# Density and Viscosity Experimental Data of the Ternary Mixtures 1-Propanol or 2-Propanol + Water + 1-Ethyl-3-methylimidazolium Ethylsulfate. Correlation and Prediction of Physical Properties of the Ternary Systems

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In this paper, we have determined densities, $\rho$, refractive indices, $n_D$, and dynamic viscosities, $\eta$, of two ternary mixtures, 1-propanol + water + 1-ethyl-3-methylimidazolium ethylsufate (EMISE) and 2-propanol + water + EMISE, at 298.15 K over the whole composition range and at atmospheric pressure. Excess molar volumes, $V^E$, refractive index deviations, $\Delta n_D$, viscosity deviations, $\Delta\eta$, and excess free energies of activation of viscous flow, $\Delta G^{*E}$, have been calculated from experimental data and fitted to Cibulka and Singh et al. equations. The excess properties have been used to test several prediction models.

## 1. Introduction

From a combination of organic cations and various anions, room-temperature ionic liquids (RTILs) are obtained. These compounds are salts with a low melting point (below 100 °C). Nowadays, very few reliable data of liquid densities and excess molar volumes of multicomponent systems on ionic liquids are available in the literature. These properties are used for the design of industrial equipment and many industrial applications. Moreover, these properties can provide an important tool to extract information about the behavior of components in pure as well as in mixed states.

In this work, density, refractive index, and viscosity of the ternary systems 1-propanol + water + EMISE and 2-propanol + water + EMISE have been measured. The binary systems 1-propanol + water and 2-propanol + water were measured too because no literature data with all needed properties were available. From the physical properties, we have determined the excess molar volumes, $V^E$, deviations of refractive index, viscosity deviations, $\Delta\eta$, and excess free energies of activation of viscous flow, $\Delta G^{*E}$. Experimental data were correlated using empirical equations[1,2] and predicted using geometrical models that assume that interactions in a ternary mixture depend on the interactions in binary systems.[3–9]

## 2. Experimental

***2.1. Chemicals.*** 1-Propanol was supplied by Sigma-Aldrich with a purity higher than 99.9 %, and 2-propanol was supplied by Merck with a purity higher than 99.7 %. They were degassed ultrasonically and dried over molecular sieves type 4Å, supplied by Aldrich. Water was bidistilled and deionized. The ionic liquid used in this work was synthesized in our laboratory.[10] Table 1 shows a comparison between experimental and literature data of pure components at 298.15 K.

***2.2. Apparatus and Procedure.*** Samples were prepared by syringing known amounts of the pure liquids into stoppered bottles, in an inert-atmosphere glovebox, using a Mettler AX-205 Delta Range balance with a precision of $\pm 10^{-5}$ g, covering the whole composition range of the mixture. A glovebox was used because the ionic liquid is a sensitive moisture.

Kinematic viscosities were determined using an automatic viscosimeter Lauda PVS1 with four Ubbelhode capillary microviscosimeters of $0.4 \cdot 10^{-3}$ m, $0.53 \cdot 10^{-3}$ m, $0.70 \cdot 10^{-3}$ m, and $1.26 \cdot 10^{-3}$ m diameter (the uncertainty in experimental measurement is ($\pm$ 0.0001, $\pm$ 0.003, $\pm$ 0.03, and $\pm$ 0.2) mPa·s, respectively). The capillary is maintained in a D20KP LAUDA thermostat with a resolution of 0.01 K.

The densities of the pure liquids and mixtures were measured using an Anton Paar DSA-5000 digital vibrating-tube densi-

**Table 1. Comparison of Measured Pure Component Property Data with Literature Values at $T = 298.15$ K**

| | $\rho$/g·cm$^{-3}$ | | $10^3\eta$/Pa·s | |
|---|---|---|---|---|
| component | exptl | lit. | exptl | lit. |
| 1-propanol | 0.7996 | 0.79960[a] | 2.017 | 1.951[b] |
| | | | | 2.0074[c] |
| 2-propanol | 0.7807 | 0.7809[d] | 2.082 | 2.081[d] |
| EMISE | 1.23763 | 1.23915[e] | 97.580 | 100.77[e] |
| | | 1.2296[f] | | |

[a] From ref 11. [b] From ref 12. [c] From ref 13. [d] From ref 14. [e] From ref 15. [f] From ref 16.

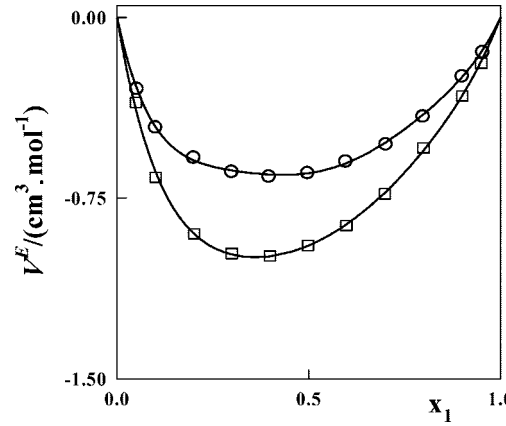


**Figure 1.** Experimental excess molar volume, $V^E$, calculated from the Redlich–Kister equation (—) plotted against mole fraction at $T = 298.15$ K for the binary mixtures: ○, 1-propanol (1) + water (2); □, 2-propanol (1) + water (2).

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10.1021/je700700f CCC: $40.75 
Published on Web 02/23/2008

**Table 2. Densities, $\rho$, Dynamic Viscosities, $\eta$, Refractive Index, $n_D$, Excess Molar Volumes, $V^E$, Deviations of Refractive Index, $\Delta n_D$, Viscosity Deviations, $\Delta\eta$, and Excess Free Energies of Activation of Viscous Flow, $\Delta G^{*E}$, for Studied Binary Systems at 298.15 K**

