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Study of Solution Properties of Some Alkali Bromides in Aqueous Binary Mixtures of 1,3-Dioxolane in View of Different Models

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Viscosity *B*-coefficients of lithium, sodium, and potassium bromide in (30, 60, and 90) % aqueous 1,3-dioxolane solutions have been determined at (298.15 and 308.15) K from density and viscosity measurements employing the Jones−Dole, Breslau−Miller, and Vand equations. *B*-coefficients thus obtained were compared. Furthermore, the applicability of the Moulik equation was shown by linear plots of relative viscosity and concentration values in their square terms. It has been found that the *B*-coefficients vary with the size of the cations, which suggests a structure-promoting tendency for all of the salts and for all of the concentrations.

Introduction

The addition of an organic solvent to an aqueous electrolytic solution alters the pattern of ion solvation and often stimulates phenomenal changes in the behavior of the dissolved electrolytes. Hence, studies on viscosity *B*-coefficients of electrolytes at infinite dilution provide valuable information regarding ion−ion, ion−solvent, and solvent−solvent interactions.[1−3] This information is of fundamental importance for a proper understanding of the behavior of electrolytes in solution. Recently we have utilized a set of equations[4,6] to study the solvation and association behavior of some electrolytes[3,7−10] in different aqueous and nonaqueous solvent media from the measurement of transport and thermodynamic properties. As viscosity *B*-coefficients of a solute reflect the cumulative effects[11] of ion−ion and ion−solvent interactions, in this paper we attempted to study these properties for lithium, sodium, and potassium bromide in aqueous binary mixtures of 1,3-dioxolane at (298.15 and 308.15) K; such data may highlight the role of the constituent ions in characterizing their behavior. Since all of the salts have a common anion, for all of the electrolytes under investigation, the present work may enable us to have a qualitative comparison of the role played by the cations in aqueous binary mixtures of 1,3-dioxolane in terms of various derived parameters obtained from density and viscosity measurements.

Experimental Section

Materials. 1,3-Dioxolane, $C_3H_6O_2$ (ethylene glycol formal or ethylene glycol methylene ether) from Merck, containing 0.3 % water and 0.005 % peroxides and sterilized with butylated hydroxytoluene, was purified by heating under reflux with PbO_2 for 2 h and then cooled and filtered. After adding xylene to the filtrate, the mixture was fractionally distilled.[11,12] The solvent obtained after purification had a boiling point of 348 K/760 mm, a density of 1058.6 $kg \cdot m^{-3}$ and a coefficient of viscosity of 0.00575 mPa·s at 298.15 K which was in good agreement with the literature values.[12,13] The purity of the solvent finally obtained was 99.0 %. Triply distilled water was used for the experiments. The purification of alkali bromides (LiBr, NaBr, KBr) has been described earlier in the literature.[5]

Apparatus and Procedure. Densities were measured with an Ostwald−Sprengel type pycnometer with a bulb volume of about 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. The pycnometer with experimental liquid was equilibrated in a glass-walled thermostatted water bath maintained within ± 0.01 K of the desired temperature. The pycnometer was then removed from the thermostat, properly dried, and weighed in an electronic balance (Mettler Toledo digital electronic balance) with a precision of ± 0.01 mg. Adequate precautions were taken to avoid evaporation losses during the time of measurements. An average of the triplicate measurements was taken into account. The uncertainty of density values is $\pm\, 3 \cdot 10^{-4}$ $g \cdot cm^{-3}$. The details of the methods and measurement techniques have been described earlier.[14] Solvent viscosities were measured by means of a suspended Ubbelohde type viscometer, calibrated at 298.15 K with doubly distilled water and purified methanol using density and viscosity values from the literature.[15−17] A thoroughly cleaned and perfectly dried viscometer filled with experimental liquid was placed vertically in the glass-walled thermostat maintained to ± 0.01 K. After attainment of thermal equilibrium, efflux times of flow were recorded with a stopwatch correct to ± 0.1 s. At least three repetitions of each datum reproducible to ± 0.1 s were taken to average the flow times. The uncertainty of viscosity values is ± 0.003 mPa·s. The details of the methods and measurement techniques have been described elsewhere.[18,19]

Results and Discussion

Density, viscosity, and relative viscosity ($\eta_r = \eta/\eta_0$) data of alkali metal bromides in the binary mixtures of aqueous 1,3-dioxolane at (298.15 and 308.15) K are recorded in Table 1. It can be seen from the data that the values of the viscosities of the solutions increase with increasing concentration of the salt and decrease with increasing temperature.

