# Excess Molar Volumes and Kinematic Viscosities for Binary Mixtures of Dipropylene Glycol Monobutyl Ether and Dipropylene Glycol *tert*-Butyl Ether with 2-Pyrrolidinone, *N*-Methyl-2-pyrrolidinone, *N*,*N*-Dimethylformamide, and *N*,*N*-Dimethylacetamide at 298.15 K

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Excess molar volumes $V_m^E$ and kinematic viscosities $\nu$ have been measured as a function of composition for binary mixtures of dipropylene glycol monobutyl ether and dipropylene glycol *tert*-butyl ether with 2-pyrrolidinone, *N*-methyl-2 -pyrrolidinone, *N*,*N*-dimethylformamide, and *N*,*N*-dimethylacetamide at 298.15 K. In all mixtures, the excess molar volumes are negative across the entire composition range. From the experimental data, the deviation in dynamic viscosity $\eta$ from $\Sigma x_i \eta_i$ had been calculated. Both excess molar volumes and viscosity deviations are fit to a Redlich−Kister-type polynomial equation to estimate the binary coefficient and standard errors. The experimental and calculated quantities were used to analyze the mixing behavior of the components. Equations such as Heric's, Auslaender's, and McAllister's four-body interaction model were used to correlate the kinematic viscosities.

## Introduction

Systematic studies of thermodynamic, transport, and acoustic properties of mixtures containing straight-chain alkoxypropanols with *n*-alkylamines,[1,2] or *n*-alkanols[3−6] have been carried out in our laboratory with the aim of a better understanding of the association behavior of the amine or alcohol and the ether at 298.15 K. In our present investigation, we report new experimental data for the excess molar volumes $V_m^E$ and dynamic viscosities $\eta$ of binary solvent mixtures containing dipropylene glycol monobutyl ether or dipropylene glycol *tert*-butyl ether with 2-pyrrolidinone, *N*-methyl-2-pyrrolidinone, *N*,*N*-dimethylformamide, and *N*,*N*-dimethylacetamide over the whole composition range at 298.15 K and atmospheric pressure. The present study was undertaken to assess whether the physicochemical properties of *n*-alkoxypropanols + amide systems resemble those of *n*-alkoxyethanols + amide systems.[7−11] An attempt is also made to compare the excess molar properties of dipropylene glycol monobutyl ether + *N*,*N*-dialkyl amides or cyclic amides with those of dipropylene glycol *tert*-butyl ether + *N*,*N*-dialkyl amides or cyclic amides. The effect of specific interactions on the excess properties, the dependence on the position of the $CH_3$ group in the dipropylene glycol monobutyl ether, and the influence of *N*,*N*-dialkyl amides or cyclic amides is analyzed. We are not aware of any volumetric and viscometric data available in the literature for the present systems.

## Experimental Section

***Materials.*** *N*,*N*-Dimethylformamide (DMF) (S. D. fine chemicals, Bombay; HPLC, GLC > 99.5 mol %), *N*-methyl-2-pyrrolidinone (NM2P) (S. D. fine chemicals, Bombay; LR, GLC > 99.7 mol %), *N*,*N* -dimethylacetamide (DMA) (S. D. fine chemicals, Bombay; HPLC, GLC > 99.5 mol %), and 2-pyrrolidinone (2P) (Merck-Schuchardt; FRG, GC > 99 mol %) were the same as those used in our earlier study.[9] Dipropylene glycol monobutyl ether (DPGBE) and dipropylene glycol *tert*-butyl ether (DPGTBE) were obtained from Aldrich with a purity better than 99 mol %. All amide samples were purified as previously described.[12] All liquids were stored and protected from atmospheric pressure and $CO_2$ and carefully dried over 0.4 nm molecular sieves to reduce water content. Prior to experimental measurements, all liquids were partially degassed with a vacuum pump. The densities and viscosities of these liquids were measured and compared at (298.15 ± 0.01) K and atmospheric pressure with their corresponding literature values[13−20] as shown in Table 1. The densities of pure liquids were measured using a single-stem pycnometer having a total volume of 8 $cm^3$ and an internal capillary diameter of about 0.1 cm. The reproducibility of the density measurements was better than $3 \times 10^{-5}$ g $cm^{-3}$.