| $x_1$ | $\rho$ / g·cm$^{-3}$ | $n_D$ | $\eta$ / mPa·s | $V^E$ / cm$^3$·mol$^{-1}$ | $\Delta n_D$ | $\Delta\eta$ / mPa·s | $\Delta G^{*E}$ / J·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|
| 1-Propanol (1) + Water (2) | | | | | | | |
| 0.0000 | 0.9972 | 1.33251 | 0.890 | 0.000 | 0.0000 | 0.000 | 0.0 |
| 0.0504 | 0.9751 | 1.34612 | 1.616 | −0.294 | 0.0111 | 0.669 | 1683.1 |
| 0.0993 | 0.9533 | 1.35475 | 2.121 | −0.454 | 0.0172 | 1.119 | 2452.7 |
| 0.1986 | 0.9149 | 1.36491 | 2.599 | −0.580 | 0.0224 | 1.485 | 2981.6 |
| 0.2979 | 0.8872 | 1.37116 | 2.691 | −0.639 | 0.0236 | 1.465 | 2953.3 |
| 0.3949 | 0.8669 | 1.37516 | 2.610 | −0.659 | 0.0227 | 1.275 | 2682.2 |
| 0.4955 | 0.8503 | 1.37805 | 2.494 | −0.644 | 0.0205 | 1.045 | 2309.4 |
| 0.5953 | 0.8369 | 1.38008 | 2.339 | −0.596 | 0.0175 | 0.778 | 1846.4 |
| 0.6995 | 0.8256 | 1.38154 | 2.221 | −0.525 | 0.0136 | 0.542 | 1369.4 |
| 0.7969 | 0.8163 | 1.38239 | 2.124 | −0.409 | 0.0096 | 0.336 | 909.3 |
| 0.8983 | 0.8077 | 1.38299 | 2.026 | −0.242 | 0.0050 | 0.124 | 405.8 |
| 0.9518 | 0.8035 | 1.38308 | 1.998 | −0.142 | 0.0024 | 0.036 | 162.3 |
| 1.0000 | 0.7996 | 1.38309 | 2.017 | 0.000 | 0.0000 | 0.000 | 0.0 |
| 2-Propanol (1) + Water (2) | | | | | | | |
| 0.0000 | 0.9972 | 1.3325 | 0.890 | 0.000 | 0.0000 | 0.000 | 0.0 |
| 0.0491 | 0.9746 | 1.3456 | 1.718 | −0.353 | 0.0110 | 0.770 | 1844.5 |
| 0.1008 | 0.9536 | 1.3557 | 2.495 | −0.664 | 0.0189 | 1.485 | 2878.1 |
| 0.2011 | 0.9126 | 1.3650 | 3.038 | −0.898 | 0.0240 | 1.908 | 3379.0 |
| 0.2994 | 0.8816 | 1.3697 | 3.087 | −0.980 | 0.0245 | 1.840 | 3296.2 |
| 0.3992 | 0.8577 | 1.3724 | 2.924 | −0.991 | 0.0229 | 1.558 | 2948.0 |
| 0.4981 | 0.8388 | 1.3740 | 2.693 | −0.946 | 0.0203 | 1.210 | 2472.5 |
| 0.5979 | 0.8235 | 1.3749 | 2.489 | −0.863 | 0.0170 | 0.886 | 1960.6 |
| 0.6987 | 0.8106 | 1.3754 | 2.274 | −0.732 | 0.0132 | 0.551 | 1380.7 |
| 0.7993 | 0.7994 | 1.3756 | 2.153 | −0.541 | 0.0091 | 0.311 | 873.0 |
| 0.8998 | 0.7898 | 1.3754 | 2.073 | −0.325 | 0.0047 | 0.110 | 386.3 |
| 0.9495 | 0.7854 | 1.3752 | 2.063 | −0.189 | 0.0024 | 0.041 | 176.6 |
| 1.0000 | 0.7807 | 1.3750 | 2.082 | 0.000 | 0.0000 | 0.000 | 0.0 |

meter. The repeatability and the uncertainty in experimental measurement have been found to be lower than ($\pm 2\cdot10^{-6}$ and $\pm 2.6\cdot10^{-5}$) g·cm$^{-3}$.

To measure refractive indices, an automatic refractometer Abbemat-HP Dr. Kernchen with a resolution of $\pm 10^{-6}$ and an uncertainty in the experimental measurements of $\pm 4\cdot10^{-5}$ was used.

## 3. Results and Discussion

***3.1. Experimental Results.*** Physical properties of binary mixtures of 1-propanol and 2-propanol with EMISE were previously published.[17] The density, $\rho$, refractive index, $n_D$, and viscosity, $\eta$, of binary systems 1-propanol (1) + water (2) and 2-propanol (1) + water (2) together with their derived properties are presented in Table 2.

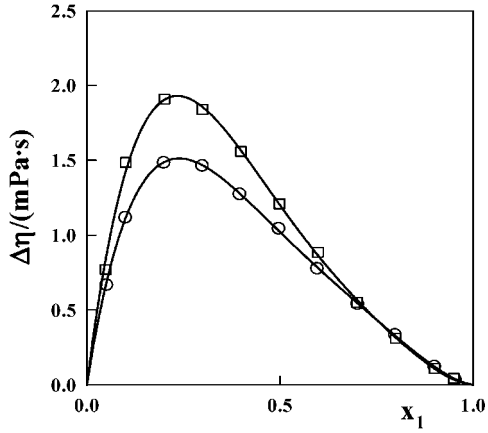


**Figure 2.** Experimental dynamic viscosity deviations, $\Delta\eta$, calculated from the Redlich–Kister equation (—) plotted against mole fraction at $T = 298.15$ K for the binary mixtures: ○, 1-propanol (1) + water (2); □, 2-propanol (1) + water (2).

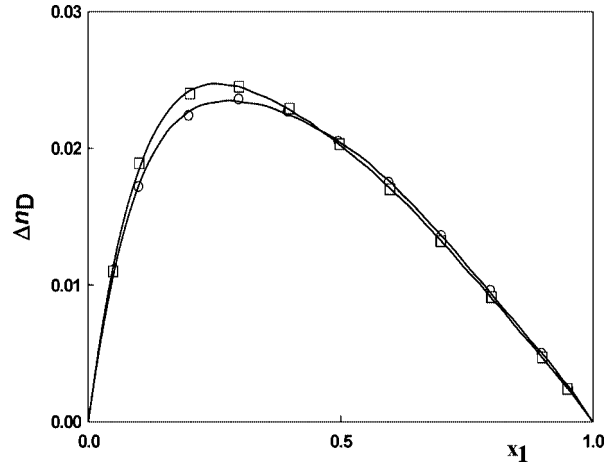


**Figure 3.** Experimental deviations of refractive index, $\Delta n_D$, calculated from the Redlich–Kister equation (—) plotted against mole fraction at $T = 298.15$ K for the binary mixtures: ○, 1-propanol (1) + water (2); □, 2-propanol (1) + water (2).

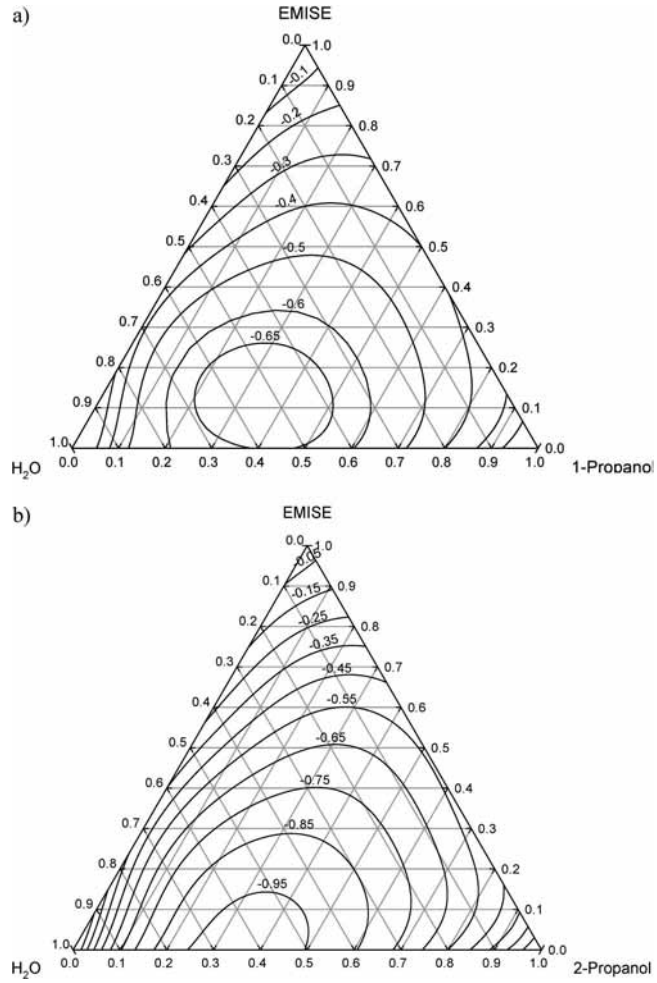


**Figure 4.** Isolines for excess molar volumes, $V^E_{123}$, in cm$^3$·mol$^{-1}$, from Cibulka and Redlich–Kister equations for the ternary systems (a) 1-propanol (1) + water (2) + EMISE (3) and (b) 2-propanol (1) + water (2) + EMISE (3).

