# Density, Viscosity, and Electrical Conductivity Measurements on the Ternary System $H_2O + C_2H_5OH + LiCl$ over the Entire Ranges of Solvent Composition and LiCl Solubility from (−5 to +50) °C

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Systematic measurements of density, viscosity, and electrical conductivity on the ternary system water + ethanol + lithium chloride over entire ranges of solvent composition and LiCl solubility from (−5 to +50) °C have been carried out. The temperature-independent aquamolality scale, $\boldsymbol{m}$ (LiCl moles per 55.5 moles of solvent) has been used to justify data comparison not only at different temperatures but also for the solutions in various solvent mixtures. Density isotherms (fitted to second-order polynomials in $\boldsymbol{m}$) provided a composition-dependent density index, $\mathbf{g}(x_1)$, which can conveniently be used to determine the density at any desired LiCl concentration, mixture composition, and temperature. Excess molar volumes ($V^E$) of the solvent mixtures have been calculated over the whole temperature range, and minima of all the $V^E$ isotherms at the same water mole fraction, $x_1 = 0.6$, suggest that no significant structural rearrangement takes place in the solvent mixtures upon temperature variation. From the density data, the "temperature average coefficient of thermal expansion" ($\beta$) is also determined for all the samples. Fitting the viscosity ($\eta$) isotherms to an extended Jones-Dole-type cubic equation in $\boldsymbol{m}$ provided coefficients related to various interactions taking place in the solutions. The temperature dependence of the entire $\eta$ data can well be explained by the Arrhenius-type equation involving single flow activation energies ($E_a$) that always increased with LiCl concentration for a given solvent but exhibited a sharper rise for the ethanol-rich mixtures; $E_a$ values tend to pass through maxima at $x_1 \approx 0.6$. In all the cases, electrical conductivity ($\kappa$) plotted against $\boldsymbol{m}$ exhibits maxima which shift from 4.5$\boldsymbol{m}$ to ~6$\boldsymbol{m}$ in the water-rich mixtures, and exhibition of the $\kappa$-maxima has been explained with a simple model according to which $\kappa$ at any given $\boldsymbol{m}$ is comprised of two sets of the oppositely acting terms, namely $\kappa_{up}$ and $\kappa_{down}$. Since all the $\kappa$ plots fit very well to third-order polynomials in $\boldsymbol{m}$ with negligibly small coefficients of the cubic terms, the other two terms involving $\boldsymbol{m}$ and $\boldsymbol{m}^2$ can be compared with $\kappa_{up}$ and $\kappa_{down}$, respectively. From temperature variation of molar conductivity, activation energies have been determined and compared with those of viscous flow.

## Introduction

Propanols and other lower alcohols are completely miscible with water in all proportions at ambient temperatures which allows preparation of their aqueous mixtures of continuously varying properties.[1–4] Structural properties of aqueous alcohols have remained the subject of many investigations.[5–8] Aqueous alcohols are also considered as interesting "solvent systems" for exhibiting relatively higher solubilities of different solutes including electrolytes.[9,10] By maintaining calculated amounts of alcohols as cosolvents in their aqueous mixtures, controlled precipitation of salts can be achieved.[11–14] The significance of the electrolyte solutions in aqueous alcohols has been reported in various investigations of biological interest as well.[15–17] Therefore, data collected on some of the basic properties like density, molar volume, viscosity, electrical conductivity, etc. for aqueous alcohol systems over wide ranges of solvent composition, electrolyte concentration, and temperature can be of great utility. Since water molecules act as a strong ligand[18,19] and tend to selectively solvate ions in the mixtures,[20–22] change of either mixture composition or electrolyte concentration or even temperature may have an influence on the above-mentioned basic properties of the ternary systems.

Besides densities of liquids, also their viscosities over a wide range around ambient temperature have immense importance in solving engineering problems related to heat or mass transfer and fluid dynamics.[23–25] Further, the viscosity variation with temperature and solvent composition are commonly employed as indices for obtaining useful information on structure and interactions within the liquid systems.[24,26,27] Temperature-dependent viscosity studies enable determination of flow activation energy, still another important parameter in chemical research.[28,29] Electrical conductance has always remained important in the investigation of electrolyte solutions, also for its significance toward understanding of the ion−ion, ion−solvent, and even solvent−solvent interactions.[30–32] Conductivity ($\kappa$) is known to depend on factors like temperature, solvent employed, nature of the electrolyte, and its concentration.[33,34] However, the molar conductivity ($\Lambda_m$) being a concentration normalized quantity should not depend on the electrolyte's concentration unless mutual interactions between the ions start operating.[35]

$H_2O + C_2H_5OH$ (*the mixture*) has shown appreciably high solubility for LiCl, while $H_2O + C_2H_5OH + LiCl$ (*the system*) could sustain up to quite low temperatures without salting out; in addition, *the system* also exhibited some interesting structural and dynamic features.[36] *The system* may have its potential applications in areas like chemical engineering, the automobile industry, biotechnology, medicine, batteries, fuel cells, etc. In

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**Table 1. Densities ($\rho$/g·cm$^{-3}$) at Different Temperatures and LiCl Concentrations in Binary Mixtures of Water (1) and Ethanol (2) Having Mole Fractions $x_1$ and $x_2$**

| concentration | | temperature, *t*/°C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| aquamolality/***m*** | molality/*m* | −5 | 0 | 5 | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 |
| $x_1 = 1.0, x_2 = 0.0$ | | | | | | | | | | | | | |
| **g**/10$^3$·g$^2$·cm$^{-3}$·mol$^{-1}$ = 0.0223 ± 0.0006 | | | | | | | | | | | | | |
| 0.0 | 0.0 | – | – | 0.9999 | 0.9997 | 0.9991 | 0.9982 | 0.9971 | 0.9956 | 0.9941 | 0.9922 | 0.9902 | 0.9881 |
| 0.5 | 0.5 | 1.0134 | 1.0132 | 1.0137 | 1.0125 | 1.0116 | 1.0108 | 1.0094 | 1.0081 | 1.0066 | 1.0051 | 1.0035 | 1.0022 |
| 1.0 | 1.0 | 1.0244 | 1.0244 | 1.0240 | 1.0233 | 1.0222 | 1.0214 | 1.0202 | 1.0187 | 1.0172 | 1.0158 | 1.0138 | 1.0125 |
| 2.0 | 2.0 | 1.0471 | 1.0469 | 1.0463 | 1.0453 | 1.0441 | 1.0430 | 1.0418 | 1.0401 | 1.0387 | 1.0371 | 1.0352 | 1.0338 |
| 3.0 | 3.0 | 1.0678 | 1.0674 | 1.0664 | 1.0654 | 1.0643 | 1.0631 | 1.0618 | 1.0603 | 1.0587 | 1.0569 | 1.0555 | 1.0390 |
| 4.5 | 4.5 | 1.0959 | 1.0950 | 1.0941 | 1.0928 | 1.0915 | 1.0905 | 1.0891 | 1.0877 | 1.0859 | 1.0844 | 1.0830 | 1.0811 |
| 6.0 | 6.0 | 1.1217 | 1.1212 | 1.1198 | 1.1188 | 1.1174 | 1.1161 | 1.1146 | 1.1131 | 1.1115 | 1.1101 | 1.1084 | 1.1064 |
| 8.0 | 8.0 | 1.1554 | 1.1537 | 1.1526 | 1.1514 | 1.1498 | 1.1485 | 1.1470 | 1.1454 | 1.1439 | 1.1423 | 1.1407 | 1.1388 |
| 11.0 | 11.0 | 1.2002 | 1.1987 | 1.1971 | 1.1956 | 1.1940 | 1.1925 | 1.1909 | 1.1893 | 1.1878 | 1.1861 | 1.1843 | 1.1826 |
| 14.0 | 14.0 | 1.2407 | 1.2391 | 1.2374 | 1.2358 | 1.2341 | 1.2326 | 1.2307 | 1.2289 | 1.2271 | 1.2253 | 1.2235 | 1.2218 |
| 17.8 | 17.8 | 1.2884 | 1.2864 | 1.2842 | 1.2822 | 1.2802 | 1.2783 | 1.2764 | 1.2746 | 1.2728 | 1.2707 | 1.2689 | 1.2669 |
| $x_1 = 0.9, x_2 = 0.1$ | | | | | | | | | | | | | |
| **g**/10$^3$·g$^2$·cm$^{-3}$·mol$^{-1}$ = 0.0201 ± 0.0006 | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.9773 | 0.9756 | 0.9732 | 0.9708 | 0.9673 | 0.9637 | 0.9603 | 0.9562 | 0.9527 | 0.9490 | 0.9455 | 0.9420 |
| 0.5 | 0.433 | 0.9848 | 0.9834 | 0.9819 | 0.9806 | 0.9786 | 0.9767 | 0.9745 | 0.9721 | 0.9698 | 0.9674 | 0.9643 | 0.9616 |
| 1.0 | 0.865 | 0.9965 | 0.9954 | 0.9932 | 0.9913 | 0.9894 | 0.9873 | 0.9851 | 0.9827 | 0.9802 | 0.9776 | 0.9749 | 0.9722 |
| 2.0 | 1.730 | 1.0163 | 1.0148 | 1.0128 | 1.0109 | 1.0090 | 1.0070 | 1.0047 | 1.0023 | 0.9999 | 0.9974 | 0.9948 | 0.9922 |
| 3.0 | 2.596 | 1.0347 | 1.0331 | 1.0312 | 1.0293 | 1.0273 | 1.0253 | 1.0233 | 1.0213 | 1.0194 | 1.0175 | 1.0154 | 1.0136 |
| 4.5 | 3.893 | 1.0606 | 1.0587 | 1.0566 | 1.0548 | 1.0530 | 1.0511 | 1.0490 | 1.0470 | 1.0450 | 1.0430 | 1.0409 | 1.0385 |
| 6.0 | 5.191 | 1.0842 | 1.0820 | 1.0800 | 1.0781 | 1.0760 | 1.0739 | 1.0718 | 1.0698 | 1.0678 | 1.0659 | 1.0639 | 1.0619 |
| 8.0 | 6.922 | 1.1121 | 1.1101 | 1.1079 | 1.1060 | 1.1039 | 1.1017 | 1.0996 | 1.0977 | 1.0955 | 1.0932 | 1.0911 | 1.0890 |
| 11.0 | 9.517 | 1.1507 | 1.1487 | 1.1464 | 1.1441 | 1.1420 | 1.1399 | 1.1376 | 1.1352 | 1.1337 | 1.1315 | 1.1295 | 1.1273 |
| 14.0 | 12.113 | 1.1865 | 1.1841 | 1.1820 | 1.1800 | 1.1779 | 1.1759 | 1.1736 | 1.1713 | 1.1693 | 1.1671 | 1.1649 | 1.1628 |
| 18.0 | 15.574 | 1.2253 | 1.2227 | 1.2204 | 1.2181 | 1.2159 | 1.2139 | 1.2116 | 1.2096 | 1.2074 | 1.2052 | 1.2030 | 1.2009 |
| $x_1 = 0.8, x_2 = 0.2$ | | | | | | | | | | | | | |
| **g**/10$^3$·g$^2$·cm$^{-3}$·mol$^{-1}$ = 0.0174 ± 0.0007 | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.9510 | 0.9480 | 0.9450 | 0.9414 | 0.9380 | 0.9346 | 0.9313 | 0.9276 | 0.9238 | 0.9195 | 0.9155 | 0.9112 |
| 0.5 | 0.381 | 0.9615 | 0.9584 | 0.9554 | 0.9518 | 0.9485 | 0.9450 | 0.9419 | 0.9382 | 0.9346 | 0.9305 | 0.9267 | 0.9227 |
| 1.0 | 0.763 | 0.9702 | 0.9673 | 0.9641 | 0.9605 | 0.9575 | 0.9541 | 0.9508 | 0.9471 | 0.9437 | 0.9396 | 0.9361 | 0.9322 |
| 2.0 | 1.525 | 0.9866 | 0.9836 | 0.9807 | 0.9772 | 0.9741 | 0.9708 | 0.9677 | 0.9642 | 0.9612 | 0.9571 | 0.9538 | 0.9500 |
| 3.0 | 2.288 | 1.0011 | 0.9977 | 0.9945 | 0.9909 | 0.9875 | 0.9840 | 0.9809 | 0.9773 | 0.9739 | 0.9697 | 0.9663 | 0.9623 |
| 4.5 | 3.431 | 1.0228 | 1.0198 | 1.0168 | 1.0130 | 1.0099 | 1.0066 | 1.0036 | 1.0002 | 0.9969 | 0.9928 | 0.9893 | 0.9855 |
| 6.0 | 4.575 | 1.0424 | 1.0394 | 1.0366 | 1.0332 | 1.0303 | 1.0275 | 1.0253 | 1.0224 | 1.0199 | 1.0163 | 1.0135 | 1.0103 |
| 8.0 | 6.100 | 1.0683 | 1.0650 | 1.0623 | 1.0590 | 1.0563 | 1.0534 | 1.0511 | 1.0478 | 1.0452 | 1.0419 | 1.0399 | 1.0369 |
| 11.0 | 8.388 | 1.1041 | 1.1013 | 1.0988 | 1.0958 | 1.0929 | 1.0903 | 1.0877 | 1.0848 | 1.0821 | 1.0786 | 1.0758 | 1.0726 |
| 14.0 | 10.675 | 1.1370 | 1.1343 | 1.1321 | 1.1289 | 1.1264 | 1.1239 | 1.1215 | 1.1186 | 1.1158 | 1.1125 | 1.1097 | 1.1066 |
| 17.5 | 13.344 | 1.1735 | 1.1699 | 1.1671 | 1.1637 | 1.1613 | 1.1585 | 1.1561 | 1.1532 | 1.1508 | 1.1477 | 1.1451 | 1.1421 |
| $x_1 = 0.7, x_2 = 0.3$ | | | | | | | | | | | | | |
| **g**/10$^3$·g$^2$·cm$^{-3}$·mol$^{-1}$ = 0.0152 ± 0.0005 | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.9316 | 0.9271 | 0.9227 | 0.9187 | 0.9145 | 0.9099 | 0.9053 | 0.9010 | 0.8965 | 0.8920 | 0.8874 | 0.8830 |
| 0.5 | 0.341 | 0.9397 | 0.9344 | 0.9300 | 0.9250 | 0.9207 | 0.9156 | 0.9096 | 0.9058 | 0.9012 | 0.8961 | 0.8909 | 0.8859 |
| 1.0 | 0.682 | 0.9488 | 0.9425 | 0.9387 | 0.9335 | 0.9289 | 0.9241 | 0.9180 | 0.9144 | 0.9104 | 0.9051 | 0.9001 | 0.8954 |
| 2.0 | 1.363 | 0.9627 | 0.9574 | 0.9532 | 0.9493 | 0.9443 | 0.9394 | 0.9336 | 0.9300 | 0.9259 | 0.9208 | 0.9156 | 0.9109 |
| 3.0 | 2.045 | 0.9726 | 0.9668 | 0.9625 | 0.9576 | 0.9532 | 0.9485 | 0.9423 | 0.9387 | 0.9343 | 0.9296 | 0.9243 | 0.9195 |
| 4.5 | 3.067 | 0.9918 | 0.9863 | 0.9822 | 0.9776 | 0.9736 | 0.9690 | 0.9634 | 0.9600 | 0.9561 | 0.9514 | 0.9467 | 0.9425 |
| 6.0 | 4.090 | 1.0116 | 1.0065 | 1.0026 | 0.9980 | 0.9942 | 0.9911 | 0.9852 | 0.9818 | 0.9778 | 0.9736 | 0.9694 | 0.9654 |
| 8.0 | 5.453 | 1.0355 | 1.0302 | 1.0266 | 1.0223 | 1.0184 | 1.0142 | 1.0088 | 1.0058 | 1.0026 | 0.9984 | 0.9942 | 0.9906 |
| 11.0 | 7.498 | 1.0675 | 1.0614 | 1.0559 | 1.0509 | 1.0477 | 1.0439 | 1.0389 | 1.0355 | 1.0323 | 1.0286 | 1.0248 | 1.0218 |
| 14.0 | 9.542 | 1.0978 | 1.0923 | 1.0887 | 1.0845 | 1.0812 | 1.0770 | 1.0718 | 1.0690 | 1.0654 | 1.0620 | 1.0578 | 1.0542 |
| 16.5 | 11.246 | 1.1215 | 1.1161 | 1.1123 | 1.1081 | 1.1047 | 1.1007 | 1.0956 | 1.0927 | 1.0895 | 1.0853 | 1.0821 | 1.0787 |
| $x_1 = 0.6, x_2 = 0.4$ | | | | | | | | | | | | | |
| **g**/10$^3$·g$^2$·cm$^{-3}$·mol$^{-1}$ = 0.0145 ± 0.0004 | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.9081 | 0.9035 | 0.8993 | 0.8946 | 0.8896 | 0.8851 | 0.8806 | 0.8761 | 0.8714 | 0.8668 | 0.8621 | 0.8573 |
| 0.5 | 0.308 | 0.9112 | 0.9065 | 0.9020 | 0.8948 | 0.8889 | 0.8843 | 0.8792 | 0.8744 | 0.8729 | 0.8712 | 0.8705 | 0.8695 |
| 1.0 | 0.616 | 0.9184 | 0.9137 | 0.9092 | 0.9022 | 0.8965 | 0.8921 | 0.8872 | 0.8825 | 0.8805 | 0.8791 | 0.8782 | 0.8771 |
| 2.0 | 1.232 | 0.9303 | 0.9252 | 0.9206 | 0.9131 | 0.9078 | 0.9047 | 0.8995 | 0.8944 | 0.8924 | 0.8953 | 0.8914 | 0.8884 |
| 3.0 | 1.849 | 0.9390 | 0.9341 | 0.9297 | 0.9225 | 0.9168 | 0.9127 | 0.9080 | 0.9034 | 0.9020 | 0.9007 | 0.8999 | 0.8995 |
| 4.5 | 2.773 | 0.9571 | 0.9529 | 0.9488 | 0.9420 | 0.9366 | 0.9326 | 0.9281 | 0.9238 | 0.9225 | 0.9215 | 0.9206 | 0.9202 |
| 6.0 | 3.697 | 0.9766 | 0.9726 | 0.9684 | 0.9626 | 0.9576 | 0.9544 | 0.9498 | 0.9448 | 0.9436 | 0.9426 | 0.9426 | 0.9424 |
| 8.0 | 4.930 | 0.9995 | 0.9954 | 0.9914 | 0.9849 | 0.9799 | 0.9762 | 0.9722 | 0.9682 | 0.9671 | 0.9669 | 0.9670 | 0.9672 |
| 11.0 | 6.778 | 1.0317 | 1.0273 | 1.0236 | 1.0169 | 1.0120 | 1.0085 | 1.0043 | 1.0009 | 1.0010 | 1.0006 | 1.0012 | 1.0019 |
| 14.0 | 8.627 | 1.0567 | 1.0527 | 1.0488 | 1.0424 | 1.0375 | 1.0346 | 1.0307 | 1.0268 | 1.0253 | 1.0249 | 1.0251 | 1.0277 |
| 16.0 | 9.859 | 1.0766 | 1.0724 | 1.0688 | 1.0624 | 1.0578 | 1.0544 | 1.0504 | 1.0466 | 1.0462 | 1.0457 | 1.0460 | 1.0466 |
| $x_1 = 0.5, x_2 = 0.5$ | | | | | | | | | | | | | |
| **g**/10$^3$·g$^2$·cm$^{-3}$·mol$^{-1}$ = 0.0131 ± 0.0006 | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.8866 | 0.8821 | 0.8777 | 0.8728 | 0.8685 | 0.8638 | 0.8592 | 0.8548 | 0.8501 | 0.8452 | 0.8405 | 0.8358 |
| 0.5 | 0.281 | 0.8880 | 0.8830 | 0.8789 | 0.8722 | 0.8657 | 0.8629 | 0.8571 | 0.8543 | 0.8493 | 0.8435 | 0.8386 | 0.8337 |
| 1.0 | 0.562 | 0.8946 | 0.8900 | 0.8867 | 0.8804 | 0.8742 | 0.8716 | 0.8663 | 0.8642 | 0.8592 | 0.8539 | 0.8490 | 0.8443 |
| 2.0 | 1.125 | 0.9073 | 0.9033 | 0.8974 | 0.8921 | 0.8863 | 0.8835 | 0.8778 | 0.8753 | 0.8706 | 0.8652 | 0.8594 | 0.8549 |
| 3.0 | 1.687 | 0.9169 | 0.9136 | 0.9092 | 0.9008 | 0.8973 | 0.8933 | 0.8876 | 0.8845 | 0.8798 | 0.8745 | 0.8668 | 0.8623 |

