Viscosity Coefficients of Alkali-Metal Bromides, Tetrabutyl Ammonium Bromide, and Sodium Tetraphenylborate in Aqueous Binary Mixtures of 2-Methoxy-1-ethanol at 298.15 K

Debasis Nandi and Dilip K. Hazra*

Department of Chemistry, North Bengal University, Darjeeling-734430, India

Viscosities of solutions of the alkali metal bromides, MBr (M^+ = Li, Na, K, Rb, and Cs), Bu_4NBr, and $NaBPh_4$ have been measured in 2-methoxy-1-ethanol (1) + water (2) mixtures at 298.15 K. The data have been analyzed using the Jones−Dole equation in the form of unassociated electrolytes when ion association does not occur in solution as found from conductance measurements, but the following equation $\eta_r = 1 + A(\alpha C)^{1/2} + B_i\alpha C + B_p(1 - \alpha)C$ has been used for the electrolytes where ion association occurs. The ionic *B* coefficients and other single-ion parameters have been determined using Bu_4NBPh_4 as the "reference electrolyte". The results have been discussed in terms of ion−solvent interactions and the structural changes of the mixed solvent as the mole fraction of organic solvent is increased in the mixture.

Introduction

Study of the transport properties of electrolytic solutions gives very useful information about ion−ion and ion−solvent interactions in solution. One of the methods employed for these investigations is to study the solution viscosity. Recently, we have reported the viscometric behaviour of tetraalkyl ammonium bromides (Hazra et al., 1989) and alkali-metal chlorides and bromides (Nandi and Hazra, 1989) in 2-methoxy-1-ethanol (ME), a solvent having potential uses for various industrial processes (Garst, 1969). Still, its practical applications have not stimulated much interest for the study of its mixtures with water (Das and Hazra, 1993). The properties of the pure liquid is reasonably well-known, but there are relatively few studies of its aqueous solutions, especially in the water-rich region.

We have studied the viscometric behavior of alkali-metal bromides, MBr (where M^+ = Li, Na, K, Rb, and Cs), and other important electrolytes, viz., Bu_4NBr and $NaBPh_4$ in ME (1) + water (2) mixtures at X_1 = 0.092, 0.262, and 0.680 at 298.15 K, and the results are reported in this communication.

Experimental Section

2-Methoxy-1-ethanol (Merck, >99% pure) was distilled twice in an all-glass distillation set, and the middle fraction distilling between 397 K and 398 K was collected. The fractionally distilled solvent had a specific conductance less than (1 to 3) × 10^{-7} S·cm^{-1}. Triply distilled water with a specific conductance less than 10^{-6} S·cm^{-1} was used for the preparation of the mixtures.

Alkali-metal bromides were Fluka's purum or puriss grade (purity >99%) and were purified as described earlier (Nandi and Hazra, 1989). Purification of tetrabutyl ammonium bromide and sodium tetraphenyl borate (Fluka, Puriss) have also been reported previously (Nandi et al., 1989).

A stock solution for each salt was prepared by mass, and the working solutions were obtained by weight dilution. The conversion of the molality into molarity was done by using density values measured with an Ostwald−sprengel-type pycnometer. The bulb volume of the pycnometer was about 25 cm^3, and the internal diameter of the capillary of about 0.1 cm. Details have been described earlier (Das and Hazra, 1993; Saha et al., 1995).

Table 1. Densities (ρ), Viscosities (η), and Dielectric Constants (*D*) for 2 Methoxy-1-ethanol (1) + Water (2) at 298.15 K

X_1[a]	*D*	ρ/g·cm^{-3}	η_0/mPa·s
0.092	64.10	1.006 57	1.9465
0.262	42.19	1.002 33	2.8849
0.680	19.01	0.975 70	2.1342
1.000	16.93	0.960 02	1.5414

[a] X_1 is the mole fraction of 2-methoxy-1-ethanol.

The densities, ρ, were measured with an Ostwald−Sprengel-type pycnometer having a bulb volume of 25 cm^3 and an internal diameter of the capillary of about 0.1 cm. The pycnometer was calibrated at 298.15 K with doubly distilled water and benzene. The temperature of the bath was controlled to ±0.01 K.

