

# Partial Molar Volumes, Viscosity *B*-Coefficients, and Adiabatic Compressibilities of Sodium Molybdate in Aqueous 1,3-Dioxolane Mixtures from 303.15 to 323.15 K

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**Abstract** Partial molar volumes ( $V_{\phi}^0$ ) and viscosity *B*-coefficients of sodium molybdate in 1,3-dioxolane + water mixtures have been determined from solution density and viscosity measurements at 303.15, 313.15, and 323.15 K and at various electrolyte concentrations. Also, the adiabatic compressibility of different solutions has been determined from the measurement of sound speeds at 303.15 K. The experimental density data were evaluated by the Masson equation, and the derived parameters were interpreted in terms of ion–solvent and ion–ion interactions. The viscosity data have been analyzed using the Jones–Dole equation, and the derived parameters, *B* and *A*, have also been interpreted in terms of ion–solvent and ion–ion interactions, respectively. The structure- making or breaking capacity of the electrolyte under investigation has been discussed in terms of the sign of  $(\delta^2 V_{\phi}^0 / \delta T^2)_P$ . The compressibility data obtained from sound speeds of different solutions indicate the electrostriction of the solvent molecules around the ions.

**Keywords** Adiabatic compressibility · Density · Electrostriction · Ion–solvent and ion–ion interactions · Sodium molybdate · Viscosity · 1,3-dioxolane

## 1 Introduction

Partial molar volumes of electrolytes provide valuable information about 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. Measurement of sound speeds of the solutions also helps in this regard. The increasing use of cyclic diethers and their aqueous mixtures in many industrial processes

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such as pharmaceuticals and cosmetics has greatly stimulated the need for extensive information on their various properties. 1,3-dioxolane is an important solvent for a number of separation processes, solution studies, Mannich reactions, and as an electrolyte in batteries [4–6]. Studies on systems containing sodium molybdate in aqueous binary mixtures of cyclic diethers are scarce in literature. Therefore in this paper, an attempt has been made to reveal the nature of various types of interactions prevailing in solutions of sodium molybdate in 1,3-dioxolane + water mixtures from volumetric, viscometric, and acoustic measurements. The partial molar volumes have been fitted with a polynomial equation in terms of temperature, and the structure-making or breaking capacities of the electrolyte have been inferred from the sign of  $(\delta^2 V_\phi^0 / \delta T^2)_P$ .

## 2 Experimental

1,3-Dioxolane (LR) was purified by standard methods. It was refluxed with  $\text{PbO}_2$  and then fractionally distilled after the addition of xylene [7]. The solvent finally obtained after purification was 99.9% pure as checked by gas chromatography.

Doubly distilled, degassed, and deionized water with a specific conductance of  $1 \times 10^{-6} \Omega^{-1} \cdot \text{cm}^{-1}$  was used. Sodium molybdate was procured from Merck, India. It was purified by re-crystallizing twice from conductivity water and was dried in a vacuum desiccator over  $\text{P}_2\text{O}_5$  for 24 h before use. The experimental values of densities ( $\rho_0$ ) and viscosities ( $\eta_0$ ) of pure 1,3-dioxolane and different aqueous 1,3-dioxolane mixtures at 303.15, 313.15, and 323.15 K are reported in Table 1 and compared with literature values [7, 8] whenever available.

The densities were measured with an Ostwald-Sprengel type pycnometer having a bulb volume of  $25 \text{ cm}^3$  and an internal diameter 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. The pycnometer with the test solution was equilibrated in a water bath maintained within  $\pm 0.01 \text{ K}$  of the desired temperatures. The pycnometer was then removed from the thermostatic bath, properly dried, and weighed. Adequate precautions were taken to avoid evaporation losses during the time of actual measurements. An average of triplicate measurements was taken into account. The mass measurements, accurate to  $\pm 0.01 \text{ mg}$ , were made on a digital electronic analytical balance (Mettler, AG 285, Switzerland). The total uncertainty of the density is  $\pm 0.0001 \text{ g} \cdot \text{cm}^{-3}$ , and that of the temperature is  $\pm 0.01 \text{ K}$ .