***Apparatus and Procedure.*** Excess molar volumes, which are reproducible to ±0.003 $cm^3$ $mol^{-1}$, were measured with a continuous dilution dilatometer per Dickinson et al.[21] Details of its calibration, the experimental setup, and measuring procedure have been described previously.[22,23] Each run covered just over half of the mole fraction range so as to give an overlap between two runs. The mole fraction of each mixture was obtained with an uncertainty of $1 \times 10^{-4}$ from the measured apparent mass of one of the components. All masses were corrected for buoyancy. All molar quantities were based upon the IUPAC table of atomic weights.[24]

The kinematic viscosities of both the pure liquids and liquids mixtures were measured at 298.15 K and atmospheric pressure using a Ubbelohde suspended-level viscometer.[25] Experimental details have been given previously.[26,27] The viscometer is filled with liquid or liquid mixtures, and its limbs are closed with Teflon caps, taking due precaution to minimize evaporation losses. The flow-time measurements were made using an electronic stopwatch with a precision of ±0.01 s. An average of four or five sets of flow times was taken for each liquid and liquid

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**Table 1. Comparison of Experimental Densities, $\rho$, and Dynamic Viscosities, $\eta$, of Pure Liquids with Literature Values at 298.15 K**

| | $\rho$/g cm$^{-3}$ | | $\eta$/mPa s | |
|---|---|---|---|---|
| liquid | exptl | lit | exptl | lit |
| dipropylene glycol monobutyl ether | 0.90865 | 0.9089[13] | 4.357 | 4.503[13] |
| dipropylene glycol *tert*-butyl ether | 0.90746 | | 6.255 | |
| 2-pyrrolidinone | 1.10682 | 1.107019[14] | 13.194 | 13.363[14] |
| *N*-methyl-2-pyrrolidinone | 1.02791 | 1.02831[14] | 1.672 | 1.663[14] |
| | | 1.0279[15] | | 1.6615[16] |
| | | 1.02794[16] | | |
| *N*,*N*-dimethylformamide | 0.94365 | 0.9439[15] | 0.804 | 0.8012[17] |
| | | 0.94389[17] | | 0.802[18] |
| *N*,*N*-dimethylacetamide | 0.93654 | 0.9366[19] | 0.944 | 0.9437[20] |

mixture. The caps of the limbs were removed during the measurements of flow times. The measured values of kinematic viscosity ($\nu$) were converted to dynamic viscosity ($\eta$) after multiplication by the density. The reproducibility of dynamic viscosity results was found to be within ±0.003 mPa s. A thermostatically controlled, well-stirred water bath whose temperature was controlled to ±0.01 K was used for all of the measurements.

## Results and Discussion

Results obtained experimentally for excess molar volumes of all binary mixtures over a range of mole fraction at 298.15 K and atmospheric pressure are given in Table 2 and are graphically represented in Figures 1 and 2. The viscosities are given in Table 3 and are graphically shown in Figure 3.

The densities of the liquid mixtures were calculated from the excess molar volumes and densities of pure liquids according to the equation

$$\rho = \frac{x_1 M_1 + x_2 M_2}{V_m^E + x_1 V_1^* + x_1 V_2^*} \quad (1)$$

where $x_1$ and $x_2$ are the mole fractions, $M_1$ and $M_2$ are the molar masses, and $V_1^*$ and $V_2^*$ are the molar volumes of ether (1) and amide (2), respectively. The accuracy of the density $\rho$ due to the estimated accuracy of the excess volume (±0.003 cm$^{-3}$ mol$^{-1}$) is $3 \times 10^{-5}$ g cm$^{-3}$.

The deviation of viscosity from a linear dependence on the average mole fraction has been calculated from the relationship

$$\Delta\eta = \eta - (x_1\eta_1 + x_2\eta_2) \quad (2)$$

where $\eta$, $\eta_1$, and $\eta_2$ are the viscosities of the mixtures and components 1 and 2, respectively. The variations of $\Delta\eta$ with mole fraction of ether at 298.15 K are shown in Figure 4.

The values of $V_m^E$ and $\Delta\eta$ for each mixture were correlated to the Redlich−Kister polynomial equation

$$Y(x) = x_1 x_2 \sum_{i=0} A_i (x_1 - x_2)^i \quad (3)$$

Values of coefficients $A_i$ and the standard deviation $\sigma$ are summarized in Table 4.

