# Densities, Viscosities, and Surface Tensions of the System Water + Diethylene Glycol

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Densities, $\rho$, viscosities, $\eta$, at temperatures (303.15 to 323.15) K, and surface tensions, $\gamma$, at temperatures (283.15 to 303.15) K for water (W) + diethylene glycol (DEG) solutions were measured. The excess molar volumes, $V_m^E$, were calculated from the measured densities. The properties, $\rho$, $\eta$, and $\gamma$ have been fitted to appropriate polynomial equation. The excess molar volumes, $V_m^E$, have been fitted to a Redlich−Kister type equation.

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## Introduction

Physico-chemical properties such as densities, viscosities, and surface tensions of liquid and liquid mixtures over the whole composition range at different temperatures contribute to the understanding of the types of interactions involved in the solutions as well as being useful for practical chemical engineering purposes. The properties of aqueous solutions of organic solutes also helps in the development of knowledge of complex but interesting natural phenomena like biological structures in the water environment and the geochemistry of hydrothermal systems via a fundamental understanding of the different modes of interactions, e.g., hydrophilic hydration, ionic hydration, hydrophobic effects, interstitial accommodation,[1,2] etc.

Glycols and their ethers are organic hydrophobic compounds, which have significant industrial applications in the manufacture of solvents, lubricants, and conditioning agents. In the recent past several works have been published in the literature on both aqueous and non aqueous solutions of glycols and glycol ethers.[3−6] Diethylene glycol is an important member of the glycols. Though some studies on the densities and transport properties of (W + DEG)[7−14] are available, to the best of our knowledge, no data on surface tensions of W + DEG system have been published.

As a continuation of our research work on aqueous solutions of organic solutes,[15−19] in this report we present surface tensions for W + DEG solutions along with volumetric and viscosity data.

## Experimental Section

***Materials.*** Diethylene glycol was obtained from Merck Schuchardt with a purity of 0.99 mass fraction and was kept over molecular sieves (0.4 nm) prior to each experiment. The purity of DEG was checked by measuring and comparing the densities and viscosities of this sample with corresponding literature values in the temperature range (298.15 to 303.15) K. The measured densities and viscosities agreed reasonably well with the literature values as shown in Table 1.

**Table 1. Comparison of Experimental Densities, $\rho$ (g·cm$^{-3}$), and Viscosities, $\eta$ (mPa·s), of Diethylene Glycol from $T$ = (298.15 to 323.15) K**

| $T$ | $\rho$ | | $\eta$ | |
|---|---|---|---|---|
| K | this work | lit. | this work | lit. |
| 298.15 | 1.1131 | 1.1123[14]<br>1.1124[11]<br>1.1135[9] | 27.93 | |
| 303.15 | 1.10948 | 1.1095[11]<br>1.1099[9] | 22.16 | 21.754[11]<br>22.091[9] |
| 308.15 | 1.10572 | 1.1057[14]<br>1.1059[11]<br>1.1063[9] | 17.93 | 17.364[11] |
| 313.15 | 1.10211 | 1.1027[9] | 14.71 | 14.70[14]<br>14.56[9] |
| 318.15 | 1.09835 | 1.0991[9] | 12.192 | |
| 323.15 | 1.09494 | 1.0955[9] | 10.195 | 10.03[9] |

***Apparatus and Procedure.*** Solutions were prepared by mass and kept in airtight stoppered glass bottles to avoid evaporation. The weighing was done with an electronic balance (Mettler Toledo) with an accuracy of $\pm 1 \times 10^{-5}$ g. The mole fraction of each mixture was obtained with an accuracy of $2 \times 10^{-5}$ from the measured masses of the components. A 10 cm$^3$ bicapillary pycnometer was used for density measurement. Before each series of measurements, the pycnometer was calibrated with redistilled water. The calibration was accepted if the measurements were within $\pm 1 \times 10^{-4}$ g cm$^{-3}$ of the published values for water. For the measurement of density at temperatures, (283.15 to 323.15) K, a thermostatic water bath of sensitivity ±0.05 K was used. Our density values for pure DEG at the studied temperature range are within 0.007% of literature values.[9,11,14]

For the measurement of the viscosity coefficient, $\eta$, A and B type Ostwald U-tube viscometers (British Standard Institution) with sufficiently long efflux times were used. These were previously calibrated with water and with 0.40 mass fraction of sugar solution in redistilled water as required. The flow times of the solutions were recorded by an electronic timer with an accuracy of 0.01 s. The same water bath as used for density measurement was used for the measurement of $\eta$ at different temperatures.