**Table 1 Continued**

| concentration | | temperature, *t*/°C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| aquamolality/***m*** | molality/*m* | −5 | 0 | 5 | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 |
| 4.5 | 2.530 | 0.9307 | 0.9261 | 0.9220 | 0.9155 | 0.9091 | 0.9067 | 0.9012 | 0.8988 | 0.8941 | 0.8888 | 0.8839 | 0.8794 |
| 6.0 | 3.374 | 0.9489 | 0.9442 | 0.9406 | 0.9347 | 0.9288 | 0.9259 | 0.9206 | 0.9185 | 0.9141 | 0.9091 | 0.9045 | 0.9004 |
| 8.0 | 4.498 | 0.9706 | 0.9663 | 0.9628 | 0.9568 | 0.9507 | 0.9488 | 0.9437 | 0.9418 | 0.9376 | 0.9325 | 0.9282 | 0.9241 |
| 11.0 | 6.185 | 0.9977 | 0.9931 | 0.9893 | 0.9837 | 0.9778 | 0.9758 | 0.9705 | 0.9688 | 0.9647 | 0.9602 | 0.9562 | 0.9528 |
| 14.0 | 7.872 | 1.0248 | 1.0206 | 1.0170 | 1.0113 | 1.0056 | 1.0036 | 0.9988 | 0.9970 | 0.9929 | 0.9882 | 0.9842 | 0.9807 |
| 15.2 | 8.547 | 1.0376 | 1.0333 | 1.0297 | 1.0239 | 1.0181 | 1.0162 | 1.0114 | 1.0097 | 1.0062 | 1.0016 | 0.9973 | 0.9943 |
| $x_1 = 0.4, x_2 = 0.6$ | | | | | | | | | | | | | |
| $\mathbf{g}/10^3 \cdot g^2 \cdot cm^{-3} \cdot mol^{-1} = 0.0133 \pm 0.0008$ | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.8677 | 0.8634 | 0.8588 | 0.8541 | 0.8495 | 0.845 | 0.8406 | 0.8361 | 0.8314 | 0.8269 | 0.8224 | 0.8177 |
| 0.5 | 0.259 | 0.8667 | 0.8620 | 0.8576 | 0.8510 | 0.8474 | 0.8428 | 0.8385 | 0.8340 | 0.8287 | 0.8242 | 0.8197 | 0.8139 |
| 1.0 | 0.517 | 0.8729 | 0.8687 | 0.8641 | 0.8578 | 0.8545 | 0.8497 | 0.8457 | 0.8409 | 0.8356 | 0.8309 | 0.8264 | 0.8213 |
| 2.0 | 1.034 | 0.8858 | 0.8811 | 0.8768 | 0.8704 | 0.8672 | 0.8625 | 0.8585 | 0.8541 | 0.8489 | 0.8446 | 0.8406 | 0.8357 |
| 3.0 | 1.551 | 0.8977 | 0.8934 | 0.8890 | 0.8829 | 0.8800 | 0.8755 | 0.8708 | 0.8676 | 0.8627 | 0.8589 | 0.8547 | 0.8496 |
| 4.5 | 2.327 | 0.9106 | 0.9052 | 0.9013 | 0.8929 | 0.8894 | 0.8858 | 0.8814 | 0.8772 | 0.8735 | 0.8682 | 0.8636 | 0.8580 |
| 6.0 | 3.102 | 0.9242 | 0.9202 | 0.9165 | 0.9101 | 0.9073 | 0.9029 | 0.8993 | 0.8952 | 0.8906 | 0.8866 | 0.8826 | 0.8778 |
| 8.0 | 4.136 | 0.9462 | 0.9418 | 0.9375 | 0.9313 | 0.9288 | 0.9247 | 0.9211 | 0.9171 | 0.9124 | 0.9084 | 0.9047 | 0.9000 |
| 11.0 | 5.687 | 0.9753 | 0.9710 | 0.9672 | 0.9613 | 0.9587 | 0.9547 | 0.9515 | 0.9481 | 0.9435 | 0.9399 | 0.9367 | 0.9321 |
| 14.0 | 7.238 | 0.9998 | 0.9955 | 0.9917 | 0.9862 | 0.9842 | 0.9804 | 0.9779 | 0.9745 | 0.9707 | 0.9675 | 0.9643 | 0.9607 |
| $x_1 = 0.3, x_2 = 0.7$ | | | | | | | | | | | | | |
| $\mathbf{g}/10^3 \cdot g^2 \cdot cm^{-3} \cdot mol^{-1} = 0.0134 \pm 0.0008$ | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.8522 | 0.8477 | 0.8429 | 0.8385 | 0.8339 | 0.8294 | 0.8251 | 0.8206 | 0.816 | 0.8111 | 0.8066 | 0.8019 |
| 0.5 | 0.239 | 0.8495 | 0.8447 | 0.8403 | 0.8358 | 0.8311 | 0.8272 | 0.8228 | 0.8175 | 0.8128 | 0.8074 | 0.8027 | 0.7978 |
| 1.0 | 0.479 | 0.8559 | 0.8509 | 0.8465 | 0.8419 | 0.8368 | 0.8332 | 0.8286 | 0.8233 | 0.8187 | 0.8138 | 0.8091 | 0.8038 |
| 2.0 | 0.957 | 0.8673 | 0.8624 | 0.8582 | 0.8539 | 0.8491 | 0.8454 | 0.8409 | 0.8359 | 0.8314 | 0.8263 | 0.8219 | 0.8175 |
| 3.0 | 1.436 | 0.8787 | 0.8743 | 0.8700 | 0.8661 | 0.8613 | 0.8575 | 0.8532 | 0.8486 | 0.8444 | 0.8396 | 0.8355 | 0.8317 |
| 4.5 | 2.153 | 0.8963 | 0.8912 | 0.8873 | 0.8833 | 0.8787 | 0.8753 | 0.8709 | 0.8662 | 0.8621 | 0.8573 | 0.8527 | 0.8487 |
| 6.0 | 2.871 | 0.9116 | 0.9074 | 0.9034 | 0.9000 | 0.8956 | 0.8921 | 0.8881 | 0.8836 | 0.8796 | 0.8750 | 0.8709 | 0.8665 |
| 8.0 | 3.828 | 0.9276 | 0.9234 | 0.9197 | 0.9160 | 0.9117 | 0.9080 | 0.9038 | 0.8999 | 0.8967 | 0.8915 | 0.8875 | 0.8836 |
| 11.0 | 5.264 | 0.9536 | 0.9492 | 0.9452 | 0.9416 | 0.9374 | 0.9343 | 0.9305 | 0.9263 | 0.9226 | 0.9186 | 0.9152 | 0.9114 |
| 13.5 | 6.460 | 0.9757 | 0.9713 | 0.9672 | 0.9634 | 0.9591 | 0.9560 | 0.9526 | 0.9486 | 0.9449 | 0.9408 | 0.9373 | 0.9332 |
| $x_1 = 0.2, x_2 = 0.8$ | | | | | | | | | | | | | |
| $\mathbf{g}/10^3 \cdot g^2 \cdot cm^{-3} \cdot mol^{-1} = 0.0132 \pm 0.0007$ | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.8382 | 0.8337 | 0.829 | 0.8247 | 0.8201 | 0.8156 | 0.811 | 0.806 | 0.8014 | 0.7968 | 0.7922 | 0.7875 |
| 0.5 | 0.223 | 0.8369 | 0.8320 | 0.8277 | 0.8229 | 0.8174 | 0.8128 | 0.8072 | 0.8019 | 0.7973 | 0.7903 | 0.7873 | 0.7842 |
| 1.0 | 0.445 | 0.8435 | 0.8381 | 0.8350 | 0.8278 | 0.8227 | 0.8188 | 0.8140 | 0.8084 | 0.8043 | 0.7976 | 0.7949 | 0.7912 |
| 2.0 | 0.891 | 0.8544 | 0.8504 | 0.8462 | 0.8404 | 0.8350 | 0.8308 | 0.8260 | 0.8210 | 0.8166 | 0.8100 | 0.8074 | 0.8040 |
| 3.0 | 1.336 | 0.8647 | 0.8605 | 0.8568 | 0.8513 | 0.8461 | 0.8422 | 0.8373 | 0.8324 | 0.8282 | 0.8222 | 0.8200 | 0.8177 |
| 4.5 | 2.004 | 0.8812 | 0.8770 | 0.8733 | 0.8681 | 0.8628 | 0.8591 | 0.8545 | 0.8499 | 0.8459 | 0.8398 | 0.8375 | 0.8343 |
| 6.0 | 2.672 | 0.8965 | 0.8924 | 0.8890 | 0.8840 | 0.8789 | 0.8752 | 0.8707 | 0.8660 | 0.8620 | 0.8558 | 0.8534 | 0.8505 |
| 8.0 | 3.563 | 0.9159 | 0.9124 | 0.9095 | 0.9045 | 0.8994 | 0.8959 | 0.8915 | 0.8870 | 0.8832 | 0.8771 | 0.8744 | 0.8719 |
| 11.0 | 4.899 | 0.9394 | 0.9335 | 0.9304 | 0.9255 | 0.9218 | 0.9170 | 0.9129 | 0.9086 | 0.9069 | 0.8977 | 0.8952 | 0.8951 |
| 13.0 | 5.789 | 0.9534 | 0.9497 | 0.9463 | 0.9414 | 0.9367 | 0.9332 | 0.9287 | 0.9242 | 0.9207 | 0.9147 | 0.9129 | 0.9104 |
| $x_1 = 0.1, x_2 = 0.9$ | | | | | | | | | | | | | |
| $\mathbf{g}/10^3 \cdot g^2 \cdot cm^{-3} \cdot mol^{-1} = 0.0133 \pm 0.0006$ | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.8249 | 0.8203 | 0.8159 | 0.8114 | 0.8069 | 0.8021 | 0.7977 | 0.793 | 0.7882 | 0.7836 | 0.779 | 0.7743 |
| 0.5 | 0.208 | 0.823 | 0.8172 | 0.8129 | 0.8084 | 0.8035 | 0.7988 | 0.7941 | 0.7891 | 0.784 | 0.7795 | 0.775 | 0.7701 |
| 1.0 | 0.416 | 0.827 | 0.8226 | 0.8185 | 0.8145 | 0.8095 | 0.8048 | 0.8002 | 0.7954 | 0.7903 | 0.786 | 0.7817 | 0.7785 |
| 2.0 | 0.833 | 0.8387 | 0.8341 | 0.8304 | 0.8258 | 0.8213 | 0.8165 | 0.8118 | 0.807 | 0.8021 | 0.7981 | 0.7937 | 0.7895 |
| 3.0 | 1.249 | 0.8506 | 0.8451 | 0.8409 | 0.8373 | 0.8322 | 0.8276 | 0.8232 | 0.8187 | 0.8139 | 0.8101 | 0.806 | 0.8022 |
| 4.5 | 1.874 | 0.8638 | 0.8576 | 0.854 | 0.8494 | 0.8443 | 0.8398 | 0.8349 | 0.8304 | 0.826 | 0.822 | 0.8195 | 0.816 |
| 6.0 | 2.499 | 0.8802 | 0.8754 | 0.8722 | 0.8685 | 0.8641 | 0.8595 | 0.8558 | 0.8514 | 0.847 | 0.8433 | 0.8394 | 0.8355 |
| 8.0 | 3.332 | 0.8983 | 0.8922 | 0.8923 | 0.8883 | 0.8842 | 0.8797 | 0.8757 | 0.8715 | 0.8671 | 0.8635 | 0.8597 | 0.8558 |
| 11.0 | 4.581 | 0.9175 | 0.9114 | 0.9079 | 0.9034 | 0.8994 | 0.8952 | 0.8915 | 0.8872 | 0.8828 | 0.8794 | 0.8759 | 0.8724 |
| $x_1 = 0.0, x_2 =1.0$ | | | | | | | | | | | | | |
| $\mathbf{g}/10^3 \cdot g^2 \cdot cm^{-3} \cdot mol^{-1} = 0.0131 \pm 0.0008$ | | | | | | | | | | | | | |
| 0.0 | 0.0 | 0.8111 | 0.8068 | 0.8026 | 0.7984 | 0.7942 | 0.7899 | 0.7857 | 0.7814 | 0.7771 | 0.7728 | 0.7679 | 0.7634 |
| 0.5 | 0.196 | 0.8109 | 0.8051 | 0.8032 | 0.7961 | 0.7897 | 0.786 | 0.7824 | 0.7765 | 0.7738 | 0.773 | 0.7722 | 0.7687 |
| 1.0 | 0.391 | 0.8179 | 0.8106 | 0.8103 | 0.8009 | 0.7959 | 0.7919 | 0.7883 | 0.7825 | 0.7797 | 0.779 | 0.7786 | 0.7743 |
| 2.0 | 0.782 | 0.8284 | 0.8213 | 0.8215 | 0.8122 | 0.8075 | 0.8033 | 0.7994 | 0.7947 | 0.7912 | 0.7907 | 0.7909 | 0.7861 |
| 3.0 | 1.173 | 0.8381 | 0.8325 | 0.8341 | 0.8234 | 0.8175 | 0.8136 | 0.8103 | 0.8048 | 0.8023 | 0.8019 | 0.8018 | 0.7966 |
| 4.5 | 1.760 | 0.8545 | 0.847 | 0.8455 | 0.8374 | 0.8316 | 0.8288 | 0.8238 | 0.8195 | 0.8166 | 0.8158 | 0.8168 | 0.811 |
| 6.0 | 2.347 | 0.8694 | 0.8631 | 0.8645 | 0.8542 | 0.8491 | 0.8454 | 0.8423 | 0.8371 | 0.8356 | 0.8352 | 0.8349 | 0.8304 |
| 8.0 | 3.129 | 0.8877 | 0.8824 | 0.8836 | 0.8757 | 0.8684 | 0.8647 | 0.8619 | 0.8567 | 0.8547 | 0.8547 | 0.8539 | 0.8513 |
| 10.0 | 3.911 | 0.9078 | 0.9011 | 0.9021 | 0.8918 | 0.8869 | 0.8834 | 0.8805 | 0.8754 | 0.8736 | 0.8737 | 0.874 | 0.8694 |

the present work, comprehensive data on density ($\rho$), viscosity ($\eta$), and electrical conductivity ($\kappa$) for *the system* have been presented over the entire ranges of solvent composition and LiCl solubility from (−5 to +50) °C. Excess molar volumes ($V^E$) for *the mixture* at a series of compositions have also been determined whose isotherms exhibit minima at the same common water mole fraction ($x_1$) of 0.6. From the temperature dependence of $\eta$ and $\Lambda_m$, respective activation energies have been determined and mutually compared. In the case that conductivity is only governed by the solution viscosity, a correspondence should exist between the activation energies determined for the viscosity and conductivity.

## Experimental Section

***Chemicals and Preparation of Samples.*** Chemicals of high grade purity were employed for the study. Water (room-temperature electrical conductivity between (1 and 2) $\mu S \cdot cm^{-1}$) used as one of the solvent components was freshly prepared after double distillation carried out in an all-quickfit apparatus in the presence of a few crystals of $KMnO_4$ and a small amount of NaOH.[37] Ethanol (Merck, purity ≥ 99 %) was used without further drying because all the LiCl solutions were to be prepared in *the mixtures* (except those in neat ethanol). A proton NMR spectrum of the ethanol measured at 32 °C exhibited no extra

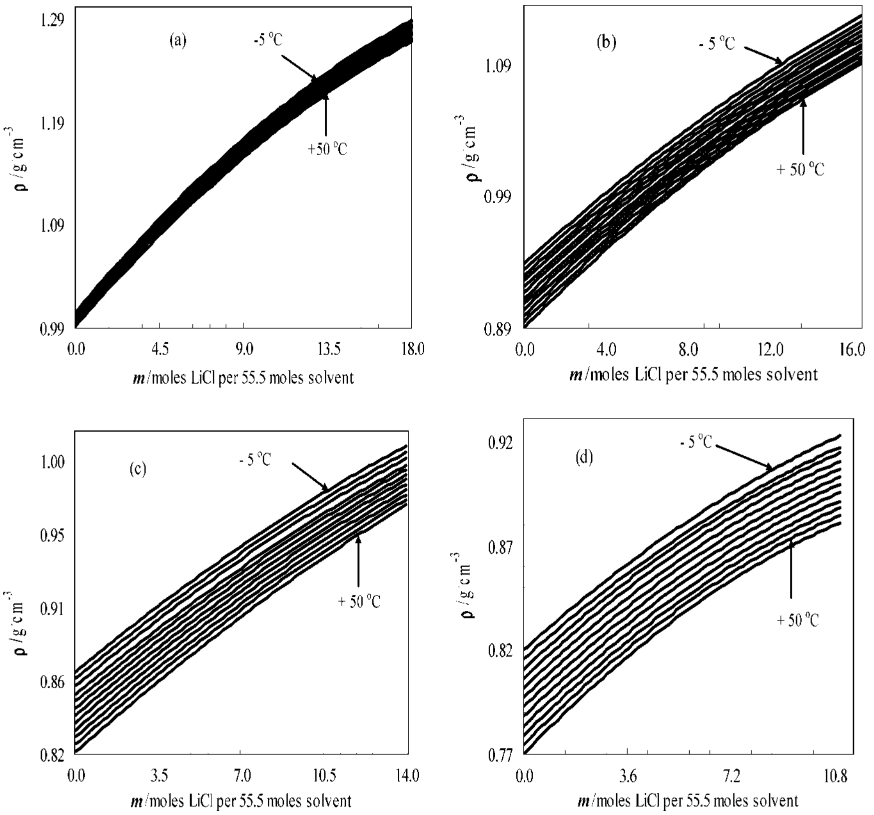

**Figure 1.** (a−d) Densities, $\rho$, fitted to LiCl aquamolality, $\boldsymbol{m}$, in the isotherm: $\rho(\boldsymbol{m}) = d_0 + d_1 \cdot \boldsymbol{m} + d_2 \cdot \boldsymbol{m}^2$ from −5 to +50 at 5 °C intervals for solutions prepared in aqueous ethanol of water mole fractions $x_1$ =: (a) 1.0, (b) 0.7, (c) 0.4, and (d) 0.1 (refer to Table 2 for $d$ values).