The reproductibility of density measurements was ±3 × 10^{-5} g·cm^{-3}. The kinematic viscosities, ν, were measured by means of a suspended level Ubbelohde viscometer. The time of efflux was measured with a stopwatch to ±0.1 s. The viscometer was kept in a vertical position in a water thermostat controlled to ±0.01 K. The absolute temperature was determined by a calibrated platinum resistance thermometer and Muller bridge. The efflux time for water at 298.15 K was about 540 s. The kinematic viscosity (ν) and the absolute viscosity (η) are given by the following equations

$$\nu = Ct - K/t \qquad (1)$$

$$\eta = \nu\rho \qquad (2)$$

where *t* is the efflux time, ρ is the density, and *C* and *K* are the characteristic constants of the viscometer. The values of the constants *C* and *K* determined by using the density and viscosity values of water and benzene (Saha et al., 1995) were found to be 1.648 × 10^{-5} cm^2 S^{-2} and −0.023 316 47 cm^2, respectively. The estimated error of the viscosity measurements was ±0.2%. The electrical

S0021-9568(97)00283-5 CCC: $15.00

Table 2. Theoretical *A*-Coefficients, Limiting Conductances ($\Lambda°$), and Association Constants (K_A) of Alkali-Metal Bromides, $NaBPh_4$, and Bu_4NBr in 2-Methoxy-1-ethanol (1) + Water (2) at 298.15 K

	$X_1 = 0.092$			$X_1 = 0.262$			$X_1 = 0.680$			$X_1 = 1.00^a$		
	A/dm$\cdot$ mol$^{-1/2}$	Λ/S$\cdot$ cm$^2\cdot$ mol^{-1}	K_A/dm$^{-3}\cdot$ mol^{-1}	A/dm$\cdot$ mol$^{-1/2}$	Λ/S$\cdot$cm$^2\cdot$ mol^{-1}	K_A/dm$^{-3}\cdot$ mol^{-1}	A/dm$\cdot$ mol$^{-1/2}$	Λ/S$\cdot$cm$^2\cdot$ mol^{-1}	K_A/dm$^{-3}\cdot$ mol^{-1}	A/dm$\cdot$ mol$^{-1/2}$	Λ/S$\cdot$cm$^2\cdot$ mol^{-1}	K_A/dm$^{-3}\cdot$ mol^{-1}
LiBr	0.0074	53.82 (±0.06)	3.59 (±0.05)	0.0108	32.51 (±0.05)	17.40 (±0.43)	0.0234	27.93 (±0.01)	25.15 (±2.11)	0.0248	38.22 (±0.26)	328.00 (±25.00)
NaBr	0.0063	60.90 (±0.05)	1.29 (±0.03)	0.0087	37.39 (±0.04)	17.32 (±0.28)	0.0201	31.54 (±0.07)	35.38 (±1.31)	0.0289	33.71 (±0.31)	127.00 (±10.00)
KBr	0.0053	72.14 (±0.04)	1.59 (±0.02)	0.0077	41.80 (±0.19)	13.94 (±0.95)	0.0182	35.35 (±0.10)	40.61 (±1.48)	0.0247	38.40 (±0.10)	175.00 (±8.00)
RbBr	0.0052	73.22 (±0.13)	1.99 (±0.08)	0.0076	42.07 (±0.11)	11.88 (±0.46)	0.0176	36.66 (±0.07)	48.68 (±1.17)	0.0229	41.22 (±0.23)	240.00 (±16.00)
CsBr	0.0052	73.80 (±0.06)	1.91 (±0.03)	0.0075	42.61 (±0.03)	8.70 (±0.14)	0.0160	40.60 (±0.20)	81.82 (±3.31)	0.0203	46.42 (±0.27)	524.00 (±34.00)
$NaBPh_4$	0.0112	35.15 (±0.15)	0.73 (±0.05)	0.0095	28.58 (±0.02)	10.39 (±0.11)	0.0260	18.97 (±0.10)	17.71 (±2.45)	0.0341	24.69 (±0.13)	24.15 (±0.90)
Bu_4NBr	0.0096	51.06 (±0.04)	12.96 (±0.09)	0.0074	46.75 (±0.35)	61.73 (±4.10)	0.0245	26.67 (±0.06)	35.90 (±1.26)	0.0260	36.74 (±0.27)	372.00 (±24.00)

[a] Nandi and Hazra, 1989.

conductivities were measured with a Pye−Unicam conductivity meter (PW 9509) (accuracy ±0.1%).