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**Table 2. Densities, $\rho$ ($g\cdot cm^{-3}$), and Excess Molar Volumes, $V_m^E$ ($cm^3\cdot mol^{-1}$), for Water (1) + Diethylene Glycol (2), from $T$ = (303.15 to 323.15) K**

| | 303.15 K | | 308.15 K | | 313.15 K | | 318.15 K | | 323.15 K | |
|---|---|---|---|---|---|---|---|---|---|---|
| $X_2$ | $\rho$ | $V_m^E$ | $\rho$ | $V_m^E$ | $\rho$ | $V_m^E$ | $\rho$ | $V_m^E$ | $\rho$ | $V_m^E$ |
| 0.00000 | 0.99560 | 0.0000 | 0.99412 | 0.0000 | 0.99225 | 0.0000 | 0.99030 | 0.0000 | 0.98810 | 0.0000 |
| 0.05006 | 1.02838 | −0.1704 | 1.02609 | −0.1640 | 1.02367 | −0.1610 | 1.02108 | −0.1550 | 1.01836 | −0.1499 |
| 0.10162 | 1.05183 | −0.3365 | 1.04894 | −0.3234 | 1.04602 | −0.3149 | 1.04303 | −0.3057 | 1.03994 | −0.2958 |
| 0.15119 | 1.06799 | −0.5061 | 1.06534 | −0.4910 | 1.06204 | −0.4765 | 1.05872 | −0.4631 | 1.05520 | −0.4441 |
| 0.20121 | 1.07871 | −0.5649 | 1.07525 | −0.5455 | 1.07177 | −0.5287 | 1.06824 | −0.5125 | 1.06450 | −0.4878 |
| 0.24774 | 1.08655 | −0.6396 | 1.08295 | −0.6188 | 1.07942 | −0.6027 | 1.07576 | −0.5848 | 1.07218 | −0.5661 |
| 0.29804 | 1.09227 | −0.6753 | 1.08852 | −0.6517 | 1.08497 | −0.6369 | 1.08158 | −0.6315 | 1.07747 | −0.5934 |
| 0.35046 | 1.09622 | −0.6736 | 1.09242 | −0.6502 | 1.08868 | −0.6294 | 1.08497 | −0.6137 | 1.08116 | −0.5866 |
| 0.44827 | 1.10117 | −0.6338 | 1.09758 | −0.6238 | 1.09368 | −0.5974 | 1.08991 | −0.5821 | 1.08610 | −0.5542 |
| 0.54213 | 1.10374 | −0.5422 | 1.10011 | −0.5344 | 1.09637 | −0.5172 | 1.09291 | −0.5215 | 1.08898 | −0.4859 |
| 0.65175 | 1.10625 | −0.4484 | 1.10258 | −0.4429 | 1.09897 | −0.4357 | 1.09529 | −0.4315 | 1.09134 | −0.3924 |
| 0.75248 | 1.10778 | −0.3432 | 1.10395 | −0.3306 | 1.10025 | −0.3196 | 1.09658 | −0.3191 | 1.09318 | −0.3164 |
| 0.78643 | 1.10794 | −0.2690 | 1.10426 | −0.2655 | 1.10056 | −0.2551 | 1.09689 | −0.2560 | 1.09337 | −0.2450 |
| 0.84987 | 1.10855 | −0.2086 | 1.10493 | −0.2146 | 1.10132 | −0.2122 | 1.09762 | −0.2125 | 1.09391 | −0.1872 |
| 0.90053 | 1.10884 | −0.1341 | 1.10516 | −0.1374 | 1.10155 | −0.1320 | 1.09787 | −0.1393 | 1.09419 | −0.1160 |
| 0.95032 | 1.10966 | −0.0741 | 1.10558 | −0.0792 | 1.10198 | −0.0799 | 1.09833 | −0.0847 | 1.09455 | −0.0553 |
| 1.00000 | 1.10948 | 0.0000 | 1.10572 | 0.0000 | 1.10211 | 0.0000 | 1.09835 | 0.0000 | 1.09494 | 0.0000 |

