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Electrical Conductances of Tetraethylammonium Bromide and Tetrapropylammonium Bromide in 2-Ethoxyethanol (1) + Water (2) Mixtures at (308.15, 313.15, 318.15, and 323.15) K

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The electrical conductances of the solutions of tetraethylammonium bromide (Et_4NBr) and tetrapropylammonium bromide (Pr_4NBr) in 2-ethoxyethanol (1) + water (2) mixed solvent media containing 0.25, 50, and 0.75 mass fractions of 2-ethoxyethanol (w_1) have been reported at (308.15, 313.15, 318.15, and 323.15) K. The conductance data have been analyzed by the 1978 Fuoss conductance−concentration equation in terms of the limiting molar conductance (Λ^0), the association constant (K_A), and the association diameter (R). These two electrolytes are found to exist essentially as free ions in the solvent mixtures with $w_1 = 0.25$ and 50 over the entire temperature range; however, slight ionic association was observed in the mixed solvent medium richest in 2-ethoxyethanol. The electrostatic ion−solvent interaction is found to be very weak for the tetraalkylammonium ions in the aqueous 2-ethoxyethanol mixtures investigated.

Introduction

Studies on the transport properties of electrolytes in different solvent media are of great importance to obtain information as to the solvation and association behavior of ions in solutions. Earlier, we have investigated[1−8] these properties for a wide variety of electrolytes in different solvents in great detail. The solvent properties such as the viscosity and the relative permittivity have also been taken into account in determining the extent of ionic association and the solute−solvent interactions that enabled many to interpret the unique structure of the solvent. The present paper reports the molar conductivities of two tetraalkylammonium salts, namely, tetraethylammonium bromide (Et_4NBr) and tetrapropylammonium bromide (Pr_4NBr), in 2-ethoxyethanol (1) + water (2) mixtures at (308.15, 313.15, 318.15, and 323.15) K to investigate the ion−ion and ion−solvent interactions in these media.

Experimental Section

2-Ethoxyethanol (G. R. E. Merck) was dried with potassium carbonate and distilled twice in an all glass distillation set immediately before use, and the middle fraction was collected. The purified solvent had a density (ρ_0) of 0.92497 $g\cdot cm^{-3}$ and a viscosity (η_0) of 1.8277 mPa·s at 298.15 K; these values are found to be in good agreement with the literature values.[9,10] Triply distilled water with a specific conductance of less than 10^{-6} $S\cdot cm^{-1}$ at 308.15 K was used for the preparation of the mixed solvents by mass. The physical properties of 2-ethoxyethanol (1) + water (2) mixed solvents used in this study at (308.15, 313.15, 318.15, and 323.15) K are reported in Table 1. The relative permittivities of 2-ethoxyethanol (1) + water (2) mixtures at the experimental temperatures were obtained with the equations as described in the literature[11] using the literature density and relative permittivity data of the pure solvents[7,12] and the densities of the mixed solvents given in Table 1.

Table 1. Properties of 2-Ethoxyethanol (1) + Water (2) Mixtures with w_1 = 0.25, 0.50, and 0.75 at (308.15, 313.15, 318.15, and 323.15) K

T/K	ρ_0/(g·cm^3)	η_0/(mPa·s)	ϵ
	$w_1 = 0.25$		
308.15	1.00354	1.8430	60.13
313.15	1.00021	1.5293	58.70
318.15	0.99781	1.2738	57.37
323.15	0.99582	1.0923	56.11
	$w_1 = 0.50$		
308.15	0.99361	1.9234	44.30
313.15	0.98514	1.7195	43.03
318.15	0.98004	1.4552	41.95
323.15	0.97610	1.2762	40.96
	$w_1 = 0.75$		
308.15	0.95451	1.7002	27.93
313.15	0.95147	1.5293	27.29
318.15	0.94873	1.3498	26.68
323.15	0.94625	1.1901	26.10

Both tetraethylammonium bromide (Et_4NBr) and tetrapropylammonium bromide (Pr_4NBr) were of Fluka purum grade and were purified by recrystallization from acetone. The recrystallized salts were dried in vacuo at 333.15 K for 48 h.