The variation of relative viscosity with concentration is usually expressed by the Jones−Dole equation[20]

$$\eta_r = 1 + Ac^{1/2} + Bc \qquad (1)$$

Since, in general, $A/B \ll 1$, the second term may be neglected at concentrations above 0.002 M, and eq 1 may be written as[21]

$$\eta_r = 1 + Bc \qquad 0.002 < M < \sim 0.1 \qquad (2)$$

In the present study, eq 2 is valid in solutions of concentration ranging between (0.01 and 0.08) M. The linearity obtained by

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Table 1. Concentration in Molarity (M), Molality (m), Density (ρ), Viscosity (η), and Relative Viscosity (η_r) of Alkali Bromides in Aqueous 1,3-Dioxolane at Different Temperatures

molarity (M) mol·dm^{-3}	molality (m) mol·kg^{-1}	$\rho \cdot 10^{-3}$ kg·m^{-3}	η mPa·s	η_r
298.15 K				
LiBr in 30 % Water + 70 % 1,3-DO, MW = 86.85, ρ = 1.0492, η = 0.9716				
0.0117	0.0090	1.0543	1.025	1.055
0.0212	0.0194	1.0549	1.034	1.064
0.0291	0.0276	1.0553	1.041	1.071
0.0382	0.0363	1.0559	1.048	1.079
0.0504	0.0479	1.0567	1.059	1.090
0.0596	0.0567	1.0572	1.067	1.098
0.0706	0.0671	1.0579	1.077	1.109
0.0784	0.0746	1.0584	1.087	1.119
308.15 K				
ρ = 1.0411, η = 0.9314				
0.0094	0.0090	1.0456	0.932	1.005
0.0202	0.0194	1.0465	0.952	1.014
0.0289	0.0276	1.0473	0.977	1.024
0.0380	0.0363	1.0482	0.993	1.032
0.0501	0.0479	1.0495	1.024	1.038
0.0592	0.0567	1.0503	1.085	1.049
0.0702	0.0671	1.0513	1.123	1.057
0.0779	0.0746	1.0519	1.158	1.063
298.15 K				
NaBr in 30 % Water + 70 % 1,3-DO, MW = 102.90, ρ = 1.0492, η = 0.9716				
0.0117	0.0111	1.0556	0.993	1.022
0.0202	0.0192	1.0558	1.012	1.042
0.0302	0.0287	1.0571	1.017	1.047
0.0401	0.0381	1.0573	1.028	1.058
0.0520	0.0494	1.0582	1.037	1.068
0.0597	0.0567	1.0589	1.044	1.075
0.0700	0.0665	1.0595	1.056	1.087
0.0786	0.0747	1.0607	1.065	1.096
308.15 K				
ρ = 1.0411, η = 0.9314				
0.0116	0.0111	1.0456	0.982	1.055
0.0200	0.0192	1.0464	0.989	1.062
0.0299	0.0287	1.0473	0.997	1.071
0.0398	0.0381	1.0483	1.006	1.080
0.0498	0.0477	1.0491	1.014	1.088
0.0592	0.0567	1.0501	1.023	1.098
0.0694	0.0665	1.0510	1.032	1.108
0.0779	0.0747	1.0518	1.040	1.117
298.15 K				
KBr in 30 % Water + 70 % 1,3-DO, MW = 119.01, ρ = 1.0492, η = 0.9716				
0.0117	0.0111	1.0560	0.990	1.019
0.0189	0.0179	1.0566	1.001	1.031
0.0301	0.0280	1.0574	1.017	1.047
0.0410	0.0389	1.0583	1.035	1.065
0.0502	0.0477	1.0590	1.048	1.079
0.0592	0.0562	1.0597	1.060	1.092
0.0693	0.0659	1.0606	1.075	1.106
0.0786	0.0753	1.0613	1.088	1.121
308.15 K				
ρ = 1.0411, η = 0.9314				
0.0116	0.0111	1.0445	0.958	1.028
0.0187	0.0179	1.0454	0.959	1.029
0.0292	0.0280	1.0469	0.973	1.044
0.0406	0.0389	1.0485	0.986	1.058
0.0498	0.0477	1.0497	1.000	1.073
0.0587	0.0562	1.0508	1.012	1.086
0.0687	0.0659	1.0520	1.026	1.101
0.0786	0.0753	1.0531	1.037	1.113
298.15 K				
LiBr in 60 % Water + 40 % 1,3-DO, MW = 86.85, ρ = 1.0391, η = 1.3572				
0.0139	0.0126	1.0404	1.406	1.036
0.0175	0.0170	1.0406	1.418	1.044
0.0294	0.0283	1.0413	1.438	1.059
0.0407	0.0392	1.0420	1.453	1.070
0.0536	0.0516	1.0427	1.472	1.084
0.0603	0.0552	1.0431	1.481	1.091
0.0706	0.0681	1.0436	1.503	1.107
0.0799	0.0770	1.0441	1.516	1.117