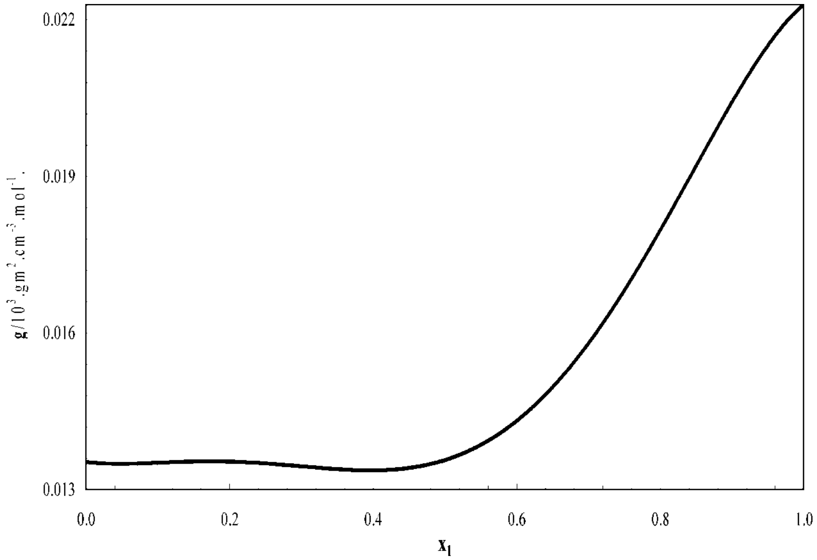

**Figure 2.** Dependence of the density index, $\mathbf{g}(x_1)$, on water mole fractions, $x_1$, for the system $H_2O$ + $C_2H_5OH$ + LiCl.

peak, and also a triplet peak was observed for its OH.[38,39] “Anhydrous” LiCl (Merck, purity > 99 % also verified by atomic absorption spectrophotometry) employed in the study was first kept in an electric oven at about 200 °C for several hours and finally stored in a desiccator under low pressure.

Suitable volumes of the samples were carefully prepared (with minimum possible atmospheric exposure) by adding together precalculated amounts of the individual components in closed vessels and subsequently stored in sealed bottles. An ER-180A digital balance from A & D, Japan (detection limit 0.1 mg) was employed for the weighings. As in many previous works,[36,40–43] the solute (LiCl) concentration has been expressed in the temperature-independent aquamolality scale, $\boldsymbol{m}$ (moles of solute per 55.5 moles of the solvent).

***Temperature Controlling.*** All the density measurements were carried out in thermostatic conditions. For the set of measurements on a given sample, temperature was successively raised through intervals of 5 °C over the employed range from (−5 to +50) °C. The required temperature of the sample (submerged in a bath of a water−ethanol mixture) was maintained by circulating the mixture using a (HAAKE F3, Germany) chiller equipped with an automatic temperature-controlling sensor having a detection limit of ± 0.02 °C. Besides a digital display on the chiller, the bath temperature was also simultaneously

**Table 2. Coefficients (in Appropriate Units) of the Equation $\rho(t, m) = d_o(t) + d_1(t)\cdot m + d_2(t)\cdot m^2$ at Different Temperatures in Binary Mixtures of Water (1) and Ethanol (2) Having Mole Fractions $x_1$ and $x_2$ with *m* Being the LiCl Aquamolality**

| | temperature, $t$/°C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| coefficients | −5 | 0 | 5 | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 |
| $x_1 = 1.0, x_2 = 0.0$ | | | | | | | | | | | | |
| $d_0$ | 1.0029 | 1.0031 | 1.0000 | 0.9996 | 0.9993 | 0.9981 | 0.9970 | 0.9958 | 0.9942 | 0.9921 | 0.9900 | 0.9882 |
| $d_1$ | 0.0218 | 0.0216 | 0.0221 | 0.0220 | 0.0217 | 0.0217 | 0.0217 | 0.0216 | 0.0216 | 0.0218 | 0.0219 | 0.0214 |
| $d_2$ | −0.0003 | −0.0003 | −0.0004 | −0.0004 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0004 | −0.0004 | −0.0003 |
| $x_1 = 0.9, x_2 = 0.1$ | | | | | | | | | | | | |
| $d_0$ | 0.9746 | 0.9733 | 0.9719 | 0.9700 | 0.9682 | 0.9660 | 0.9641 | 0.9614 | 0.9589 | 0.9563 | 0.9532 | 0.9501 |
| $d_1$ | 0.0202 | 0.0201 | 0.0199 | 0.0200 | 0.0199 | 0.0199 | 0.0199 | 0.0200 | 0.0202 | 0.0203 | 0.0205 | 0.0207 |
| $d_2$ | −0.0004 | −0.0004 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0004 | −0.0004 | −0.0004 | −0.0004 | −0.0004 |
| $x_1 = 0.8, x_2 = 0.2$ | | | | | | | | | | | | |
| $d_0$ | 0.9519 | 0.9489 | 0.9459 | 0.9423 | 0.9389 | 0.9355 | 0.9322 | 0.9285 | 0.9247 | 0.9204 | 0.9164 | 0.9121 |
| $d_1$ | 0.0164 | 0.0165 | 0.0165 | 0.0165 | 0.0166 | 0.0167 | 0.0168 | 0.0169 | 0.0171 | 0.0173 | 0.0175 | 0.0177 |
| $d_2$ | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0003 | −0.0003 |
| $x_1 = 0.7, x_2 = 0.3$ | | | | | | | | | | | | |
| $d_0$ | 0.9325 | 0.9280 | 0.9236 | 0.9196 | 0.9154 | 0.9108 | 0.9062 | 0.9019 | 0.8974 | 0.8929 | 0.8883 | 0.8839 |
| $d_1$ | 0.0143 | 0.0143 | 0.0145 | 0.0145 | 0.0146 | 0.0148 | 0.0149 | 0.0151 | 0.0154 | 0.0156 | 0.0157 | 0.0160 |
| $d_2$ | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 |
| $x_1 = 0.6, x_2 = 0.4$ | | | | | | | | | | | | |
| $d_0$ | 0.9090 | 0.9044 | 0.9002 | 0.8955 | 0.8905 | 0.8860 | 0.8815 | 0.8770 | 0.8723 | 0.8677 | 0.8630 | 0.8582 |
| $d_1$ | 0.0126 | 0.0128 | 0.0128 | 0.0127 | 0.0129 | 0.0132 | 0.0133 | 0.0134 | 0.0140 | 0.0149 | 0.0155 | 0.0163 |
| $d_2$ | −0.0001 | −0.0001 | −0.0001 | −0.0001 | −0.0001 | −0.0001 | −0.0001 | −0.0001 | −0.0002 | −0.0002 | −0.0002 | −0.0003 |
| $x_1 = 0.5, x_2 = 0.5$ | | | | | | | | | | | | |
| $d_0$ | 0.8866 | 0.8821 | 0.8777 | 0.8728 | 0.8685 | 0.8638 | 0.8592 | 0.8548 | 0.8501 | 0.8452 | 0.8405 | 0.8358 |
| $d_1$ | 0.0121 | 0.0123 | 0.0124 | 0.0123 | 0.0123 | 0.0127 | 0.0127 | 0.0131 | 0.0132 | 0.0134 | 0.0133 | 0.0135 |
| $d_2$ | −0.0001 | −0.0001 | −0.0001 | −0.0001 | −0.0001 | −0.0001 | −0.0001 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 |
| $x_1 = 0.4, x_2 = 0.6$ | | | | | | | | | | | | |
| $d_0$ | 0.8677 | 0.8634 | 0.8588 | 0.8541 | 0.8495 | 0.8450 | 0.8406 | 0.8361 | 0.8314 | 0.8269 | 0.8224 | 0.8177 |
| $d_1$ | 0.0124 | 0.0124 | 0.0126 | 0.0123 | 0.0126 | 0.0128 | 0.0128 | 0.013 | 0.0132 | 0.0133 | 0.0134 | 0.0134 |
| $d_2$ | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 |
| $x_1 = 0.3, x_2 = 0.7$ | | | | | | | | | | | | |
| $d_0$ | 0.8522 | 0.8477 | 0.8429 | 0.8385 | 0.8339 | 0.8294 | 0.8251 | 0.8206 | 0.8160 | 0.8111 | 0.8066 | 0.8019 |
| $d_1$ | 0.0122 | 0.0123 | 0.0125 | 0.0127 | 0.0129 | 0.0131 | 0.0131 | 0.0132 | 0.0134 | 0.0136 | 0.0137 | 0.0139 |
| $d_2$ | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0003 |
| $x_1 = 0.2, x_2 = 0.8$ | | | | | | | | | | | | |
| $d_0$ | 0.8382 | 0.8337 | 0.8290 | 0.8247 | 0.8201 | 0.8156 | 0.8110 | 0.8060 | 0.8014 | 0.7968 | 0.7922 | 0.7875 |
| $d_1$ | 0.0125 | 0.0126 | 0.0129 | 0.0128 | 0.0128 | 0.0130 | 0.0131 | 0.0134 | 0.0135 | 0.0134 | 0.0138 | 0.0143 |
| $d_2$ | −0.0002 | −0.0002 | −0.0003 | −0.0002 | −0.0002 | −0.0002 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 |
| $x_1 = 0.1, x_2 = 0.9$ | | | | | | | | | | | | |
| $d_0$ | 0.8249 | 0.8203 | 0.8159 | 0.8114 | 0.8069 | 0.8021 | 0.7977 | 0.7930 | 0.7882 | 0.7836 | 0.7790 | 0.7743 |
| $d_1$ | 0.0120 | 0.0121 | 0.0124 | 0.0127 | 0.0127 | 0.0129 | 0.0129 | 0.0131 | 0.0133 | 0.0135 | 0.0139 | 0.0142 |
| $d_2$ | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0003 | −0.0004 | −0.0004 |
| $x_1 = 0.0, x_2 = 1.0$ | | | | | | | | | | | | |
| $d_0$ | 0.8111 | 0.8068 | 0.8026 | 0.7984 | 0.7942 | 0.7899 | 0.7857 | 0.7814 | 0.7771 | 0.7728 | 0.7679 | 0.7634 |
| $d_1$ | 0.0125 | 0.0120 | 0.0127 | 0.0120 | 0.0116 | 0.0119 | 0.0119 | 0.0119 | 0.0122 | 0.0130 | 0.0143 | 0.0141 |
| $d_2$ | −0.0002 | −0.0002 | −0.0002 | −0.0002 | −0.0001 | −0.0002 | −0.0001 | −0.0001 | −0.0002 | −0.0002 | −0.0003 | −0.0003 |

monitored using an ASTM-calibrated thermometer [(0 to 100) °C] having a graduation of 0.01 °C. Although samples attained the required temperatures in about 10 min, they were allowed to thermally equilibrate in the bath for at least 20 min before starting the measurements.

***Density Measurement.*** Densities were determined using 10 mL Pyrex density bottles which were calibrated at each working temperature by employing water and toluene (Merck, purity > 99.9 %) as references. Toluene was used without further purification as it did not exhibit any extra peak in its NMR spectrum. The sample and the density bottles were allowed to attain equilibrium in the preadjusted thermal bath. Since the laboratory temperature was maintained around 25 °C throughout the experiments, the colder baths were accordingly adjusted at slightly lower than the required temperatures to allow compensation due to heat exchange during weighing of the density bottle (and conversely at higher temperatures for the hotter baths); nevertheless, weighings were done swiftly, and all readings were repeated at least three times. Whereas the maximum difference of the required and actual temperatures could be ± 0.1 °C, uncertainty in the density value could be ± 0.0002 $\text{g}\cdot\text{cm}^{-3}$. Some selected density values at various temperatures were also monitored on a DMA-4500, Anton Paar (Austria) vibrating tube density meter having a resolution[44] of $5\cdot10^{-5}$ $\text{g}\cdot\text{cm}^{-3}$. The density values up to the fourth decimal position were in agreement with our two kinds of measurements.

***Viscosity Measurement.*** Viscosities at different temperatures were measured in a specially designed transparent glass bath which ensured placing of the viscometer and an ASTM-calibrated thermometer at their same respective positions during each measurement. The bath temperature was maintained by circulating the thermostatted ethanol−water mixture through it. For each measurement, a 20 mL sample loaded in the viscometer was first allowed to attain the required temperature in the bath. Since viscosity values covered a considerably wide range (from (0.55 to 181) mPa·s) on varying temperature and LiCl concentration in our study, Ubbelhode viscometers (each ~20 mL capacity) of three different capillary bores were employed. The

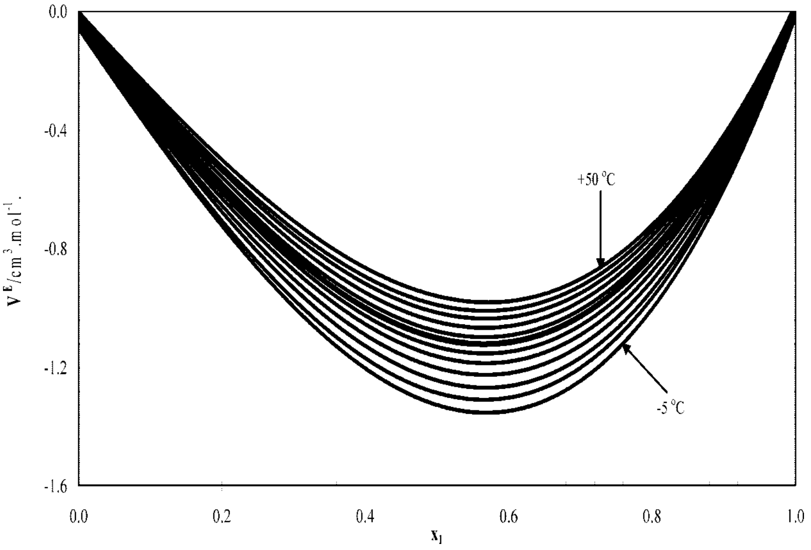

**Figure 3.** Excess molar volume, $V^E$, plotted against water mole fraction, $x_1$, for the aqueous mixtures of ethanol.

**Table 3. Excess Molar Volume ($V^E$/cm$^3$·mol$^{-1}$) for the Aqueous Mixtures of Ethanol at Different Temperatures and Water Mole Fractions ($x_1$)[a]**

| temp | $x_1$ | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| °C | 0.9 | 0.8 | 0.7 | 0.6 | 0.5 | 0.4 | 0.3 | 0.2 | 0.1 |
| −5 | −0.601 | −0.938 | −1.287 | −1.340 | −1.236 | −1.087 | −0.937 | −0.724 | −0.417 |
| 0 | −0.586 | −0.913 | −1.234 | −1.292 | −1.198 | −1.065 | −0.911 | −0.703 | −0.393 |
| 5 | −0.561 | −0.892 | −1.186 | −1.259 | −1.164 | −1.027 | −0.866 | −0.666 | −0.377 |
| 10 | −0.542 | −0.860 | −1.154 | −1.211 | −1.111 | −0.986 | −0.844 | −0.654 | −0.355 |
| 15 | −0.504 | −0.839 | −1.121 | −1.156 | −1.087 | −0.952 | −0.812 | −0.624 | −0.332 |
| 20 | −0.470 | −0.823 | −1.081 | −1.125 | −1.052 | −0.929 | −0.793 | −0.607 | −0.296 |
| 25 | −0.443(−0.454) | −0.812 | −1.041 | −1.093(−1.096) | −1.019 | −0.907 | −0.780 | −0.486 | −0.281(−0.267) |
| 30 | −0.407 | −0.797 | −1.018 | −1.068 | −1.001 | −0.888 | −0.763 | −0.563 | −0.252 |
| 35 | −0.384 | −0.779 | −0.987 | −1.035 | −0.970 | −0.859 | −0.740 | −0.541 | −0.229 |
| 40 | −0.363 | −0.753 | −0.962 | −1.009 | −0.934 | −0.842 | −0.702 | −0.520 | −0.214 |
| 45 | −0.348 | −0.737 | −0.934 | −0.981 | −0.906 | −0.826 | −0.687 | −0.493 | −0.205 |
| 50 | −0.335 | −0.714 | −0.914 | −0.949 | −0.879 | −0.799 | −0.661 | −0.465 | −0.183 |

[a] Some literature values are also given in parentheses.

**Table 4. Values of ($-\beta\cdot10^4$/°C$^{-1}$) as Determined from $\rho(t) = \rho(25)\cdot e^{\beta(t-25)}$for the System (LiCl + Water + Ethanol) with $x_1$ and $m$ Being Water Mole Fraction in the Solvent Mixture and LiCl Aquamolality, Respectively[a]**

| | $x_1$ | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| ***m*** | 1.0 | 0.9 | 0.8 | 0.7 | 0.6 | 0.5 | 0.4 | 0.3 | 0.2 | 0.1 | 0.0 |
| 0.0 | 2.71 | 6.89 | 7.70 | 9.69 | 10.37 | 10.67 | 10.76 | 11.01 | 11.42 | 11.5 | 11.4 |
| 0.5 | 2.23 | 6.70 | 7.97 | 9.28 | 8.86 | 10.32 | 10.41 | 10.57 | 11.33 | 11.2 | 10.7 |
| 1.0 | 2.24 | 6.88 | 7.33 | 9.01 | 8.69 | 9.32 | 10.20 | 10.63 | 11.05 | 10.5 | 10.5 |
| 2.0 | 2.45 | 6.72 | 6.84 | 8.73 | 8.24 | 9.61 | 9.69 | 10.02 | 10.46 | 10.4 | 9.9 |
| 3.0 | 3.51 | 6.13 | 7.09 | 8.80 | 8.25 | 10.03 | 9.12 | 9.37 | 9.77 | 10.0 | 9.8 |
| 4.5 | 2.50 | 6.02 | 6.72 | 7.93 | 7.54 | 9.12 | 9.73 | 9.22 | 9.43 | 9.8 | 9.6 |
| 6.0 | 2.53 | 5.87 | 5.63 | 7.21 | 6.90 | 8.45 | 8.52 | 8.48 | 9.14 | 8.8 | 8.8 |
| 8.0 | 2.60 | 5.90 | 5.40 | 6.78 | 6.32 | 7.76 | 8.22 | 8.16 | 8.51 | 8.2 | 8.5 |
| 11.0 | 2.72 | 5.83 | 5.25 | 6.63 | 5.67 | 7.28 | 7.38 | 7.56 | 8.30 | – | – |
| 14.0 | 2.81 | 5.86 | 4.90 | 6.12 | 5.59 | 7.01 | – | – | – | – | – |
| | 3.01 (17.8) | 5.50 (18.0) | 4.82 (17.5) | 5.91 (16.5) | 5.52 (16.0) | 6.80 (15.2) | 6.42 (14.0) | 7.40 (13.5) | 8.02 (13.0) | 8.4 (11.0) | 8.2 (10.0) |
| $\sigma$ | ± 0.09 | ± 0.09 | ± 0.08 | ± 0.09 | ± 0.09 | ± 0.09 | ± 0.07 | ± 0.06 | ± 0.08 | ± 0.1 | ± 0.32 |

[a] Highest ***m*** values employed for the mixtures are given in parentheses, and the corresponding standard deviations ($\sigma$) of the $\beta$-values are given at the bottom of each column.

viscometers were standardized using 20 °C literature values[1,45] of water (1.002), nitrobenzene (1.980), and aniline (4.467 mPa·s). Viscosities of the references were also measured from time to time as a check. For still higher viscosities, a previously measured solution served as the reference for the next one. For each measurement, the flow time taken was an average of at least three runs, and the time could be read with an estimated uncertainty within ± 0.02 s. The maximum uncertainties in the viscosity measurements (as could be estimated from the 20 °C reported data of the solvent mixtures[1]) are within ± 1 %.