For surface tension measurements a Langmuir trough (type 601) from Nima Technology was used and the surface tension was determined via the Wilhelmy paper plate method. The trough was connected to a thermostat (Julabo F 12), which maintained the temperature at a constant value to within ±0.1 K. Before each experiment, the accuracy of the method was ensured by calibration of the pressure sensor using deionized water. Each Wilhelmy paper plate was washed gently with the experimental liquid to be measured and was carefully attached to the balance via three S-shaped hooks so that there was no additional pressure on the hook of the balance and so that the paper was suspended perpendicularly across the liquid surface. The surface tension was measured by immersing the plate into the liquid and then withdrawing it very slowly. For each particular temperature, this step was repeated for 5−6 times and then the average of the surface tensions was taken. The accuracy of the measurements was found ±0.2 mN/m.

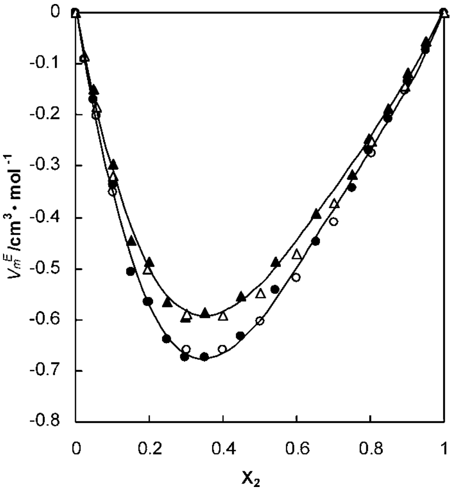

**Figure 1.** Excess molar volume of water (1) + diethylene glycol (2) at 303.15 K (●) and 323.15 K (▲). Experimental data of Garcia et al.[9] at 303.15 K (○) and 323.15 K (Δ).

**Table 3. Experimental Viscosities, $\eta$ (mPa·s), for Water (1) + Diethylene Glycol (2)**

| | $\eta$ | | | | |
|---|---|---|---|---|---|
| $X_2$ | 303.15 K | 308.15 K | 313.15 K | 318.15 K | 323.15 K |
| 0.00000 | 0.801 | 0.722 | 0.656 | 0.598 | 0.549 |
| 0.05006 | 1.556 | 1.370 | 1.222 | 1.091 | 0.985 |
| 0.10162 | 2.582 | 2.242 | 1.962 | 1.729 | 1.535 |
| 0.15119 | 3.830 | 3.286 | 2.833 | 2.459 | 2.161 |
| 0.20121 | 5.261 | 4.448 | 3.806 | 3.286 | 2.846 |
| 0.24774 | 6.653 | 5.591 | 4.739 | 4.052 | 3.506 |
| 0.29804 | 8.301 | 6.910 | 5.820 | 4.962 | 4.252 |
| 0.35046 | 9.851 | 8.154 | 6.821 | 5.768 | 4.929 |
| 0.44827 | 12.538 | 10.291 | 8.514 | 7.148 | 6.071 |
| 0.54213 | 15.060 | 12.284 | 10.145 | 8.478 | 7.143 |
| 0.65175 | 17.460 | 14.112 | 11.634 | 9.672 | 8.114 |
| 0.75248 | 19.297 | 15.645 | 12.824 | 10.647 | 8.937 |
| 0.78643 | 19.903 | 16.124 | 13.229 | 10.956 | 9.190 |
| 0.84987 | 20.610 | 16.738 | 13.728 | 11.423 | 9.461 |
| 0.90053 | 21.307 | 17.230 | 14.081 | 11.689 | 9.799 |
| 0.95032 | 21.741 | 17.529 | 14.408 | 11.921 | 9.999 |
| 1.00000 | 22.156 | 17.934 | 14.706 | 12.191 | 10.195 |

## Results and Discussion

Diethylene glycol is a liquid which is extensively associated through H-bonding, as is evident from its high dielectric constant (31.69 × 3.335641 × E$^{-30}$ coulomb meter) and high boiling point (518.15K). The measured density and viscosity values of this compound also reflect the high degree of association. Densities, $\rho$, and excess molar volumes, $V_m^E$, for the system, W + DEG, at temperatures (303.15 to 323.15) K are listed in Table 2.