Table 1 Continued

molarity (M) $\text{mol}\cdot\text{dm}^{-3}$	molality (m) $\text{mol}\cdot\text{kg}^{-1}$	$\rho\cdot 10^{-3}$ $\text{kg}\cdot\text{m}^{-3}$	η $\text{mPa}\cdot\text{s}$	η_r
308.15 K				
$\rho = 1.0307$, $\eta = 1.1328$				
0.0132	0.0126	1.0316	1.198	1.057
0.0178	0.0170	1.0318	1.213	1.071
0.0295	0.0283	1.0323	1.226	1.082
0.0408	0.0392	1.0329	1.240	1.094
0.0537	0.0516	1.0335	1.253	1.106
0.0574	0.0552	1.0337	1.258	1.110
0.0707	0.0681	1.0344	1.272	1.122
0.0800	0.0770	1.0350	1.282	1.132
298.15 K				
NaBr in 60 % Water + 40 % 1,3-DO, MW = 102.90, $\rho = 1.0391$, $\eta = 1.3572$				
0.0111	0.0109	1.0396	1.384	1.020
0.0199	0.0192	1.0404	1.402	1.033
0.0297	0.0286	1.0410	1.424	1.049
0.0390	0.0376	1.0416	1.441	1.061
0.0508	0.0490	1.0423	1.463	1.078
0.0628	0.0606	1.0431	1.483	1.093
0.0697	0.0672	1.0437	1.496	1.102
0.0764	0.0737	1.0443	1.510	1.112
308.15 K				
$\rho = 1.0307$, $\eta = 1.1328$				
0.0112	0.0109	1.0309	1.157	1.021
0.0197	0.0192	1.0316	1.173	1.035
0.0295	0.0286	1.0329	1.184	1.045
0.0387	0.0376	1.0338	1.195	1.055
0.0505	0.0490	1.0357	1.209	1.067
0.0624	0.0606	1.0359	1.225	1.081
0.0692	0.0672	1.0360	1.234	1.089
0.0758	0.0737	1.0366	1.242	1.097
298.15 K				
KBr in 60 % Water + 40 % 1,3-DO, MW = 119.01, $\rho = 1.0391$, $\eta = 1.3572$				
0.0105	0.0111	1.0397	1.383	1.019
0.0211	0.0203	1.0405	1.404	1.034
0.0309	0.0298	1.0413	1.426	1.050
0.0405	0.0390	1.0421	1.448	1.066
0.0514	0.0496	1.0430	1.472	1.084
0.0599	0.0578	1.0437	1.491	1.098
0.0696	0.0672	1.0446	1.512	1.114
0.0772	0.0745	1.0453	1.527	1.125
308.15 K				
$\rho = 1.0307$, $\eta = 1.1328$				
0.0114	0.0111	1.0307	1.156	1.020
0.0209	0.0203	1.0318	1.174	1.037
0.0306	0.0298	1.0327	1.192	1.052
0.0402	0.0390	1.0335	1.208	1.066
0.0510	0.0496	1.0344	1.225	1.082
0.0594	0.0578	1.0352	1.241	1.096
0.0691	0.0672	1.0359	1.256	1.108
0.0765	0.0745	1.0365	1.268	1.119
298.15 K				
LiBr in 90 % Water + 10 % 1,3-DO, MW = 86.85, $\rho = 1.0092$, $\eta = 0.9415$				
0.0136	0.0135	1.0100	1.006	1.069
0.0192	0.0190	1.0103	1.013	1.076
0.0300	0.0249	1.0109	1.024	1.088
0.0425	0.0426	1.0116	1.049	1.115
0.0492	0.0488	1.0119	1.057	1.123
0.0621	0.0626	1.0125	1.072	1.139
0.0688	0.0695	1.0128	1.081	1.149
0.0791	0.0786	1.0133	1.093	1.161
308.15 K				
$\rho = 1.0033$, $\eta = 0.8220$				
0.0132	0.0135	1.0039	0.971	1.181
0.0191	0.0190	1.0042	0.981	1.193
0.0249	0.0249	1.0045	0.988	1.203
0.0436	0.0426	1.0055	1.008	1.226
0.0489	0.0488	1.0059	1.015	1.235
0.0626	0.0626	1.0067	1.029	1.252
0.0696	0.0695	1.0070	1.035	1.260
0.0787	0.0786	1.0076	1.043	1.269