The measured densities, $\rho$, refractive indices, $n_D$, and dynamic viscosities, $\eta$, and their derived properties for the ternary mixtures 1-propanol (1) + water (2) + EMISE (3) and 2-propanol (1) + water (2) + EMISE (3) at 298.15 K and at atmospheric pressure are listed in Tables 3 and 4.

**Table 3. Densities, $\rho$, Dynamic Viscosities, $\eta$, Refractive Index, $n_D$, Excess Molar Volumes, $V^E$, Deviations of Refractive Index, $\Delta n_D$, Viscosity Deviations, $\Delta\eta$, and Excess Free Energies of Activation of Viscous Flow, $\Delta G^{*E}$, for the Ternary System 1-Propanol (1) + Water (2) + EMISE (3) at 298.15 K**

| $x_1$ | $x_2$ | $\rho$ / g·cm$^{-3}$ | $n_D$ | $\eta$ / mPa·s | $V^E$ / cm$^3$·mol$^{-1}$ | $\Delta n_D$ | $\Delta\eta$ / mPa·s | $\Delta G^{*E}$ / J·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|---|
| 0.1280 | 0.6147 | 1.1486 | 1.44560 | 10.128 | −0.580 | 0.069 | −15.785 | 4563.0 |
| 0.2412 | 0.5349 | 1.1022 | 1.43789 | 8.497 | −0.645 | 0.060 | −14.315 | 4058.6 |
| 0.3498 | 0.4584 | 1.0575 | 1.43022 | 7.028 | −0.643 | 0.052 | −12.809 | 3518.8 |
| 0.4649 | 0.3772 | 1.0108 | 1.42206 | 5.826 | −0.629 | 0.043 | −10.855 | 2983.9 |
| 0.5725 | 0.3013 | 0.9678 | 1.41456 | 4.938 | −0.599 | 0.035 | −8.795 | 2511.3 |
| 0.6642 | 0.2367 | 0.9314 | 1.40804 | 3.908 | −0.543 | 0.027 | −7.312 | 1856.0 |
| 0.0826 | 0.7820 | 1.1180 | 1.42504 | 5.717 | −0.530 | 0.068 | −8.353 | 4490.2 |
| 0.1695 | 0.7080 | 1.0745 | 1.42032 | 5.451 | −0.628 | 0.061 | −7.478 | 4229.2 |
| 0.2588 | 0.6318 | 1.0332 | 1.41545 | 5.009 | −0.665 | 0.054 | −6.747 | 3858.4 |
| 0.3494 | 0.5546 | 0.9953 | 1.41067 | 4.634 | −0.683 | 0.046 | −5.931 | 3496.9 |
| 0.4500 | 0.4688 | 0.9573 | 1.40580 | 4.260 | −0.682 | 0.039 | −4.983 | 3093.6 |
| 0.6559 | 0.2933 | 0.8899 | 1.39678 | 3.103 | −0.576 | 0.024 | −3.435 | 1852.6 |
| 0.0662 | 0.8408 | 1.0970 | 1.41152 | 4.244 | −0.454 | 0.062 | −5.714 | 4054.3 |
| 0.1383 | 0.7759 | 1.0585 | 1.40610 | 4.326 | −0.581 | 0.054 | −5.019 | 3996.6 |
| 0.2141 | 0.7076 | 1.0216 | 1.40625 | 4.247 | −0.642 | 0.051 | −4.453 | 3821.3 |
| 0.2999 | 0.6304 | 0.9848 | 1.40315 | 4.063 | −0.678 | 0.045 | −3.907 | 3546.3 |
| 0.3963 | 0.5436 | 0.9487 | 1.39982 | 3.873 | −0.683 | 0.038 | −3.277 | 3226.9 |
| 0.5062 | 0.4446 | 0.9132 | 1.39642 | 3.283 | −0.658 | 0.031 | −2.933 | 2542.7 |
| 0.7206 | 0.2516 | 0.8569 | 1.39049 | 3.041 | −0.517 | 0.017 | −1.352 | 1834.3 |
| 0.0559 | 0.8764 | 1.0792 | 1.40006 | 3.394 | −0.395 | 0.055 | −4.104 | 3612.1 |
| 0.1139 | 0.8226 | 1.0471 | 1.39969 | 3.913 | −0.524 | 0.052 | −3.248 | 3924.7 |
| 0.1845 | 0.7571 | 1.0117 | 1.39839 | 3.683 | −0.609 | 0.048 | −3.069 | 3653.7 |
| 0.2669 | 0.6805 | 0.9758 | 1.39664 | 3.675 | −0.661 | 0.043 | −2.598 | 3505.4 |
| 0.3557 | 0.5981 | 0.9428 | 1.39485 | 3.481 | −0.680 | 0.038 | −2.277 | 3183.5 |
| 0.5658 | 0.4030 | 0.8820 | 1.39098 | 2.887 | −0.626 | 0.025 | −1.650 | 2190.4 |
| 0.8287 | 0.1590 | 0.8281 | 1.38633 | 2.391 | −0.398 | 0.010 | −0.620 | 966.9 |
| 0.0474 | 0.9071 | 1.0577 | 1.38662 | 2.604 | −0.330 | 0.045 | −2.737 | 2986.7 |
| 0.1023 | 0.8549 | 1.0278 | 1.38842 | 3.042 | −0.496 | 0.044 | −2.108 | 3357.6 |