***Electrical Conductivity Measurement.*** A CyberScan CON500 conductivity meter from Eutech Instruments, Singapore (with reported resolution of 0.05 % and inaccuracy within ± 1 % of the given full scale), served to measure electrical conductivity. The conductivity meter was equipped with a thermal sensor along with provision of an automatic compensation for temperature variation. The $\kappa$ values were directly measured employing any one of the five scales of the instrument covering the range from 1 $\mu$S·cm$^{-1}$ to 199.9 mS·cm$^{-1}$ (also the maximum readable value). Although

**Table 5. Viscosities ($\eta$/mPa·s) at Different Temperatures and LiCl Aquamolalities ($m$) in Binary Mixtures of Water (1) and Ethanol (2) Having Mole Fractions $x_1$ and $x_2$**

| concentration | temperature, $t$/°C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $m$ | −5 | 0 | 5 | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 |
| | | | | | $x_1 = 1.0, x_2 = 0.0$ | | | | | | | |
| 0.0 | – | – | 1.519 | 1.308 | 1.140 | 1.005 | 0.894 | 0.799 | 0.723 | 0.655 | 0.599 | 0.548 |
| 0.5 | 2.273 | 1.944 | 1.663 | 1.424 | 1.243 | 1.087 | 0.970 | 0.866 | 0.782 | 0.722 | 0.668 | 0.625 |
| 1.0 | 2.554 | 2.107 | 1.784 | 1.518 | 1.346 | 1.160 | 1.052 | 0.929 | 0.834 | 0.774 | 0.714 | 0.665 |
| 2.0 | 2.831 | 2.370 | 2.000 | 1.701 | 1.502 | 1.308 | 1.165 | 1.047 | 0.941 | 0.866 | 0.802 | 0.747 |
| 3.0 | 3.191 | 2.681 | 2.266 | 1.936 | 1.729 | 1.495 | 1.339 | 1.239 | 1.083 | 0.977 | 0.910 | 0.859 |
| 4.5 | 3.649 | 3.133 | 2.690 | 2.303 | 2.034 | 1.777 | 1.586 | 1.422 | 1.280 | 1.177 | 1.076 | 1.003 |
| 6.0 | 4.461 | 3.815 | 3.258 | 2.777 | 2.452 | 2.131 | 1.906 | 1.713 | 1.532 | 1.406 | 1.293 | 1.194 |
| 8.0 | 6.735 | 5.744 | 4.899 | 4.170 | 3.602 | 3.199 | 2.800 | 2.486 | 2.256 | 2.071 | 1.919 | 1.742 |
| 11.0 | 10.848 | 9.191 | 7.784 | 6.579 | 5.630 | 4.898 | 4.292 | 3.812 | 3.407 | 3.091 | 2.869 | 2.690 |
| 14.0 | 18.985 | 15.816 | 13.169 | 10.970 | 9.142 | 7.966 | 6.899 | 6.073 | 5.332 | 4.812 | 4.412 | 4.123 |
| 17.8 | 28.660 | 24.988 | 21.788 | 19.662 | 17.145 | 14.889 | 12.680 | 11.538 | 10.462 | 9.741 | 8.984 | 8.265 |
| | | | | | $x_1 = 0.9, x_2 = 0.1$ | | | | | | | |
| 0.0 | 6.025 | 4.997 | 4.089 | 3.340 | 2.706 | 2.268 | 1.920 | 1.656 | 1.448 | 1.271 | 1.121 | 0.975 |
| 0.5 | 6.601 | 5.372 | 4.406 | 3.614 | 2.924 | 2.473 | 2.090 | 1.811 | 1.551 | 1.361 | 1.185 | 1.041 |
| 1.0 | 7.317 | 5.872 | 4.703 | 3.852 | 3.181 | 2.652 | 2.247 | 1.950 | 1.708 | 1.459 | 1.288 | 1.115 |
| 2.0 | 7.802 | 6.321 | 5.118 | 4.196 | 3.425 | 2.875 | 2.569 | 2.094 | 1.809 | 1.533 | 1.369 | 1.203 |
| 3.0 | 8.355 | 6.895 | 5.751 | 4.695 | 3.832 | 3.207 | 2.744 | 2.352 | 1.985 | 1.669 | 1.472 | 1.296 |
| 4.5 | 10.453 | 8.678 | 7.206 | 5.853 | 5.086 | 4.261 | 3.707 | 3.207 | 2.852 | 2.506 | 2.315 | 2.048 |
| 6.0 | 11.283 | 9.824 | 8.554 | 7.283 | 6.125 | 5.115 | 4.387 | 3.864 | 3.372 | 3.075 | 2.879 | 2.492 |
| 8.0 | 17.824 | 14.459 | 11.724 | 9.516 | 7.844 | 6.608 | 5.629 | 5.221 | 4.252 | 3.750 | 3.429 | 3.154 |
| 11.0 | 33.686 | 26.232 | 20.525 | 16.479 | 12.406 | 10.551 | 8.894 | 7.648 | 6.640 | 5.833 | 5.280 | 4.782 |
| 14.0 | 60.400 | 46.020 | 35.471 | 27.642 | 20.615 | 17.491 | 14.560 | 12.697 | 11.231 | 9.266 | 8.272 | 7.470 |
| 18.0 | 115.642 | 88.450 | 67.556 | 51.851 | 37.369 | 31.211 | 25.390 | 21.732 | 18.914 | 15.286 | 13.943 | 12.183 |
| | | | | | $x_1 = 0.8, x_2 = 0.2$ | | | | | | | |
| 0.0 | 7.549 | 6.169 | 5.037 | 4.102 | 3.433 | 2.841 | 2.450 | 2.101 | 1.772 | 1.557 | 1.369 | 1.202 |
| 0.5 | 8.076 | 6.618 | 5.425 | 4.471 | 3.704 | 3.085 | 2.595 | 2.257 | 1.907 | 1.605 | 1.437 | 1.285 |
| 1.0 | 8.690 | 7.115 | 5.839 | 4.821 | 4.038 | 3.400 | 2.744 | 2.392 | 2.072 | 1.761 | 1.532 | 1.314 |
| 2.0 | 9.689 | 7.892 | 6.501 | 5.256 | 4.342 | 3.634 | 3.068 | 2.601 | 2.251 | 1.901 | 1.691 | 1.438 |
| 3.0 | 13.300 | 10.558 | 8.398 | 6.656 | 5.348 | 4.466 | 3.759 | 3.238 | 2.793 | 2.460 | 2.156 | 1.924 |
| 4.5 | 15.579 | 12.442 | 9.976 | 7.943 | 6.425 | 5.380 | 4.546 | 3.915 | 3.323 | 2.936 | 2.498 | 2.185 |
| 6.0 | 20.010 | 15.577 | 12.128 | 9.704 | 7.872 | 6.598 | 5.565 | 4.805 | 4.115 | 3.582 | 3.135 | 2.701 |
| 8.0 | 29.226 | 22.508 | 17.425 | 13.738 | 10.239 | 8.634 | 7.305 | 6.609 | 5.419 | 4.759 | 4.201 | 3.787 |
| 11.0 | 50.673 | 38.717 | 29.934 | 23.195 | 17.380 | 14.550 | 12.092 | 10.332 | 8.820 | 7.707 | 6.782 | 6.157 |
| 14.0 | 93.315 | 69.558 | 52.975 | 40.751 | 29.682 | 24.752 | 20.307 | 17.455 | 15.191 | 12.407 | 11.300 | 9.624 |
| 17.5 | 184.015 | 137.485 | 102.739 | 76.793 | 54.108 | 42.847 | 35.508 | 29.811 | 25.788 | 20.559 | 18.467 | 15.922 |
| | | | | | $x_1 = 0.7, x_2 = 0.3$ | | | | | | | |
| 0.0 | 7.172 | 5.947 | 4.933 | 4.093 | 3.372 | 2.842 | 2.350 | 2.040 | 1.780 | 1.560 | 1.371 | 1.170 |
| 0.5 | 7.668 | 6.365 | 5.428 | 4.487 | 3.632 | 3.051 | 2.610 | 2.234 | 1.935 | 1.669 | 1.435 | 1.251 |
| 1.0 | 8.276 | 6.898 | 5.778 | 4.759 | 3.928 | 3.217 | 2.738 | 2.395 | 2.019 | 1.724 | 1.539 | 1.361 |
| 2.0 | 9.651 | 7.889 | 6.554 | 5.341 | 4.340 | 3.653 | 3.134 | 2.720 | 2.321 | 1.975 | 1.695 | 1.523 |
| 3.0 | 13.241 | 10.698 | 8.599 | 6.926 | 5.683 | 4.765 | 4.114 | 3.582 | 3.002 | 2.641 | 2.319 | 1.975 |
| 4.5 | 16.264 | 13.142 | 10.598 | 8.468 | 6.918 | 5.826 | 4.941 | 4.270 | 3.671 | 3.189 | 2.791 | 2.413 |
| 6.0 | 20.370 | 16.269 | 13.050 | 10.400 | 8.486 | 7.137 | 6.003 | 5.108 | 4.502 | 3.921 | 3.435 | 2.980 |
| 8.0 | 32.411 | 25.185 | 19.554 | 15.375 | 11.527 | 9.708 | 8.265 | 7.143 | 6.157 | 5.419 | 4.832 | 4.174 |
| 11.0 | 57.668 | 46.354 | 34.910 | 27.140 | 20.262 | 17.087 | 14.171 | 12.292 | 10.841 | 8.913 | 7.857 | 7.000 |
| 14.0 | 128.627 | 96.395 | 72.165 | 54.823 | 39.567 | 32.693 | 26.659 | 22.650 | 19.673 | 15.779 | 14.234 | 12.361 |
| 16.5 | 214.294 | 159.709 | 119.043 | 88.746 | 62.740 | 51.068 | 40.735 | 34.304 | 29.270 | 23.151 | 20.749 | 17.691 |
| | | | | | $x_1 = 0.6, x_2 = 0.4$ | | | | | | | |
| 0.0 | 5.931 | 4.978 | 4.176 | 3.499 | 2.922 | 2.463 | 2.089 | 1.812 | 1.582 | 1.430 | 1.270 | 1.140 |
| 0.5 | 6.377 | 5.358 | 4.497 | 3.767 | 3.127 | 2.641 | 2.250 | 1.953 | 1.706 | 1.492 | 1.341 | 1.186 |
| 1.0 | 6.889 | 5.819 | 4.841 | 4.046 | 3.379 | 2.838 | 2.419 | 2.100 | 1.825 | 1.591 | 1.423 | 1.255 |
| 2.0 | 7.975 | 6.711 | 5.621 | 4.660 | 3.896 | 3.275 | 2.794 | 2.420 | 2.108 | 1.803 | 1.610 | 1.421 |
| 3.0 | 11.736 | 9.552 | 7.769 | 6.321 | 5.182 | 4.367 | 3.708 | 3.191 | 2.760 | 2.422 | 2.185 | 1.964 |
| 4.5 | 15.485 | 12.347 | 9.834 | 8.011 | 6.568 | 5.551 | 4.691 | 4.063 | 3.495 | 3.074 | 2.770 | 2.490 |
| 6.0 | 21.833 | 17.208 | 13.529 | 10.918 | 8.530 | 7.378 | 6.146 | 5.070 | 4.368 | 3.820 | 3.497 | 3.059 |
| 8.0 | 32.700 | 25.512 | 19.860 | 15.586 | 11.686 | 10.045 | 8.413 | 7.227 | 6.210 | 5.454 | 4.902 | 4.289 |
| 11.0 | 66.130 | 50.701 | 38.662 | 29.881 | 22.074 | 18.596 | 15.216 | 13.179 | 11.672 | 9.429 | 8.456 | 7.319 |
| 14.0 | 147.813 | 111.617 | 84.103 | 63.494 | 45.490 | 37.249 | 30.118 | 25.190 | 22.061 | 17.464 | 15.544 | 13.483 |
| 16.0 | 300.611 | 222.805 | 165.149 | 122.426 | 84.843 | 69.094 | 54.459 | 44.644 | 37.475 | 29.651 | 26.232 | 22.153 |
| | | | | | $x_1 = 0.5, x_2 = 0.5$ | | | | | | | |
| 0.0 | 4.829 | 4.123 | 3.518 | 2.995 | 2.539 | 2.153 | 1.840 | 1.598 | 1.407 | 1.270 | 1.140 | 1.020 |
| 0.5 | 5.148 | 4.441 | 3.829 | 3.262 | 2.755 | 2.340 | 2.007 | 1.754 | 1.541 | 1.348 | 1.211 | 1.074 |
| 1.0 | 5.821 | 4.915 | 4.158 | 3.527 | 2.980 | 2.571 | 2.191 | 1.947 | 1.733 | 1.476 | 1.341 | 1.148 |
| 2.0 | 6.835 | 5.771 | 4.947 | 4.119 | 3.487 | 2.934 | 2.530 | 2.214 | 1.939 | 1.711 | 1.529 | 1.358 |
| 3.0 | 10.402 | 8.541 | 7.012 | 5.753 | 4.755 | 4.065 | 3.463 | 3.011 | 2.606 | 2.312 | 2.082 | 1.724 |
| 4.5 | 13.961 | 11.341 | 9.199 | 7.473 | 6.242 | 5.300 | 4.516 | 3.878 | 3.391 | 2.971 | 2.613 | 2.300 |

**Table 5 Continued**

| concentration | temperature, $t$/°C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $m$ | −5 | 0 | 5 | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 |
| 6.0 | 20.305 | 15.899 | 12.727 | 10.234 | 7.997 | 6.692 | 5.985 | 4.930 | 4.262 | 3.752 | 3.366 | 2.911 |
| 8.0 | 31.501 | 24.866 | 19.598 | 15.599 | 12.519 | 10.103 | 8.447 | 7.266 | 6.180 | 5.454 | 4.866 | 4.184 |
| 11.0 | 79.870 | 60.159 | 46.474 | 35.686 | 26.401 | 21.962 | 18.209 | 15.734 | 13.743 | 10.983 | 10.021 | 8.455 |
| 14.0 | 180.863 | 132.010 | 95.569 | 71.990 | 51.472 | 41.370 | 33.179 | 28.115 | 24.131 | 19.012 | 16.993 | 14.556 |
| 15.2 | 333.404 | 245.142 | 180.224 | 134.336 | 93.211 | 75.946 | 60.253 | 49.001 | 41.068 | 32.397 | 28.465 | 22.821 |
| $x_1 = 0.4, x_2 = 0.6$ | | | | | | | | | | | | |
| 0.0 | 3.991 | 3.451 | 2.980 | 2.570 | 2.212 | 1.901 | 1.651 | 1.446 | 1.296 | 1.149 | 1.039 | 0.932 |
| 0.5 | 4.501 | 3.955 | 3.349 | 2.837 | 2.429 | 2.090 | 1.818 | 1.609 | 1.444 | 1.271 | 1.126 | 1.006 |
| 1.0 | 4.921 | 4.211 | 3.625 | 3.113 | 2.638 | 2.284 | 2.010 | 1.755 | 1.505 | 1.325 | 1.217 | 1.100 |
| 2.0 | 5.811 | 5.020 | 4.285 | 3.653 | 3.152 | 2.692 | 2.331 | 2.019 | 1.795 | 1.575 | 1.367 | 1.251 |
| 3.0 | 6.959 | 6.001 | 5.115 | 4.354 | 3.713 | 3.187 | 2.755 | 2.336 | 2.145 | 1.887 | 1.622 | 1.435 |
| 4.5 | 12.770 | 10.499 | 8.686 | 7.138 | 5.880 | 5.087 | 4.409 | 3.833 | 3.347 | 2.931 | 2.629 | 2.352 |
| 6.0 | 19.116 | 15.185 | 12.106 | 9.770 | 7.715 | 6.536 | 5.546 | 4.832 | 4.199 | 3.691 | 3.299 | 2.902 |
| 8.0 | 30.193 | 24.008 | 19.051 | 15.238 | 11.601 | 9.930 | 8.460 | 7.339 | 6.312 | 5.565 | 4.936 | 4.313 |
| 11.0 | 67.538 | 52.155 | 40.710 | 31.932 | 23.587 | 19.735 | 16.242 | 14.135 | 12.430 | 10.129 | 9.193 | 7.865 |
| 14.0 | 123.648 | 98.860 | 78.949 | 62.320 | 43.351 | 33.894 | 28.536 | 24.844 | 21.169 | 16.393 | 14.945 | 12.853 |
| $x_1 = 0.3, x_2 = 0.7$ | | | | | | | | | | | | |
| 0.0 | 3.345 | 2.938 | 2.579 | 2.260 | 1.980 | 1.715 | 1.513 | 1.356 | 1.215 | 1.096 | 0.998 | 0.902 |
| 0.5 | 3.741 | 3.357 | 2.874 | 2.513 | 2.194 | 1.902 | 1.686 | 1.498 | 1.331 | 1.181 | 1.072 | 0.982 |
| 1.0 | 4.194 | 3.635 | 3.173 | 2.751 | 2.457 | 2.126 | 1.849 | 1.621 | 1.489 | 1.311 | 1.175 | 1.035 |
| 2.0 | 5.009 | 4.390 | 3.799 | 3.286 | 2.843 | 2.469 | 2.165 | 1.932 | 1.726 | 1.521 | 1.389 | 1.239 |
| 3.0 | 6.100 | 5.381 | 4.631 | 3.996 | 3.400 | 2.954 | 2.594 | 2.302 | 2.045 | 1.822 | 1.675 | 1.469 |
| 4.5 | 9.056 | 7.495 | 6.206 | 5.238 | 4.486 | 3.877 | 3.363 | 2.996 | 2.639 | 2.362 | 2.157 | 1.798 |
| 6.0 | 12.910 | 10.622 | 8.726 | 7.178 | 5.920 | 5.127 | 4.449 | 3.873 | 3.387 | 2.985 | 2.702 | 2.492 |
| 8.0 | 22.497 | 18.653 | 15.460 | 12.816 | 10.651 | 9.067 | 7.704 | 6.684 | 5.764 | 5.118 | 4.594 | 4.180 |
| 11.0 | 67.128 | 52.393 | 40.820 | 32.134 | 23.727 | 19.960 | 16.605 | 14.327 | 12.652 | 10.269 | 9.301 | 8.108 |
| 13.5 | 139.112 | 104.463 | 78.306 | 59.415 | 42.780 | 35.153 | 28.478 | 24.163 | 20.882 | 16.555 | 14.885 | 12.766 |
| $x_1 = 0.2, x_2 = 0.8$ | | | | | | | | | | | | |
| 0.0 | 2.805 | 2.491 | 2.210 | 1.959 | 1.721 | 1.515 | 1.353 | 1.218 | 1.110 | 1.004 | 0.925 | 0.839 |
| 0.5 | 3.126 | 2.787 | 2.485 | 2.213 | 1.939 | 1.709 | 1.521 | 1.358 | 1.223 | 1.105 | 1.015 | 0.925 |
| 1.0 | 3.529 | 3.120 | 2.763 | 2.435 | 2.125 | 1.869 | 1.654 | 1.489 | 1.345 | 1.208 | 1.103 | 1.007 |
| 2.0 | 4.411 | 3.852 | 3.360 | 2.925 | 2.524 | 2.254 | 2.003 | 1.794 | 1.602 | 1.420 | 1.328 | 1.225 |
| 3.0 | 5.248 | 4.613 | 4.054 | 3.554 | 3.100 | 2.703 | 2.381 | 2.130 | 1.925 | 1.714 | 1.564 | 1.420 |
| 4.5 | 7.139 | 6.237 | 5.448 | 4.748 | 4.119 | 3.574 | 3.131 | 2.789 | 2.487 | 2.252 | 2.072 | 1.823 |
| 6.0 | 11.375 | 9.575 | 8.021 | 6.713 | 5.598 | 4.830 | 4.237 | 3.728 | 3.259 | 2.955 | 2.691 | 2.514 |
| 8.0 | 16.025 | 13.729 | 11.165 | 8.999 | 7.965 | 7.070 | 6.265 | 5.506 | 4.765 | 4.077 | 3.569 | 3.101 |
| 11.0 | 63.702 | 49.786 | 39.279 | 30.949 | 23.481 | 19.622 | 16.284 | 14.187 | 12.501 | 10.199 | 9.135 | 7.575 |
| 13.0 | 123.911 | 91.252 | 69.106 | 53.332 | 38.717 | 31.596 | 26.141 | 22.233 | 19.276 | 15.356 | 13.709 | 11.483 |
| $x_1 = 0.1, x_2 = 0.9$ | | | | | | | | | | | | |
| 0.0 | 2.307 | 2.059 | 1.837 | 1.639 | 1.510 | 1.355 | 1.249 | 1.156 | 1.067 | 0.920 | 0.860 | 0.760 |
| 0.5 | 2.871 | 2.595 | 2.386 | 2.149 | 1.887 | 1.710 | 1.537 | 1.393 | 1.291 | 1.158 | 1.074 | 0.991 |
| 1.0 | 3.245 | 2.918 | 2.615 | 2.340 | 2.096 | 1.892 | 1.690 | 1.546 | 1.408 | 1.285 | 1.189 | 1.095 |
| 2.0 | 3.911 | 3.512 | 3.150 | 2.822 | 2.503 | 2.233 | 2.005 | 1.817 | 1.673 | 1.530 | 1.413 | 1.295 |
| 3.0 | 4.839 | 4.313 | 3.842 | 3.423 | 3.047 | 2.684 | 2.399 | 2.176 | 1.980 | 1.796 | 1.662 | 1.521 |
| 4.5 | 6.701 | 5.883 | 5.166 | 4.529 | 3.950 | 3.496 | 3.103 | 2.820 | 2.529 | 2.287 | 2.133 | 1.909 |
| 6.0 | 10.288 | 8.711 | 7.372 | 6.334 | 5.456 | 4.771 | 4.290 | 3.792 | 3.335 | 3.034 | 2.790 | 2.608 |
| 8.0 | 16.193 | 13.643 | 11.483 | 9.681 | 8.225 | 7.103 | 6.253 | 5.524 | 4.879 | 4.342 | 3.992 | 3.687 |
| 11.0 | 86.485 | 68.040 | 53.466 | 41.529 | 31.392 | 26.382 | 21.428 | 18.652 | 16.420 | 13.181 | 12.033 | 10.423 |
| $x_1 = 0.0, x_2 = 1.0$ | | | | | | | | | | | | |
| 0.0 | 1.975 | 1.775 | 1.629 | 1.468 | 1.341 | 1.207 | 1.105 | 1.010 | 0.920 | 0.840 | 0.772 | 0.713 |
| 0.5 | 2.208 | 2.043 | 1.863 | 1.695 | 1.495 | 1.357 | 1.218 | 1.089 | 1.006 | 0.928 | 0.866 | 0.807 |
| 1.0 | 2.546 | 2.313 | 2.099 | 1.908 | 1.715 | 1.524 | 1.412 | 1.306 | 1.129 | 1.046 | 0.968 | 0.884 |
| 2.0 | 3.283 | 2.943 | 2.638 | 2.366 | 2.100 | 1.869 | 1.677 | 1.527 | 1.369 | 1.267 | 1.164 | 1.070 |
| 3.0 | 4.167 | 3.722 | 3.323 | 2.959 | 2.603 | 2.307 | 2.057 | 1.846 | 1.671 | 1.524 | 1.414 | 1.285 |
| 4.5 | 5.819 | 5.158 | 4.570 | 4.045 | 3.546 | 3.124 | 2.767 | 2.469 | 2.223 | 2.010 | 1.842 | 1.677 |
| 6.0 | 9.082 | 7.821 | 6.735 | 5.802 | 4.988 | 4.395 | 3.869 | 3.436 | 3.077 | 2.762 | 2.538 | 2.335 |
| 8.0 | 14.963 | 12.709 | 10.782 | 9.203 | 7.807 | 6.784 | 5.908 | 5.171 | 4.578 | 4.089 | 3.735 | 3.437 |
| 10.0 | 25.437 | 21.163 | 17.617 | 14.676 | 12.335 | 10.561 | 9.071 | 7.924 | 6.884 | 6.076 | 5.603 | 5.009 |