Table 1 Continued

molarity (M) mol·dm^{-3}	molality (m) mol·kg^{-1}	$\rho\cdot10^{-3}$ kg·m^{-3}	η mPa·s	η_r
298.15 K				
NaBr in 90 % Water + 10 % 1,3-DO, MW = 102.90, $\rho = 1.0092$, $\eta = 0.9415$				
0.0109	0.0092	1.0104	1.111	1.180
0.0198	0.0196	1.0110	1.128	1.199
0.0296	0.0293	1.0118	1.145	1.216
0.0394	0.0391	1.0125	1.161	1.233
0.0506	0.0508	1.0134	1.180	1.253
0.0599	0.0594	1.0141	1.194	1.268
0.0716	0.0693	1.0151	1.209	1.285
0.0787	0.0734	1.0157	1.215	1.291
308.15 K				
$\rho = 1.0033$, $\eta = 0.8220$				
0.0092	0.0092	1.0041	1.058	1.201
0.0197	0.0196	1.0047	1.076	1.220
0.0294	0.0293	1.0053	1.089	1.235
0.0392	0.0391	1.0064	1.103	1.251
0.0509	0.0508	1.0070	1.116	1.266
0.0595	0.0594	1.0073	1.128	1.279
0.0694	0.0693	1.0088	1.140	1.293
0.0735	0.0734	1.0092	1.144	1.298
298.15 K				
KBr in 90 % Water + 10 % 1,3-DO, MW = 119.01, $\rho = 1.0092$, $\eta = 0.9415$				
0.0072	0.0091	1.0095	0.985	1.047
0.0171	0.0198	1.0105	1.002	1.065
0.0288	0.0298	1.0115	1.018	1.081
0.0406	0.0403	1.0126	1.035	1.099
0.0486	0.0500	1.0136	1.050	1.116
0.0595	0.0608	1.0145	1.068	1.135
0.0671	0.0694	1.0152	1.083	1.150
0.0800	0.0792	1.0162	1.099	1.168
308.15 K				
$\rho = 1.0033$, $\eta = 0.8220$				
0.0095	0.0091	1.0040	0.947	1.152
0.0192	0.0198	1.0049	0.965	1.174
0.0310	0.0298	1.0058	0.978	1.190
0.0419	0.0403	1.0068	0.993	1.208
0.0519	0.0500	1.0077	1.006	1.224
0.0630	0.0608	1.0087	1.022	1.244
0.0719	0.0694	1.0095	1.035	1.259
0.0819	0.0792	1.0104	1.049	1.277

the plotting of $\eta_r - 1$ versus c is shown in Figures 1 and 2. This type of characteristic is an indication of the applicability of eq 2. The values of B obtained from the slopes of the given plots are given in Table 2.

The relative viscosity data of solutions have been fitted to the following equations:

(i) Moulik equation:[21]

$$\eta_r^{\,2} = M + k^1 c^2 \tag{3}$$

where M and k^1 are constants. The applicability of the Moulik equation is shown by the linear plots of $\eta_r^{\,2}$ versus c^2 (Figures 3 and 4).

(ii) Breslau−Miller equation:[20]

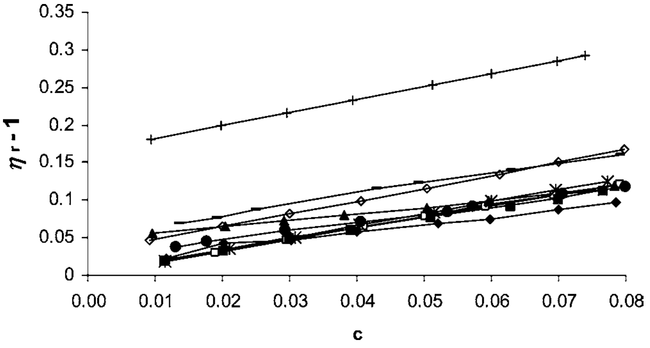

Figure 1. Plots of $\eta_r - 1$ vs c for different alkali bromides at 298.15 K. LiBr: ▲, 30 %; ●, 60 %; −, 90 %. NaBr: ♦, 30 %; ■, 60 %; +, 90 %. KBr: □, 30 %; ◊, 60 %; ∗, 90 %.