Figures 1 and 2 show that, for all of the mixture studied, the $V_m^E$ values are negative over the whole mole fraction range. In all mixtures, the minimum occurs around $x_1 = 0.5$. With each amide except *N*,*N*-dimethylacetamide, $V_m^E$ is more negative with dipropylene glycol *tert*-butyl ether than with dipropylene glycol monobutyl ether. This contrasts with the behavior of the $V_m^E$ values for 2-propoxyethanol or 2-isopropoxyethanol + 1-propanol or 2-propanol[28] and + propylamine or dipropylamine,[29] where the

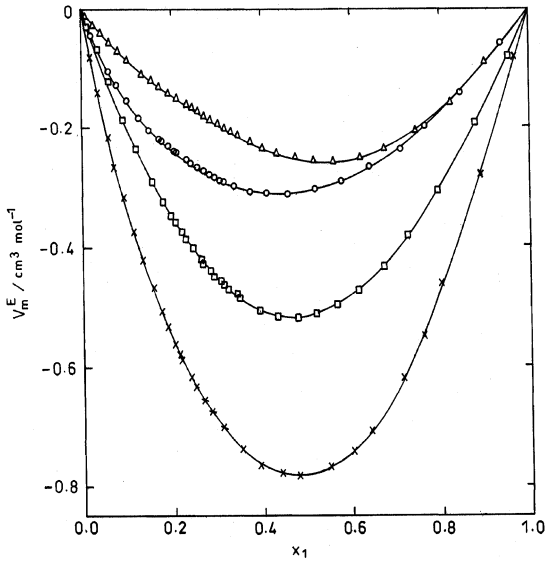


**Figure 1.** Comparison of the excess molar volumes $V_m^E$ at 298.15 K for dipropylene glycol monobutyl ether (1) + *N*,*N*-dimethylformamide (2) ○, + *N*,*N*-dimethylacetamide (2) △, + *N*-methyl-2-pyrrolidinone (2) □, and + 2-pyrrolidinone (2) ×. The solid curves have been drawn from eq 3.

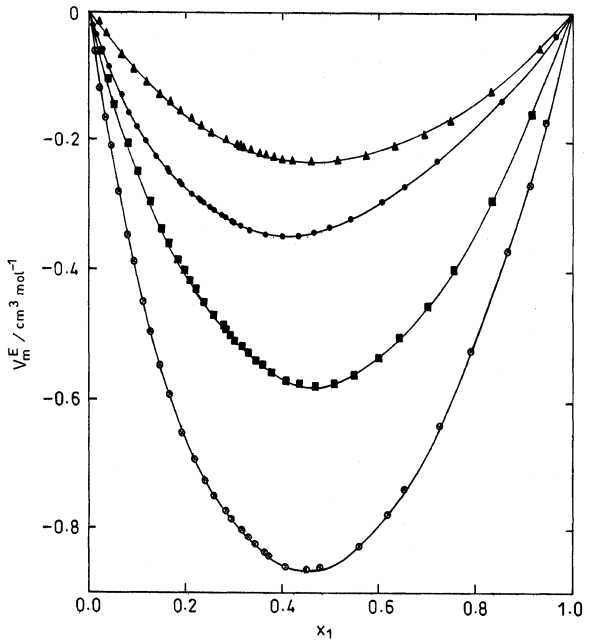


**Figure 2.** Comparison of the excess molar volumes $V_m^E$ at 298.15 K for dipropylene glycol *tert*-butyl ether (1) + *N*,*N*-dimethylformamide (2) ●, + *N*,*N*-dimethylacetamide (2) ▲, + *N*-methyl-2-pyrrolidinone (2) ■, and + 2-pyrrolidinone (2) ⊗. The solid curves have been drawn from eq 3.

