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Electrical Conductances of Tetrabutylammonium Bromide, Sodium Tetraphenylborate, and Sodium Bromide in Methanol (1) + Water (2) Mixtures at (298.15, 308.15, and 318.15) K

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The electrical conductances of the solutions of tetrabutylammonium bromide (Bu_4NBr), sodium tetraphenylborate ($NaBPh_4$), and sodium bromide (NaBr) in methanol (1) + water (2) mixed solvent media containing 0.10, 0.20, and 0.30 volume fractions of methanol have been reported at (298.15, 308.15, and 318.15) 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). The ionic contributions to the limiting molar conductance have been estimated using tetrabutylammonium tetraphenylborate (Bu_4NBPh_4) as the "reference electrolyte". All of these three electrolytes are found to exist as free ions in the present solvent mixtures within the temperature range (298.15 to 318.15) K. Besides the relative permittivity and the viscosity of the media, specific interactions of the ions with the solvent media have been found to play a profound influence on their mobilities. Although the association constants of the electrolytes do not vary significantly as a function of temperature, the limiting molar conductances of the electrolytes as well as the single-ion conductivity values increase appreciably with temperature.

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[1−7] 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 tetrabutylammonium bromide (Bu_4NBr), sodium tetraphenylborate ($NaBPh_4$), and sodium bromide (NaBr) in methanol (1) + water (2) mixtures at (298.15, 308.15, and 318.15) K in order to obtain precise temperature-dependent single-ion conductivities since such data are practically nonexistent in mixed solvent media.[8,9]

Experimental Section

Methanol (Sisco, India, 99 % pure) was dried over molecular sieves and distilled fractionally. The middle fraction was collected and redistilled. The physical properties of the purified solvent were found to be in good agreement with the literature values.[10] Table 1 shows the comparison of the experimental values of density and viscosity of methanol with literature data.[10] Triply distilled water with a specific conductance of less than 10^{-6} S·cm^{-1} at 298.15 K was used for the preparation of the mixed solvents. The densities and viscosities of methanol (1) + water (2) mixed solvents used in this study at (298.15, 308.15, and 318.15) K are reported in Table 1 along with those (at 298.15 K) obtained from the literature.[11] The relative permittivities of methanol (1) + water (2) mixtures were obtained by regressing the relative permittivity data as a function of solvent composition from the literature[12] and are included in Table 1.

Table 1. Properties of Methanol (1) + Water (2) Mixtures Containing 0.10, 0.20, and 0.30 Volume Fractions of Methanol at (298.15, 308.15, and 318.15) K

	ρ_0/g·cm^3		η_0/mPa·s		
T/K	this work	lit.[a]	this work	lit.[a]	ϵ^c
		$\phi_1 = 0.01$			
298.15	0.98297	0.98292[a]	1.0844	1.080[a]	75.09
308.15	0.97973		0.8665		71.57
318.15	0.97604		0.7020		68.18
		$\phi_1 = 0.20$			
298.15	0.96963	0.96996[a]	1.3106	1.310[a]	71.61
308.15	0.96632		1.0217		68.14
318.15	0.96162		0.8075		64.80
		$\phi_1 = 0.30$			
298.15	0.95620	0.95689[a]	1.4712	1.472[a]	67.65
308.15	0.95160		1.1418		64.25
318.15	0.94626		0.8957		60.99
		$\phi_1 = 1.00$			
298.15	0.78659	0.78656[b]	0.5446	0.5422[b]	32.66
308.15	0.77728	0.77720[b]	0.4747	0.4742[b]	30.74
318.15	0.76774	0.76770[b]	0.4185	0.4174[b]	28.92

[a] From ref 11. [b] From ref 10. [c] From ref 12.

All of these salts were of Fluka purum or puriss grade. Tetrabutylammonium bromide (Bu_4NBr) was purified by recrystallization from acetone, and the recrystallized salt was dried in vacuo at 333.15 K for 48 h. Sodium tetraphenylborate ($NaBPh_4$) was recrystallized three times from acetone and then dried under vacuum at 353.15 K for 72 h. Sodium bromide (NaBr) was dried in vacuo for 72 h immediately prior to use and was used without further purification.