different options of cell constants were available, for uniformity sake, the cell of 1 $\mathrm{cm}^{-1}$ was employed throughout. The instrument was first calibrated at 25 °C with standard aqueous KCl solutions,[1] and the calibration was also monitored from time to time during the experimentation. Previously reported[46] values for concentrated aqueous solutions of $Na_2SO_4$ at different temperatures could well be reproduced within ± 0.04 %. For the conductivity measurement, equal portions of the sample, taken in two identical narrow-necked cells, were first thermostatted in a bath of required temperature for about 20 min. One of them served for the conductivity measurement, while the ASTM-calibrated ther-

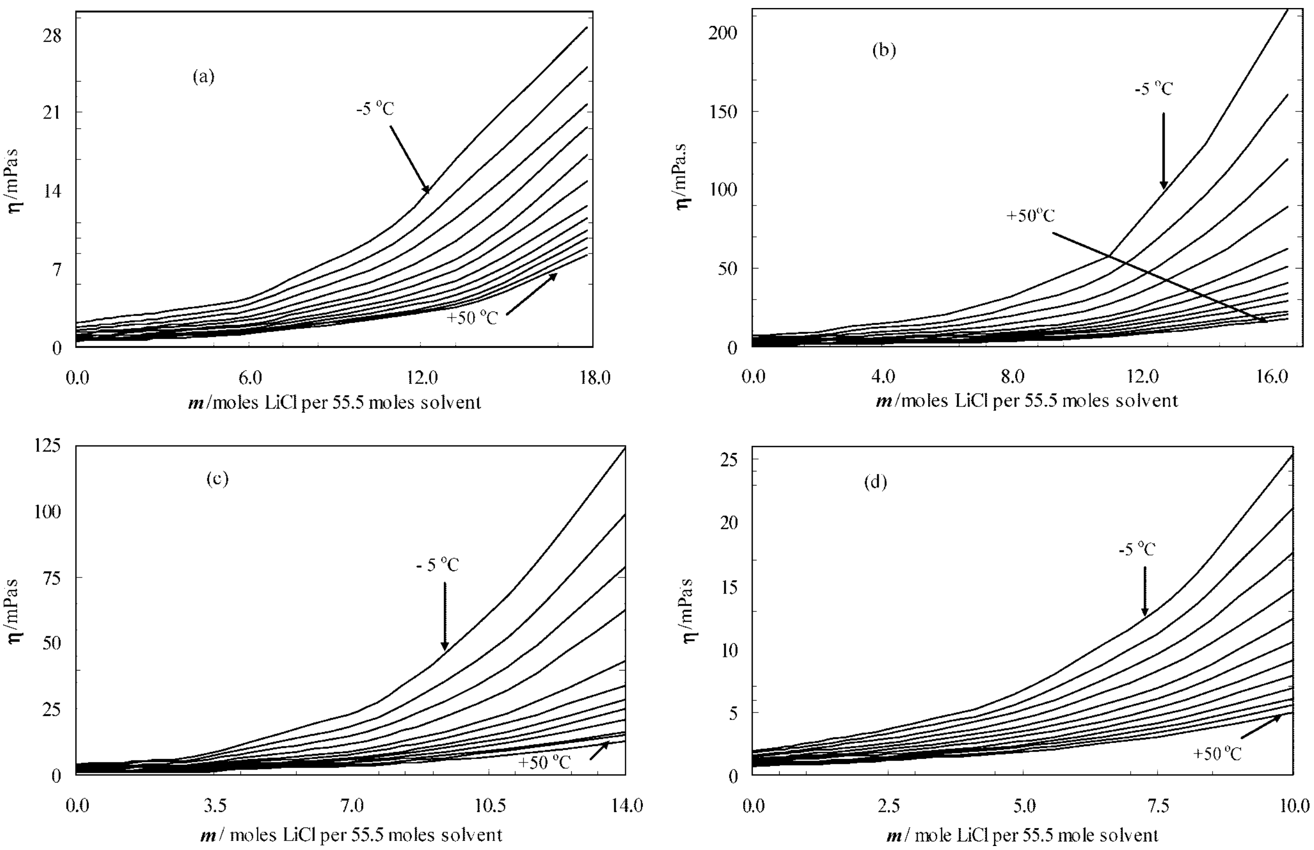

**Figure 4.** (a−d) Viscosity, $\eta$, fitted to LiCl aquamolality, $\boldsymbol{m}$, in the isotherm $\eta(\boldsymbol{m}) = \eta^\circ + a_1 \cdot \boldsymbol{m} + a_2 \cdot \boldsymbol{m}^2$ from −5 °C to +50 °C at 5 °C intervals. The binary ethanol mixtures of water mole fractions, $x_1$; $\eta^\circ$ refers to viscosity of the solvent mixtures (refer to Table 1) (a) $x_1 = 1.0$, (b) $x_1 = 0.7$, (c) $x_1 = 0.4$, and (d) $x_1 = 0.0$.

**Table 6. Coefficients (in Appropriate Units) of the Polynomial $\eta/\eta^\circ = 1 + B \cdot m + C \cdot m^2 + D \cdot m^3$ at Different Temperatures in Aqueous Mixtures of Ethanol of Various Water Mole Fractions $x_1$ (Other Details in the Text)**

| | | temperature, *t*/°C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $x_1$ | parameters | −5 | 0 | 5 | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 |
| 0.0 | *B* | 0.347 | 0.345 | 0.325 | 0.317 | 0.291 | 0.270 | 0.252 | 0.253 | 0.245 | 0.234 | 0.249 | 0.239 |
| | *C* | −0.029 | −0.026 | −0.021 | −0.017 | −0.014 | −0.007 | −0.004 | −0.006 | −0.002 | −0.002 | −0.004 | −0.002 |
| | *D* | 0.011 | 0.010 | 0.009 | 0.008 | 0.007 | 0.006 | 0.005 | 0.005 | 0.004 | 0.004 | 0.004 | 0.003 |
| 0.1 | *B* | 2.026 | 1.799 | 1.605 | 1.411 | 1.123 | 1.063 | 0.898 | 0.849 | 0.826 | 0.797 | 0.777 | 0.760 |
| | *C* | −0.771 | −0.667 | −0.577 | −0.487 | −0.369 | −0.343 | −0.278 | −0.261 | −0.254 | −0.222 | −0.216 | −0.198 |
| | *D* | 0.080 | 0.070 | 0.060 | 0.051 | 0.039 | 0.036 | 0.030 | 0.028 | 0.027 | 0.024 | 0.023 | 0.021 |
| 0.2 | *B* | 1.263 | 0.927 | 0.753 | 0.668 | 0.498 | 0.429 | 0.399 | 0.352 | 0.321 | 0.292 | 0.304 | 0.333 |
| | *C* | −0.433 | −0.304 | −0.236 | −0.200 | −0.131 | −0.104 | −0.090 | −0.074 | −0.066 | −0.051 | −0.055 | −0.058 |
| | *D* | 0.045 | 0.034 | 0.028 | 0.023 | 0.017 | 0.015 | 0.013 | 0.012 | 0.011 | 0.009 | 0.009 | 0.008 |
| 0.3 | *B* | 0.802 | 0.626 | 0.472 | 0.363 | 0.300 | 0.267 | 0.229 | 0.204 | 0.177 | 0.147 | 0.145 | 0.098 |
| | *C* | −0.251 | −0.185 | −0.129 | −0.088 | −0.057 | −0.044 | −0.030 | −0.022 | −0.015 | −0.001 | −0.001 | −0.016 |
| | *D* | 0.031 | 0.024 | 0.019 | 0.015 | 0.011 | 0.010 | 0.008 | 0.007 | 0.007 | 0.005 | 0.005 | 0.008 |
| 0.4 | *B* | 0.223 | 0.284 | 0.331 | 0.337 | 0.267 | 0.211 | 0.225 | 0.249 | 0.202 | 0.161 | 0.161 | 0.181 |
| | *C* | −0.013 | −0.019 | −0.038 | −0.042 | −0.019 | −0.009 | −0.001 | −0.007 | −0.004 | −0.022 | −0.021 | −0.014 |
| | *D* | 0.027 | 0.010 | 0.010 | 0.010 | 0.007 | 0.006 | 0.005 | 0.005 | 0.003 | 0.002 | 0.003 | 0.003 |
| 0.5 | *B* | 2.322 | 2.020 | 1.738 | 1.508 | 1.211 | 1.178 | 1.087 | 0.938 | 0.843 | 0.770 | 0.706 | 0.558 |
| | *C* | −0.641 | −0.549 | −0.461 | −0.394 | −0.303 | −0.292 | −0.260 | −0.219 | −0.193 | −0.169 | −0.151 | −0.112 |
| | *D* | 0.050 | 0.043 | 0.036 | 0.031 | 0.024 | 0.023 | 0.021 | 0.018 | 0.017 | 0.014 | 0.013 | 0.011 |
| 0.6 | *B* | 0.486 | 0.416 | 0.400 | 0.342 | 0.312 | 0.282 | 0.283 | 0.262 | 0.215 | 0.207 | 0.193 | 0.167 |
| | *C* | −0.096 | −0.076 | −0.072 | −0.057 | −0.047 | −0.038 | −0.033 | −0.027 | −0.017 | −0.012 | −0.010 | −0.002 |
| | *D* | 0.011 | 0.009 | 0.008 | 0.007 | 0.006 | 0.005 | 0.005 | 0.004 | 0.004 | 0.003 | 0.003 | 0.003 |
| 0.7 | *B* | 0.481 | 0.412 | 0.398 | 0.340 | 0.312 | 0.282 | 0.283 | 0.262 | 0.216 | 0.210 | 0.201 | 0.180 |
| | *C* | −0.096 | −0.076 | −0.072 | −0.057 | −0.047 | −0.038 | −0.033 | −0.027 | −0.017 | −0.012 | −0.011 | −0.003 |
| | *D* | 0.010 | 0.009 | 0.008 | 0.007 | 0.006 | 0.005 | 0.005 | 0.004 | 0.004 | 0.003 | 0.003 | 0.003 |
| 0.8 | *B* | 0.362 | 0.347 | 0.310 | 0.274 | 0.234 | 0.197 | 0.193 | 0.184 | 0.186 | 0.200 | 0.186 | 0.200 |
| | *C* | −0.055 | −0.052 | −0.044 | −0.035 | −0.026 | −0.014 | −0.015 | −0.010 | −0.011 | −0.013 | −0.011 | −0.013 |
| | *D* | 0.006 | 0.006 | 0.005 | 0.004 | 0.003 | 0.003 | 0.003 | 0.002 | 0.003 | 0.002 | 0.003 | 0.003 |
| 0.9 | *B* | 0.197 | 0.185 | 0.179 | 0.153 | 0.181 | 0.171 | 0.215 | 0.192 | 0.169 | 0.157 | 0.153 | 0.152 |
| | *C* | −0.017 | −0.016 | −0.014 | −0.009 | −0.010 | −0.007 | −0.015 | −0.011 | −0.008 | −0.009 | −0.009 | −0.011 |
| | *D* | 0.003 | 0.003 | 0.003 | 0.003 | 0.002 | 0.002 | 0.002 | 0.002 | 0.002 | 0.002 | 0.002 | 0.002 |
| 1.0 | *B* | 0.092 | 0.154 | 0.209 | 0.199 | 0.194 | 0.228 | 0.245 | 0.280 | 0.290 | 0.287 | 0.099 | 0.154 |
| | *C* | 0.011 | −0.006 | −0.018 | −0.016 | −0.014 | −0.022 | −0.027 | −0.035 | −0.036 | −0.034 | −0.011 | −0.006 |
| | *D* | 0.001 | 0.002 | 0.003 | 0.003 | 0.003 | 0.003 | 0.003 | 0.004 | 0.004 | 0.003 | 0.004 | 0.002 |

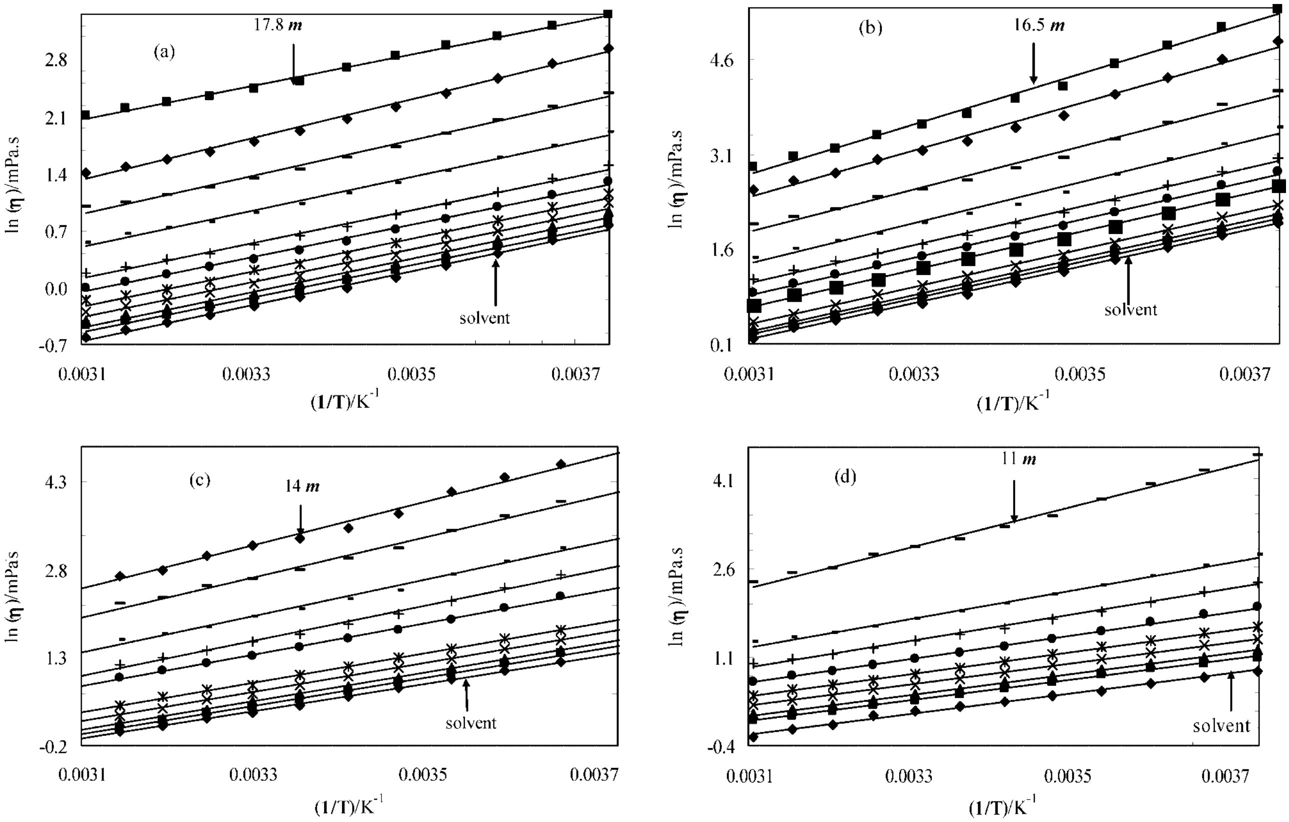

**Figure 5.** (a−d) ln($\eta$) vs 1/$T$ plots to determine flow activation energies ($E_a$) of the LiCl solutions in aqueous ethanol of different water mole fractions, $x_1$: (a) $x_1 = 1.0$, (b) $x_1 = 0.7$, (c) $x_1 = 0.4$, and (d) $x_1 = 0.1$.

