# Articles

# Electrical Conductances of Some Symmetrical Tetraalkylammonium Salts in Methanol, Acetonitrile, and Methanol (1) + Acetonitrile (2) Mixtures at 298.15 K

**Bijan Das* and Nirmal Saha**

Department of Chemistry, North Bengal University, Darjeeling 734 430, India

Electrical conductance measurements are reported for tetraethylammonium bromide ($Et_4NBr$), tetrapropylammonium bromide ($Pr_4NBr$), tetrabutylammonium bromide ($Bu_4NBr$), and tetrapentylammonium bromide ($Pen_4NBr$) in methanol, acetonitrile, and methanol (1) + acetonitrile (2) binary mixtures at 298.15 K. The conductance data have been analyzed by the Fuoss conductance−concentration equation in terms of the limiting molar conductance ($\Lambda°$), the association constant ($K_A$), and the distance of closest approach of ions ($R$). The results have been interpreted in terms of ion−ion and ion−solvent interactions.

## Introduction

Extensive studies on electrical conductivities in various mixed organic solvents have been performed in recent years (Muhuri and Hazra, 1995; Srivastava and Samant, 1994; Doe et al., 1990; Taniewska-Osinska et al., 1989; Islam et al., 1989; Papadopulos, 1989; Niazi, 1988) to examine the nature and magnitude of ion−ion and ion−solvent interactions. Such solvent properties as the viscosity and the relative permittivity have also been taken into consideration, which help determine the extent of ion association and the solute−solvent interactions.

The present study deals with mixtures of methanol—a dipolar protic solvent—and acetonitrile—a dipolar aprotic solvent. Methanol is known to be extensively self-associated through hydrogen-bonding in the pure state (Pikkarainen, 1988). In the case of acetonitrile, on the other hand, there is a lack of strong specific intermolecular forces, while dipole−dipole forces predominate (Moumouzias et al., 1991). Recently, we have shown (Saha et al., 1995) that, in the mixture containing methanol and acetonitrile, there is a breakdown of hydrogen bonds in methanol and formation of specific complexes between the unlike molecules through hydrogen bonds. In view of the complex nature of methanol−acetonitrile mixtures, it is of interest to investigate the influence of the solvent composition on the ionic association and the solvation behavior of some tetraalkylammonium bromides using conductance measurements.

In this paper, we report the molar conductivities of four selected symmetrical tetraalkylammonium bromides, viz., tetraethylammonium bromide ($Et_4NBr$), tetrapropylammonium bromide ($Pr_4NBr$), tetrabutylammonium bromide ($Bu_4NBr$), and tetrapentylammonium bromide ($Pen_4NBr$) in methanol, acetonitrile, and methanol (1) + acetonitrile (2) ranging in composition from 0.20 to 0.80 mole fraction of methanol at 298.15 K.

## Experimental Section

Methanol (E. Merck, India, uvasol grade, 99.5% pure) was dried over 3 Å molecular sieves and distilled fractionally. The middle fraction was collected and redistilled. The purified solvent had a density of 0.78654 $g\cdot cm^{-3}$ and a viscosity of 0.545 mPa·s at 298.15 K. These values agree well with the literature values (Moumouzias et al., 1991; Singh et al., 1994).

Acetonitrile (E. Merck, India, 99% pure) was distilled with $P_2O_5$ and then redistilled over $CaH_2$. The purified solvent had a density of 0.77686 $g\cdot cm^{-3}$ and a coefficient of viscosity of 0.345 mPa·s at 298.15 K; these values are also found to be in good agreement with literature values (Doe et al., 1984).

The purities as checked by gas chromatography were found to be better than 99.8% for methanol and 99.5% for acetonitrile.

Tetraalkylammonium bromides were of purum or puriss grade (Fluka) and were purified as described earlier by us (Das and Hazra, 1991; Muhuri et al., 1996).

A stock solution for each salt was prepared by mass, and the working solutions were obtained by mass dilution. The conversion of molality to molarity was done using the density values.