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

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Table 2. Equivalent Conductances and Corresponding Molarities of Electrolytes in Methanol (1) + Water (2) Mixtures Containing 0.10, 0.20, and 0.30 Volume Fractions of Methanol at (298.15, 308.15, and 318.15) K

T = 298.15 K		T = 308.15 K		T = 318.15 K		T = 298.15 K		T = 308.15 K		T = 318.15 K	
c	Λ	c	Λ	c	Λ	c	Λ	c	Λ	c	Λ
$\mathrm{mol{\cdot}dm^3}$	$\mathrm{S{\cdot}cm^2{\cdot}mol^{-1}}$	$\mathrm{mol{\cdot}dm^3}$	$\mathrm{S{\cdot}cm^2{\cdot}mol^{-1}}$	$\mathrm{mol{\cdot}dm^3}$	$\mathrm{S{\cdot}cm^2{\cdot}mol^{-1}}$	$\mathrm{mol{\cdot}dm^3}$	$\mathrm{S{\cdot}cm^2{\cdot}mol^{-1}}$	$\mathrm{mol{\cdot}dm^3}$	$\mathrm{S{\cdot}cm^2{\cdot}mol^{-1}}$	$\mathrm{mol{\cdot}dm^3}$	$\mathrm{S{\cdot}cm^2{\cdot}mol^{-1}}$
$\phi_1 = 0.10$											
Bu_4NBr											
0.00896	74.88	0.00893	87.61	0.00508	92.09	0.01793	72.69	0.01767	84.47	0.01271	102.78
0.01024	74.70	0.01021	87.03	0.03819	94.80	0.02560	71.13	0.02552	82.50	0.01144	103.37
0.01152	74.32	0.01149	86.54	0.02542	98.18	0.03842	69.07	0.03830	79.88	0.01017	103.99
0.01280	73.96	0.01276	86.08	0.01780	100.72	0.05121	67.41	0.05105	77.77	0.00890	105.44
$NaPh_4B$											
0.00798	60.64	0.00796	68.03	0.00792	82.29	0.00976	59.98	0.00972	67.28	0.00969	81.29
0.00843	60.46	0.00840	67.85	0.00836	82.03	0.01020	59.83	0.01017	67.10	0.01013	81.06
0.00887	60.30	0.00884	67.65	0.00881	81.77	0.01064	59.68	0.01061	66.93	0.01057	80.82
0.00931	60.14	0.00928	67.47	0.00925	81.53	0.01116	59.50	0.01105	66.76	0.01101	80.60
NaBr											
0.00909	107.42	0.00906	125.94	0.00903	152.38	0.04049	100.75	0.04027	118.02	0.04012	142.61
0.01010	106.90	0.01007	125.65	0.01003	152.05	0.05051	99.43	0.05034	116.37	0.05015	140.56
0.02019	104.25	0.02014	122.36	0.02006	147.98	0.06061	98.26	0.06041	114.91	0.06017	138.77
0.03031	102.32	0.03020	119.96	0.03009	145.01	0.07577	96.73	0.07551	113.01	0.07521	136.41
$\phi_1 = 0.20$											
Bu_4NBr											
0.00905	60.42	0.00899	73.62	0.00897	92.29	0.02264	57.73	0.02251	70.51	0.02244	88.26
0.01127	59.93	0.01120	73.05	0.01118	91.46	0.03389	56.15	0.03372	68.71	0.03360	85.99
0.01285	59.56	0.01350	72.44	0.01347	90.69	0.04514	54.87	0.04493	67.26	0.04474	84.18
0.01580	58.94	0.01570	71.91	0.01566	90.02	0.05640	53.78	0.05616	66.05	0.05590	82.65
$NaPh_4B$											
0.00720	48.27	0.00717	61.28	0.00714	77.10	0.00879	47.78	0.00876	60.78	0.00872	76.27
0.00760	48.14	0.00756	61.17	0.07533	76.89	0.00919	47.66	0.00916	60.63	0.00911	76.08
0.00800	48.02	0.00796	61.06	0.00793	76.67	0.00959	47.54	0.00956	60.46	0.00951	75.89
0.00840	47.90	0.00836	60.93	0.00832	76.47	0.00999	47.43	0.00999	60.28	0.00990	75.71
NaBr											
0.00930	87.68	0.00924	110.00	0.00923	141.36	0.04133	82.41	0.04108	102.61	0.04102	128.59
0.01033	87.51	0.01027	109.71	0.01025	140.37	0.05172	81.30	0.05135	101.05	0.0513	125.92
0.02067	85.30	0.02054	106.67	0.0205	135.42	0.06197	80.35	0.06163	99.55	0.0615	123.37
0.03101	83.70	0.03081	104.43	0.03075	131.68	0.07231	79.49	0.07189	98.58	0.0717	121.47
$\phi_1 = 0.30$											
Bu_4NBr											
0.01104	52.63	0.01099	61.72	0.01092	80.69	0.02650	48.33	0.02638	56.35	0.02622	73.59
0.01547	51.16	0.01540	59.89	0.01531	78.29	0.03313	46.96	0.03297	54.65	0.03278	71.33
0.01987	49.92	0.01978	58.34	0.01966	76.24	0.05521	43.41	0.05495	50.27	0.05462	65.53
0.02208	49.36	0.02198	57.63	0.02185	75.30						
$NaPh_4B$											
0.00726	39.86	0.00723	56.78	0.00719	70.31	0.00841	39.53	0.00912	56.01	0.00833	69.72
0.00745	39.81	0.00742	56.70	0.00738	70.20	0.00916	39.31	0.00913	56.01	0.00908	69.35
0.00764	39.75	0.00799	56.48	0.00757	70.10	0.00917	39.31	0.00952	55.86	0.00946	69.17
0.00802	39.64	0.008373	56.30	0.00794	69.90	0.00956	39.20				
NaBr											
0.01008	77.88	0.01003	98.80	0.00998	129.09	0.04036	73.75	0.04013	93.11	0.04001	116.72
0.01513	76.98	0.01505	97.59	0.01498	126.29	0.05045	72.86	0.05016	91.84	0.05003	113.92
0.02017	76.15	0.02006	96.44	0.01997	123.92	0.06057	72.08	0.06020	90.74	0.06009	111.44
0.03026	74.82	0.03010	94.59	0.02999	119.98	0.06940	71.47	0.06897	89.87	0.06889	109.48