| 0.1670 | 0.7932 | 0.9949 | 1.38901 | 3.216 | −0.579 | 0.042 | −1.708 | 3432.0 |
| 0.2397 | 0.7240 | 0.9640 | 1.38906 | 3.406 | −0.655 | 0.039 | −1.264 | 3477.2 |
| 0.3161 | 0.6512 | 0.9356 | 1.38884 | 3.146 | −0.667 | 0.036 | −1.258 | 3121.7 |
| 0.4188 | 0.5534 | 0.9044 | 1.38834 | 3.184 | −0.670 | 0.031 | −0.862 | 2934.1 |
| 0.0422 | 0.9290 | 1.0361 | 1.37367 | 2.255 | −0.283 | 0.035 | −1.469 | 2627.4 |
| 0.0901 | 0.8825 | 1.0108 | 1.37757 | 2.506 | −0.457 | 0.036 | −1.133 | 2889.0 |
| 0.1467 | 0.8276 | 0.9815 | 1.37995 | 2.968 | −0.525 | 0.036 | −0.570 | 3307.8 |
| 0.2082 | 0.7680 | 0.9555 | 1.38151 | 2.933 | −0.605 | 0.035 | −0.495 | 3201.3 |
| 0.2888 | 0.6898 | 0.9268 | 1.38288 | 3.140 | −0.663 | 0.033 | −0.144 | 3257.7 |
| 0.4746 | 0.5095 | 0.8770 | 1.38445 | 2.712 | −0.646 | 0.026 | −0.241 | 2452.0 |
| 0.0384 | 0.9460 | 1.0140 | 1.36098 | 1.865 | −0.241 | 0.024 | −0.569 | 2106.6 |
| 0.0813 | 0.9039 | 0.9930 | 1.36670 | 2.179 | −0.424 | 0.028 | −0.236 | 2527.9 |
| 0.1286 | 0.8573 | 0.9697 | 1.37051 | 2.642 | −0.515 | 0.029 | 0.248 | 3038.4 |
| 0.1909 | 0.7961 | 0.9435 | 1.37393 | 2.701 | −0.592 | 0.030 | 0.333 | 3047.6 |
| 0.2591 | 0.7289 | 0.9189 | 1.37668 | 2.963 | −0.615 | 0.029 | 0.625 | 3214.0 |
| 0.3408 | 0.6485 | 0.8958 | 1.37895 | 2.936 | −0.650 | 0.028 | 0.634 | 3043.6 |
| 0.0375 | 0.9551 | 0.9973 | 1.35204 | 1.651 | −0.220 | 0.017 | 0.004 | 1757.5 |
| 0.1198 | 0.8735 | 0.9590 | 1.36371 | 2.308 | −0.493 | 0.024 | 0.629 | 2683.8 |
| 0.2472 | 0.7471 | 0.9110 | 1.37225 | 2.852 | −0.599 | 0.026 | 1.125 | 3160.4 |
| 0.0961 | 0.1388 | 1.2153 | 1.47318 | 50.905 | −0.242 | 0.023 | −24.069 | 1598.4 |
| 0.1971 | 0.1233 | 1.1923 | 1.46844 | 38.682 | −0.310 | 0.026 | −28.139 | 1712.5 |
| 0.0981 | 0.2695 | 1.2078 | 1.46965 | 34.718 | −0.354 | 0.039 | −27.433 | 2660.1 |
| 0.2061 | 0.2372 | 1.1798 | 1.46404 | 26.738 | −0.447 | 0.039 | −28.206 | 2596.3 |
| 0.3040 | 0.2080 | 1.1513 | 1.45823 | 20.220 | −0.499 | 0.039 | −28.197 | 2415.1 |
| 0.3994 | 0.1795 | 1.1201 | 1.45178 | 15.510 | −0.511 | 0.037 | −26.550 | 2250.9 |
| 0.5003 | 0.1493 | 1.0830 | 1.44410 | 11.477 | −0.503 | 0.035 | −23.856 | 2013.2 |
| 0.6001 | 0.1195 | 1.0414 | 1.43535 | 8.367 | −0.479 | 0.031 | −20.313 | 1718.9 |
| 0.7013 | 0.0893 | 0.9937 | 1.42523 | 6.086 | −0.472 | 0.026 | −15.847 | 1414.7 |
| 0.8010 | 0.0595 | 0.9388 | 1.41352 | 4.291 | −0.379 | 0.020 | −10.995 | 1007.3 |
| 0.9011 | 0.0296 | 0.8753 | 1.39981 | 3.013 | −0.300 | 0.012 | −5.601 | 575.9 |
| 0.0962 | 0.4557 | 1.1924 | 1.46221 | 20.230 | −0.483 | 0.059 | −24.087 | 3990.4 |
| 0.2001 | 0.4034 | 1.1592 | 1.45593 | 16.021 | −0.559 | 0.055 | −23.433 | 3625.5 |
| 0.2998 | 0.3531 | 1.1247 | 1.44926 | 12.695 | −0.581 | 0.051 | −22.094 | 3250.2 |
| 0.4954 | 0.2544 | 1.0500 | 1.43456 | 8.001 | −0.579 | 0.040 | −17.631 | 2494.0 |
| 0.5960 | 0.2037 | 1.0076 | 1.42611 | 6.313 | −0.558 | 0.034 | −14.613 | 2102.7 |
| 0.6982 | 0.1522 | 0.9612 | 1.41674 | 4.821 | −0.507 | 0.027 | −11.319 | 1624.6 |
| 0.8022 | 0.0997 | 0.9101 | 1.40633 | 3.660 | −0.407 | 0.019 | −7.615 | 1132.9 |
| 0.8979 | 0.0515 | 0.8593 | 1.39585 | 2.770 | −0.284 | 0.010 | −4.026 | 609.6 |