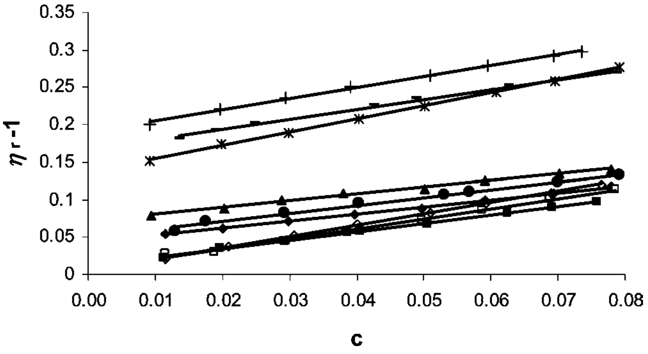

Figure 2. Plots of $\eta_r - 1$ vs c for different alkali bromides at 308.15 K. LiBr: ▲, 30 %; ●, 60 %; −, 90 %. NaBr: ♦, 30 %; ■, 60 %; +, 90 %. KBr: □, 30 %; ◊, 60 %; ∗, 90 %.

Table 2. *B*-Values from Various Equations at Different Temperatures

		temperatures					
		298.15 K			308.15 K		
salt		B^a	B^b	B^c	B^b	B^b	B^c
LiBr	30 % water + 70 % 1,3-DO	0.930	1.061	0.960	0.904	1.031	0.572
	60 % water + 40 % 1,3-DO	1.182	1.353	1.902	1.053	1.203	0.961
	90 % water + 10 % 1,3-DO	1.453	1.667	0.910	1.328	1.522	0.216
NaBr	30 % water + 70 % 1,3-DO	1.023	1.168	2.602	0.935	1.067	1.108
	60 % water + 40 % 1,3-DO	1.394	1.599	3.167	1.133	1.297	2.869
	90 % water + 10 % 1,3-DO	1.486	1.706	0.165	1.655	1.902	0.241
KBr	30 % water + 70 % 1,3-DO	1.518	1.743	3.451	1.341	1.538	2.969
	60 % water + 40 % 1,3-DO	1.611	1.851	3.453	1.508	1.731	3.116
	90 % water + 10 % 1,3-DO	1.715	1.971	1.309	1.758	2.021	2.445

[a] *B*-values obtained from the Jones−Dole equation. [b] *B*-values obtained from the Breslau−Miller equation. [c] *B*-values obtained from the Vand equation.

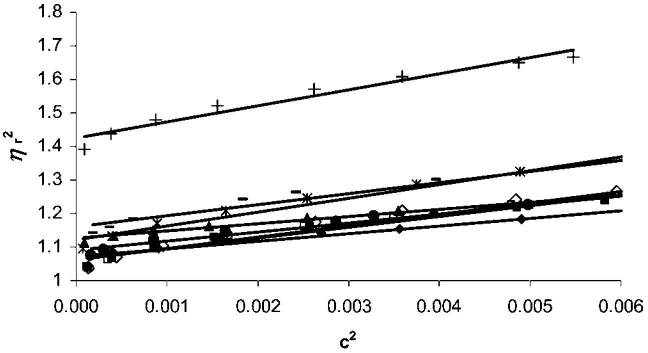

Figure 3. Plots of η_r^2 vs c^2 for different alkali bromides at 298.15 K. LiBr: ▲, 30 %; ●, 60 %; −, 90 %. NaBr: ♦, 30 %; ■, 60 %; +, 90 %. KBr: □, 30 %; ◇, 60 %; ∗, 90 %.