an uncertainty of 0.01 %. 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 $\mathrm{cm^3}$ 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 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. 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 methanol (1) + water (2) mixtures with volume fractions of 0.10, 0.20, and 0.30 of methanol at (298.15, 308.15, and 318.15) 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, and 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, which 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 the salts investigated here, since a preliminary scan using incremental 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

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

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.

To investigate the specific behavior of the individual ions comprising these electrolytes, it is necessary to split the limiting molar electrolyte conductances into their ionic components. The limiting ionic conductivities have been evaluated from the division of the Λ^0 values of Bu_4NBPh_4 using the following relationship:

$$\lambda^0(Bu_4N^+) = 0.517\Lambda^0(Bu_4NPh_4B) \tag{9}$$

as described in the literature.[19,20]

The limiting molar conductances of NaBr in some selected methanol (1) + water (2) mixtures at 298.15 K have also been reported by Zhang et al.[11] They analyzed their data with the Fuoss−Onsager equation. Consequently, a direct comparison is not possible.

Table 3. Derived Conductivity Parameters of Electrolytes in Methanol (1) + Water (2) Mixtures Containing 0.10, 0.20, and 0.30 Volume Fractions of Methanol at (298.15, 308.15, and 318.15) K

T/K	Λ^0/S·cm^2·mol^{-1}	K_A/dm^3·mol^{-1}	R/Å	σ/%^{a}
		$\phi_1 = 0.10$		
		Bu_4NBr		
298.15	81.16 ± 0.07	2.72 ± 0.06	9.99	0.10
308.15	95.30 ± 0.07	3.20 ± 0.05	10.09	0.08
318.15	114.75 ± 0.28	3.81 ± 0.18	10.18	0.27
		$NaPh_4B$		
298.15	66.87 ± 0.02	4.88 ± 0.04	9.50	0.01
308.15	75.45 ± 0.02	4.83 ± 0.05	9.59	0.01
318.15	91.86 ± 0.02	5.79 ± 0.03	9.68	0.01
		NaBr		
298.15	114.46 ± .04	1.41 ± 0.01	5.95	0.05
308.15	134.88 ± 0.05	1.50 ± 0.02	5.95	0.05
318.15	163.50 ± 0.07	1.61 ±0.02	5.96	0.05
		$\phi_1 = 0.20$		
		Bu_4NBr		
298.15	65.56 ± 0.05	2.74 ± 0.05	10.09	0.08
308.15	79.87 ± 0.06	2.34 ± 0.05	10.09	0.09
318.15	100.19 ± 0.09	2.40 ± 0.06	10.10	0.10
		$NaPh_4B$		
298.15	53.14 ± 0.02	4.87 ± 0.06	9.59	0.01
308.15	67.33 ± 0.19	4.16 ± 0.06	9.59	0.07
318.15	85.18 ± 0.02	5.30 ± 0.03	9.68	0.01
		NaBr		
298.15	93.85 ± 0.04	1.41 ± 0.02	6.04	0.05
308.15	118.20 ± 0.08	1.82 ± 0.03	6.04	0.08
318.15	153.22 ± 0.12	3.38 ± 0.04	6.05	0.08
		$\phi_1 = 0.30$		
		Bu_4NBr		
298.15	59.94 ± 0.04	8.05 ± 0.06	10.18	0.05
308.15	71.05 ± 0.04	8.95 ± 0.05	10.18	0.04
318.15	93.08 ± 0.04	9.41 ± 0.04	10.19	0.03
		$NaPh_4B$		
298.15	44.46 ± 0.02	6.25 ± 0.09	9.68	0.01
308.15	63.08 ± 0.05	6.44 ± 0.13	9.68	0.01
318.15	78.17 ± 0.03	6.28 ± 0.06	9.69	0.01
		NaBr		
298.15	83.67 ± 0.05	1.22 ± 0.03	6.13	0.07
308.15	106.55 ± 0.08	1.55 ± 0.04	6.13	0.08
318.15	142.04 ± 0.10	4.60 ± 0.04	6.14	0.07