**Table 4. Densities, $\rho$, Dynamic Viscosities, $\eta$, Refractive Index, $n_D$, Excess Molar Volumes, $V^E$, Deviations of Refractive Index, $\Delta n_D$, Viscosity Deviations, $\Delta\eta$, and Excess Free Energies of Activation of Viscous Flow, $\Delta G^{*E}$, for the Ternary System 2-Propanol (1) + Water (2) + EMISE (3) at 298.15 K**

| $x_1$ | $x_2$ | $\rho$ / g·cm$^{-3}$ | $n_D$ | $\eta$ / mPa·s | $V^E$ / cm$^3$·mol$^{-1}$ | $\Delta n_D$ | $\Delta\eta$ / mPa·s | $\Delta G^{*E}$ / J·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|---|
| 0.0949 | 0.1426 | 1.2147 | 1.47289 | 50.594 | −0.324 | 0.024 | −24.138 | 1616.8 |
| 0.2002 | 0.1260 | 1.1896 | 1.46762 | 38.893 | −0.472 | 0.028 | −27.380 | 1778.5 |
| 0.2964 | 0.1109 | 1.1633 | 1.46203 | 29.818 | −0.563 | 0.030 | −28.735 | 1847.3 |
| 0.4843 | 0.0813 | 1.1005 | 1.44825 | 16.603 | −0.633 | 0.031 | −26.872 | 1771.8 |
| 0.6918 | 0.0486 | 1.0069 | 1.42737 | 8.040 | −0.615 | 0.027 | −18.775 | 1418.4 |
| 0.7943 | 0.0324 | 0.9467 | 1.41360 | 5.437 | −0.524 | 0.022 | −13.152 | 1127.1 |
| 0.0907 | 0.2737 | 1.2086 | 1.46971 | 36.644 | −0.424 | 0.040 | −25.814 | 2781.8 |
| 0.1902 | 0.2437 | 1.1818 | 1.46423 | 28.198 | −0.556 | 0.041 | −27.653 | 2649.3 |
| 0.2845 | 0.2154 | 1.1535 | 1.45842 | 22.538 | −0.644 | 0.040 | −27.050 | 2584.6 |
| 0.4839 | 0.1553 | 1.0821 | 1.44315 | 12.918 | −0.699 | 0.037 | −23.428 | 2202.4 |
| 0.5849 | 0.1249 | 1.0388 | 1.43349 | 9.527 | −0.670 | 0.034 | −20.117 | 1935.2 |
| 0.6881 | 0.0939 | 0.9887 | 1.42243 | 6.871 | −0.643 | 0.029 | −15.922 | 1609.0 |
| 0.8921 | 0.0325 | 0.8651 | 1.39461 | 3.401 | −0.405 | 0.013 | −5.845 | 777.4 |
| 0.0834 | 0.4648 | 1.1950 | 1.46251 | 21.619 | −0.540 | 0.060 | −23.056 | 4165.0 |
| 0.1860 | 0.4128 | 1.1609 | 1.45589 | 17.075 | −0.679 | 0.057 | −22.830 | 3773.2 |
| 0.3755 | 0.3167 | 1.0907 | 1.44179 | 11.400 | −0.791 | 0.048 | −19.700 | 3134.3 |
| 0.4723 | 0.2676 | 1.0511 | 1.43361 | 9.141 | −0.788 | 0.043 | −17.461 | 2767.3 |
| 0.5715 | 0.2173 | 1.0082 | 1.42468 | 7.252 | −0.795 | 0.037 | −14.741 | 2374.3 |
| 0.6749 | 0.1649 | 0.9596 | 1.41446 | 5.595 | −0.702 | 0.030 | −11.596 | 1915.7 |
| 0.8849 | 0.0584 | 0.8500 | 1.39062 | 3.105 | −0.431 | 0.012 | −4.327 | 810.2 |
| 0.1126 | 0.6808 | 1.1365 | 1.43874 | 8.626 | −0.713 | 0.071 | −12.372 | 4772.9 |
| 0.2236 | 0.5957 | 1.0853 | 1.43083 | 7.597 | −0.860 | 0.062 | −11.035 | 4314.2 |
| 0.3310 | 0.5133 | 1.0372 | 1.42265 | 6.616 | −0.916 | 0.053 | −9.726 | 3829.7 |
| 0.4354 | 0.4332 | 0.9926 | 1.41490 | 5.704 | −0.925 | 0.045 | −8.413 | 3320.2 |
| 0.5330 | 0.3583 | 0.9527 | 1.40784 | 4.891 | −0.898 | 0.037 | −7.146 | 2801.8 |
| 0.6305 | 0.2835 | 0.9144 | 1.40090 | 4.158 | −0.826 | 0.029 | −5.800 | 2259.7 |
| 0.0850 | 0.7786 | 1.1168 | 1.42522 | 6.055 | −0.656 | 0.069 | −8.132 | 4619.5 |
| 0.1722 | 0.7044 | 1.0719 | 1.41990 | 5.791 | −0.840 | 0.062 | −7.244 | 4356.9 |
| 0.2632 | 0.6269 | 1.0279 | 1.41416 | 5.431 | −0.931 | 0.054 | −6.398 | 4031.5 |
| 0.3595 | 0.5450 | 0.9855 | 1.40819 | 4.964 | −0.966 | 0.046 | −5.592 | 3620.4 |
| 0.4571 | 0.4619 | 0.9467 | 1.40248 | 4.409 | −0.976 | 0.039 | −4.856 | 3120.9 |
| 0.6619 | 0.2877 | 0.8755 | 1.39153 | 3.380 | −0.809 | 0.024 | −3.175 | 2008.5 |
| 0.7683 | 0.1972 | 0.8432 | 1.38623 | 2.878 | −0.647 | 0.016 | −2.270 | 1358.4 |
| 0.8792 | 0.1028 | 0.8124 | 1.38090 | 2.445 | −0.421 | 0.008 | −1.235 | 687.6 |
| 0.0688 | 0.8355 | 1.0977 | 1.41265 | 4.554 | −0.583 | 0.063 | −5.668 | 4212.8 |
| 0.1434 | 0.7686 | 1.0570 | 1.40970 | 4.720 | −0.798 | 0.058 | −4.850 | 4187.6 |
| 0.2231 | 0.6971 | 1.0165 | 1.40584 | 4.649 | −0.910 | 0.052 | −4.225 | 4006.8 |
| 0.3084 | 0.6205 | 0.9781 | 1.40180 | 4.418 | −0.976 | 0.046 | −3.709 | 3706.9 |
| 0.4012 | 0.5373 | 0.9411 | 1.39752 | 4.076 | −0.983 | 0.039 | −3.240 | 3299.3 |
| 0.5022 | 0.4466 | 0.9059 | 1.39324 | 3.690 | −0.952 | 0.032 | −2.744 | 2807.7 |
| 0.0579 | 0.8750 | 1.0776 | 1.39993 | 3.573 | −0.504 | 0.055 | −3.877 | 3743.5 |
| 0.1209 | 0.8165 | 1.0427 | 1.39946 | 3.937 | −0.744 | 0.053 | −3.154 | 3916.4 |
| 0.1919 | 0.7505 | 1.0060 | 1.39761 | 4.048 | −0.895 | 0.048 | −2.639 | 3877.6 |
| 0.2695 | 0.6784 | 0.9699 | 1.39515 | 4.008 | −0.957 | 0.044 | −2.236 | 3712.1 |
| 0.3575 | 0.5968 | 0.9349 | 1.39241 | 3.843 | −0.991 | 0.038 | −1.900 | 3418.0 |
| 0.5641 | 0.4049 | 0.8705 | 1.38635 | 3.160 | −0.915 | 0.025 | −1.406 | 2393.6 |
| 0.0505 | 0.9031 | 1.0575 | 1.38754 | 2.901 | −0.439 | 0.046 | −2.532 | 3267.7 |
| 0.1066 | 0.8498 | 1.0270 | 1.38930 | 3.332 | −0.706 | 0.046 | −1.903 | 3580.2 |
| 0.1684 | 0.7910 | 0.9943 | 1.38931 | 3.601 | −0.849 | 0.044 | −1.416 | 3711.3 |
| 0.2400 | 0.7229 | 0.9612 | 1.38851 | 3.690 | −0.944 | 0.040 | −1.074 | 3663.6 |
| 0.3223 | 0.6446 | 0.9289 | 1.38722 | 3.614 | −1.005 | 0.036 | −0.859 | 3449.6 |
| 0.4151 | 0.5563 | 0.8979 | 1.38560 | 3.380 | −0.998 | 0.031 | −0.766 | 3062.4 |
| 0.0434 | 0.9286 | 1.0337 | 1.37318 | 2.306 | −0.359 | 0.035 | −1.339 | 2682.8 |
| 0.0925 | 0.8810 | 1.0087 | 1.37750 | 2.885 | −0.642 | 0.037 | −0.680 | 3258.3 |
| 0.1486 | 0.8265 | 0.9798 | 1.37965 | 3.214 | −0.809 | 0.037 | −0.260 | 3502.2 |
| 0.2130 | 0.7640 | 0.9502 | 1.38069 | 3.393 | −0.915 | 0.036 | 0.025 | 3569.3 |
| 0.2891 | 0.6901 | 0.9206 | 1.38109 | 3.412 | −0.981 | 0.033 | 0.169 | 3459.1 |
| 0.4888 | 0.4963 | 0.8632 | 1.38037 | 2.979 | −0.967 | 0.025 | 0.061 | 2633.7 |
| 0.0389 | 0.9432 | 1.0179 | 1.36330 | 1.977 | −0.305 | 0.027 | −0.691 | 2263.9 |
| 0.0834 | 0.8995 | 0.9967 | 1.36925 | 2.578 | −0.596 | 0.031 | −0.063 | 2970.9 |
| 0.1346 | 0.8492 | 0.9712 | 1.37292 | 2.994 | −0.777 | 0.032 | 0.385 | 3349.6 |
| 0.1967 | 0.7884 | 0.9431 | 1.37527 | 3.248 | −0.898 | 0.032 | 0.677 | 3511.8 |
| 0.3555 | 0.6325 | 0.8884 | 1.37773 | 3.179 | −1.016 | 0.028 | 0.704 | 3183.7 |
| 0.5934 | 0.3990 | 0.8359 | 1.37794 | 2.643 | −0.927 | 0.019 | 0.314 | 2057.9 |
| 0.0353 | 0.9565 | 0.9994 | 1.35229 | 1.663 | −0.256 | 0.017 | −0.069 | 1776.4 |
| 0.0754 | 0.9166 | 0.9823 | 1.35984 | 2.321 | −0.546 | 0.023 | 0.575 | 2686.5 |
| 0.1225 | 0.8699 | 0.9602 | 1.36520 | 2.780 | −0.742 | 0.026 | 1.017 | 3169.5 |