The densities ($\rho$) were measured with an Ostwald−Sprengel type pycnometer having a bulb volume of 25 $cm^3$ and an internal diaimeter of the capillary of about 0.1 cm. The pycnometer was calibrated at (298.15, 308.15, and 318.15) K with doubly distilled water and benzene. Details have been described earlier (Das and Hazra, 1993, 1995a; Saha et al., 1995).

Conductance measurements were carried out on a Pye−Unicam PW 9509 conductivity meter at a frequency of 2000 Hz using a dip-type cell (cell constant = 0.751 $cm^{-1}$) with an accuracy of 0.01%. The cell was calibrated by the method of Lind and co-workers (Lind et al., 1959) using aqueous potassium chloride solutions. Measurements were made in an oil bath maintained at (298.15 ± 0.005) K, as described earlier (Das and Hazra, 1992, 1995b). Several independent solutions were prepared, and conductance measurements were performed with each of these to ensure the reproduc-

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**Table 1. Densities ($\rho$), Viscosities ($\eta$), and Relative Permittivities ($D$) for Methanol (1) + Acetonitrile (2) at 298.15 K**

| $x_1{}^a$ | $\rho^b$/g·cm$^{-3}$ | $\eta^b$/mPa·s | $D^c$ |
|---|---|---|---|
| 0.00 | 0.776 86 | 0.346 | 35.95 |
| 0.20 | 0.779 52 | 0.334 | 35.47 |
| 0.40 | 0.782 12 | 0.343 | 34.92 |
| 0.60 | 0.784 65 | 0.373 | 34.30 |
| 0.80 | 0.786 80 | 0.435 | 33.58 |
| 1.00 | 0.786 54 | 0.545 | 32.62 |

[a] $x_1$ is the mole fraction of methanol. [b] From Saha et al., 1995. [c] From Gill and Chauhan, 1984.

ibility of the results. Corrections were made for the specific conductances of the solvent.

The experimental values of densities ($\rho$), viscosities ($\eta$), and relative permittivities ($D$) of methanol (1) + acetonitrile (2) at 298.15 K are not available at the relevant compositions in which conductance measurements have been carried out. The available values of $\rho$ and $\eta$ (from Saha et al., 1995) and those of $D$ (from Gill and Chauhan, 1984) are, therefore, plotted against the mole fraction of methanol, and the values of $\rho$, $\eta$, and $D$ at all the desired compositions have been generated from the smooth master curves and are given in Table 1.

## Results

Molar conductances ($\Lambda$) of electrolyte solutions as a function of molar concentration ($c$) are given in Table 2 in different solvent mixtures at 298.15 K.

The conductance data have been analyzed by the 1978 Fuoss conductance−concentration equation (Fuoss, 1978a,b). For a given set of conductivity values ($c_j$, $\Lambda_j$; $j = 1, ..., n$), three adjustable parameters, the limiting molar conductivity ($\Lambda°$), the association constant ($K_A$), and the distance of closest approach of ions ($R$), are derived from the following set of equations:

$$\Lambda = p[\Lambda°(1 + R_X) + E_L] \tag{1}$$

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

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

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

$$\beta = e^2/Dk_BT \tag{5}$$

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

where $R_X$ is the relaxation field effect, $E_L$ is the electrophoretic countercurrent, $k^{-1}$ is the radius of the ion atmosphere, $D$ is the relative permittivity of the solvent, $e$ is the electron charge, $k_B$ is the Boltzmann constant, $\gamma$ is the fraction of solute present as unpaired ion, $c$ is the molarity of the solution, $f$ is the activity coefficient, $T$ is the absolute temperature, and $\beta$ is twice the Bjerrum distance. The computations were performed on a computer using the program suggested by Fuoss. The initial $\Lambda°$ values for the iteration procedure were obtained from Shedlovsky extrapolation of the data. Input for the program is the set ($c_j$, $\Lambda_j$; $j = 1, ..., n$), $n$, $D$, $\eta$, $T$, initial value of $\Lambda°$, and an instruction to cover a preselected range of $R$ values.