Conductance measurements were carried out on a Pye-Unicam PW 9509 conductivity meter at a frequency of 2000 Hz using a dip-type cell of cell constant 1.15 cm^{-1} and having a precision of 0.10 %. The cell was calibrated by the method of Lind et al.[13] using aqueous potassium chloride solutions. The measurements were made in a water bath maintained within ± 0.005 K of the desired temperature. The details of the experimental procedure have been described earlier.[14,15] Solutions were prepared by mass for the conductance runs, the molalities being converted to molarities by the use of densities measured with an Ostwald-Sprengel type pycnometer of about 25 cm^3 capacity. Several independent solutions were prepared, and runs were performed to ensure the reproducibility of the results. Due correction was made for the specific conductance of the solvent by subtracting the specific conductance of the relevant solvent medium from those of the salt

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Table 2. Equivalent Conductances and Corresponding Molarities of Electrolytes in 2-Ethoxyethanol (1) + Water (2) Mixtures with w_1 = 0.25, 50, and 0.75 at (308.15, 313.15, 318.15, and 323.15) K

Et_4NBr						Pr_4NBr					
$w_1 = 0.25$		$w_1 = 0.50$		$w_1 = 0.75$		$w_1 = 0.25$		$w_1 = 0.50$		$w_1 = 0.75$	
c	Λ	c	Λ	c	Λ	c	Λ	c	Λ	c	Λ
$mol\cdot dm^{-3}$	$S\cdot cm^2\cdot mol^{-1}$	$mol\cdot dm^{-3}$	$S\cdot cm^2\cdot mol^{-1}$	$mol\cdot dm^{-3}$	$S\cdot cm^2\cdot mol^{-1}$	$mol\cdot dm^{-3}$	$S\cdot cm^2\cdot mol^{-1}$	$mol\cdot dm^{-3}$	$/(S\cdot cm^2\cdot mol^{-1})$	$mol\cdot dm^{-3}$	$S\cdot cm^2\cdot mol^{-1}$
T = 308.15 K											
0.005122	66.77	0.002120	58.49	0.000501	49.08	0.005026	63.87	0.002030	55.66	0.000508	44.71
0.007683	64.95	0.003181	57.55	0.000752	48.54	0.007359	62.21	0.003045	55.17	0.000763	44.07
0.010244	63.64	0.004240	56.84	0.001002	48.18	0.010052	60.78	0.004060	54.43	0.001017	43.44
0.012055	62.39	0.005300	56.23	0.001253	47.80	0.012565	59.61	0.005075	53.79	0.001271	42.88
0.015367	61.24	0.006358	55.66	0.001504	47.55	0.015078	58.50	0.006090	53.37	0.001525	42.38
0.017928	60.19	0.007420	55.12	0.001754	47.37	0.017591	57.47	0.007105	52.78	0.001780	41.93
0.020489	59.30	0.008482	54.60	0.002005	47.08	0.020104	56.51	0.008120	52.34	0.002034	41.55
0.025611	57.59	0.010600	53.77	0.002506	46.68	0.025130	54.75	0.010150	51.63	0.002542	40.94
T = 313.15 K											