**Table 5. Redlich–Kister Parameters for the Binary Systems and Root-Mean-Square Deviations**

| property | $a_0$ | $a_1$ | $a_2$ | $a_3$ | $a_4$ | $\sigma$ |
|---|---|---|---|---|---|---|
| | | | 1-Propanol (1) + EMISE (2) | | | |
| $V^E$/cm$^3$·mol$^{-1}$ | −1.6090 | −0.3029 | −0.1465 | −0.6409 | −1.4859 | 0.007 |
| $\Delta n_D$ | 0.0896 | 0.0393 | 0.0172 | 0.0118 | 0.0093 | $2.9\cdot10^{-5}$ |
| $10^3\Delta\eta$/Pa·s | −122.81 | 47.16 | −11.25 | −3.39 | 8.83 | 0.077 |
| $\Delta G^{*E}$/J·mol$^{-1}$ | 4461.2 | 565.53 | −370.19 | 817.80 | 1880.4 | 36.02 |
| | | | 2-Propanol (1) + EMISE (2) | | | |
| $V^E$/cm$^3$·mol$^{-1}$ | −2.1355 | −0.2885 | −0.1942 | −2.0258 | −1.3439 | 0.012 |
| $\Delta n_D$ | 0.0977 | 0.0407 | 0.0163 | 0.0169 | 0.0112 | $5.3\cdot10^{-5}$ |
| $10^3\Delta\eta$/Pa·s | −114.96 | 39.22 | −22.07 | 3.19 | 18.26 | 0.238 |
| $\Delta G^{*E}$/J·mol$^{-1}$ | 5303.3 | 1256.5 | −1581.1 | 6.3960 | 2461.3 | 16.6 |
| | | | 1-Propanol (1) + Water (2) | | | |
| $V^E$/cm$^3$·mol$^{-1}$ | −2.5832 | 0.4464 | −0.6835 | 1.5432 | −2.1059 | 0.006 |
| $\Delta n_D$ | 0.0817 | −0.0495 | 0.0321 | −0.0560 | 0.0516 | $1.4\cdot10^{-4}$ |
| $10^3\Delta\eta$/Pa·s | 4.08 | −5.00 | 4.46 | −2.96 | −0.20 | 0.007 |
| $\Delta G^{*E}$/J·mol$^{-1}$ | 9164.4 | −7491.2 | 5170.2 | −10878 | 8679.6 | 35.4 |
| | | | 2-Propanol (1) + Water (2) | | | |
| $V^E$/cm$^3$·mol$^{-1}$ | −3.7862 | 1.3329 | −1.5360 | 1.3259 | −1.4134 | 0.011 |
| $\Delta n_D$ | 0.0810 | −0.0578 | 0.0481 | −0.0586 | 0.0414 | $1.7\cdot10^{-4}$ |
| $10^3\Delta\eta$/Pa·s | 4.80 | −7.21 | 6.12 | −2.98 | −0.57 | 0.028 |
| $\Delta G^{*E}$/J·mol$^{-1}$ | 9920.2 | −9238.1 | 5542.1 | −12232 | 10991 | 29.9 |
| | | | Water (1) + EMISE (2) | | | |
| $V^E$/cm$^3$·mol$^{-1}$ | −1.1585 | −1.3068 | −1.1589 | 0.8021 | 1.6604 | 0.013 |
| $\Delta n_D$ | 0.2491 | 0.1774 | 0.1139 | 0.2931 | 0.2864 | $8.8\cdot10^{-4}$ |
| $10^3\Delta\eta$/Pa·s | −97.494 | 31.556 | 7.923 | 7.091 | −17.981 | 0.203 |
| $\Delta G^{*E}$/J·mol$^{-1}$ | 17064.5 | 10198.7 | 8018.4 | 17882.9 | 15108.6 | 49.1 |

**Table 6. Fit Parameters and Root-Mean-Square Deviations for Empirical Equations**

| | $V^E$ / cm$^3$·mol$^{-1}$ | $\Delta n_D$ | $10^3\Delta\eta$ / Pa·s | $\Delta G^{*E}$ / J·mol$^{-1}$ |
|---|---|---|---|---|
| | 1-Propanol (1) + Water (2) + EMISE (3) | | | |
| Cibulka | | | | |
| *A* | −1.636 | −0.1682 | 59.30 | −4200.8 |
| *B* | 4.012 | 0.3956 | 23.89 | 34626 |
| *C* | 0.346 | 0.3451 | −13.77 | −49261 |
| $\sigma$ | $1.4\cdot10^{-2}$ | $7.3\cdot10^{-4}$ | $1.8\cdot10^{-1}$ | 168.73 |
| Singh et al. | | | | |
| *A* | −0.086 | 0.0776 | 63.98 | −8525.7 |
| *B* | −0.004 | 0.0045 | −0.2541 | −529.30 |
| *C* | 0.037 | 0.0160 | −0.7957 | 635.15 |
| $\sigma$ | $1.6\cdot10^{-2}$ | $8.2\cdot10^{-4}$ | $1.9\cdot10^{-1}$ | 223.16 |
| | 2-Propanol (1) + Water (2) + EMISE (3) | | | |
| Cibulka | | | | |
| *A* | −3.2073 | −0.1559 | 48.86 | −5613.1 |
| *B* | 3.6773 | 0.4171 | 35.40 | 44383 |
| *C* | 4.0159 | 0.3254 | −8.406 | −62556 |
| $\sigma$ | $1.9\cdot10^{-2}$ | $6.3\cdot10^{-4}$ | $2.3\cdot10^{-1}$ | 182.30 |
| Singh et al. | | | | |
| *A* | −0.3477 | 0.0975 | 59.70 | −10379 |
| *B* | 0.0058 | 0.0034 | −0.1551 | −641.85 |
| *C* | −0.1588 | 0.0084 | −0.9129 | 77.854 |
| $\sigma$ | $2.0\cdot10^{-2}$ | $8.4\cdot10^{-4}$ | $2.4\cdot10^{-1}$ | 256.90 |