[a] σ/% = 100σ/Λ^0.

The limiting molar conductivity (Λ^0) of the "reference electrolyte" Bu_4NBPh_4 was obtained by considering the Kohlrausch rule that allows the calculation of the Λ^0 value for a given electrolyte by the appropriate combination of others. The Λ^0 values of Bu_4NBr, $NaBPh_4$, and NaBr obtained in the present solvent media have been used to obtain the Λ^0 value of Bu_4NBPh_4 through the following equation:

$$\Lambda^0(Bu_4NBPh_4) = \Lambda^0(Bu_4NBr) + \Lambda^0(NaPh_4B) - \Lambda^0(NaBr) \tag{10}$$

The limiting ionic conductances calculated from the above equations are recorded in Table 4.

The association constants (K_A) listed in Table 3 for all these systems are practically negligible (i.e., $K_A < 10$). So, the numerical values of K_A should not be taken seriously.[21] One can only conclude that all of these three electrolytes exist as free ions in the present methanol−water solvent mixtures within the temperature range of (298.15 to 318.15) K. This is expected because the relative permittivities of the solvent mixtures are

Table 4. Limiting Ionic Conductances in Methanol (1) + Water (2) Mixtures Containing 0.10, 0.20, and 0.30 Volume Fractions of Methanol at (298.15, 308.15, and 318.15) K

	$\lambda_{\pm}^{0}$/S·cm^2·mol^{-1}			
T/K	Na^+	Bu_4N^+	Br^-	Ph_4B^-
		$\phi_1 = 0.10$		
298.15	50.66	17.36	63.80	16.21
308.15	58.12	18.54	76.76	17.33
318.15	71.04	22.29	92.46	20.82
		$\phi_1 = 0.20$		
298.15	41.14	12.85	52.71	12.00
308.15	53.32	14.99	64.88	14.01
318.15	69.65	16.62	83.57	15.53
		$\phi_1 = 0.30$		
298.15	34.45	10.72	49.22	10.01
308.15	49.76	14.26	56.79	13.32
318.15	64.06	15.10	77.98	14.11

fairly high ($66.75 \leq \epsilon \leq 76.78$), thus promoting dissociation of electrolytes.

In all the mixed solvent media, the limiting ionic equivalent conductances decrease in the order: $\lambda_{Br^-}^{0} > \lambda_{Na^+}^{0} > \lambda_{Bu_4N^+}^{0} > \lambda_{Ph_4B^-}^{0}$ at each temperature, indicating that the sizes of these ions as they exist in solutions follow the order: $Br^- < Na^+ < Bu_4N^+ < Ph_4B^-$. It is also observed that the limiting ionic equivalent conductances of all the ions decrease in going from 0.10 volume fraction of methanol to 0.30 volume fraction of methanol in the mixture at all temperatures investigated.

The limiting equivalent conductances of the electrolytes as well as the single-ion conductivity values increase appreciably with temperature. The effect of temperature on $\lambda_{Na^+}^{0}$ and $\lambda_{Br^-}^{0}$ ions is more pronounced as compared to that as $\lambda_{Bu_4N^+}^{0}$ and $\lambda_{Ph_4B^-}^{0}$ in the present mixed solvent media.

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