0.005099	71.97	0.002110	63.03	0.000499	53.32	0.005003	68.95	0.002020	60.88	0.000506	49.20
0.007649	70.34	0.003165	62.55	0.000748	52.92	0.007505	67.15	0.003031	60.05	0.000759	48.34
0.010198	68.93	0.004220	61.61	0.000998	52.52	0.010007	65.75	0.004041	59.39	0.001012	47.72
0.012748	67.70	0.005275	61.04	0.001247	52.12	0.012509	64.75	0.005052	58.99	0.001265	47.11
0.015298	66.61	0.006330	60.34	0.001496	51.85	0.015012	63.55	0.006062	58.40	0.001518	46.50
0.017848	65.50	0.007386	59.98	0.001746	51.49	0.017514	62.52	0.007072	57.97	0.001771	46.01
0.020398	64.52	0.008441	59.47	0.001995	51.22	0.020016	61.50	0.008083	57.53	0.002024	45.50
0.025498	62.87	0.010551	58.48	0.002494	50.71	0.025021	59.71	0.010104	56.81	0.002530	44.92
T = 318.15 K											
0.005079	77.18	0.002099	68.60	0.000496	58.68	0.004984	73.84	0.002010	65.67	0.000503	54.84
0.007619	75.47	0.003149	67.64	0.000744	57.94	0.007476	72.37	0.003015	65.00	0.000755	53.91
0.010158	74.03	0.004198	66.69	0.000992	57.37	0.009968	70.83	0.004020	64.42	0.001007	53.15
0.012698	72.77	0.005248	66.12	0.001240	56.86	0.012461	69.58	0.005025	63.88	0.001258	52.54
0.015238	71.60	0.006298	65.58	0.001488	56.46	0.014953	68.48	0.006031	63.34	0.001510	51.99
0.017778	70.65	0.007347	65.06	0.001736	56.11	0.017446	67.41	0.007036	62.96	0.001762	51.38
0.020318	69.64	0.008397	64.55	0.001984	55.70	0.019939	66.40	0.008041	62.43	0.002013	50.81
0.025398	67.80	0.010496	63.55	0.002480	55.01	0.024925	64.63	0.010051	61.58	0.002516	49.91
T = 323.15 K											
0.005060	83.00	0.002087	73.79	0.000494	64.23	0.004965	79.15	0.001998	71.06	0.000501	61.32
0.007590	80.76	0.003130	72.83	0.000740	63.35	0.007448	77.47	0.002998	70.39	0.000751	60.39
0.010120	79.24	0.004174	72.11	0.000987	62.74	0.009931	75.82	0.003997	69.80	0.001001	59.62
0.012651	78.02	0.005217	71.49	0.001234	62.01	0.012414	74.67	0.004996	69.25	0.001252	58.76
0.015181	76.80	0.006261	70.91	0.001481	61.53	0.014898	73.57	0.005996	68.72	0.001502	58.19
0.017712	75.77	0.007305	70.37	0.001727	61.02	0.017382	72.49	0.006995	68.19	0.001752	57.61
0.020243	74.69	0.008483	69.83	0.001974	60.54	0.019866	71.43	0.007994	67.80	0.002003	56.98
0.025304	72.87	0.010436	68.99	0.002468	59.65	0.024834	69.70	0.009994	66.94	0.002503	55.77

Table 3. Derived Conductivity Parameters of Electrolytes in 2-Ethoxyethanol (1) + Water (2) Mixtures with w_1 = 0.25, 0.50, and 0.75 at (308.15, 313.15, 318.15, and 323.15) K