**Table 7. Root-Mean-Square Deviations of Predictions of Excess Molar Volumes, $V^E$, Deviations of Refractive Index, $\Delta n_D$, Viscosity Deviations $\Delta\eta$, and Excess Free Energies of Activation of Viscous Flow, $\Delta G^{*E}$, for 1-Propanol (1) + Water (2) + EMISE (3) and 2-Propanol (1) + Water (2) + EMISE (3) at 298.15 K**

| | $V^E_{123}$ / cm$^3$·mol$^{-1}$ | $\Delta n_D$ | $\Delta\eta$ / mPa·s$^{-1}$ | $\Delta G^{*E}_{123}$ / J·mol$^{-1}$ |
|---|---|---|---|---|
| | 1-Propanol (1) + Water (2) + EMISE (3) | | | |
| Rastogi | 0.203 | 0.0120 | 3.341 | 785.61 |
| Radojkovic | 0.018 | 0.0019 | 0.991 | 316.69 |
| Jacob and Fitzner | 0.055 | 0.0064 | 1.428 | 263.87 |
| Toop[a] | 0.069 | 0.0065 | 2.267 | 841.28 |
| Toop[b] | 0.085 | 0.0095 | 2.242 | 459.09 |
| Toop[c] | 0.081 | 0.0138 | 3.410 | 1146.3 |
| Kohler | 0.026 | 0.0053 | 0.978 | 698.61 |
| Scatchard[a] | 0.132 | 0.0342 | 10.088 | 2522.5 |
| Scatchard[b] | 0.200 | 0.0069 | 14.569 | 511.49 |
| Scatchard[c] | 0.228 | 0.0165 | 3.382 | 1609.4 |
| Tsao and Smith[a] | 0.080 | 0.0137 | 3.914 | 1291.0 |
| Tsao and Smith[b] | 0.086 | 0.0080 | 4.485 | 456.03 |
| Tsao and Smith[c] | 0.123 | 0.0147 | 3.369 | 1305.7 |
| | 2-Propanol (1) + Water (2) + EMISE (3) | | | |
| Rastogi | 0.296 | 0.0122 | 3.695 | 795.93 |
| Radojkovic | 0.025 | 0.0020 | 0.912 | 373.22 |
| Jacob and Fitzner | 0.075 | 0.0065 | 1.446 | 277.78 |
| Toop[a] | 0.100 | 0.0067 | 2.013 | 948.82 |
| Toop[b] | 0.102 | 0.0096 | 2.282 | 420.35 |
| Toop[c] | 0.104 | 0.0144 | 3.325 | 1259.7 |
| Kohler | 0.036 | 0.0054 | 0.933 | 790.62 |
| Scatchard[a] | 0.138 | 0.0343 | 9.394 | 2577.9 |
| Scatchard[b] | 0.272 | 0.0074 | 13.688 | 584.08 |
| Scatchard[c] | 0.318 | 0.0171 | 3.321 | 1764.2 |
| Tsao and Smith[a] | 0.106 | 0.0140 | 3.659 | 1393.7 |
| Tsao and Smith[b] | 0.101 | 0.0079 | 4.185 | 425.57 |
| Tsao and Smith[c] | 0.161 | 0.0154 | 3.284 | 1434.1 |

[a] Ethanol is the asymmetric component. [b] Water is the asymmetric component. [c] EMISE is the asymmetric component.

The excess molar volumes, viscosity deviations, and deviations in the refractive index were calculated from experimental values as follows

$$V^E = \sum_{i=1}^{N} x_i M_i(\rho^{-1} - \rho_i^{-1}) \quad (1)$$

$$\Delta\eta = \eta - \sum_{i}^{N} x_i\eta_i \quad (2)$$

$$\Delta n_D = n_D - \sum_{i}^{N} x_i n_{D,i} \quad (3)$$

where $\rho$ and $\rho_i$ are the density of the mixture and the density of the pure components, respectively; $x_i$ represents the mole fraction of the $i$ component; $\eta$ and $\eta_i$ are the dynamic viscosity of the mixture and the pure components, respectively; and $n_D$ and $n_{D,i}$ are the refractive index of the mixture and the refractive index of the pure components, respectively.

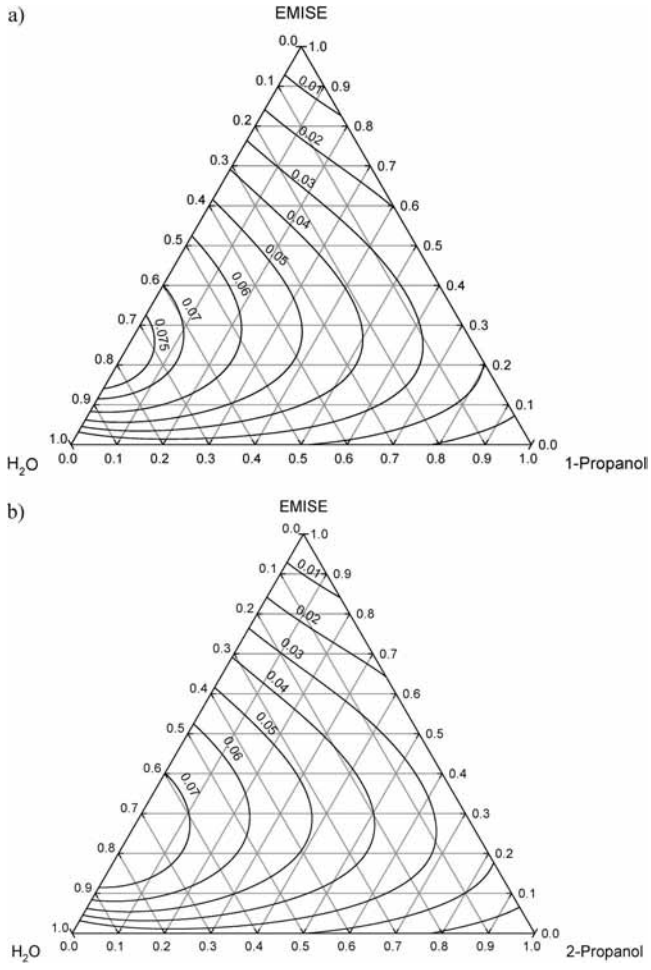


**Figure 5.** Isolines for the deviation of refractive index, $\Delta n_{D123}$, from Cibulka and Redlich–Kister equations for the ternary systems (a) 1-propanol (1) + water (2) + EMISE (3) and (b) 2-propanol (1) + water (2) + EMISE (3).