In all cases, the experiments were performed at least in five replicates for each composition and the results were averaged. The density (ρ) and viscosity (η_0) of the mixed solvents are given in Table 1. The dielectric constants (D) of the solvent mixtures have been taken from the literature (Sadek et al., 1971).

Results

In the case of unassociated electrolyte solutions, the viscosity data can be analyzed by the following semiempirical equation of Jones and Dole (Jones and Dole, 1929) as described earlier by us (Muhuri and Hazra, 1993).

$$\eta/\eta_0 = 1 + A\sqrt{C} + BC \tag{3}$$

where η and η_0 are the viscosities of the solution and the solvent, respectively, and the other terms have their usual significance.

Since in the present study, except for CsBr and sodium tetraphenylborate ($NaBPh_4$) at 0.262 mole fraction of ME, in all other cases electrolytes were found to be strongly associated by the conductance measurements, the corresponding viscosities data were analyzed in the form of associated electrolytes (Lawrence et al., 1986) by eq 4 as given below:

$$\eta_r = 1 + A(\alpha C)^{1/2} + B_i\alpha C + B_p(1 - \alpha)C \tag{4}$$

Here A, B_i, and B_p are the characteristic constants and α is the degree of dissociation of the ion pair. In case of CsBr and $NaBPh_4$ at 0.262 mole fraction of ME−H_2O mixture, the viscosity data were analyzed by using eq 1.

For analysis of viscosity data by eq 4, first the equivalent conductances, Λ, of the electrolytes at various concentrations were used in an iterative procedure to find both $\Lambda°$ and the association constant, K_A, of the ion pair. The degree of association has been evaluated in the following way using eqs 5−7:

$$K_A = \frac{1 - \alpha}{\alpha^2 C \nu_\pm{}^2} \tag{5}$$

$$\log_{10} \nu_\pm = \frac{-A_y\sqrt{\alpha C}}{1 + qB_y\sqrt{\alpha C}} \tag{6}$$

and

$$\Lambda = \Lambda° - \left(\frac{A_\Lambda + B_\Lambda\Lambda°}{1 + qB_y\sqrt{\alpha C}}\right)(\sqrt{\alpha C}) \tag{7}$$

Here Λ is the conductivity of the ionized fraction of the electrolyte, and other terms have their usual significance (Lawrence et al., 1986).

Conductance measurements was made for each system at molarities of the electrolytes in the range 0.009−0.05 mol dm^{-3}. The data were analyzed by the Fuoss conductance equation (Fuoss, 1978), and the values of $\Lambda°$, the limiting equivalent conductance of the electrolyte, were obtained. The limiting equivalent conductances of the ions were calculated from the "reference electrolyte" Bu_4NBPh_4 (Hazra et al., 1989).

For analytical purpose, eq 4 was rearranged to give

$$(\eta_r - 1 - \mathrm{A}\sqrt{\alpha C})/\alpha C = B_i + B_p\left(\frac{1 - \alpha}{\alpha}\right) \tag{8}$$

and when $(\eta_r - 1 - A(\alpha C)^{1/2})/\alpha C$ was plotted against $(1 - \alpha)/\alpha$, the B_i (viscosity coefficient of ion−solvent interaction) was calculated from the intercept of the curve. The A-values, as required for the above calculation, was obtained from the physical parameters of the solvent and the limiting ionic equivalent conductances (Das and Hazra, 1996) using Falkenhagen and Vernon equation and are given in Table 2. These A-values have been used for the analysis of the data.

The B-coefficients (B_i-values) for the electrolytes at 298.15 K obtained by the least-squares method are presented in Table 3.

It may be seen from Table 1 that the viscosity of ME + water mixtures is greater compared to the viscosity data of pure ME. This indicates increased structure formation of a mixed solvent system, which is maximum at about 0.262 mole fraction of ME. After this the downward trend is the reflection of the usual structure-breaking effect owing to packing imbalance at higher compositions of the cosolvent.