In practice, calculations are performed by finding the values of $\Lambda°$ and $\alpha$ which minimize the standard deviation $\sigma$

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

for a sequence of $R$ values and then plotting $\sigma$ against $R$; the best-fit $R$ corresponds to the minimum of the $\sigma$ versus $R$ curve. First, approximate runs over a fairly wide range of $R$ values are made to locate the minimum, and then, a fine scan around the minimum is made. Finally, with this minimizing value of $R$, the corresponding $\Lambda°$ and $K_A$ are calculated.

The values of $\Lambda°$, $K_A$, and $R$ obtained by this procedure are reported in Table 3.

## Discussion

The conductances of solutions of $Pr_4NBr$, and $Bu_4NBr$ in acetonitrile and of $Et_4NBr$, $Pr_4NBr$, $Bu_4NBr$, and $Pen_4NBr$ in methanol have also been reported earlier by other workers (Evans et al., 1965; Kay et al., 1965). They analyzed their data with the Fuoss−Onsager conductance equation. Conductance data of sufficient precision, with the exception of the works mentioned above, for any of the systems investigated here do not exist in the literature. Consequently, we compared the $\Lambda°$ and $K_A$ values for the above-mentioned systems with those from the present work (Table 3). It has been observed that the values of $\Lambda°$ calculated from the previous conductance equation do not differ significantly from the values of $\Lambda°$ calculated from the 1978 Fuoss conductance−concentration equation. The values of $K_A$ obtained from our work are, however, generally higher. This discrepancy may be attributed to the different models used for analysis of the experimental data. We have reanalyzed the literature experimental data with the 1978 Fuoss conductance equation, and the following results were obtained: $\Lambda° = 170.60$, $K_A = 25.89$ for $Pr_4NBr$, and $\Lambda° = 162.26$, $K_A = 20.60$ for $Bu_4NBr$ in acetonitrile, and $\Lambda° = 117.01$, $K_A = 26.12$ for $Et_4NBr$, and $\Lambda° = 102.61$, $K_A = 28.69$ for $Pr_4NBr$, and $\Lambda° = 95.48$, $K_A = 29.34$ for $Bu_4NBr$, and $\Lambda° = 91.47$, $K_A = 30.98$ for $Pen_4NBr$ in methanol. In each case a better fit of the conductance data to the 1978 Fuoss equation than to the Fuoss−Onsager equation has been observed. These $\Lambda°$ and $K_A$ values are in good agreement with our results.

The association constants ($K_A$) in Table 3 indicate that all the salts are slightly associated in acetonitrile and methanol (1) + acetonitrile (2) mixtures. The $K_A$ values for all the $R_4NBr$ salts in a given solvent medium decrease with increasing size of the cation in the order $Et_4N^+ > Pr_4N^+ > Bu_4N^+ > Pen_4N^+$, similar to those observed earlier, due to an apparent decrease in the ion−dipole interaction (Kay et al., 1965; Islam et al., 1989). For each salt, however, ionic association increases slowly with increasing methanol content in the solvent mixtures. This is quite expected due to the gradual decrease in the dielectric constant of the medium with increasing amount of methanol in the mixtures.

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**Table 2. Molar Conductances ($\Lambda$) and Corresponding Molarities ($c$) of Tetraalkylammonium Bromides in Methanol (1) + Acetonitrile (2) at 298.15 K**