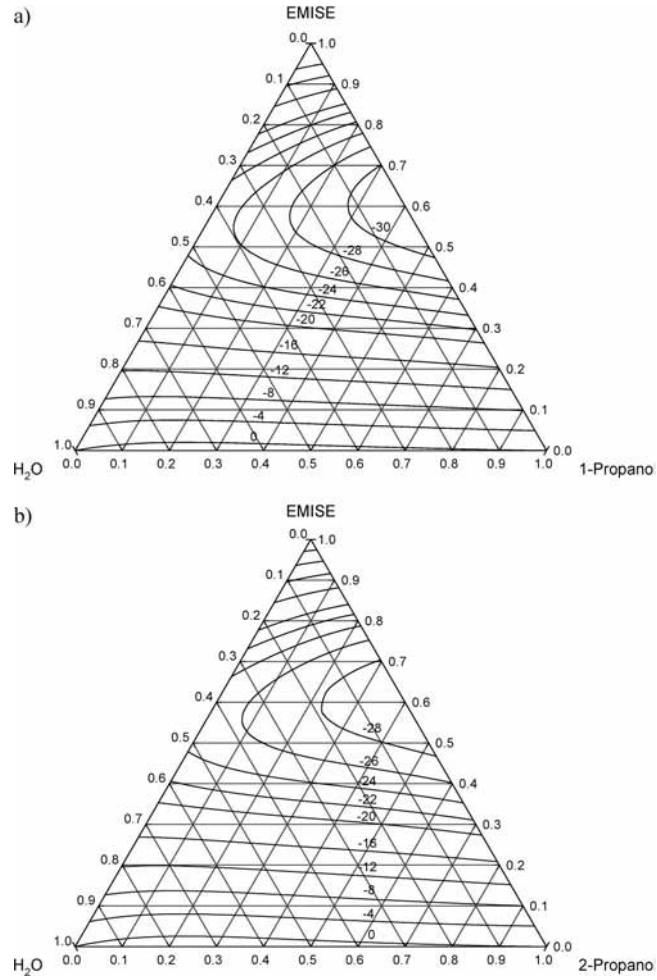


**Figure 6.** Isolines for the deviation of dynamic viscosity, $\Delta n_{D123}$, in mPa·s, from Cibulka and Redlich–Kister equations for the ternary systems (a) 1-propanol (1) + water (2) + EMISE (3) and (b) 2-propanol (1) + water (2) + EMISE (3).

***3.2. Correlation of Physical Properties.*** The contribution to the excess property of the constituent binary mixtures was evaluated by the Redlich–Kister equation[18]

$$\Delta Q_{12} = x_1 x_2 \sum_{p=0}^{M} B_p (x_1 - x_2)^p \qquad (4)$$

where $\Delta Q_{12}$ is the excess property; $x_1$ and $x_2$ are the mole fraction of component 1 and 2, respectively; $B_p$ is the fitting parameter; and M is the degree of the polynomic expansion. The parameters for the excess properties of all the binary mixtures involved in the ternary system are listed in Table 5 together with the root-mean-square deviations

$$\sigma = \left\{ \sum_{i}^{n_{dat}} (z_{exptl} - z_{calcd})^2 / n_{dat} \right\} \qquad (5)$$

where $z_{exptl}$, $z_{calcd}$, and $n_{dat}$ are the values of the experimental and calculated property and the number of experimental data, respectively.

The binary systems 1-propanol (1) + EMISE (2), 2-propanol (1) + EMISE (2), and water (1) + EMISE (2) were determined in a previous paper.[17] Figures 1 to 3 show the fitted curves of excess properties $V^E$, $\Delta\eta$, and $\Delta n_D$, of the binary systems 1-propanol (1) + water (2) and 2-propanol (1) + water (2).

The $V^E$, $\Delta n_D$, $\Delta\eta$, and $\Delta G^{*E}$ data of the ternary systems were correlated using the equations proposed by Cibulka[1] and Singh et al.[2]

The Cibulka equation:

$$Q^E_{123} = Q^E_{12} + Q^E_{13} + Q^E_{23} + x_1 x_2 x_3 (A + B x_1 + C x_2) \qquad (6)$$

The Singh et al. equation:

$$Q^E_{123} = Q^E_{12} + Q^E_{13} + Q^E_{23} + A x_1 x_2 x_3 + B x_1 (x_2 - x_3) + C x_1^2 (x_2 - x_3)^2 \qquad (7)$$

where $A$, $B$, and $C$ are fit parameters and $Q^E_{ij}$ is the contribution to the excess property of the constituent binary mixtures evaluated by the Redlich–Kister equation (eq 4).

The fit parameters and root-mean-square deviations (eq 5) are given in Table 6. As can be observed in this table, for $\Delta n_D$ and $\Delta G^{*E}$, both equations correlate in a similar way the behavior of the two ternary systems, but for the other studied properties, the better fitting is given by Cibulka, where the correlated values are in good agreement with the experimental data. In Figures 4 to 6, the isolines of the excess properties $V^E$, $\Delta n_D$, and $\Delta\eta$ calculated from Cibulka for both ternary systems are shown.

***3.3. Prediction of Physical Properties.*** The predictive methods can be divided into symmetric and asymmetric, depending on whether the assumption of the three binary mixtures contributing equally to the ternary mixture magnitude is accepted or not.

To predict the excess properties ($V^E$, $\Delta n_D$, $\Delta\eta$, $\Delta G^{*E}$), we have used symmetric (Radojkovic et al.,[3] Rastogi,[4] Jacob and

Fitzer,[5] and Kohler [6]) and asymmetric (Toop,[7] Tsao and Smith,[8] and Scatchard et al.[9]) geometrical solution models.

Table 7 lists the root-mean-square deviations of fit for each dependent variable and equation. The root-mean-square deviations, $\sigma$, presented were calculated as eq 5.

## 4. Conclusions

Densities, refractive indices and viscosities for the binary and ternary mixtures 1-propanol + water, 2-propanol + water, 1-propanol + water + EMISE, and 2-propanol + water + EMISE were measured at 298.15 K and at atmospheric pressure over the whole range of compositions. From these physical properties, the excess properties were calculated.

For both binary systems, the excess molar volumes show negative deviations over the whole range of compositions with a minimum over 0.4 mol fraction. Dynamic viscosity deviations and refractive index deviations are both positive over the whole range of compositions with a maximum over (0.2 and 0.3) mol fraction, respectively.

For the studied ternary systems, the excess molar volumes and dynamic viscosity deviations are negative for both systems. For the ternary system containing 1-propanol, we can observe a minimum over 0.7 mol fraction. Refractive index deviation for both systems is positive over the whole range of compositions.

Seven geometrical solution models were used to predict the excess properties. The one of Radojkovic gives reasonable good results for all properties for the two studied systems. Also the Kohler model describes quite well the behavior of all the excess properties of the ternary system, except for the excess free energies of activation of viscous flow. The best prediction of the excess free energy of activation of the two studied ternary systems is calculated by the Jacob and Fitzner model. In general, the symmetric equations give better predictive results, especially the ones of Radojkovic, Kohler, and Jacob and Fitzner. The predictions of the asymmetric equations of Tsao and Smith and Scatchard disagree significantly with experimental data. It is noteworthy that in all asymmetric models the best results for the $\Delta G^{*E}$ are obtained when water is the asymmetric component.

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Received for review November 28, 2007. Accepted January 11, 2008. The authors are grateful to the Ministerio de Ciencia y Tecnología of Spain (project CTQ2004-00454) and Xunta de Galicia (PGIDIT05PXIC-38303PN) for financial support.

JE700700F