The B-coefficients shown in Table 3 in general are large and positive, and the values increase in ME + H_2O mixtures as the crystal radius decreases (except for sodium and potassium bromide). In 0.092 mole fraction of ME, B-coefficient values in some cases are not far from zero (Table 3). In such cases it might be that the viscosity of

Table 3. Viscosity *B*-coefficients in 2-Methoxy-1-ethanol (1) + Water (2) at 298.15 K

salt	X_1	$B/\mathrm{dm^3 \cdot mol^{-1}}$
LiBr	0.092	0.130(±0.01)
	0.262	0.251(±0.001)
	0.680	0.597(±0.002)
	1.000	0.302(±0.03)
NaBr	0.092	0.007(±0.002)
	0.262	0.193(±0.001)
	0.680	0.495(±0.002)
	1.000	0.435(±0.005)
KBr	0.092	0.065(±0.001)
	0.262	0.154(±0.001)
	0.680	0.359(±0.002)
	1.000	0.289(±0.002)
RbBr	0.092	0.084(±0.001)
	0.262	0.121(±0.001)
	0.680	0.291(±0.001)
	1.000	0.246(±0.003)
CsBr	0.092	0.011(±0.0002)
	0.262	0.101(±0.001)
	0.680	0.256(±0.001)
	1.000	0.202(±0.001)
$NaBPh_4$	0.092	0.020(±0.0003)
	0.262	0.513(±0.002)
	0.680	1.181(±0.001)
	1.000	0.687(±0.003)
Bu_4NBr	0.092	0.016(±0.001)
	0.262	0.416(±0.002)
	0.680	0.928(±0.002)
	1.000	0.425(±0.002)

Table 4. Ionic *B*-coefficients in 2-Methoxy-1-ethanol (1) + Water (2) at 298.15 K

ion	X_1	$B_\pm/\mathrm{dm^3 \cdot mol^{-1}}$
Li^+	0.000	−0.043[a]
	0.092	0.127
	0.262	0.168
	0.680	0.394
	1.000	0.181
Na^+	0.000	−0.106[a]
	0.092	0.004
	0.262	0.108
	0.680	0.293
	1.000	0.314
K^+	0.000	−0.199[a]
	0.092	0.062
	0.262	0.069
	0.680	0.156
	1.000	0.168
Rb^+	0.000	−0.221[a]
	0.092	0.081
	0.262	0.036
	0.680	0.088
	1.000	0.125
Cs^+	0.000	−0.237[a]
	0.092	0.008
	0.262	0.016
	0.680	0.053
	1.000	0.081
Bu_4N^+	0.000	1.074[b]
	0.092	0.013
	0.262	0.331
	0.680	0.725
	1.000	0.304
Ph_4B^-	0.000	1.316[b]
	0.092	0.016
	0.262	0.405
	0.680	0.888
	1.000	0.373
Br^-	0.000	0.160[b]
	0.092	0.003
	0.262	0.085
	0.680	0.203
	1.000	0.121

[a] *B*-values for the electrolytes taken from: Stokes, R. H.; Mills, R. *Viscosity of electrolytes and related properties*; Pergamon Press: Oxford, 1965. [b] *B*-values taken from: Sacco, A.; Giglio, A. D.; Dell'Atti, A.; Petrella, M. *J. Chem. Soc., Faraday Trans. 1* **1981**, *77*, 2693.

the solvent is very little modified by the presence of these solutes in solution. According to Gurney (Gurney, 1962), this can happen when the cosphere of the positive ions causes a local increase in viscosity, while the cosphere of the negative ion causes a nearly equal decrease in the viscosity, or vice versa. And whenever such a cancellation occurs, the *B*-coefficient of the solute will have a value not far from zero.

The ionic *B*-values were calculated by eqs 9−11 using Bu_4NBPh_4 as the "reference electrolyte" (Tuan and Fuoss, 1963; Gill and Sharma, 1982).

$$\frac{B_{\mathrm{Ph_4B^-}}}{B_{\mathrm{Bu_4N^+}}} = \frac{r^3{}_{\mathrm{Ph_4B^-}}}{r^3{}_{\mathrm{Bu_4N^+}}} = \left(\frac{5.35}{5.00}\right)^3 \tag{9}$$

$$B_{\mathrm{Bu_4NBPh_4}} = B_{\mathrm{Ph_4B^-}} + B_{\mathrm{Bu_4N^+}} \tag{10}$$

$$B_{\mathrm{Bu_4NBPh_4}} = B_{\mathrm{Bu_4NBr}} + B_{\mathrm{NaBPh_4}} - B_{\mathrm{NaBr}} \tag{11}$$

The average B_{ion}-values of alkali-metal ions are recorded in Table 4. The ionic *B*-coefficients of alkali−metal ions are found to decrease from Li^+ to Cs^+ in ME + H_2O mixtures except at 0.092 mole fraction of ME and for Na^+ in pure ME.