**Table 2. Excess Molar Volumes $V_m^E$ for the Binary Mixtures at 298.15 K**

| $x_1$ | $V_m^E$ / cm$^3$ mol$^{-1}$ | $x_1$ | $V_m^E$ / cm$^3$ mol$^{-1}$ | $x_1$ | $V_m^E$ / cm$^3$ mol$^{-1}$ | $x_1$ | $V_m^E$ / cm$^3$ mol$^{-1}$ | $x_1$ | $V_m^E$ / cm$^3$ mol$^{-1}$ | $x_1$ | $V_m^E$ / cm$^3$ mol$^{-1}$ |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Dipropylene Glycol Monobutyl Ether (1) + *N*,*N*-Dimethylformamide (2) | | | | | | | | | | | |
| 0.0058 | −0.013 | 0.1290 | −0.185 | 0.2078 | −0.244 | 0.2826 | −0.281 | 0.3721 | −0.309 | 0.6418 | −0.266 |
| 0.0218 | −0.047 | 0.1501 | −0.204 | 0.2310 | −0.257 | 0.2928 | −0.284 | 0.4099 | −0.311 | 0.7139 | −0.233 |
| 0.0320 | −0.066 | 0.1738 | −0.220 | 0.2371 | −0.261 | 0.3085 | −0.289 | 0.4612 | −0.310 | 0.7679 | −0.196 |
| 0.0579 | −0.104 | 0.1758 | −0.221 | 0.2517 | −0.268 | 0.3100 | −0.292 | 0.5176 | −0.302 | 0.8499 | −0.138 |
| 0.0760 | −0.129 | 0.1920 | −0.235 | 0.2631 | −0.274 | 0.3370 | −0.301 | 0.5768 | −0.289 | 0.9375 | −0.057 |
| 0.0989 | −0.155 | 0.2069 | −0.242 | 0.2773 | −0.277 | | | | | | |
| Dipropylene Glycol Monobutyl Ether (1) + *N*,*N*-Dimethylacetamide (2) | | | | | | | | | | | |
| 0.0079 | −0.008 | 0.1334 | −0.108 | 0.2382 | −0.164 | 0.3107 | −0.199 | 0.4344 | −0.245 | 0.6730 | −0.235 |
| 0.0244 | −0.024 | 0.1530 | −0.121 | 0.2508 | −0.171 | 0.3238 | −0.205 | 0.4715 | −0.250 | 0.7481 | −0.203 |
| 0.0419 | −0.039 | 0.1693 | −0.129 | 0.2628 | −0.177 | 0.3433 | −0.214 | 0.5144 | −0.255 | 0.8235 | −0.157 |
| 0.0604 | −0.056 | 0.1920 | −0.141 | 0.2729 | −0.182 | 0.3732 | −0.226 | 0.5624 | −0.256 | 0.8983 | −0.090 |
| 0.0800 | −0.070 | 0.2092 | −0.150 | 0.2804 | −0.185 | 0.4020 | −0.235 | 0.6216 | −0.248 | | |
| 0.0984 | −0.084 | 0.2302 | −0.160 | 0.2988 | −0.194 | | | | | | |
| Dipropylene Glycol Monobutyl Ether (1) + *N*-Methyl-2-pyrrolidinone (2) | | | | | | | | | | | |
| 0.0113 | −0.027 | 0.1805 | −0.325 | 0.2431 | −0.401 | 0.3068 | −0.456 | 0.4356 | −0.514 | 0.6744 | −0.430 |
| 0.0302 | −0.064 | 0.1987 | −0.353 | 0.2607 | −0.421 | 0.3224 | −0.462 | 0.4833 | −0.517 | 0.7292 | −0.380 |
| 0.0588 | −0.121 | 0.2020 | −0.356 | 0.2698 | −0.426 | 0.3456 | −0.480 | 0.5204 | −0.510 | 0.7908 | −0.304 |
| 0.0892 | −0.185 | 0.2207 | −0.379 | 0.2858 | −0.441 | 0.3461 | −0.482 | 0.5640 | −0.497 | 0.8817 | −0.188 |
| 0.1187 | −0.235 | 0.2246 | −0.381 | 0.2892 | −0.447 | 0.3954 | −0.504 | 0.6160 | −0.470 | 0.9563 | −0.079 |
| 0.1547 | −0.288 | 0.2415 | −0.397 | 0.3063 | −0.455 | | | | | | |
| Dipropylene Glycol Monobutyl Ether (1) + 2-Pyrrolidinone (2) | | | | | | | | | | | |
| 0.0060 | −0.024 | 0.1148 | −0.373 | 0.2176 | −0.583 | 0.3107 | −0.705 | 0.5443 | −0.765 | 0.8021 | −0.461 |
| 0.0203 | −0.081 | 0.1361 | −0.420 | 0.2222 | −0.587 | 0.3500 | −0.737 | 0.5982 | −0.738 | 0.8888 | −0.271 |
| 0.0372 | −0.142 | 0.1568 | −0.465 | 0.2417 | −0.621 | 0.3953 | −0.766 | 0.6378 | −0.706 | 0.9690 | −0.078 |
| 0.0606 | −0.213 | 0.1749 | −0.504 | 0.2489 | −0.628 | 0.4441 | −0.777 | 0.7158 | −0.614 | | |