| $10^4c$/mol·dm$^{-3}$ | $\Lambda$/S·cm$^2$·mol$^{-1}$ | $10^4c$/mol·dm$^{-3}$ | $\Lambda$/S·cm$^2$·mol$^{-1}$ | $10^4c$/mol·dm$^{-3}$ | $\Lambda$/S·cm$^2$·mol$^{-1}$ | $10^4c$/mol·dm$^{-3}$ | $\Lambda$/S·cm$^2$·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|
| | | | | $x_1 = 0.00$ | | | |
| Et$_4$NBr | | Pr$_4$NBr | | Bu$_4$NBr | | Pen$_4$NBr | |
| 5.011 | 176.55 | 5.005 | 162.55 | 5.500 | 153.44 | 4.883 | 148.56 |
| 7.499 | 174.50 | 11.110 | 158.30 | 10.100 | 150.40 | 7.712 | 146.58 |
| 10.207 | 172.59 | 14.546 | 156.42 | 15.043 | 147.82 | 10.102 | 145.13 |
| 14.990 | 169.76 | 20.249 | 153.75 | 19.226 | 145.99 | 15.408 | 142.49 |
| 25.535 | 164.83 | 35.307 | 148.24 | 25.120 | 143.73 | 20.090 | 140.54 |
| 35.412 | 161.14 | 40.010 | 146.80 | 30.234 | 142.00 | 24.996 | 138.75 |
| 40.008 | 159.62 | 42.111 | 146.16 | 34.950 | 140.54 | 30.108 | 137.08 |
| 45.163 | 158.01 | 46.124 | 145.08 | 40.111 | 139.07 | 35.311 | 135.54 |
| | | | | 42.000 | 138.55 | 39.416 | 134.39 |
| | | | | 45.461 | 137.67 | 45.919 | 132.78 |
| | | | | $x_1 = 0.20$ | | | |
| Et$_4$NBr | | Pr$_4$NBr | | Bu$_4$NBr | | Pen$_4$NBr | |
| 4.505 | 171.98 | 4.603 | 155.79 | 4.566 | 147.06 | 4.514 | 141.67 |
| 9.514 | 167.95 | 10.005 | 151.82 | 9.862 | 143.34 | 9.965 | 139.97 |
| 12.162 | 166.25 | 12.016 | 150.63 | 11.924 | 142.19 | 12.055 | 136.85 |
| 15.036 | 164.61 | 14.996 | 149.05 | 14.900 | 140.70 | 15.022 | 135.42 |
| 19.992 | 162.12 | 19.988 | 146.73 | 20.001 | 138.48 | 19.886 | 133.38 |
| 25.003 | 159.91 | 24.980 | 144.71 | 25.010 | 136.59 | 25.112 | 131.48 |
| 30.107 | 157.89 | 30.006 | 142.89 | 29.979 | 134.90 | 29.982 | 129.89 |
| 34.935 | 156.15 | 35.105 | 141.20 | 34.998 | 133.36 | 35.114 | 128.38 |
| 38.849 | 154.51 | 40.008 | 139.71 | 40.110 | 131.91 | 40.027 | 127.04 |
| 44.666 | 153.01 | 44.945 | 138.31 | 45.001 | 130.63 | 44.925 | 125.80 |
| | | | | $x_1 = 0.40$ | | | |
| Et$_4$NBr | | Pr$_4$NBr | | Bu$_4$NBr | | Pen$_4$NBr | |
| 4.500 | 166.17 | 4.418 | 150.25 | 4.432 | 140.54 | 4.530 | 136.15 |
| 10.012 | 161.88 | 10.104 | 146.16 | 9.994 | 136.73 | 9.998 | 132.52 |
| 12.116 | 160.57 | 11.990 | 145.05 | 12.016 | 135.63 | 12.171 | 131.38 |
| 15.108 | 158.89 | 15.032 | 143.47 | 15.034 | 134.16 | 15.123 | 130.00 |
| 20.308 | 156.34 | 20.146 | 141.16 | 19.976 | 132.07 | 20.315 | 127.88 |
| 24.996 | 154.32 | 24.969 | 139.26 | 25.126 | 130.17 | 24.992 | 126.22 |
| 30.132 | 152.33 | 30.125 | 137.44 | 30.039 | 128.56 | 30.096 | 124.59 |
| 34.988 | 150.62 | 35.077 | 135.84 | 34.999 | 127.07 | 35.016 | 123.17 |