**Table 7. Conductivity ($E_c$) and Flow ($E_a$) Activation Energies (kJ·mol$^{-1}$) for LiCl Solutions of Different Concentrations (*m*) in Aqueous Mixtures of Ethanol Having Water Mole Fractions, $x_1$[a]**

| | | aquamolality (***m***) | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| $x_1$ | *E*'s | 0.5 | 1.0 | 2.0 | 3.0 | 4.5 | 6.0 | 8.0 | 11.0 | 14.0 | 18.0 |
| 1.0 | $E_c$ | 15.31 | 14.32 | 14.83 | 15.46 | 18.01 | 16.84 | 15.93 | 15.92 | 17.15 | 20.04 (17.8) |
| | $E_a$ | 17.91 | 18.12 | 18.30 | 18.72 | 18.98 | 19.24 | 19.49 | 19.74 | 20.58 | 21.35 |
| 0.9 | $E_c$ | 19.63 | 20.67 | 19.80 | 19.58 | 19.07 | 18.87 | 19.07 | 19.83 | 21.50 | 24.16 (18.0) |
| | $E_a$ | 24.32 | 24.40 | 24.47 | 24.71 | 24.97 | 25.21 | 25.62 | 26.50 | 27.74 | 28.09 |
| 0.8 | $E_c$ | 23.34 | 22.96 | 21.23 | 20.66 | 20.09 | 20.10 | 21.51 | 22.40 | 23.67 | 25.42 (17.5) |
| | $E_a$ | 24.50 | 24.73 | 25.01 | 25.36 | 25.58 | 25.81 | 26.82 | 27.91 | 29.47 | 31.50 |
| 0.7 | $E_c$ | 21.74 | 20.91 | 20.63 | 20.51 | 20.41 | 20.52 | 22.05 | 21.68 | 25.18 | 26.85 (16.5) |
| | $E_a$ | 23.91 | 24.12 | 24.40 | 24.57 | 24.82 | 25.51 | 26.72 | 27.91 | 30.74 | 32.92 |
| 0.6 | $E_c$ | 20.38 | 19.76 | 19.58 | 19.48 | 19.51 | 20.09 | 20.59 | 22.25 | 25.39 | 27.51 (16.0) |
| | $E_a$ | 22.33 | 22.61 | 22.88 | 23.73 | 24.02 | 25.89 | 26.61 | 28.72 | 31.63 | 34.51 |
| 0.5 | $E_c$ | 20.02 | 18.71 | 18.25 | 18.70 | 18.61 | 18.48 | 20.54 | 22.66 | 25.56 | 26.63 (15.2) |
| | $E_a$ | 20.92 | 21.08 | 21.43 | 23.14 | 23.51 | 25.23 | 26.50 | 29.12 | 32.90 | 34.93 |
| 0.4 | $E_c$ | 17.57 | 17.43 | 17.52 | 17.57 | 18.17 | 18.57 | 19.75 | 22.23 | 25.75 (14.0) | |
| | $E_a$ | 19.90 | 19.96 | 20.50 | 20.83 | 22.20 | 24.63 | 25.41 | 28.07 | 31.21 | |
| 0.3 | $E_c$ | 16.96 | 16.25 | 16.39 | 16.72 | 17.38 | 18.32 | 19.61 | 22.50 | 26.61 (13.5) | |
| | $E_a$ | 17.97 | 18.31 | 18.50 | 18.90 | 20.52 | 21.81 | 22.50 | 27.82 | 31.30 | |
| 0.2 | $E_c$ | 15.69 | 15.18 | 15.68 | 16.14 | 16.83 | 17.60 | 19.05 | 21.94 | 25.46 (13.0) | |
| | $E_a$ | 16.30 | 16.67 | 17.14 | 17.41 | 18.02 | 20.01 | 21.00 | 27.48 | 30.71 | |
| 0.1 | $E_c$ | 15.93 | 14.10 | 14.82 | 15.34 | 16.50 | 17.43 | 18.95 | 23.86 (11.0) | | |
| | $E_a$ | 14.42 | 14.81 | 15.66 | 16.31 | 17.05 | 18.91 | 20.20 | 27.32 | | |
| 0.0 | $E_c$ | 14.86 | 13.26 | 14.45 | 15.21 | 16.66 | 17.54 | 18.78 | 20.23 (10.0) | | |
| | $E_a$ | 13.83 | 14.02 | 14.91 | 15.74 | 16.62 | 18.13 | 19.60 | 21.62 | | |

[a] Values in parentheses correspond to the highest value of ***m***.

mometer was dipped in the other in such a way that positions of the electrode and thermometer were maintained each time. Subsequently, the mouths of the cells were closed with paraffin film to minimize any variation of the sample composition. Conductivity values could be reproduced within ± 1 % for some randomly selected samples in the subsequent sessions.

## Results and Discussion

Table 1 presents density data for the solvent systems and their LiCl solutions, while the density isotherms at some selected compositions have been plotted against LiCl aquamolality (***m***) in Figure 1a−d. Our measured densities (at 20 °C) for *the mixtures* ($\rho°$) and aqueous LiCl solutions fall

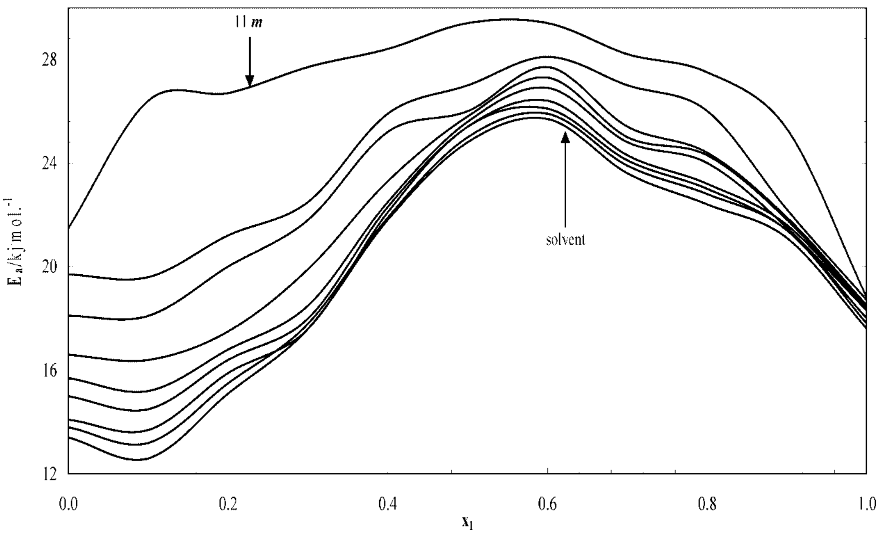

**Figure 6.** Variation trend of the flow activation energy ($E_a$) with water mole fraction, $x_1$, in aqueous ethanol at different LiCl*m* (from bottom to top): 0.0, 0.5, 1.0, 2.0, 3.0, 4.5, 6.0, 8.0, and 11.0 *m*.

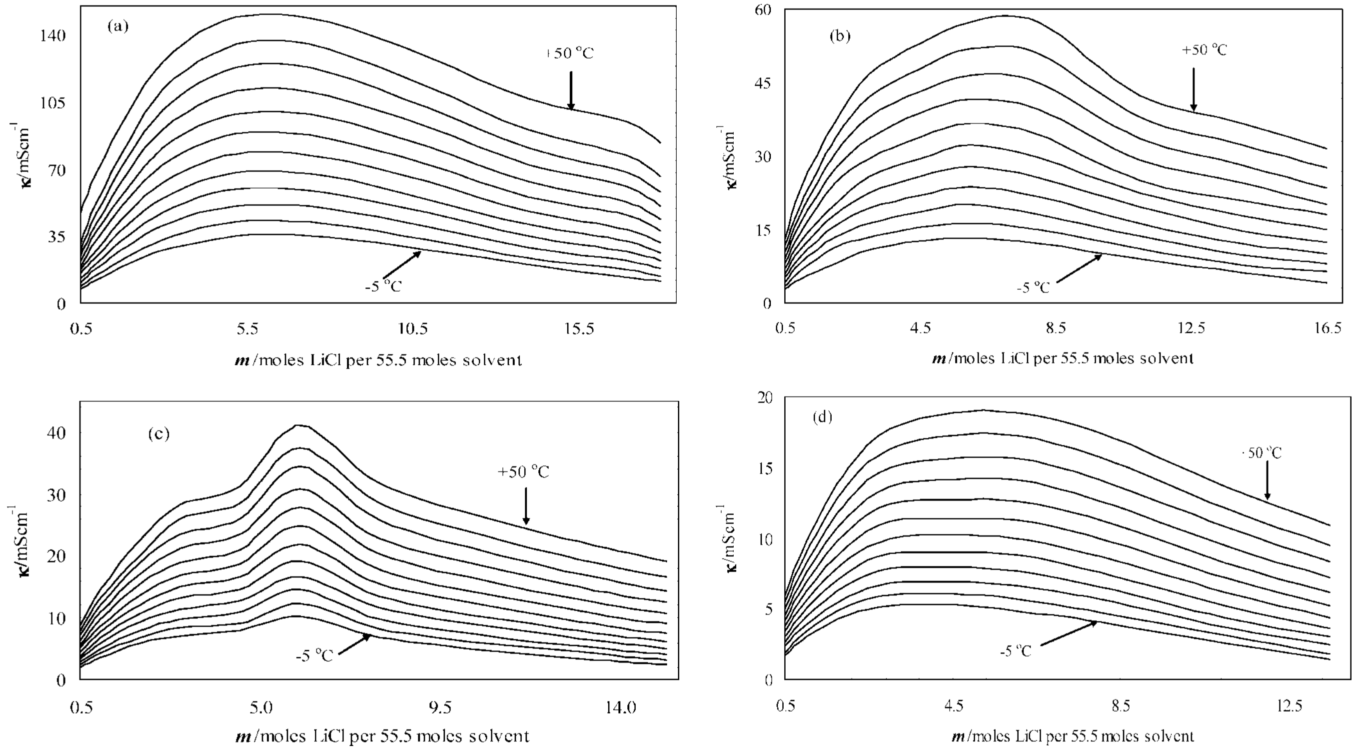

**Figure 7.** (a−d) Electrical conductivity isotherms plotted against LiCl aquamolality (*m*) in aqueous ethanol of different water mole fractions, $x_1$: (a) $x_1 = 0.9$, (b) $x_1 = 0.7$, (c) $x_1 = 0.5$, and (d) $x_1 = 0.3$.

within ± 0.05 % of the corresponding literature values.[1,2,40,47,48] As expected, the density appreciably changed with both solvent composition and LiCl concentration. All the solutions exhibited almost a linear density increase with *m* making the $\rho$-isotherms fit a second order eq 1.

$$\rho(t,\boldsymbol{m}) = d_o(t) + d_1(t)\cdot\boldsymbol{m} + d_2(t)\cdot\boldsymbol{m}^2 \qquad (1)$$

where $d_o$ is the fitted density of the neat solvent or mixture, whose values have deviated only up to 0.1 % from the respective measured values, $\rho_o$ (ref Table 1) and $d_1$ and $d_2$ are the corresponding density coefficients whose values are also listed in Table 2. A closer inspection of the plots suggests that change of density with LiCl concentration is greater when water is in excess, but on the contrary, the variation with temperature (though minor) is more pronounced when ethanol is in excess. Since the concentration gradient $\mathbf{g}(x_1)$ of eq 1 does not appreciably change with temperature, it can be regarded as a specific parameter for a given solute−solvent system and can conveniently be used as a density index. Due to a negligible contribution of the term involving $\boldsymbol{m}^2$ for *the system*, $\mathbf{g}(x_1)$ can simply be compared with $d_1$ of eq 1. The **g**-values (included in Table 1 and also plotted against the mixture compositions in Figure 2) can be employed for fairly accurate determination of solution density at any LiCl concentration and solvent composition within the given temperature range. Consequently, the density of *the system*

**Table 8. Electrical Conductivity ($\kappa$/mS·cm$^{-1}$) at Different Temperatures and LiCl Aquamolalities ($m$) in Binary Mixtures of Water (1) and Ethanol (2) Having Mole Fractions $x_1$ and $x_2$**

| concentration | temperature, $t$/°C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| aquamolality/$m$ | −5 | 0 | 5 | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 |
| $x_1 = 1.0$[a], $x_2 = 0.0$ | | | | | | | | | | | | |
| 0.5 | – | – | 26.100 | 29.900 | 33.500 | 37.400 | 41.500 | 45.800 | 49.900 | 54.300 | 58.500 | 62.800 |
| 1.0 | – | – | 46.400 | 52.000 | 58.700 | 65.300 | 72.900 | 80.000 | 87.000 | 94.500 | 102.200 | 110.100 |
| 2.0 | – | – | 76.400 | 86.100 | 96.200 | 107.400 | 120.400 | 132.000 | 144.200 | 156.400 | 168.300 | 180.800 |
| 3.0 | – | – | 95.900 | 109.600 | 122.600 | 137.100 | 152.400 | 166.800 | 181.900 | 197.400 | [b] | [b] |
| 4.5 | – | – | 114.800 | 130.000 | 144.600 | 160.800 | 176.800 | 194.200 | [b] | [b] | [b] | [b] |
| 6.0 | – | – | 120.400 | 136.200 | 152.300 | 168.900 | 186.600 | [b] | [b] | [b] | [b] | [b] |
| 8.0 | – | – | 115.300 | 130.700 | 146.100 | 162.700 | 179.500 | 197.300 | [b] | [b] | [b] | [b] |
| 11.0 | – | – | 93.700 | 106.900 | 120.800 | 135.600 | 151.000 | 167.000 | 182.600 | 199.700 | [b] | [b] |
| 14.0 | – | – | 69.900 | 80.100 | 90.900 | 103.600 | 117.000 | 131.100 | 145.400 | 160.400 | 175.400 | 191.400 |
| 17.8 | – | – | 47.100 | 55.200 | 64.500 | 74.400 | 85.000 | 96.100 | 108.600 | 122.600 | 136.400 | 150.600 |
| $x_1 = 0.9$, $x_2 = 0.1$ | | | | | | | | | | | | |
| 0.5 | 7.340 | 9.020 | 10.980 | 13.140 | 15.340 | 17.840 | 19.200 | 21.800 | 24.550 | 27.300 | 30.400 | 33.100 |
| 1.0 | 12.120 | 14.820 | 17.950 | 21.040 | 24.550 | 28.050 | 33.200 | 37.650 | 42.100 | 47.400 | 52.500 | 57.400 |
| 2.0 | 20.800 | 25.300 | 30.400 | 36.100 | 41.800 | 48.200 | 55.200 | 61.900 | 69.800 | 77.600 | 85.300 | 93.500 |
| 3.0 | 27.600 | 33.500 | 40.300 | 47.500 | 55.100 | 63.000 | 72.400 | 81.300 | 90.700 | 100.600 | 111.600 | 122.200 |
| 4.5 | 33.300 | 40.700 | 48.600 | 57.300 | 65.900 | 74.800 | 85.100 | 95.600 | 107.000 | 118.700 | 130.100 | 143.300 |
| 6.0 | 36.100 | 43.200 | 51.200 | 60.000 | 69.100 | 78.900 | 89.300 | 100.500 | 112.600 | 124.900 | 137.600 | 150.300 |
| 8.0 | 34.500 | 41.100 | 49.300 | 57.500 | 66.500 | 75.900 | 85.800 | 96.800 | 108.600 | 120.800 | 133.000 | 146.100 |
| 11.0 | 28.000 | 33.500 | 40.200 | 47.300 | 54.400 | 63.300 | 72.500 | 81.700 | 92.000 | 102.200 | 113.000 | 125.500 |
| 14.0 | 20.100 | 24.000 | 29.100 | 35.100 | 41.000 | 48.100 | 55.500 | 63.400 | 72.300 | 81.500 | 90.600 | 101.000 |
| 18.0 | 11.620 | 14.480 | 18.030 | 22.100 | 26.700 | 32.000 | 37.800 | 44.200 | 51.100 | 58.300 | 66.100 | 74.400 |
| $x_1 = 0.8$, $x_2 = 0.2$ | | | | | | | | | | | | |
| 0.5 | 3.300 | 4.220 | 5.240 | 6.380 | 7.650 | 9.040 | 10.53 | 12.11 | 13.79 | 15.53 | 17.35 | 19.23 |
| 1.0 | 6.210 | 8.000 | 9.830 | 11.91 | 14.21 | 16.70 | 19.38 | 22.40 | 25.40 | 28.70 | 32.00 | 35.30 |
| 2.0 | 10.54 | 15.04 | 18.30 | 21.60 | 26.20 | 29.50 | 33.80 | 37.70 | 42.50 | 47.10 | 51.80 | 57.00 |
| 3.0 | 13.76 | 19.25 | 23.90 | 28.40 | 32.70 | 37.50 | 42.40 | 47.80 | 53.50 | 59.50 | 64.70 | 70.90 |
| 4.5 | 16.85 | 23.70 | 29.80 | 32.70 | 39.40 | 44.40 | 50.60 | 56.70 | 63.40 | 70.00 | 76.90 | 83.50 |
| 6.0 | 17.93 | 26.80 | 32.00 | 36.60 | 42.80 | 48.90 | 55.70 | 62.20 | 69.50 | 76.80 | 84.40 | 91.80 |
| 8.0 | 15.30 | 20.00 | 24.30 | 28.90 | 34.10 | 40.70 | 45.60 | 52.00 | 58.60 | 65.50 | 73.20 | 80.20 |
| 11.0 | 11.62 | 14.64 | 17.86 | 21.20 | 25.30 | 29.70 | 34.60 | 39.70 | 45.20 | 51.10 | 57.10 | 63.60 |
| 14.0 | 8.940 | 11.38 | 14.05 | 17.05 | 20.20 | 24.00 | 28.10 | 32.60 | 37.50 | 42.70 | 48.30 | 54.30 |
| 17.5 | 6.700 | 8.680 | 10.90 | 13.42 | 16.31 | 19.58 | 23.00 | 27.20 | 31.30 | 36.20 | 41.10 | 46.50 |
| $x_1 = 0.7$, $x_2 = 0.3$ | | | | | | | | | | | | |
| 0.5 | 2.790 | 3.480 | 4.220 | 5.070 | 5.970 | 6.950 | 8.000 | 9.090 | 10.32 | 11.61 | 12.91 | 14.30 |
| 1.0 | 4.900 | 6.820 | 8.240 | 9.670 | 11.13 | 12.80 | 14.59 | 16.45 | 18.34 | 20.30 | 23.60 | 25.60 |
| 2.0 | 8.040 | 11.28 | 13.39 | 15.67 | 18.18 | 20.20 | 24.60 | 27.30 | 30.50 | 33.60 | 36.90 | 40.50 |
| 3.0 | 10.97 | 13.56 | 16.34 | 19.32 | 22.40 | 25.90 | 29.60 | 33.70 | 38.00 | 42.20 | 46.70 | 51.20 |
| 4.5 | 12.61 | 15.40 | 18.60 | 22.00 | 25.80 | 29.60 | 34.00 | 38.90 | 43.20 | 47.90 | 52.90 | 58.60 |
| 6.0 | 13.12 | 16.39 | 20.30 | 23.90 | 27.90 | 32.30 | 36.70 | 41.60 | 46.50 | 51.90 | 57.40 | 63.20 |
| 8.0 | 12.11 | 14.96 | 18.08 | 21.60 | 25.50 | 29.80 | 34.50 | 39.60 | 45.10 | 51.00 | 57.30 | 64.00 |
| 11.0 | 9.100 | 11.18 | 13.72 | 16.42 | 19.57 | 22.40 | 25.20 | 29.10 | 33.50 | 37.90 | 42.40 | 47.40 |
| 14.0 | 6.170 | 7.840 | 9.860 | 12.12 | 14.82 | 17.76 | 21.00 | 24.50 | 28.20 | 32.30 | 36.70 | 41.40 |
| 16.5 | 4.180 | 6.500 | 8.140 | 10.13 | 12.47 | 15.07 | 18.08 | 20.10 | 23.60 | 27.60 | 31.50 | 36.00 |
| $x_1 = 0.6$, $x_2 = 0.4$ | | | | | | | | | | | | |
| 0.5 | 2.390 | 2.880 | 3.440 | 4.060 | 4.750 | 5.470 | 6.250 | 7.100 | 7.960 | 8.890 | 9.850 | 10.83 |
| 1.0 | 4.150 | 5.070 | 6.050 | 7.090 | 8.240 | 9.490 | 10.82 | 12.22 | 13.69 | 15.25 | 16.87 | 17.62 |
| 2.0 | 6.830 | 8.290 | 9.960 | 11.63 | 13.48 | 15.50 | 17.58 | 19.84 | 22.10 | 24.50 | 27.10 | 29.60 |
| 3.0 | 8.460 | 10.19 | 12.23 | 14.33 | 16.60 | 19.07 | 21.60 | 24.40 | 27.00 | 30.10 | 33.00 | 36.30 |
| 4.5 | 9.840 | 11.84 | 14.20 | 16.60 | 19.30 | 22.10 | 25.10 | 28.20 | 31.40 | 35.10 | 38.70 | 42.40 |
| 6.0 | 9.850 | 12.02 | 14.30 | 16.85 | 19.72 | 22.70 | 25.80 | 29.20 | 32.80 | 36.60 | 40.40 | 44.60 |
| 8.0 | 8.670 | 10.55 | 12.77 | 15.08 | 17.69 | 20.00 | 23.00 | 26.20 | 29.60 | 33.30 | 37.20 | 41.00 |
| 11.0 | 6.770 | 8.100 | 9.890 | 11.91 | 14.22 | 16.65 | 19.37 | 21.80 | 25.00 | 28.40 | 32.00 | 35.80 |
| 14.0 | 4.100 | 5.160 | 6.570 | 8.100 | 9.780 | 11.72 | 13.95 | 16.43 | 19.08 | 21.30 | 24.50 | 27.90 |
| 16.0 | 3.030 | 3.870 | 5.030 | 6.290 | 7.730 | 9.400 | 11.34 | 13.44 | 15.81 | 18.46 | 21.10 | 24.10 |
| $x_1 = 0.5$, $x_2 = 0.5$ | | | | | | | | | | | | |
| 0.5 | 2.120 | 2.510 | 2.970 | 3.450 | 3.990 | 4.560 | 5.160 | 5.800 | 6.500 | 7.220 | 8.010 | 8.800 |
| 1.0 | 3.630 | 4.300 | 5.100 | 5.940 | 6.780 | 7.710 | 8.720 | 9.790 | 10.92 | 12.12 | 13.34 | 14.58 |
| 2.0 | 5.830 | 6.890 | 8.060 | 9.360 | 10.76 | 12.16 | 13.76 | 15.44 | 17.23 | 19.02 | 20.10 | 22.70 |
| 3.0 | 7.060 | 8.410 | 9.900 | 11.46 | 13.18 | 15.00 | 16.94 | 19.00 | 21.20 | 23.50 | 26.00 | 28.30 |
| 4.5 | 7.940 | 9.250 | 10.91 | 12.70 | 14.67 | 16.64 | 18.84 | 20.98 | 23.50 | 26.00 | 28.60 | 31.40 |
| 6.0 | 10.39 | 12.37 | 14.51 | 16.77 | 19.27 | 21.90 | 24.90 | 27.90 | 30.90 | 34.40 | 37.50 | 41.20 |
| 8.0 | 6.830 | 8.210 | 9.830 | 11.54 | 13.45 | 15.61 | 17.84 | 20.30 | 22.90 | 25.70 | 28.60 | 31.70 |
| 11.0 | 4.650 | 5.690 | 6.930 | 8.370 | 9.920 | 11.64 | 13.51 | 15.52 | 17.80 | 20.10 | 22.70 | 25.50 |
| 14.0 | 3.030 | 3.790 | 4.730 | 5.860 | 7.110 | 8.520 | 10.11 | 11.85 | 13.73 | 15.87 | 18.15 | 20.70 |
| 15.2 | 2.570 | 3.250 | 4.170 | 5.090 | 6.230 | 7.550 | 9.030 | 10.71 | 12.59 | 14.47 | 16.67 | 19.13 |
| $x_1 = 0.4$, $x_2 = 0.6$ | | | | | | | | | | | | |
| 0.5 | 1.980 | 2.290 | 2.650 | 3.060 | 3.470 | 3.910 | 4.390 | 4.910 | 5.440 | 5.990 | 6.570 | 7.180 |
| 1.0 | 3.340 | 3.850 | 4.480 | 5.140 | 5.830 | 6.590 | 7.380 | 8.220 | 9.100 | 10.02 | 11.00 | 11.98 |
| 2.0 | 5.160 | 6.020 | 6.950 | 7.970 | 9.090 | 10.25 | 11.52 | 12.84 | 14.24 | 15.68 | 17.18 | 18.74 |
| 3.0 | 6.160 | 7.220 | 8.360 | 9.600 | 10.93 | 12.35 | 13.85 | 15.48 | 17.17 | 18.91 | 20.70 | 22.60 |
| 4.5 | 6.770 | 7.940 | 9.300 | 10.70 | 12.27 | 13.83 | 15.60 | 17.48 | 19.47 | 21.60 | 23.70 | 26.10 |
| 6.0 | 6.550 | 7.700 | 8.990 | 10.46 | 12.03 | 13.69 | 15.49 | 17.35 | 19.38 | 21.40 | 23.60 | 26.00 |
| 8.0 | 5.530 | 6.560 | 7.750 | 9.080 | 10.54 | 12.11 | 13.79 | 15.61 | 17.58 | 19.64 | 21.70 | 23.90 |