| 0.0747 | −0.264 | 0.1874 | −0.530 | 0.2685 | −0.658 | 0.4832 | −0.779 | 0.7578 | −0.548 | | |
| 0.0946 | −0.322 | 0.2034 | −0.560 | 0.2856 | −0.672 | | | | | | |
| Dipropylene Glycol *tert*-Butyl Ether (1) + *N*,*N*-Dimethylformamide (2) | | | | | | | | | | | |
| 0.0092 | −0.018 | 0.1196 | −0.202 | 0.2112 | −0.282 | 0.2613 | −0.309 | 0.3680 | −0.344 | 0.6048 | −0.294 |
| 0.0265 | −0.058 | 0.1424 | −0.227 | 0.2123 | −0.283 | 0.2806 | −0.317 | 0.4006 | −0.346 | 0.6525 | −0.271 |
| 0.0425 | −0.085 | 0.1666 | −0.249 | 0.2267 | −0.290 | 0.2944 | −0.324 | 0.4304 | −0.343 | 0.7196 | −0.231 |
| 0.0658 | −0.129 | 0.1686 | −0.250 | 0.2321 | −0.294 | 0.2961 | −0.325 | 0.4660 | −0.338 | 0.8554 | −0.135 |
| 0.0818 | −0.155 | 0.1897 | −0.268 | 0.2411 | −0.298 | 0.3163 | −0.333 | 0.5007 | −0.332 | 0.9679 | −0.035 |
| 0.1029 | −0.182 | 0.1911 | −0.269 | 0.2576 | −0.309 | 0.3367 | −0.340 | 0.5447 | −0.319 | | |
| Dipropylene Glycol *tert*-Butyl Ether (1) + *N*,*N*-Dimethylacetamamide (2) | | | | | | | | | | | |
| 0.0171 | −0.010 | 0.1479 | −0.129 | 0.2502 | −0.185 | 0.3349 | −0.214 | 0.4207 | −0.230 | 0.6903 | −0.189 |
| 0.0359 | −0.032 | 0.1642 | −0.140 | 0.2847 | −0.197 | 0.3552 | −0.218 | 0.4627 | −0.233 | 0.7493 | −0.167 |
| 0.0651 | −0.066 | 0.1943 | −0.157 | 0.3064 | −0.205 | 0.3609 | −0.219 | 0.5101 | −0.231 | 0.8305 | −0.125 |
| 0.0919 | −0.091 | 0.2116 | −0.165 | 0.3119 | −0.207 | 0.3789 | −0.223 | 0.5774 | −0.218 | 0.9311 | −0.054 |
| 0.1182 | −0.110 | 0.2363 | −0.179 | 0.3297 | −0.211 | 0.4027 | −0.228 | 0.6328 | −0.206 | | |
| Dipropylene Glycol *tert*-Butyl Ether (1) + *N*-Methyl-2-pyrrolidinone (2) | | | | | | | | | | | |
| 0.0113 | −0.036 | 0.1496 | −0.338 | 0.2391 | −0.451 | 0.3179 | −0.521 | 0.4357 | −0.576 | 0.6997 | −0.457 |
| 0.0374 | −0.107 | 0.1666 | −0.361 | 0.2577 | −0.472 | 0.3242 | −0.526 | 0.4659 | −0.579 | 0.7550 | −0.400 |
| 0.0520 | −0.143 | 0.1850 | −0.385 | 0.2771 | −0.490 | 0.3457 | −0.541 | 0.5097 | −0.573 | 0.8334 | −0.294 |
| 0.0778 | −0.204 | 0.1994 | −0.406 | 0.2783 | −0.492 | 0.3541 | −0.546 | 0.5486 | −0.562 | 0.9114 | −0.162 |
| 0.0968 | −0.248 | 0.2089 | −0.420 | 0.2944 | −0.506 | 0.3772 | −0.560 | 0.5980 | −0.535 | | |
| 0.1241 | −0.296 | 0.2230 | −0.432 | 0.2989 | −0.509 | 0.4059 | −0.569 | 0.6398 | −0.507 | | |
| Dipropylene Glycol *tert*-Butyl Ether (1) + 2-Pyrrolidinone (2) | | | | | | | | | | | |
| 0.0108 | −0.061 | 0.0911 | −0.388 | 0.2179 | −0.688 | 0.3323 | −0.816 | 0.4512 | −0.863 | 0.7259 | −0.640 |
| 0.0218 | −0.115 | 0.1111 | −0.448 | 0.2404 | −0.723 | 0.3488 | −0.825 | 0.4825 | −0.858 | 0.7942 | −0.523 |
| 0.0330 | −0.167 | 0.1286 | −0.495 | 0.2624 | −0.752 | 0.3685 | −0.840 | 0.5577 | −0.823 | 0.8681 | −0.372 |
| 0.0440 | −0.212 | 0.1481 | −0.545 | 0.2842 | −0.775 | 0.3764 | −0.841 | 0.6167 | −0.778 | 0.9110 | −0.267 |
| 0.0608 | −0.282 | 0.1669 | −0.589 | 0.2960 | −0.785 | 0.4076 | −0.859 | 0.6546 | −0.737 | 0.9456 | −0.170 |
| 0.0785 | −0.344 | 0.1958 | −0.648 | 0.3183 | −0.805 | | | | | | |