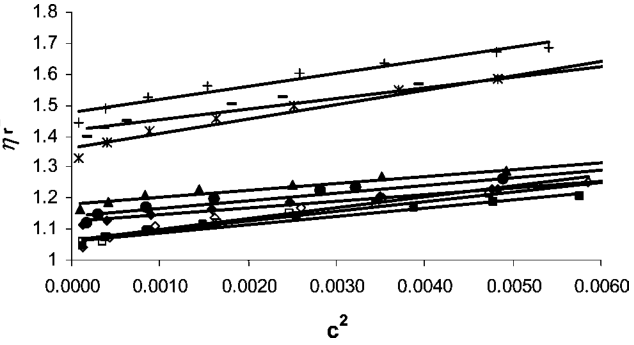

Figure 4. Plots of η_r^2 vs c^2 for different alkali bromides at 308.15 K. LiBr: ▲, 30 %; ●, 60 %; −, 90 %. NaBr: ♦, 30 %; ■, 60 %; +, 90 %. KBr: □, 30 %; ◇, 60 %; ∗, 90 %.

$$V_e = (-2.5c + [(2.5c)^2 - 4(10.05c^2)(1 - \eta r)]^{1/2})/2(10.05)c^2 \tag{4}$$

where V_e is the average effective rigid molar volume. The values of V_e thus obtained were used for calculating the *B*-coefficient, employing the following relation:

$$B = 2.90V_e - 0.018 \tag{5}$$

(iii) Vand equation:[22]

$$\ln \eta_r = 2.5\Phi/(1 - \kappa\Phi) \tag{6}$$

where Φ is the volume fraction and *k* the generalized particle interaction coefficient. Substituting $\Phi = cv'$ and rearranging eq 6, we have,

$$c/\log \eta_r = (2.303/2.5v') - (2.303kc/2.5) \tag{7}$$

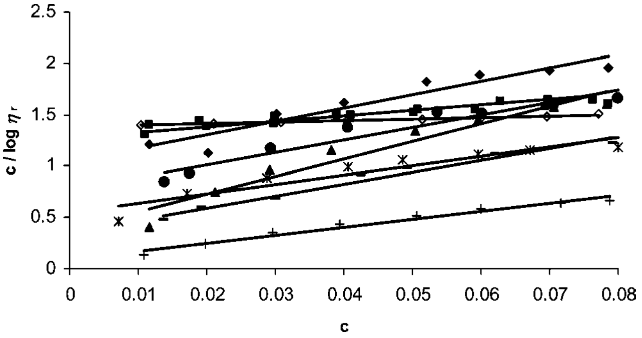

Figure 5. Plots of c/log η_r vs c for different alkali bromides at 298.15 K. LiBr: ▲, 30 %; ●, 60 %; −, 90 %. NaBr: ♦, 30 %; ■, 60 %; +, 90 %. KBr: □, 30 %; ◇, 60 %; ∗, 90 %.

where v' is the effective flowing volume. v' has been obtained from the intercept of the linear plots of c/log η_r versus c, shown in Figure 5. *B*-coefficients were calculated from the relation,

$$B = 2.5v' \tag{8}$$

B-values thus obtained from eqs 2, 5, and 8 are listed in Table 2 for comparison. *B*-values obtained from the Jones−Dole and Breslau−Miller equations are more or less near to each other; however, mostly they differ considerably from those obtained from the Vand eq 7. This discrepancy, in regards to the magnitude of *B*, may be attributed to the concentration dependence validity of the viscosity equation and variable ion−solvent interaction with the varying concentration of electrolyte. The familiar viscosity *B*-coefficients and the related ionic molar contribution to the free energy, enthalpy, and entropy of activation are re-examined on the compensation principle by Feakins et al.[23] Particular attention is paid to the nature of the solvent in the transition state. If compensation between enthalpic and entropic contributions from solute-induced structural changes occurs in both ground and transition-state solvents, then the free energy of activation, and hence *B*, cannot be influenced by changes in the structure of the solvents of the type proposed.[24]

From Table 2 it is seen that the *B*-values always increase from lithium salts to potassium salts in all compositions. This fact indicates that the larger cations are scarcely solvated in this solvent medium.

Table 2 also shows that *B*-values are positive and decrease with a rise in temperature (negative d*B*/d*T*), suggesting[25] a structure-promoting tendency of alkali metal bromides. The positive *B*-values and negative d*B*/d*T* show the absence of a firm layer of solvent molecules around the ion in their cosphere.

Conclusion

The investigation thus indicates that all of the salts studied here exhibit strong ionic interactions and they have structure-promoting tendencies in these solvent systems.[26]

Glossary

List of Symbols

M	concentration in molarity
m	concentration in molality
V_e	average effective rigid molar volume
υ'	effective flowing volume

Greek Letters

ρ	density
η_r	relative viscosity
η	viscosity
η_0	viscosity of solvent

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