Et_4NBr						Pr_4NBr					
T	Λ^0	K_A	$\Lambda^0\eta_0$	R	100σ	T	Λ^0	K_A	$\Lambda^0\eta_0$	R	100σ
K	$S\cdot cm^2\cdot mol^{-1}$	$dm^3\cdot mol^{-1}$	$S\cdot cm^2\cdot mol^{-1}\cdot Pa\cdot s$	Λ^0	Λ^0	K	$S\cdot cm^2\cdot mol^{-1}$	$dm^3\cdot mol^{-1}$	$S\cdot cm^2\cdot mol^{-1}\cdot Pa\cdot s$	Λ^0	Λ^0
$w_1 = 0.25$											
308.15	72.90 ± 0.06	11.77 ± 0.12	0.1343	9.29	0.07	308.15	69.93 ± 0.05	12.76 ± 0.11	0.1288	10.81	0.06
313.15	78.45 ± 0.06	10.16 ± 0.09	0.1200	9.31	0.06	313.15	75.22 ± 0.13	11.12 ± 0.23	0.1150	10.83	0.14
318.15	84.05 ± 0.0	9.10 ± 0.09	0.1071	9.31	0.06	318.15	80.64 ± 0.06	9.94 ± 0.14	0.1027	10.83	0.09
323.15	90.28 ± 0.17	8.85 ± 0.24	0.0986	9.30	0.15	323.15	86.22 ± 0.08	8.94 ± 0.12	0.0942	10.82	0.08
$w_1 = 0.50$											
308.15	62.13 ± 0.03	11.39 ± 0.13	0.1195	9.63	0.04	308.15	59.17 ± 0.11	10.41 ± 0.49	0.1138	10.15	0.16
313.15	67.06 ± 0.14	9.57 ± 0.57	0.1153	9.65	0.19	313.15	64.35 ± 0.05	8.21 ± 0.22	0.1106	10.17	0.08
318.15	72.82 ± 0.08	9.01 ± 0.30	0.1060	9.65	0.10	318.15	69.60 ± 0.10	6.83 ± 0.39	0.1013	10.17	0.14
323.15	78.26 ± 0.03	6.97 ± 0.11	0.0942	9.66	0.04	323.15	75.30 ± 0.10	5.74 ± 0.35	0.0961	10.18	0.12
$w_1 = 0.75$											
308.15	51.38 ± 0.04	11.52 ± 0.77	0.0874	10.23	0.07	308.15	49.63 ± 0.11	49.58 ± 1.32	0.0894	10.75	0.10
313.15	56.02 ± 0.07	12.38 ± 1.29	0.0857	10.23	0.12	313.15	52.44 ± 0.09	52.69 ± 1.94	0.0802	10.75	0.14
318.15	61.82 ± 0.04	23.72 ± 0.66	0.0834	10.23	0.06	318.15	58.59 ± 0.05	53.17 ± 1.11	0.0791	10.75	0.08
323.15	68.00 ± 0.05	32.84 ± 0.88	0.0809	10.23	0.07	323.15	65.56 ± 0.12	53.31 ± 2.15	0.0780	10.76	0.15

solutions. The kinematic viscosities were measured using a suspended level Ubbelohde-type viscometer.

To avoid moisture pickup, all solutions were prepared in a dehumidified room with utmost care. In all cases, the experiments were performed at least in five replicates for each solution and at each temperature, and the results were averaged (repeatabilities were always within ± 0.10 $S\cdot cm^2\cdot mol^{-1}$). The experimental uncertainties in density, viscosity, and conductivity were always within 0.01 %, 0.08 %, and 0.12 %, respectively.

Results and Discussion

The measured molar conductances (Λ) of electrolyte solutions as functions of molar concentration (c) in 2-ethoxyethanol (1) + water (2) mixtures with w_1 = 0.25, 0.50, and 0.75 at (308.15, 313.15, 318.15, and 323.15) K are given in Table 2.

The conductance data have been analyzed by the 1978 Fuoss conductance−concentration equation.[16,17] For a given set of conductivity values (c_j, Λ_j; j = 1, ..., n), three adjustable parameters—the limiting molar conductivity (Λ^0), the association constant (K_A), and the association diameter (R)—are derived from the following set of equations:

$$\Lambda = p[\Lambda^0(1 + \mathrm{RX} + \mathrm{EL}] \tag{1}$$

$$p = 1 - \alpha(1 - \gamma) \tag{2}$$

$$\gamma = 1 - K_A c \gamma^2 f^2 \tag{3}$$

$$-\ln f = \frac{\beta k}{2(1 + kR)} \tag{4}$$

$$\beta = \frac{e^2}{\epsilon k_B T} \tag{5}$$

$$K_A = K_R(1 + K_S) \tag{6}$$

where RX is the relaxation field effect, EL is the electrophoretic countercurrent, γ is the fraction of unpaired ions, α is the fraction of contact pairs, K_A is the overall pairing constant evaluated from the association constants of contact pairs (K_S) of solvent-separated pairs (K_R), ϵ is the relative permittivity of the solvent, e is the electronic charge, k_B is the Boltzmann constant, k^{-1} is the radius of the ion atmosphere, c is the molarity of the solution, f is the activity coefficient, T is the temperature in absolute scale, and β is twice the Bjerrum distance. The computations were performed on a computer using the program as suggested by Fuoss. The initial Λ^0 values for the iteration procedure were obtained from Shedlovsky extrapolation[18] of the data. Input for the program is the set (c_j, Λ_j; j = 1, ..., n), n, ϵ, η, T, initial value of Λ^0, and an instruction to cover a preselected range of R values.

In practice, calculations are made by finding the values of Λ^0 and α that minimize the standard deviation (σ):

$$\sigma = [\sum[\Lambda_j\,(\mathrm{calcd}) - \Lambda_j\,(\mathrm{obsd})]^2/(n - 2)]^{1/2} \tag{7}$$

for a sequence of R values and then plotting σ against R; the best-fit R corresponds to the minimum in σ versus R curve. However, for these two electrolytes investigated, since a preliminary scan using a unit increment of R values from 4 to 20 produced no significant minima in the σ versus R curves, the R value was assumed to be $R = a + d$, where a is the sum of the ionic crystallographic radii and d is given by[17]

$$d = 1.183(M/\rho_0)^{1/3} \tag{8}$$

where M is the molecular weight of the solvent and ρ_0 is its density.

The values of Λ^0, K_A, and R obtained by this procedure are reported in Table 3. Table 3 shows that the equivalent conductivity values (Λ^0) of the two tetraalkylammonium salts increase as the temperature increases in all 2-ethoxyethanol (1) + water (2) mixtures. The Λ^0values have been fitted to the following polynomial in T:

$$\Lambda^0/\mathrm{S\cdot cm^2\cdot mol^{-1}} = a_0 + a_1(308.15 - T/\mathrm{K}) + a_2(308.15 - T/\mathrm{K})^2 \tag{9}$$

and the coefficients of these fits along with the standard deviations (σ) are given in Table 4.

The limiting molar conductivity values (Λ^0) of tetraethylammonium bromide are always found to be higher than those of tetrapropylammonium bromide (see Figure 1). This means that the mobility of the tetraethylammonium ion (Et_4N^+) is greater than that of the tetrapropylammonium ion (Pr_4N^+) (anion being common) in all of the mixed solvent media over the entire temperature range investigated. Now, a comparison of this trend in mobility with the crystallographic sizes of these ions, which is in the order[19] $Et_4N^+ < Pr_4N^+$, shows that the larger the size of the bare ion, the smaller is its ionic mobility. This indicates that the relative actual sizes of these ions as they exist in solutions follow the order: $Et_4N^+ < Pr_4N^+$. This observation clearly demonstrates that the electrostatic ion−solvent

Table 4. Coefficients of Eq 9 and the Standard Deviations (σ)

w_1	electrolyte	a_0	$-a_1$	a_2	σ
0.25	Et_4NBr	72.93 ± 0.13	1.0528 ± 0.0406	0.0068 ± 0.0026	0.13
	Pr_4NBr	69.93 ± 0.01	1.0423 ± 0.0021	0.0029 ± 0.0001	0.01
0.50	Et_4NBr	62.07 ± 0.25	1.0065 ± 0.0805	0.0051 ± 0.0041	0.26
	Pr_4NBr	59.19 ± 0.08	0.9948 ± 0.0266	0.0052 ± 0.0017	0.08
0.75	Et_4NBr	51.34 ± 0.17	0.8822 ± 0.0546	0.0154 ± 0.0035	0.17
	Pr_4NBr	49.50 ± 0.55	0.4548 ± 0.1764	0.0416 ± 0.0113	0.56