The viscosity was measured by means of a suspended Ubbelohde type viscometer, which was calibrated at 303.15, 313.15, and 323.15 K with triply distilled water and purified methanol. The flow times were accurate to  $\pm 0.1 \text{ s}$ , and the precision of the viscosity measurements, based on our work on several pure liquids, was  $\pm 0.003 \text{ mPa} \cdot \text{s}$ ; the total uncertainty in the measured viscosities was estimated to be  $\pm 0.05\%$ . Sound speeds were determined with an uncertainty of 0.3% using a single-crystal variable-path ultrasonic interferometer (Mittal Enterprises, New Delhi, India) operating at 2 MHz which was calibrated with water, methanol, and benzene at 303.15 K, as described in detail elsewhere [9, 10]. Details of the methods and techniques of density, viscosity, and sound speed measurements have been described elsewhere [7, 10–12].

**Table 1** Physical properties of pure 1,3-Dioxolane and 1,3-Dioxolane + water mixtures at different temperatures

Temperature (K)	$\rho$ ( $\text{g} \cdot \text{cm}^{-3}$ )		$\eta$ ( $\text{mPa} \cdot \text{s}$ )	
	This work	Literature	This work	Literature
10 mass% of 1,3-dioxolane + water				
303.15	1.0072	–	0.8133	–
313.15	1.0012	–	0.6555	–
323.15	0.9977	–	0.5673	–
20 mass% of 1,3-dioxolane + water				
303.15	1.0185	–	0.8766	–
313.15	1.0125	–	0.7152	–
323.15	1.0076	–	0.6053	–
30 mass% of 1,3-dioxolane + water				
303.15	1.0280	–	0.9304	–
313.15	1.0210	–	0.7618	–
323.15	1.0135	–	0.6297	–
Pure 1,3-dioxolane				
303.15	1.0518	1.0518 [7]	0.5486	0.5486 [7]
313.15	1.0422	1.0399 [8]	0.4986	0.4985 [8]
323.15	1.0311	–	0.4183	–

The electrolyte solutions studied here were prepared by mass and the conversion of molality to molarity was accomplished [13] using experimental density values. The experimental values of concentrations ( $c$ ), densities ( $\rho$ ), viscosities ( $\eta$ ), and derived parameters at various temperatures are reported in Table 2.

### 3 Discussion

The apparent molar volumes ( $V_\phi$ ) were determined from the solution densities using the following equation [3]:

$$V_\phi = \frac{M}{\rho_0} - \frac{1,000(\rho - \rho_0)}{c\rho_0} \quad (1)$$

where  $M$  is the molar mass of the solute,  $c$  is the molarity of the solution,  $\rho_0$  and  $\rho$  are the densities of the solvent and solution, respectively. The limiting apparent molar volumes ( $V_\phi^0$ ) were calculated applying a least-squares technique to the plots of  $V_\phi$  vs.  $\sqrt{c}$  using the following Masson equation [14]:

$$V_\phi = V_\phi^0 + S_V^* \sqrt{c} \quad (2)$$

**Table 2** Concentration ( $c$ ), density ( $\rho$ ), viscosity ( $\eta$ ), apparent molar volume ( $V_\phi$ ), and  $(\eta/\eta_0 - 1)/\sqrt{c}$  of sodium molybdate in different aqueous 1,3-Dioxolane mixtures at different temperatures