An analysis of $B_\pm$-coefficients can be made on the basis of Einstein's equation (Das and Hazra, 1992).

$$B_\pm = 2.5(4/3)\pi\frac{R_\pm{}^3N}{1000} \tag{12}$$

where $R_\pm$ is the radius of the ion assumed as a rigid sphere moving in a continuum, *N* is the Avogadro number, and 2.5 is the shape factor for a sphere. The number n_s of solvent molecules bound to the ion in the primary shell of solvation can be calculated by combining the Jones−Dole equation with that of Einstein.

$$B_\pm = (2.5/1000)\,(V_i + n_sV_s) \tag{13}$$

where V_i represents the bare ion molar volume and is related to the crystallographic radius, r_c, of the ion. V_s is the solvent molar volume. The values of *R* and n_s are given in Table 5. The calculation of solvation numbers is dependent on the choice of r_c-values. We have reported the solvation numbers of alkali-metal ions based on the r_c-values of Gourary and Adrian (Criss and Solomn, 1973). For cations in mixed solvents, Li^+ is found to be maximum solvated followed by Na^+, whereas in other cases r_c-values are slightly greater or almost equal to *R*-values, indicating that these ions are either partially solvated or behave as spherical entities in ME + H_2O mixtures. In pure solvent Na^+ is maximum solvated, and solvation number decreases as the size of the alkali-metal ions increases. The negative values of solvation number (n_s) listed in Table 5 are physically unacceptable. They seem to indicate that the determination of solvation numbers on the basis of eq 13 does not appear to be correct. This probably arises from the fact that the electrolytic solutions are different from the model that underlines eq 13.

The low $B_\pm$ values of alkali metal ions and Br^- ions in water (Table 4) are due to the breakdown of the tetrahedral structure of water and formation of strongly structured solvated ion. In ME, both the cations and anions have $B_\pm$-

Table 5. Ionic Radii ($R_\pm$) and Solvation Number (n_s) of Ions in 2-Methoxy-1-ethanol (1) + Water (2) at 298.15 K

ion	X_1	$r_c{}^a$/Å	$R_\pm$/Å	n_s
Li^+	0.092	0.93	2.72	2.10
	0.262		2.99	1.97
	0.680		3.97	2.64
	1.000		3.06	0.88
Na^+	0.092	1.17	0.86	−0.11
	0.262		2.58	1.18
	0.680		3.59	1.92
	1.000		3.68	1.50
K^+	0.092	1.49	2.14	0.71
	0.262		2.22	0.58
	0.680		2.91	1.06
	1.000		2.99	0.72
Rb^+	0.092	1.64	2.32	0.92
	0.262		1.79	0.10
	0.680		2.41	0.60
	1.000		2.71	0.48
Cs^+	0.092	1.83	1.08	−0.53
	0.262		1.36	−0.27
	0.680		2.03	0.36
	1.000		2.34	0.21
Bu_4N^+	0.092	4.94	1.27	−12.71
	0.262		3.75	−5.18
	0.680		4.86	−0.24
	1.000		3.64	−2.25
Br^-	0.092	1.80	0.78	−0.58
	0.262		2.38	0.58
	0.680		3.18	1.13
	1.000		2.68	0.42
Ph_4B^-	0.092	4.20	1.36	−7.77
	0.262		4.01	−0.75
	0.680		5.20	−0.53
	1.000		3.90	−0.46

[a] Criss and Salomn (1973).

values considerably higher than those in water and can reasonably be ascribed to increased ion−solvent interactions. From the result, it is apparent that Na^+ ion is strongly solvated by ME. So disruption of hydrated structure and formation of Na^+−ME complex increases the B_{Na^+}-value. Thus, the higher $B_\pm$-values in ME compare to those in water of the alkali-metal ions can be ascribed to ion−solvent interactions or increase in solvodynamic entity due to the bigger molecular size of ME.

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Received for review November 25, 1996. Accepted March 5, 1998. D.N. thanks the U.G.C., New Delhi, for the award of a research fellowship.

JE970283J