$V_m^E$ values are more negative with 2-propoxyethanol. Also, their behavior may be compared with the $V_m^E$ results for mixtures of 2-propoxyethanol or 2-isopropoxyethanol with water[30,31] or 1-propanol and 2-propanol with water:[32] branching of the alkyl chain at the α-carbon leads to more negative excess molar volumes. The most striking features in Figures 1 and 2 are the strong similarities between the composition-dependence curves of the two alkoxypropanols + cyclic amides or *N*,*N*-dialkyl amides, which is in sharp contrast to the differences between the two alkoxypropanols + *N*,*N*-dimethylacetamide curves. However, similar characteristic behavior of $V_m^E$ for mixtures of dipropylene glycol monobutyl ether or dipropylene *tert*-butyl ether with amides is obtained with 2-isopropoxyethanol + amides.[9] The $V_m^E$ is more negative for mixtures containing alkoxypropanols with cyclic amides than for those containing *N*,*N*-dialkyl amides. The behavior is consistent with that of the negative $V_m^E$ results for dipropylene glycol monobutyl ether + *n*-alkanol.[6] These results and those from our earlier data of diethylene glycol monobutyl ether with 2-pyrrolidinone[7] or *N*-methyl-2-pyrrolidinone[11] show that with each cyclic amide $V_m^E$ decreases in the order dipropylene glycol *tert*-butyl ether > dipropylene glycol monobutyl ether > diethylene glycol monobutyl ether. With each alkoxypropanol, $V_m^E$ is more negative with 2-pyrrolidinone