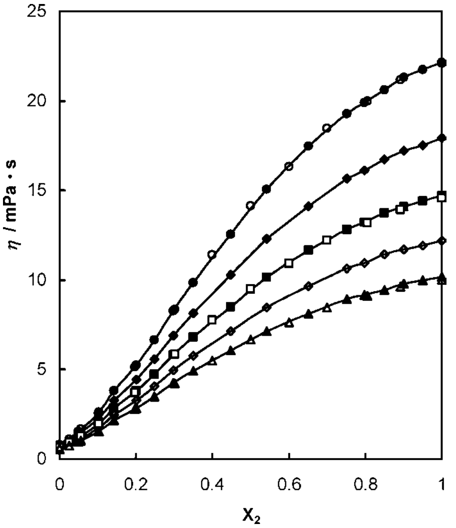

**Figure 2.** Viscosities of water (1) + diethylene glycol (2) at 303.15 K (●), 308.15 K (♦), 313.15 K (■), 318.15 K (◇), and 323.15 K (▲). Experimental data of Garcia et al.[9] at 303.15 K (○), 313.15 K (□), and 323.15 K (Δ).

**Table 4. Coefficients, $A_i$, and Square of Regression Coefficient, $r^2$ of eq 2, Expressing Density, $\rho$ (g·cm$^{-3}$), Viscosity, $\eta$ (mPa·s), and Surface Tension, $\gamma$, (mN·m$^{-1}$), for Water (1) + Diethylene Glycol (2)**

| $T$/K | $A_0$ | $A_1$ | $A_2$ | $A_3$ | $A_4$ | $A_5$ | $A_6$ | $r^2$ |
|---|---|---|---|---|---|---|---|---|
| Coefficients for Density | | | | | | | | |
| 303.15 | 0.9955 | 0.7807 | −2.6032 | 4.9404 | −5.3554 | 3.0790 | −0.7275 | 0.9997 |
| 308.15 | 0.9939 | 0.7694 | −2.6091 | 5.0822 | −5.6855 | 3.3841 | −0.8293 | 0.9996 |
| 313.15 | 0.9920 | 0.7547 | −2.5581 | 4.9749 | −5.5434 | 3.2799 | −0.7979 | 0.9996 |
| 318.15 | 0.9901 | 0.7390 | −2.5061 | 4.9032 | −5.5176 | 3.3050 | −0.8152 | 0.9996 |
| 323.15 | 0.9880 | 0.7230 | −2.4318 | 4.6723 | −5.0990 | 2.9279 | −0.6854 | 0.9996 |
| Coefficients for Viscosity | | | | | | | | |
| 303.15 | 7.9511 | 115.71 | 805.73 | −1521.4 | 1235.3 | −500.65 | 79.010 | 0.9999 |
| 308.15 | 7.2236 | 96.658 | 723.19 | −1639.1 | 1788.6 | −1065.8 | 268.43 | 0.9999 |
| 313.15 | 6.5450 | 86.810 | 588.99 | −1439.6 | 1718.7 | −11230 | 308.57 | 0.9999 |
| 318.15 | 5.9797 | 76.280 | 496.06 | −1275.6 | 1569.4 | −1026.8 | 276.60 | 0.9999 |
| 323.15 | 5.4760 | 70.729 | 380.52 | −964.33 | 1122.7 | −687.61 | 174.52 | 0.9999 |
| Coefficients for Surface Tension | | | | | | | | |
| 283.15 | 73.12 | −257.50 | 1509.0 | −4763.3 | 7787.9 | −6235.0 | 1932.1 | 0 0.9961 |
| 288.15 | 72.29 | −255.60 | 1514.2 | −4771.7 | 7774.4 | −6144.3 | 1886.8 | 0 0.9949 |
| 293.15 | 71.44 | −251.16 | 1508.5 | −4834.7 | 7972.5 | −6415.7 | 1994.7 | 0 0.9951 |
| 298.15 | 70.72 | −242.49 | 1430.3 | −4528.3 | 7405.3 | −5922.6 | 1832.5 | 0 0.9957 |
| 303.15 | 69.93 | −236.24 | 1372.3 | −4284.2 | 6927.7 | −5489.6 | 1685.0 | 0 0.9947 |

Excess molar volumes were calculated using the following relation:

$$V_m^E = (X_1M_1 + X_2M_2)/\rho - (X_1M_1/\rho_1 + X_2M_2/\rho_2) \qquad (1)$$

where $\rho$, $\rho_1$, and $\rho_2$ represent the densities of the solution, water (1), and diethylene glycol (2) respectively. $M_1$ and $M_2$ and $X_1$ and $X_2$ represent the molar masses and mole fractions of the corresponding components.