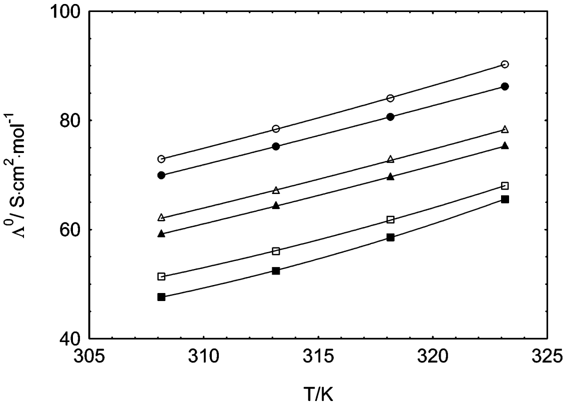

Figure 1. Temperature dependence of the limiting molar conductances of tetraethylammonium bromide (○, $w_1 = 0.25$; △, $w_1 = 0.50$; □, $w_1 = 0.75$) and tetrapropylammonium bromide (●, $w_1 = 0.25$; ▲, $w_1 = 0.50$; ■, $w_1 = 0.75$) in 2-ethoxyethanol (1) + water (2) mixtures.

interaction is very weak for these ions in aqueous 2-ethoxyethanol solutions, which is to be expected because of their large crystallographic radii[19] and hence low surface charge density. Had these interactions been very strong in this medium, the limiting molar conductivity values should have been in the reverse order: $Et_4NBr < Pr_4NBr$, because a smaller ion with greater surface charge density is expected to associate more solvent molecules thus resulting in a bigger solvodynamic entity, which is obviously not the case here. A similar trend in the Walden products, $\Lambda^0\eta_0$ (Table 3), also points to the same conclusion.

The Walden product values ($\Lambda^0\eta_0$) for the electrolytes studied here show pronounced variations with increasing temperature (Table 3). Therefore, the Stokes law cannot be applied in 2-ethoxyethanol (1) + water (2) because the $\Lambda^0\eta_0$ values, according to this law, would be expected to be independent of temperature.[20] Since the ions are often far from being spherical and since they are of the same order of magnitude as the solvent molecules, it is questionable whether the retarding effect of the later can be accurately described by the macroscopic viscosity as has been done in the derivation of the Stokes law. Hence, the Stokes law cannot be considered quantitatively reliable. Such failure of this law has also been observed earlier in other solvent media.[21,22]

The association constants (K_A) listed in Table 3 for these two electrolytes are practically negligible (i.e., the K_A values are either very close to or less than 10) in the mixed solvent media with $w_1 = 0.25$ and 0.50 over the entire temperature range. So, the numerical values of K_A should not be taken seriously.[23] One can only conclude that these two electrolytes exist essentially as free ions in both the solvent mixtures in the temperature range (308.15 to 323.15) K. This is expected because the relative permittivities of the solvent mixtures are fairly high ($40.96 \leq \epsilon \leq 60.13$). In the solvent mixture with $w_1 = 0.75$ with comparatively lower relative permittivity, these salts are found to be slightly associated.

Thus, it can be concluded that two tetraalkylammonium bromides, namely, tetraethylammonium bromide and tetrapropylammonium bromide, exist essentially in the form of free ions aqueous 2-ethoxyethanol (1) solutions with $w_1 = 0.25$ and 0.50 over the entire temperature range investigated. Slight ionic association was observed in the mixed solvent medium with $w_1 = 0.75$. The electrostatic ion–solvent interaction is found to be very weak for the tetraalkylammonium ions in the aqueous 2-ethoxyethanol mixtures investigated.

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