| 40.026 | 148.97 | 40.005 | 134.38 | 39.989 | 125.70 | 40.107 | 121.82 |
| 44.008 | 147.76 | 43.992 | 133.28 | 43.999 | 124.67 | 44.003 | 120.86 |
| | | | | $x_1 = 0.60$ | | | |
| Et$_4$NBr | | Pr$_4$NBr | | Bu$_4$NBr | | Pen$_4$NBr | |
| 4.301 | 155.39 | 4.343 | 139.17 | 4.306 | 130.52 | 4.333 | 125.51 |
| 9.784 | 151.30 | 10.012 | 135.34 | 10.107 | 126.81 | 9.998 | 121.95 |
| 12.006 | 149.99 | 11.956 | 134.27 | 12.315 | 125.68 | 12.036 | 120.95 |
| 15.133 | 148.33 | 15.018 | 132.79 | 15.228 | 124.36 | 15.102 | 119.60 |
| 20.198 | 146.01 | 20.333 | 130.51 | 20.112 | 122.38 | 20.006 | 117.72 |
| 24.821 | 144.09 | 25.006 | 128.87 | 24.996 | 120.77 | 25.016 | 116.05 |
| 30.002 | 142.21 | 29.998 | 127.18 | 30.121 | 119.15 | 30.008 | 114.57 |
| 35.119 | 140.52 | 35.016 | 125.67 | 34.995 | 117.78 | 34.998 | 113.22 |
| 39.903 | 139.05 | 40.105 | 124.25 | 40.303 | 116.37 | 40.101 | 111.96 |
| 42.996 | 138.15 | 43.000 | 123.53 | 43.011 | 115.79 | 43.001 | 111.28 |
| | | | | $x_1 = 0.80$ | | | |
| Et$_4$NBr | | Pr$_4$NBr | | Bu$_4$NBr | | Pen$_4$NBr | |
| 4.001 | 136.16 | 4.018 | 121.28 | 4.038 | 112.89 | 4.000 | 108.40 |
| 9.503 | 132.46 | 9.515 | 117.88 | 9.495 | 109.70 | 9.501 | 105.30 |
| 12.516 | 130.89 | 12.498 | 116.47 | 12.499 | 108.36 | 12.496 | 103.99 |
| 15.003 | 129.79 | 15.001 | 115.41 | 15.001 | 107.36 | 14.998 | 103.04 |
| 20.011 | 127.70 | 20.028 | 113.52 | 20.103 | 105.57 | 19.989 | 101.34 |
| 25.001 | 125.87 | 24.998 | 111.95 | 25.006 | 104.12 | 25.013 | 99.92 |
| 29.998 | 124.28 | 30.928 | 110.22 | 29.999 | 102.75 | 30.016 | 98.59 |
| 35.010 | 122.81 | 34.990 | 109.16 | 34.998 | 101.52 | 35.001 | 97.42 |
| 37.500 | 122.11 | 37.503 | 108.53 | 37.510 | 100.93 | 37.520 | 96.87 |
| 40.001 | 121.49 | 39.996 | 107.95 | 40.010 | 100.40 | 39.998 | 96.32 |
| | | | | $x_1 = 1.00$ | | | |
| Et$_4$NBr | | Pr$_4$NBr | | Bu$_4$NBr | | Pen$_4$NBr | |
| 3.501 | 111.88 | 3.499 | 97.83 | 4.002 | 90.52 | 3.501 | 87.04 |
| 5.115 | 110.81 | 5.000 | 96.93 | 7.497 | 88.73 | 5.071 | 86.10 |
| 7.500 | 109.49 | 9.120 | 94.89 | 10.101 | 87.65 | 10.210 | 83.92 |
| 12.001 | 107.36 | 10.003 | 94.53 | 16.001 | 85.66 | 12.503 | 83.14 |
| 14.216 | 106.49 | 16.006 | 92.37 | 21.978 | 84.02 | 14.001 | 82.66 |
| 20.006 | 104.49 | 22.215 | 90.59 | 25.000 | 83.29 | 19.004 | 81.26 |
| 22.100 | 103.85 | 28.008 | 89.14 | 29.986 | 82.18 | 24.999 | 79.83 |
| 27.998 | 102.18 | 33.512 | 87.91 | 33.059 | 81.53 | 30.017 | 78.78 |
| 34.165 | 100.66 | 35.000 | 87.60 | 35.003 | 81.19 | 32.992 | 78.21 |