Figure 1 represents the dependence of $V_m^E$ on the mole fraction of DEG at (303.15 and 323.15) K. It displays a well-defined minima at ~0.34 mole fraction of DEG. Throughout the whole range of composition, $V_m^E$ was found to be negative and $\delta V_m^E/\delta T$ positive. The values of $V_m^E$ for this system reported by Garcia et al.[9] have been plotted in Figure 1 for comparison. The agreement between our $V_m^E$ values and those reported by Garcia et al. at (303.15 and 323.15) K is excellent.

Table 3 lists the viscosities, $\eta$, of the system W + DEG at different temperatures. Figure 2 represents the variation of $\eta$ against mole fraction of DEG at different temperatures. Values of $\eta$ for different mole fractions of DEG at temperatures (303.15, 313.15, and 323.15) K from the recent work of Garcia et al.[9] are plotted together with our experimental values. Both sets of data are found to be in good agreement with one another.

The $\rho$ and $\eta$ values of the system were fitted to a polynomial equation of the following type:

$$Y = \sum_{i=0}^{n} A_i X_2^i \qquad (2)$$

where $X_2$ is the mole fraction of the solute and $A_i$ is the $i$th fitting coefficient of the polynomial equation. The coefficients of this equation and square of regression coefficient, $r^2$, are listed in Table 4.

The excess molar volumes, $V_m^E$, were fitted to the following type of Redlich−Kister equation:

$$Y^E = X_2(1 - X_2)\sum_{i=0}^{n} A_i(2X_2 - 1)^i \qquad (3)$$

where $X_2$ is the mole fraction of the solute and $A_i$ is the $i$th coefficient of the equation. The coefficients of this equation and standard deviations, $\sigma$, are listed in Table 5.

**Table 5. Coefficients, $A_i$, of Redlich−Kister Equation, (eq 3), Expressing Excess Molar Volumes, $V_m^E$ (cm$^3$·mol$^{-1}$), and Standard Deviation, $\sigma$, for Water (1) + Diethylene Glycol (2)**

| $T$/K | $A_0$ | $A_1$ | $A_2$ | $A_3$ | $A_4$ | $A_5$ | $\sigma$ |
|---|---|---|---|---|---|---|---|
| Coefficients for Excess Molar Volume | | | | | | | |
| 303.15 | −2.3513 | 1.5743 | −1.3435 | 0.8300 | 1.2729 | −1.5727 | 0.0181 |
| 308.15 | −2.3122 | 1.4648 | −1.0954 | 0.9688 | 0.8800 | −1.8470 | 0.0182 |
| 313.15 | −2.2351 | 1.3767 | −1.1371 | 1.2020 | 0.9240 | −2.0921 | 0.0181 |
| 318.15 | −2.2143 | 1.2698 | −1.0050 | 1.4018 | 0.7378 | −2.4013 | 0.0180 |
| 323.15 | −2.0666 | 1.3547 | −1.3432 | 0.4038 | 1.4243 | −0.7991 | 0.0174 |

**Table 6. Surface Tensions, $\gamma$ (m N·m$^{-1}$), for Water (1) + Diethylene Glycol (2) from $T$ = (283.15 to 303.15) K**