$c$ (mol · dm <sup>-3</sup> )	$\rho$ (g · cm <sup>-3</sup> )	$\eta$ (mPa · s)	$V_\phi$ (cm <sup>3</sup> · mol <sup>-1</sup> )	$\frac{(\eta/\eta_0 - 1)}{\sqrt{c}}$
10 mass% of 1,3-dioxolane + water				
$T = 303.15$ K				
0.0238	1.0114	0.8248	65.8457	0.0920
0.0318	1.0128	0.8297	65.3786	0.1133
0.0556	1.0170	0.8412	64.5071	0.1455
0.0715	1.0199	0.8499	64.1455	0.1683
0.0874	1.0227	0.8582	63.8018	0.1867
0.0993	1.0249	0.8662	63.4469	0.2063
$T = 313.15$ K				
0.0237	1.0053	0.6599	68.8716	0.0435
0.0316	1.0067	0.6639	67.8180	0.0720
0.0553	1.0110	0.6769	64.6572	0.1391
0.0711	1.0139	0.6864	63.2524	0.1766
0.0869	1.0169	0.6967	61.2091	0.2134
0.0987	1.0191	0.7052	60.5197	0.2416
$T = 323.15$ K				
0.0236	1.0016	0.5710	76.8726	0.0423
0.0316	1.0029	0.5746	77.5715	0.0726
0.0551	1.0070	0.5871	73.3346	0.1487
0.0708	1.0098	0.5960	71.2098	0.1902
0.0866	1.0127	0.6049	68.8983	0.2250
0.0983	1.0149	0.6115	67.1298	0.2485
20 mass% of 1,3-dioxolane + water				
$T = 303.15$ K				
0.0242	1.0226	0.8893	71.2111	0.0932
0.0323	1.0240	0.8963	70.3695	0.1252
0.0565	1.0284	0.9216	65.5167	0.2158
0.0727	1.0313	0.9380	64.6873	0.2600
0.0888	1.0344	0.9557	61.7535	0.3030
0.1009	1.0366	0.9696	61.4281	0.3340
$T = 313.15$ K				
0.0240	1.0162	0.7198	86.6996	0.0413
0.0322	1.0176	0.7249	82.5332	0.0758
0.0562	1.0217	0.7423	77.2829	0.1599
0.0722	1.0245	0.7555	74.8099	0.2099
0.0882	1.0274	0.7700	72.1143	0.2579
0.1003	1.0296	0.7816	70.5792	0.2930

**Table 2** continued

<i>T</i> = 323.15 K				
0.0239	1.0109	0.6069	103.0912	0.0175
0.032	1.0122	0.6118	97.4593	0.0598
0.0558	1.0159	0.6262	92.5015	0.1461
0.0718	1.0185	0.6370	89.4595	0.1952
0.0877	1.0211	0.6495	87.3523	0.2465
0.0997	1.0233	0.6586	83.8404	0.2790
30 mass% of 1,3-dioxolane + water				
<i>T</i> = 303.15 K				
0.0249	1.0319	0.9335	82.9995	0.0209
0.0320	1.0332	0.9387	77.2860	0.0498
0.0560	1.0373	0.9614	73.8118	0.1407
0.0720	1.0402	0.9786	70.5307	0.1934
0.0880	1.0430	0.9963	69.5481	0.2387
0.1000	1.0452	1.0079	68.0447	0.2634
<i>T</i> = 313.15 K				
0.0248	1.0251	0.7622	75.0513	0.0032
0.0318	1.0264	0.7654	70.6549	0.0264
0.0557	1.0311	0.7831	59.3746	0.1186
0.0716	1.0344	0.7961	53.6720	0.1685
0.0874	1.0379	0.8106	47.5868	0.2168
0.0994	1.0404	0.8218	45.8168	0.2499
<i>T</i> = 323.15 K				
0.0246	1.0156	0.6301	154.4984	0.0045
0.0304	1.0163	0.6328	147.8488	0.0287
0.0553	1.0192	0.6499	137.0260	0.1365
0.0711	1.0216	0.6620	126.3206	0.1924
0.0868	1.0240	0.6731	119.3708	0.2342
0.0988	1.0260	0.6828	113.8942	0.2685

where  $V_{\phi}^0$  is the partial molar volume at infinite dilution and  $S_V^*$  is the experimental slope. The plots of  $V_{\phi}$  against the square root of the molar concentration ( $\sqrt{c}$ ) were found to be linear with negative slopes. The values of  $V_{\phi}^0$  and  $S_V^*$  along with the standard errors are reported in Table 3. Table 3 shows that  $V_{\phi}^0$  values are generally positive and increase with a rise in both the temperature and amount of 1,3-dioxolane in the mixtures. This indicates the presence of strong ion–solvent interactions and these interactions are further strengthened at higher temperatures and higher concentrations of 1,3-dioxolane in the mixtures, suggesting larger electrostriction at higher temperatures and in enhanced amounts of the cyclic diether. A perusal of Table 3 also reveals that  $S_V^*$  values are negative for all the solutions and at all the experimental temperatures, and  $S_V^*$  values decrease as the temperature of the solutions and amount

