 (*23*)	−0.053
I^-	−0.078 (KI)	−0.069 (*17*) −0.081 (*20*)[a]	−0.088[b]
	−0.078 (NaI)		
	−0.073 (CsI)	−0.073 (*21*)[a]	

[a] The references in these cases represent the source of the datum for the appropriate salt. [b] An average of values derived from the *B* coefficients of KI, NaI, and CsI.

That the *B* coefficient for each of the alkali halides in the aqueous sucrose solution is an additive property of the constituent ions is indicated by the results presented in Table V. Differences between the *B* coefficients of the chloride and iodide of sodium, potassium, and cesium are shown to be constant within experimental error. The constancy of the corresponding differences for the aqueous solutions is also confirmed and, in addition, the difference *B*(NaCl) − *B*(KCl) is in agreement with that obtained by Kaminsky (*16*).

The electrical mobilities of the potassium and chloride ions in 20% sucrose solution differ by only 4% (*13*). We therefore adopt the practice used for aqueous solutions (*17*) and assign equal values to the *B* coefficients of these two ions. Individual ionic *B* coefficients calculated on this basis are presented in Table VI. It may be seen that dissolved ions have a greater effect upon the viscosity of the sucrose solution than they do upon the viscosity of water at 25 °C. Positive *B* coefficients are more positive in the sucrose solution and negative *B* coefficients are more negative in this solvent. This trend is similar to that which occurs in aqueous solutions when the temperature is lowered (*17*).

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