**Table 8 Continued**

| concentration | temperature, $t$/°C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| aquamolality/$\boldsymbol{m}$ | −5 | 0 | 5 | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 |
| 11.0 | 3.680 | 4.450 | 5.400 | 6.390 | 7.570 | 8.870 | 10.32 | 11.90 | 13.57 | 15.34 | 17.25 | 19.32 |
| 14.0 | 2.200 | 2.750 | 3.420 | 4.230 | 5.140 | 6.160 | 7.320 | 8.640 | 10.00 | 11.66 | 13.36 | 15.21 |
| | | | | | | $x_1 = 0.3$, $x_2 = 0.7$ | | | | | | |
| 0.5 | 1.691 | 1.933 | 2.380 | 2.690 | 3.030 | 3.400 | 3.780 | 4.170 | 4.590 | 5.030 | 5.500 | 5.950 |
| 1.0 | 2.990 | 3.420 | 3.910 | 4.450 | 5.010 | 5.590 | 6.230 | 6.900 | 7.600 | 8.290 | 9.000 | 9.780 |
| 2.0 | 4.520 | 5.200 | 5.920 | 6.740 | 7.600 | 8.510 | 9.470 | 10.52 | 11.56 | 12.68 | 13.80 | 15.01 |
| 3.0 | 5.210 | 5.990 | 6.870 | 7.830 | 8.870 | 9.940 | 11.20 | 12.41 | 13.61 | 14.93 | 16.33 | 17.75 |
| 4.5 | 5.290 | 6.020 | 6.920 | 7.960 | 9.030 | 10.19 | 11.41 | 12.74 | 14.16 | 15.68 | 17.29 | 18.90 |
| 6.0 | 4.860 | 5.700 | 6.640 | 7.680 | 8.790 | 9.990 | 11.27 | 12.64 | 14.13 | 15.62 | 17.22 | 18.88 |
| 8.0 | 4.070 | 4.760 | 5.660 | 6.560 | 7.580 | 8.710 | 9.940 | 11.27 | 12.63 | 14.13 | 15.70 | 17.40 |
| 11.0 | 2.560 | 3.090 | 3.710 | 4.400 | 5.240 | 6.150 | 7.160 | 8.260 | 9.460 | 10.78 | 12.17 | 13.63 |
| 13.5 | 1.460 | 1.830 | 2.440 | 2.990 | 3.640 | 4.360 | 5.210 | 6.140 | 7.200 | 8.300 | 9.500 | 10.87 |
| | | | | | | $x_1 = 0.2$, $x_2 = 0.8$ | | | | | | |
| 0.5 | 1.480 | 1.670 | 1.880 | 2.210 | 2.460 | 2.730 | 3.010 | 3.310 | 3.640 | 3.950 | 4.250 | 4.590 |
| 1.0 | 2.470 | 2.810 | 3.170 | 3.550 | 3.970 | 4.410 | 4.860 | 5.360 | 5.850 | 6.370 | 6.910 | 7.450 |
| 2.0 | 3.580 | 4.110 | 4.640 | 5.250 | 5.860 | 6.540 | 7.180 | 7.990 | 8.780 | 9.590 | 10.43 | 11.27 |
| 3.0 | 4.150 | 4.750 | 5.430 | 6.150 | 6.890 | 7.710 | 8.590 | 9.500 | 10.46 | 11.45 | 12.48 | 13.56 |
| 4.5 | 4.350 | 5.040 | 5.760 | 6.550 | 7.410 | 8.340 | 9.310 | 10.35 | 11.47 | 12.56 | 13.80 | 15.06 |
| 6.0 | 4.060 | 4.700 | 5.400 | 6.210 | 7.070 | 7.990 | 8.990 | 10.03 | 11.17 | 12.35 | 13.54 | 14.84 |
| 8.0 | 3.260 | 3.820 | 4.460 | 5.180 | 5.960 | 6.770 | 7.740 | 8.730 | 9.770 | 10.90 | 12.11 | 13.37 |
| 11.0 | 2.070 | 2.450 | 2.940 | 3.500 | 4.120 | 4.790 | 5.570 | 6.430 | 7.330 | 8.310 | 9.370 | 10.51 |
| 13.0 | 1.315 | 1.619 | 1.989 | 2.530 | 3.020 | 3.620 | 4.280 | 5.020 | 5.830 | 6.750 | 7.710 | 8.760 |
| | | | | | | $x_1 = 0.1$, $x_2 = 0.9$ | | | | | | |
| 0.5 | 1.305 | 1.461 | 1.644 | 1.821 | 2.300 | 2.510 | 2.740 | 2.980 | 3.230 | 3.500 | 3.760 | 4.040 |
| 1.0 | 2.410 | 2.710 | 3.030 | 3.390 | 3.730 | 4.110 | 4.510 | 4.930 | 5.360 | 5.790 | 6.240 | 6.690 |
| 2.0 | 3.310 | 3.750 | 4.210 | 4.710 | 5.240 | 5.820 | 6.430 | 7.050 | 7.670 | 8.350 | 9.010 | 9.710 |
| 3.0 | 3.780 | 4.260 | 4.820 | 5.430 | 6.040 | 6.720 | 7.440 | 8.200 | 9.000 | 9.780 | 10.66 | 11.56 |
| 4.5 | 3.820 | 4.360 | 4.960 | 5.610 | 6.340 | 7.200 | 7.970 | 8.830 | 9.770 | 10.69 | 11.70 | 12.75 |
| 6.0 | 3.510 | 4.030 | 4.650 | 5.341 | 6.030 | 6.830 | 7.660 | 8.530 | 9.510 | 10.49 | 11.58 | 12.68 |
| 8.0 | 2.740 | 3.210 | 3.760 | 4.310 | 4.950 | 5.660 | 6.440 | 7.280 | 8.120 | 9.090 | 10.07 | 11.17 |
| 11.0 | 1.527 | 1.890 | 2.560 | 3.020 | 3.540 | 4.120 | 4.760 | 5.540 | 6.310 | 7.190 | 8.070 | 9.110 |
| | | | | | | $x_1 = 0.0$, $x_2 = 1.0$ | | | | | | |
| 0.5 | 1.172 | 1.303 | 1.438 | 1.580 | 1.720 | 1.880 | 2.310 | 2.500 | 2.700 | 2.890 | 3.090 | 3.270 |
| 1.0 | 1.970 | 2.210 | 2.450 | 2.700 | 2.950 | 3.250 | 3.540 | 3.840 | 4.200 | 4.470 | 4.800 | 5.120 |
| 2.0 | 2.690 | 3.000 | 3.360 | 3.700 | 4.150 | 4.590 | 5.060 | 5.540 | 6.050 | 6.520 | 7.080 | 7.620 |
| 3.0 | 3.000 | 3.350 | 3.830 | 4.410 | 4.770 | 5.330 | 5.890 | 6.490 | 7.110 | 7.730 | 8.430 | 9.110 |
| 4.5 | 3.050 | 3.430 | 3.940 | 4.450 | 5.050 | 5.650 | 6.290 | 6.970 | 7.720 | 8.520 | 9.310 | 10.20 |
| 6.0 | 2.760 | 3.150 | 3.620 | 4.170 | 4.720 | 5.290 | 6.000 | 6.700 | 7.460 | 8.220 | 9.100 | 9.980 |
| 8.0 | 2.220 | 2.580 | 3.000 | 3.480 | 3.960 | 4.500 | 5.140 | 5.750 | 6.440 | 7.120 | 7.920 | 8.750 |
| 10.0 | 1.620 | 1.980 | 2.380 | 2.760 | 3.220 | 3.670 | 4.270 | 4.870 | 5.510 | 6.240 | 6.970 | 7.830 |

[a] Conductivities of the aqueous solutions at (−5 and 0) °C were not readable on the instrument due to fluctuations. [b] Values exceeded instrument's maximum range of 200 mS·cm$^{-1}$.

can well be summarized as the following general expression

$$\rho(x_1,\boldsymbol{m},t) = \rho_o(x_1,t) + \mathbf{g}(x_1)\cdot\boldsymbol{m} \qquad (2)$$

Rapid increase of the **g**-value on increasing $x_1$ in water-rich solutions may be attributed to both preferential solvation by water molecules and its making compacter solvates.[48]

A rather useful thermodynamic property related to density of any solvent mixture is its excess molar volume, $V^E(T)$, which for binary mixtures may be expressed as

$$V^E = [(x_1M_1 + x_2M_2)/\rho] - [(x_1M_1/\rho_1) + (x_2M_2/\rho_2)] \qquad (3)$$

where $x_{1,2}$, $M_{1,2}$, and $\rho_{1,2}$ are the mole fractions, molar masses, and densities of the two components (here water and ethanol, respectively), and $\rho$ is the experimentally measured density of *the mixture*. Excess molar volume, $V^E$, of *the mixture* has been found to depend not only on its composition but also on temperature. $V^E$ is an important quantity because it is a sensitive function of intermolecular interactions; therefore, its values for a given system should be measured over the entire composition and an extended temperature range. $V^E$ for *the mixture* at different temperatures and water mole fractions have been listed in Table 3, while $V^E$ has been plotted against mixture composition at a series of temperatures in Figure 3. The $V^E$ isotherms remain in the negative domain over the entire composition range and exhibit minima as well (the 25 °C plot in good agreement with the reported results[47–49]). Minima of all the $V^E$ isotherms at the same water mole fraction of about 0.6 (Figure 3) suggest that temperature variation does not bring about any significant structural rearrangement in the solvent mixtures. The $V^E$ values collected in Table 3 can be reproduced within 5 % at any desired water mole fraction ($x_1$) and temperature ($T$/K) by using the following expression

$$\begin{aligned} V^E(T) = {} & [0.29 \pm 0.02 - (93 \pm 5)/T] + \\ & [1.56 \pm 0.09 - (1370 \pm 80)/T]x_1 + \\ & [-0.47 \pm 0.03 + (270 \pm 15)/T]x_1^{\,2} + \\ & [-1.10 \pm 0.07 + (1110 \pm 70)/T]x_1^{\,3} \qquad (4) \end{aligned}$$

Variation of volume with temperature is normally related to the thermal expansion coefficient,[46] $\alpha(t) \equiv -d(\ln \rho)/dt$. How-

**Table 9. Coefficients (in Appropriate Units) of the Equation $\kappa(T) = k_0 + k_1 \cdot m + k_2 \cdot m^2 + k_3 \cdot m^3$ at Different Temperatures in Binary Mixtures of Water (1) and Ethanol (2) Having Mole Fractions $x_1$ and $x_2$ ($k_0$ Has Been Adjusted to Zero)**