| | 283.15 K | 288.15 K | 293.15 K | 298.15 K | 303.15 K |
|---|---|---|---|---|---|
| $X_2$ | $\gamma$ | $\gamma$ | $\gamma$ | $\gamma$ | $\gamma$ |
| 0.00000 | 74.00 | 73.20 | 72.40 | 71.6 | 70.90 |
| 0.02449 | 66.40 | 65.32 | 64.62 | 64.28 | 63.45 |
| 0.05997 | 62.04 | 61.47 | 60.90 | 60.34 | 59.78 |
| 0.09899 | 58.65 | 58.00 | 57.70 | 57.30 | 56.85 |
| 0.15199 | 56.50 | 55.87 | 55.40 | 55.00 | 54.50 |
| 0.20058 | 54.60 | 54.10 | 53.70 | 53.40 | 52.70 |
| 0.23862 | 53.50 | 53.25 | 52.87 | 52.42 | 51.95 |
| 0.29102 | 52.23 | 52.10 | 51.72 | 51.25 | 50.83 |
| 0.39008 | 50.10 | 49.70 | 49.38 | 49.10 | 48.70 |
| 0.48676 | 48.70 | 48.52 | 48.03 | 47.80 | 47.55 |
| 0.58271 | 48.00 | 47.60 | 47.24 | 47.00 | 46.75 |
| 0.69238 | 47.60 | 47.20 | 47.02 | 46.75 | 46.40 |
| 0.79508 | 47.00 | 46.60 | 46.32 | 45.98 | 45.70 |
| 0.87544 | 46.70 | 46.55 | 46.00 | 45.82 | 45.63 |
| 1.00000 | 46.23 | 45.93 | 45.55 | 45.22 | 44.80 |

The surface tensions, $\gamma$, for the above system are listed in Table 6. Fitting coefficients of eq 2 with square of regression coefficient, $r^2$, for surface tensions are listed in Table 4.

Figure 3 shows the variation of $\gamma$ for the system W+DEG at (283.15, 293.15, and 303.15) K. The surface tension decreases rapidly on addition of small amounts of DEG to water, which is typical for aqueous systems of surface-active substances.[20−26] Unfortunately, because of the lack of surface tension data of our studied system we are unable to compare the values at this stage. The surface tension data of (water + ethylene glycol) at 303.15 K, from literature,[27] have been presented in the figure for comparison. As expected, it was found that the surface tension values for the aqueous system containing DEG, having two ($-CH_2-CH_2-$) groups, are lower than those of EG, which has a single ($-CH_2-CH_2-$) group. Most of the total decrease in surface tension for W + DEG system occurs within a very narrow range of compositions, within ~0.10 mol fraction of DEG; in the remaining major part of the composition range, the surface tension decreases only slowly to reach the value of the pure component. It

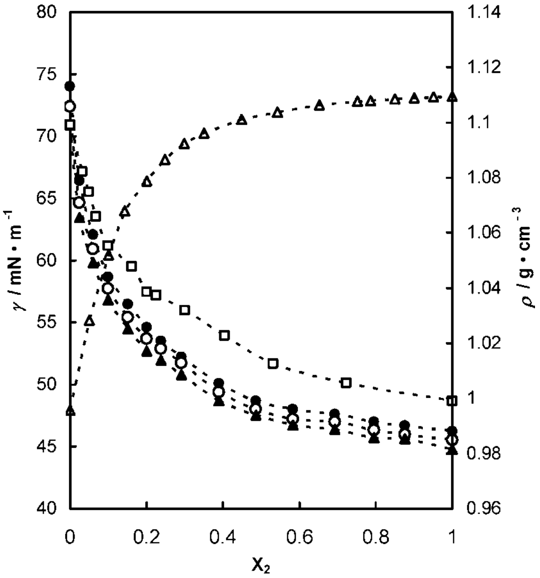

**Figure 3.** Surface tensions of water (1) + diethylene glycol (2) at 283.15 K (●), 293.15 K (○), 303.15 K (▲), and densities at 303.15 K (Δ). Experimental surface tension data of water (1) + ethylene glycol (2) at 303.15 K (□) from Tsierkezos et al.[27]

is thought that in the dilute region many competing processes occur at the surface, such as hydrophobic hydration, repulsion of solute molecules by solvent having a higher surface tension, formation of aggregates of solute at the interface by attraction of unscreened hydrophobic parts, dimerization of the solute, formation of micelle-like aggregates and clathrate hydrate-like aggregates, structural change, etc. At higher concentrations the surface phenomena possibly remain unchanged. A plot of our density data on the same graph shows an interesting correlation of surface and bulk properties for this system. This observation is strongly supported by data reported by Yano[28] for aqueous solutions of alcohols, where it was found that the mole fraction of maximum surface excess coincides with that corresponding to the minimum partial molar volumes of alcohols, such as, methanol, ethanol, 1-propanol, 2-propanol, *t*-butanol and 1-butanol - providing good examples of correlation between surface and bulk properties.

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