**Table 3** Limiting partial molar volume ( $V_{\phi}^0$ ) and experimental slope ( $S_V^*$ ) for sodium molybdate in different aqueous 1,3-dioxolane mixtures at different temperatures (standard errors are given in parentheses)

Mass% of 1,3-dioxolane	$V_{\phi}^0$ ( $\text{cm}^3 \cdot \text{mol}^{-1}$ )			$S_V^*$ ( $\text{cm}^2 \cdot \text{dm}^{1/2} \cdot \text{mol}^{-3/2}$ )		
	303.15 K	313.15 K	323.15 K	303.15 K	313.15 K	323.15 K
10	68.0 ( $\pm 0.02$ )	77.2 ( $\pm 0.01$ )	88.1 ( $\pm 0.01$ )	-14.4 ( $\pm 0.02$ )	-53.2 ( $\pm 0.03$ )	-64.8 ( $\pm 0.02$ )
20	81.4 ( $\pm 0.02$ )	100.6 ( $\pm 0.01$ )	118.4 ( $\pm 0.03$ )	-64.0 ( $\pm 0.04$ )	-95.9 ( $\pm 0.01$ )	-108.0 ( $\pm 0.01$ )
30	94.5 ( $\pm 0.01$ )	104.5 ( $\pm 0.02$ )	193.3 ( $\pm 0.01$ )	-85.8 ( $\pm 0.03$ )	-189.5 ( $\pm 0.04$ )	-250.5 ( $\pm 0.02$ )

**Table 4** Values of various coefficients of Eq. (3) for sodium molybdate in different aqueous 1,3-dioxolane mixtures

Mass% of 1,3-dioxolane	$a_0$	$a_1$	$a_2$
	( $\text{cm}^3 \cdot \text{mol}^{-1}$ )	( $\text{cm}^3 \cdot \text{mol}^{-1} \text{K}^{-1}$ )	( $\text{cm}^3 \cdot \text{mol}^{-1} \cdot \text{K}^{-2}$ )
10	596.02	-4.32	0.008
20	-1165.17	6.23	-0.007
30	37194.43	-241.82	0.394

of 1,3-dioxolane in the mixtures increases. Since  $S_V^*$  is a measure of ion–ion interactions, the results indicate the presence of weak ion–ion interactions in the solutions at all experimental temperatures, and these interactions further decrease with a rise in temperature and concentration of 1,3-dioxolane in the mixtures. In other words, it may be said that the solvation of electrolyte/ions increases with an increase of 1,3-dioxolane content in water. This is probably due to more effective and efficient thermal agitation at higher temperatures and moderate dielectric constants [15] of the aqueous 1,3-dioxolane mixtures, resulting in diminishing ion–ion interactions (ionic dissociation) [16]. A quantitative comparison of the magnitude of values shows  $V_{\phi}^0$  values are much greater in magnitude than  $S_V^*$  values, for all the solutions. This suggests that ion–solvent interactions dominate over ion–ion interactions in all the solutions and at all experimental temperatures.

The partial molar volumes ( $V_{\phi}^0$ ) were fitted to a polynomial of the following type in terms of absolute temperature ( $T$ ):

$$V_{\phi}^0 = a_0 + a_1T + a_2T^2 \quad (3)$$

Values of the coefficients  $a_0$ ,  $a_1$ , and  $a_2$  of the above equation for different sodium molybdate solutions are reported in Table 4.

**Table 5** Limiting partial molar expansibilities ( $\phi_E^0$ ) for sodium molybdate in various aqueous 1,3-dioxolane mixtures at different temperatures

Mass% of 1,3-dioxolane	$\phi_E^0$ (cm <sup>3</sup> · mol <sup>-1</sup> · K <sup>-1</sup> )			$\left(\frac{\delta\phi_E^0}{\delta T}\right)_P$ (cm <sup>3</sup> · mol <sup>-1</sup> · K <sup>-2</sup> )
	303.15 K	313.15 K	323.15 K	
10	0.530	0.690	0.850	Small positive
20	1.986	1.846	1.706	Negative
30	-2.938	4.942	12.822	Positive

The partial molar expansibilities ( $\phi_E^0$ ) can be obtained by the following equation:

$$\phi_E^0 = \left(\delta V_\phi^0 / \delta T\right)_P = a_1 + 2a_2 T \quad (4)$$

The values of  $\phi_E^0$  for different solutions of the studied electrolyte at 303.15, 313.15, and 323.15 K are reported in Table 5.