| | temperature, $t$/°C | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $k$'s | −5 | 0 | 5 | 10 | 15 | 20 | 25 | 30 | 35 | 40 | 45 | 50 |
| | | | | | | $x_1 = 1.0, x_2 = 0.0$ | | | | | | |
| $k_1$ | 34.25 | 39.14 | 44.62 | 50.54 | 56.38 | 62.62 | 69.23 | 76.09 | 85.37 | 98.23 | 112.13 | 128.73 |
| $k_2$ | −3.64 | −4.18 | −4.78 | −5.41 | −6.03 | −6.68 | −7.38 | −8.09 | −9.14 | −10.62 | −12.22 | −14.10 |
| $k_3$ | 0.100 | 0.120 | 0.140 | 0.150 | 0.170 | 0.190 | 0.210 | 0.230 | 0.260 | 0.310 | 0.360 | 0.410 |
| | | | | | | $x_1 = 0.9, x_2 = 0.1$ | | | | | | |
| $k_1$ | 12.54 | 15.26 | 18.24 | 21.43 | 24.79 | 28.19 | 32.13 | 36.11 | 40.37 | 44.8 | 49.36 | 53.96 |
| $k_2$ | −1.29 | −1.59 | −1.90 | −2.24 | −2.60 | −2.94 | −3.36 | −3.77 | −4.21 | −4.67 | −5.15 | −5.60 |
| $k_3$ | 0.035 | 0.044 | 0.053 | 0.062 | 0.073 | 0.082 | 0.095 | 0.110 | 0.120 | 0.132 | 0.146 | 0.159 |
| | | | | | | $x_1 = 0.8, x_2 = 0.2$ | | | | | | |
| $k_1$ | 6.65 | 9.62 | 11.86 | 13.6 | 16.11 | 18.29 | 20.75 | 23.24 | 25.97 | 28.73 | 31.43 | 34.22 |
| $k_2$ | −0.76 | −1.14 | −1.42 | −1.62 | −1.92 | −2.15 | −2.44 | −2.72 | −3.03 | −3.34 | −3.63 | −3.94 |
| $k_3$ | 0.020 | 0.040 | 0.050 | 0.050 | 0.060 | 0.070 | 0.080 | 0.090 | 0.100 | 0.110 | 0.110 | 0.120 |
| | | | | | | $x_1 = 0.7, x_2 = 0.3$ | | | | | | |
| $k_1$ | 5.06 | 6.54 | 7.88 | 9.27 | 10.74 | 12.31 | 14.29 | 16.09 | 17.90 | 19.83 | 21.94 | 24.09 |
| $k_2$ | −0.57 | −0.77 | −0.92 | −1.08 | −1.24 | −1.42 | −1.66 | −1.84 | −2.04 | −2.24 | −2.47 | −2.70 |
| $k_3$ | 0.017 | 0.024 | 0.029 | 0.034 | 0.039 | 0.044 | 0.052 | 0.057 | 0.063 | 0.070 | 0.077 | 0.083 |
| | | | | | | $x_1 = 0.6, x_2 = 0.4$ | | | | | | |
| $k_1$ | 4.25 | 5.16 | 6.18 | 7.21 | 8.33 | 9.58 | 10.83 | 12.26 | 13.58 | 15.03 | 16.51 | 17.98 |
| $k_2$ | −0.53 | −0.65 | −0.78 | −0.91 | −1.04 | −1.20 | −1.35 | −1.53 | −1.68 | −1.84 | −2.01 | −2.18 |
| $k_3$ | 0.018 | 0.022 | 0.027 | 0.031 | 0.035 | 0.041 | 0.046 | 0.053 | 0.058 | 0.063 | 0.069 | 0.074 |
| | | | | | | $x_1 = 0.5, x_2 = 0.5$ | | | | | | |
| $k_1$ | 3.79 | 4.46 | 5.23 | 6.02 | 6.9 | 7.8 | 8.8 | 9.81 | 10.9 | 12.04 | 13.06 | 14.34 |
| $k_2$ | −0.49 | −0.57 | −0.66 | −0.75 | −0.86 | −0.96 | −1.08 | −1.20 | −1.33 | −1.46 | −1.56 | −1.71 |
| $k_3$ | 0.018 | 0.020 | 0.023 | 0.025 | 0.029 | 0.032 | 0.035 | 0.040 | 0.044 | 0.048 | 0.051 | 0.056 |
| | | | | | | $x_1 = 0.4, x_2 = 0.6$ | | | | | | |
| $k_1$ | 3.25 | 3.78 | 4.37 | 5.02 | 5.70 | 6.40 | 7.17 | 7.98 | 8.83 | 9.72 | 10.63 | 11.60 |
| $k_2$ | −0.45 | −0.52 | −0.59 | −0.67 | −0.76 | −0.85 | −0.95 | −1.05 | −1.15 | −1.27 | −1.38 | −1.50 |
| $k_3$ | 0.016 | 0.019 | 0.022 | 0.025 | 0.028 | 0.031 | 0.034 | 0.038 | 0.041 | 0.045 | 0.049 | 0.054 |
| | | | | | | $x_1 = 0.3, x_2 = 0.7$ | | | | | | |
| $k_1$ | 3.11 | 3.56 | 4.05 | 4.60 | 5.18 | 5.78 | 6.44 | 7.12 | 7.81 | 8.54 | 9.30 | 10.07 |
| $k_2$ | −0.52 | −0.59 | −0.66 | −0.75 | −0.84 | −0.93 | −1.03 | −1.13 | −1.23 | −1.34 | −1.44 | −1.55 |
| $k_3$ | 0.023 | 0.027 | 0.030 | 0.034 | 0.038 | 0.041 | 0.046 | 0.050 | 0.054 | 0.059 | 0.064 | 0.068 |
| | | | | | | $x_1 = 0.2, x_2 = 0.8$ | | | | | | |
| $k_1$ | 2.51 | 2.87 | 3.24 | 3.66 | 4.07 | 4.54 | 5.00 | 5.51 | 6.04 | 6.57 | 7.13 | 7.70 |
| $k_2$ | −0.42 | −0.47 | −0.53 | −0.59 | −0.65 | −0.72 | −0.78 | −0.86 | −0.94 | −1.01 | −1.09 | −1.17 |
| $k_3$ | 0.019 | 0.021 | 0.024 | 0.026 | 0.029 | 0.032 | 0.034 | 0.038 | 0.041 | 0.044 | 0.048 | 0.051 |
| | | | | | | $x_1 = 0.1, x_2 = 0.9$ | | | | | | |
| $k_1$ | 2.34 | 2.63 | 2.96 | 3.31 | 3.70 | 4.09 | 4.49 | 4.91 | 5.36 | 5.79 | 6.25 | 6.72 |
| $k_2$ | −0.40 | −0.45 | −0.50 | −0.56 | −0.62 | −0.67 | −0.73 | −0.80 | −0.86 | −0.92 | −0.98 | −1.04 |
| $k_3$ | 0.018 | 0.02 | 0.023 | 0.025 | 0.028 | 0.031 | 0.033 | 0.036 | 0.039 | 0.041 | 0.044 | 0.046 |
| | | | | | | $x_1 = 0.0, x_2 = 1.0$ | | | | | | |
| $k_1$ | 1.97 | 2.19 | 2.46 | 2.73 | 3.00 | 3.31 | 3.65 | 4.00 | 4.35 | 4.69 | 5.05 | 5.42 |
| $k_2$ | −0.36 | −0.39 | −0.44 | −0.48 | −0.52 | −0.57 | −0.62 | −0.67 | −0.73 | −0.78 | −0.83 | −0.88 |
| $k_3$ | 0.018 | 0.02 | 0.022 | 0.024 | 0.025 | 0.028 | 0.030 | 0.032 | 0.035 | 0.037 | 0.039 | 0.041 |

ever, a rather more useful empirical working expression proposed in this regard (at room temperature) is[1,50]

$$\rho(t) = \rho(25) \cdot e^{\beta(t-25)} \tag{5}$$

in which $\rho(t)$ and $\rho(25)$ are densities measured at $t$ and 25 °C, respectively. Parameter $\beta$ (closely related to α) can be regarded as the temperature average coefficient of thermal expansion whose values for the entire system have been determined and collected in Table 4. The credibility of the $\beta$-values given in Table 4 may be judged from comparison of our determined value of $-2.70 \cdot 10^{-4}$ per degree for water with the corresponding literature[1] value of $-2.63 \cdot 10^{-4}$.

Viscosities at a series of temperatures and LiCl aquamolalities (***m***) are presented in Table 5 for each solvent mixture separately. Also the $\eta$ values for some selected mixtures have been plotted as isotherms against LiCl concentration in Figure 4a−d. The data fit very well to polynomials involving up to cubic terms in ***m***. For the viscosity investigations, rather more useful parameters commonly employed are the relative viscosity ($\eta/\eta°$) and the relative viscosity change ($[\eta - \eta°]/\eta° \equiv \Delta\eta/\eta°$) which have been differently correlated with electrolyte molarity (***c***) or molality (***m***) by various workers.[26,51,52] A cubic polynomial (isotherm) in ***m*** can readily be transformed into the Jones−Dole[52−54] type relation as below

$$\frac{\eta - \eta^{\circ}}{\eta^{\circ}} \equiv \frac{\Delta\eta}{\eta^{\circ}} \equiv B \cdot \boldsymbol{m} + C \cdot \boldsymbol{m}^2 + D \cdot m^3 \tag{6}$$

where $\eta°$ and $\eta$ are, respectively, the viscosities of the solvent (without LiCl) and solution at any given temperature. Various terms on the right-hand side of eq 6 contribute toward $\equiv \eta/\eta°$, each becoming relatively more significant at higher LiCl concentrations ($\boldsymbol{m} \geqslant 4.5$). Coefficients $B$, $C$, and D are listed

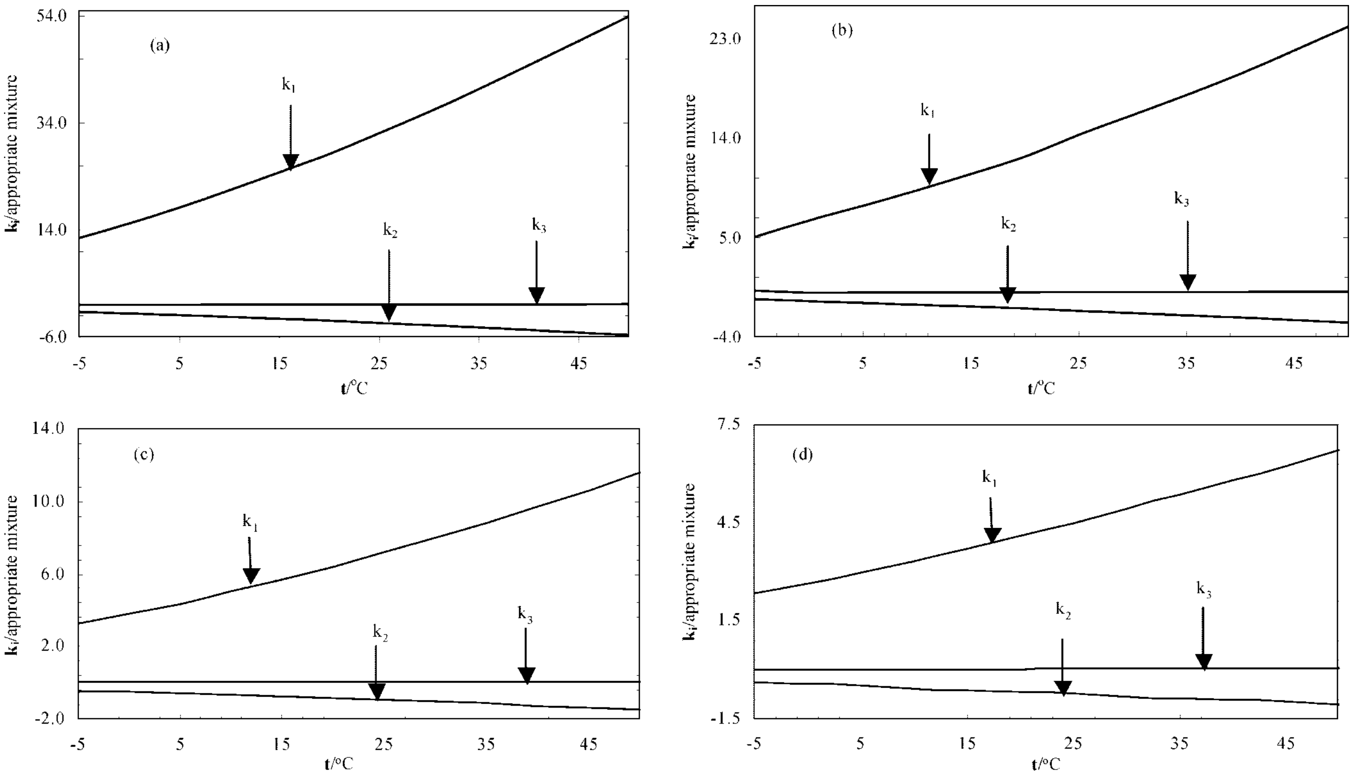

**Figure 8.** (a−d) Temperature variation of coefficients $k_i$ of the isotherm $\kappa(T) = k_1 \cdot \boldsymbol{m} + k_2 \cdot \boldsymbol{m}^2 + k_3 \cdot \boldsymbol{m}^3$ for aqueous ethanol of different water mole fractions, $x_1$. (a) $x_1 = 0.9$, (b) $x_1 = 0.7$, (c) $x_1 = 0.4$, and (d) $x_1 = 0.1$.

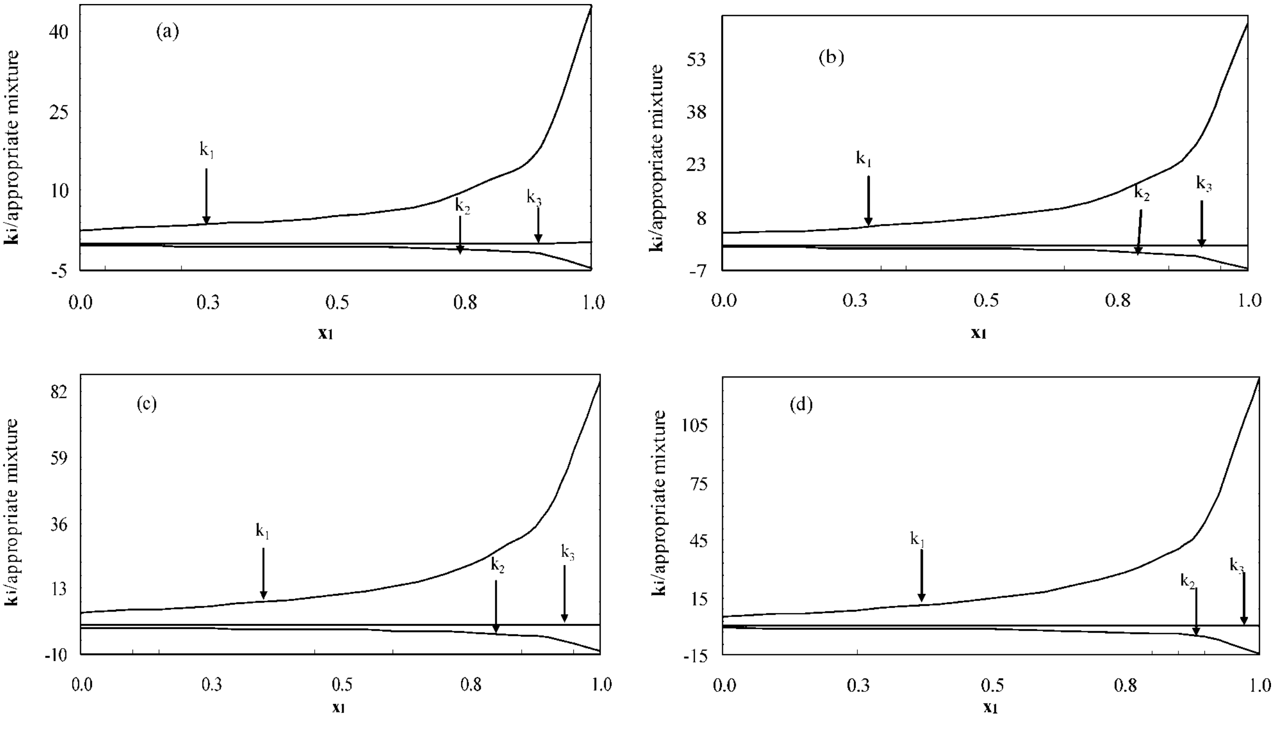

**Figure 9.** (a-d) Coefficients $k_i$ of Figure 8 plotted against water mole fraction, $x_1$, at different temperatures: (a) 5 °C, (b) 20 °C, (c) 35 °C, and (d) 50 °C.

in Table 6 which apparently do not exhibit any regular trend and correlation for different solvent mixtures; however, for a given mixture, temperature variation of the coefficients is quite consistent. *B* takes into account the solute−solvent interactions (ion−solvent for the electrolyte solutions) making it rather more important over the entire concentration range of the solute, while *C* signifies the solute−solute interactions.[24,54−57] It has been suggested regarding *D* that it should reflect variation in the solvent (1)−solvent (2) interactions, more importantly as a consequence of selective solvation of solute by one of the solvent components.[58] For all the mixtures, the solution viscosity throughout increased with LiCl concentration, a typical behavior exhibited by structure-making electrolytes also recognized from the positive *B*-values.[53]

In the selected temperature range of study, a significant viscosity decrease has been observed on raising the temperature, particularly for the solutions having higher LiCl concentrations. Viscosity is generally correlated with temperature by the Arrhenius-type relationship

$$\ln \eta = \ln \eta_{\infty} + \frac{E_a}{RT} \tag{7}$$

where $\eta_{\infty}$, $E_a$, and $R$ are, respectively, the "infinite temperature" viscosity, flow activation energy, and the gas constant. In accordance with eq 7, our ln $\eta$ versus $1/T$ plots for *the system* are fairly straight (plots for solutions in some selected mixtures are given in Figure 5a−d), while all the determined activation energies have been listed in Table 7. $E_a$ for the solvent mixtures are in agreement with the reported values,[59] but the estimated uncertainty of our $E_a$ values could be ± 5 %. For a given solvent (mixture), $E_a$ always increased with LiCl concentration; however, the increase appears to be more prominent for the ethanol-rich mixtures. Plots of $E_a$ against water mole fraction ($x_1$) at various LiCl concentrations are shown in Figure 6 which tend to pass through maxima at $x_1 \approx 0.6$, the composition at which the $V^E$ isotherms exhibit their minima. Belda and co-workers[59,60] have, however, reported the $E_a$ maximum at a slightly higher water mole fraction of ∼0.7.

The entire electrical conductivity data have been collected in Table 8, and the data for some selected solvent mixtures are plotted as isotherms against the LiCl aquamolality in Figure 7a−d. Each $\kappa$ vs $\boldsymbol{m}$ plot passes through a maximum, and such results have been previously reported for other systems as well.[61–63] The initial sharp rise of conductivity with LiCl concentration is obviously dominated by an increased number of ions. However, on further increasing the concentration, besides higher viscosities, phenomena involving ionic interactions[33] cause a decrease of conductivities after passing through the maxima. As a simplified model, it may be considered that conductivity results from two sets of oppositely acting terms, namely, $\kappa_{\text{up}}$ and $\kappa_{\text{down}}$. While there is a direct relation between $\kappa_{\text{up}}$ and number of solvates, $\kappa_{\text{down}}$ should include contributions arising due to ion−ion interactions, sharing of solvation spheres by the same solvent molecules, etc. Higher conductivities have been measured at higher temperatures in all cases mainly due to the decrease of the viscosity values. The conductivity data fit very well to the following polynomial in LiCl aquamolality, $\boldsymbol{m}$ ($k_0$ has to be zero to justify a zero value of $\kappa$ at $\boldsymbol{m} = 0$)

$$\kappa(T) = k_0 + k_1 \cdot \boldsymbol{m} + k_2 \cdot \boldsymbol{m}^2 + k_3 \cdot \boldsymbol{m}^3 \tag{8}$$

where the coefficients $k_i$ (collected in Table 9) vary with both temperature and solvent composition. Some selected plots to demonstrate typical dependence of $k_i$ on temperature and solvent composition are presented in Figure 8a−d and Figure 9a−d, respectively. The term involving $k_3$ can generally be ignored due to its negligible contribution (except in water). Consequently eq 8 simplifies as

$$\kappa(T) = k_1 \cdot \boldsymbol{m} + k_2 \cdot \boldsymbol{m}^2 \tag{9}$$

The two terms on the right-hand side may well be regarded as $\kappa_{\text{up}}$ and $\kappa_{\text{down}}$, respectively. A sharper increase of $k_1$ beyond a water mole fraction of 0.8 (Figure 9) suggests higher mobility of the aquo-solvates,[64] which is further supported by shifting of the conductivity maximum from ∼4.5$\boldsymbol{m}$ in the ethanol-rich solvents ($x_1 < 0.5$) to ∼6$\boldsymbol{m}$ in the water-rich mixtures ($x_1 > 0.5$).

Conductivity and viscosity, both being transport properties, are also related to each other in different ways.[65] Accordingly the molar conductivity, $\Lambda_m$, may also be considered to follow the Arrhenius relationship (eq 10). The corresponding conductivity activation energy, $E_c$, can readily be determined from the temperature dependent measurements.

$$\Lambda_m(T) = \Lambda^{\infty} e^{-E_c/RT} \tag{10}$$

$\Lambda^{\infty}$ is the molar conductivity at the so-called "infinite temperature", and the other symbols have the usual meanings. $E_c$ determined from the slopes of ln $\Lambda_m$ vs $1/T$ plots are also given in Table 7. The $E_c$ values come out smaller than the corresponding viscosity $E_a$ values. A possible explanation is that the ion transport under the influence of the applied electrical field involves movement of the individual solvates in the solution, whereas in the case of viscous flow under the influence of mechanical force, the whole layer of liquid has to move experiencing a higher barrier. Like $E_a$, also the $E_c$ values tend to pass through a maximum for *the system* but at a water mole fraction of ∼0.8.

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