**Table 3. Derived Conductivity Parameters for Tetaalkylammonium Bromides in Methanol (1) + Acetonitrile (2) at 298.15 K**

| $x_1$ | $\Lambda°$/S·cm$^2$·mol$^{-1}$ | $K_A$/dm$^{-3}$·mol$^{-1}$ | $R$/Å | $100\sigma/\Lambda°$ |
|---|---|---|---|---|
| | | $Et_4NBr$ | | |
| 0.00 | 185.53 ± 0.01 | 29.38 | 15.80 | 0.01 |
| 0.20 | 180.61 ± 0.01 | 30.78 | 16.10 | 0.01 |
| 0.40 | 174.67 ± 0.01 | 31.77 | 16.20 | 0.01 |
| 0.60 | 163.21± 0.01 | 32.72 | 16.30 | 0.01 |
| 0.80 | 142.80 ± 0.02 | 34.17 | 16.90 | 0.02 |
| 1.00 | 117.04 ± 0.01 (117.01[a]) | 35.77 (26.12[a]) | 17.00 | 0.01 |
| | | $Pr_4NBr$ | | |
| 0.00 | 171.11 ± 0.02 (170.60[b]) | 28.76 (25.89[b]) | 15.80 | 0.02 |
| 0.20 | 164.08 ± 0.01 | 30.30 | 16.00 | 0.01 |
| 0.40 | 158.22 ± 0.01 | 31.14 | 16.10 | 0.01 |
| 0.60 | 146.58 ± 0.02 | 32.07 | 16.30 | 0.02 |
| 0.80 | 127.53 ± 0.01 | 33.35 | 16.50 | 0.01 |
| 1.00 | 102.61 ± 0.01 (102.61[a]) | 35.51 (28.69[a]) | 17.50 | 0.01 |
| | | $Bu_4NBr$ | | |
| 0.00 | 162.08 ± 0.02 (162.26[b]) | 27.17 (20.60[b]) | 15.60 | 0.02 |
| 0.20 | 155.00 ± 0.01 | 28.87 | 15.90 | 0.01 |
| 0.40 | 148.22 ± 0.01 | 27.97 | 16.00 | 0.01 |
| 0.60 | 137.63 ± 0.03 | 31.23 | 16.30 | 0.03 |
| 0.80 | 118.92 ± 0.01 | 32.56 | 16.40 | 0.01 |
| 1.00 | 95.45 ± 0.01 (95.48[a]) | 35.01 (29.34[a]) | 17.00 | 0.02 |
| | | $Pen_4NBr$ | | |
| 0.00 | 156.54 ± 0.02 | 26.59 | 15.60 | 0.02 |
| 0.20 | 149.41 ± 0.01 | 28.39 | 16.00 | 0.01 |
| 0.40 | 143.77 ± 0.02 | 29.46 | 16.10 | 0.02 |
| 0.60 | 132.48 ± 0.03 | 30.58 | 16.10 | 0.01 |
| 0.80 | 114.26 ± 0.01 | 32.34 | 16.80 | 0.02 |
| 1.00 | 91.50 ± 0.01 (91.47[a]) | 33.18 (30.98[a]) | 16.60 | 0.02 |

[a] Values obtained from recalculation of the data from Kay et al. (1965) using the Fuoss conductance equation. [b] Values obtained from recalculation of the data from Evans et al. (1965) using Fuoss conductance equation.

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