According to Hepler [17], the sign of  $(\delta^2 V_\phi^0 / \delta T^2)_P$  is a better criterion than  $S_V^*$  in characterizing the long-range structure-making and breaking capacity of the electrolytes in solution. The general thermodynamic expression is as follows:

$$(\delta C_P / \delta P)_T = - \left(\delta^2 V_\phi^0 / \delta T^2\right)_P \quad (5)$$

If the sign of  $(\delta^2 V_\phi^0 / \delta T^2)_P$  is small positive or negative, the electrolyte is a structure breaker; otherwise, it is a structure maker. As evident from Table 5, the electrolyte under investigation is predominantly a structure breaker in all the experimental solvent mixtures except for 30 mass% of aqueous 1,3-dioxolane solutions. This may be attributed to the absence of caging or packing effects [16, 18] at a lower concentration of 1,3-dioxolane.

The viscosity data of solutions for sodium molybdate in 10, 20, and 30 mass% of 1,3-dioxolane + water mixtures have been analyzed using the Jones–Dole [19] equation;

$$\frac{\eta}{\eta_0} = 1 + A\sqrt{c} + Bc \quad (6)$$

$$\frac{(\eta/\eta_0 - 1)}{\sqrt{c}} = A + B\sqrt{c} \quad (7)$$

where  $\eta_0$  and  $\eta$  are the viscosities of solvent/solvent mixtures and solution, respectively.  $c$  is the molar concentration of a solution,  $A$  and  $B$  are the Jones–Dole constants estimated by a least-squares method and reported in Table 6.

**Table 6** Values of Jones–Dole  $A$  and  $B$  coefficients for sodium molybdate in different aqueous 1,3-dioxolane mixtures at different temperatures (standard errors are given in parentheses)

Mass% of 1,3-dioxolane	$A$ ( $\text{cm}^3/2 \cdot \text{mol}^{-1/2}$ )			$B$ ( $\text{cm}^3 \cdot \text{mol}^{-1}$ )		
	303.15 K	313.15 K	323.15 K	303.15 K	313.15 K	323.15 K
10	−0.01 (±0.01)	−0.15 (±0.03)	−0.16 (±0.02)	0.68 (±0.01)	1.22 (±0.02)	1.30 (±0.04)
20	−0.14 (±0.01)	−0.20 (±0.03)	−0.23 (±0.02)	1.49 (±0.03)	1.55 (±0.01)	1.60 (±0.02)
30	−0.23 (±0.01)	−0.25 (±0.04)	−0.26 (±0.03)	1.56 (±0.05)	1.58 (±0.02)	1.69 (±0.01)

A perusal of Table 6 shows that the values of the  $A$  coefficient are negative for all the solutions under investigation at all experimental temperatures. These results indicate the presence of weak ion–ion interactions, and these interactions further decrease with an increase of the amount of 1,3-dioxolane to the mixture.

The  $B$ -coefficient [20] reflects the effects of ion–solvent interactions on the solution viscosity. The viscosity  $B$ -coefficient is a valuable tool to provide information concerning the solvation of the solutes and their effects on the structure of the solvent in the local vicinity of the solute molecules. Table 6 illustrates that the values of the  $B$ -coefficient of sodium molybdate in the studied solvent systems are positive, thereby suggesting the presence of strong ion–solvent interactions and these types of interactions are strengthened with an increase in both the temperature and concentration of 1,3-dioxolane in the mixtures. These conclusions are in excellent agreement with those drawn from  $V_\phi^0$  values discussed earlier.

It has been reported in a number of studies [21, 22] that  $dB/dT$  is a better criterion for determining the structure-making breaking nature of any solute rather than the  $B$ -coefficient. It is found from Table 6 that the values of the  $B$ -coefficients increase with a rise in temperature (positive  $dB/dT$ ), suggesting the structure-breaking tendency of sodium molybdate in the studied solvent systems. A similar result was reported in a study [23] for the case of the viscosity of some salts in propionic acid + ethanol mixtures.

The adiabatic compressibility ( $\beta$ ) was evaluated from the following equation:

$$\beta = u^{-2} \rho^{-1} \quad (8)$$

where  $\rho$  is the solution density and  $u$  is the sound speed of the solution. The partial molal adiabatic compressibility ( $\phi_K$ ) of the solutions was determined from the relation

$$\phi_K = \frac{M\beta}{\rho_0} + \frac{1,000(\beta\rho_0 - \beta_0\rho)}{m\rho\rho_0} \quad (9)$$

where  $\beta_0$  and  $\beta$  are the adiabatic compressibilities of the solvent and solution, respectively, and  $m$  is the molality of the solution. The limiting partial molal adiabatic



**Table 7** Molality ( $m$ ), density ( $\rho$ ), sound speed ( $u$ ), adiabatic compressibility ( $\beta$ ), partial molal adiabatic compressibility ( $\phi_K$ ), limiting partial adiabatic compressibility ( $\phi_K^0$ ), and experimental slope ( $S_K^*$ ) of sodium molybdate in different aqueous 1,3-dioxolane mixtures at 303.15 K

$m$ (mol·kg <sup>-1</sup> )	$u$ (m·s <sup>-1</sup> )	$\beta \times 10^{10}$ (Pa <sup>-1</sup> )	$\phi_K \times 10^{10}$ (m <sup>3</sup> ·mol <sup>-1</sup> ·Pa <sup>-1</sup> )	$\phi_K^0 \times 10^{10a}$ (m <sup>3</sup> ·mol <sup>-1</sup> ·Pa <sup>-1</sup> )	$S_K^* \times 10^{10a}$ (m <sup>3</sup> ·mol <sup>-3/2</sup> ·Pa <sup>-1</sup> ·kg <sup>1/2</sup> )
10 mass% of 1,3-dioxolane + water					
0.0237	1510.0	4.3362	-1.106		
0.0316	1512.2	4.3175	-1.345		
0.0554	1519.9	4.2565	-1.737	0.05	-7.65
0.0713	1527.1	4.2044	-2.017	(±0.02)	(±0.04)
0.0872	1535.3	4.1480	-2.241		
0.0992	1541.0	4.1088	-2.332		
20 mass% of 1,3-dioxolane + water					
0.0238	1541.7	4.1141	-0.459		
0.0318	1542.8	4.1026	-0.627		
0.0557	1548.6	4.0546	-1.098	0.85	-8.35
0.0717	1554.7	4.0114	-1.386	(±0.03)	(±0.01)
0.0877	1561.9	3.9628	-1.640		
0.0997	1567.8	3.9248	-1.787		
30 mass% of 1,3-dioxolane + water					
0.0243	1543.4	4.0681	-0.664		
0.0312	1545.3	4.0531	-0.933		
0.0547	1555.6	3.9840	-1.639	1.18	-11.91
0.0704	1564.3	3.9288	-1.983	(±0.03)	(±0.012)
0.0861	1574.8	3.8659	-2.286		
0.0979	1585.1	3.8077	-2.563		

<sup>a</sup>Standard errors are given in parentheses

compressibility ( $\phi_K^0$ ) and the experimental slope ( $S_K^*$ ) were obtained by fitting  $\phi_K$  against the square root of molality of the electrolyte ( $\sqrt{m}$ ) by the method of least squares:

$$\phi_K = \phi_K^0 + S_K^* \sqrt{m} \quad (10)$$

The values of  $m$ ,  $u$ ,  $\beta$ ,  $\phi_K$ ,  $\phi_K^0$ , and  $S_K^*$  are presented in Table 7. A perusal of Table 7 shows that  $\phi_K^0$  values are positive and increase whereas  $S_K^*$  values are negative and decrease for all the solutions with an increase in both the amount of 1,3-dioxane in the mixtures and in the temperature. Since the values of  $\phi_K^0$  and  $S_K^*$  are measures of the ion–solvent and ion–ion interactions, respectively, the results are in good agreement with those drawn from the values of  $V_\phi^0$  and  $S_V^*$  discussed earlier.

## 4 Conclusion

In summary, this study reveals that ion–solvent interactions are predominant over ion–ion interactions for sodium molybdate in different aqueous binary mixtures of 1,3-dioxolane at all experimental temperatures. Also, the electrolyte under study was found to act as a structure breaker in the solvent mixtures studied.

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