**Table 3. Kinematic Viscosities, $\nu$, and Dynamic Viscosities, $\eta$, for the Binary Mixtures at 298.15 K**

| $x_1$ | $\nu$ / mm$^2$ s$^{-1}$ | $\eta$ / mPa s | $x_1$ | $\nu$ / mm$^2$ s$^{-1}$ | $\eta$ / mPa s | $x_1$ | $\nu$ / mm$^2$ s$^{-1}$ | $\eta$ / mPa s | $x_1$ | $\nu$ / mm$^2$ s$^{-1}$ | $\eta$ / mPa s |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Dipropylene Glycol Monobutyl Ether (1) + *N*,*N*-Dimethylformamide (2) | | | | | | | | | | | |
| 0.0203 | 0.912 | 0.859 | 0.1946 | 1.514 | 1.411 | 0.4601 | 2.626 | 2.419 | 0.7733 | 3.951 | 3.608 |
| 0.0316 | 0.943 | 0.888 | 0.2263 | 1.639 | 1.525 | 0.5238 | 2.896 | 2.663 | 0.8089 | 4.097 | 3.738 |
| 0.0442 | 0.985 | 0.927 | 0.2659 | 1.801 | 1.672 | 0.5815 | 3.142 | 2.883 | 0.8697 | 4.330 | 3.945 |
| 0.0726 | 1.077 | 1.011 | 0.3235 | 2.041 | 1.890 | 0.6234 | 3.319 | 3.042 | 0.9487 | 4.618 | 4.200 |
| 0.1120 | 1.211 | 1.134 | 0.3649 | 2.217 | 2.050 | 0.6745 | 3.535 | 3.236 | 0.9957 | 4.780 | 4.344 |
| 0.1677 | 1.414 | 1.320 | 0.4142 | 2.428 | 2.241 | | | | | | |
| Dipropylene Glycol Monobutyl Ether (1) + *N*,*N*-Dimethylacetamide (2) | | | | | | | | | | | |
| 0.0097 | 1.037 | 0.981 | 0.1900 | 1.752 | 1.637 | 0.4086 | 2.649 | 2.450 | 0.7039 | 3.852 | 3.527 |
| 0.0316 | 1.125 | 1.062 | 0.2090 | 1.828 | 1.706 | 0.4671 | 2.892 | 2.669 | 0.8068 | 4.207 | 3.841 |
| 0.0451 | 1.177 | 1.110 | 0.2338 | 1.927 | 1.797 | 0.5388 | 3.196 | 2.942 | 0.8549 | 4.360 | 3.976 |
| 0.0678 | 1.266 | 1.192 | 0.2568 | 2.022 | 1.883 | 0.5983 | 3.444 | 3.164 | 0.8942 | 4.480 | 4.081 |
| 0.0936 | 1.369 | 1.287 | 0.3330 | 2.335 | 2.167 | 0.6417 | 3.616 | 3.318 | 0.9487 | 4.650 | 4.230 |
| 0.1567 | 1.617 | 1.514 | 0.3687 | 2.484 | 2.301 | | | | | | |
| Dipropylene Glycol Monobutyl Ether (1) + *N*-Methyl-2-pyrrolidinone (2) | | | | | | | | | | | |
| 0.0136 | 1.672 | 1.713 | 0.2264 | 2.579 | 2.552 | 0.4766 | 3.628 | 3.454 | 0.7310 | 4.374 | 4.059 |
| 0.0391 | 1.769 | 1.803 | 0.2504 | 2.700 | 2.649 | 0.5287 | 3.813 | 3.609 | 0.8089 | 4.538 | 4.183 |
| 0.0742 | 1.913 | 1.935 | 0.2730 | 2.798 | 2.735 | 0.5827 | 3.985 | 3.750 | 0.8762 | 4.642 | 4.256 |
| 0.0896 | 1.970 | 1.987 | 0.3222 | 3.014 | 2.926 | 0.6327 | 4.129 | 3.866 | 0.9164 | 4.694 | 4.291 |
| 0.1435 | 2.214 | 2.211 | 0.3627 | 3.184 | 3.074 | 0.6667 | 4.222 | 3.940 | 0.9545 | 4.738 | 4.319 |
| 0.2008 | 2.471 | 2.443 | 0.4195 | 3.416 | 3.274 | | | | | | |
| Dipropylene Glycol Monobutyl Ether (1) + 2-Pyrrolidinone (2) | | | | | | | | | | | |
| 0.0073 | 11.828 | 13.050 | 0.2157 | 9.665 | 9.932 | 0.5256 | 7.255 | 6.987 | 0.8431 | 5.534 | 5.109 |
| 0.0183 | 11.705 | 12.854 | 0.2721 | 9.172 | 9.289 | 0.5808 | 6.890 | 6.579 | 0.8832 | 5.351 | 4.919 |
| 0.0575 | 11.242 | 12.156 | 0.3227 | 8.745 | 8.753 | 0.6403 | 6.540 | 6.191 | 0.9551 | 5.019 | 4.580 |
| 0.1050 | 10.737 | 11.412 | 0.3637 | 8.422 | 8.355 | 0.6774 | 6.332 | 5.964 | 0.9838 | 4.878 | 4.439 |
| 0.1680 | 10.103 | 10.524 | 0.4090 | 8.076 | 7.939 | 0.7760 | 5.842 | 5.435 | | | |
| 0.1922 | 9.876 | 10.215 | 0.4725 | 7.618 | 7.402 | | | | | | |
| Dipropylene Glycol *tert*-Butyl Ether (1) + *N*,*N*-Dimethylformamide (2) | | | | | | | | | | | |
| 0.0108 | 0.885 | 0.834 | 0.1934 | 1.587 | 1.479 | 0.4346 | 2.858 | 2.634 | 0.7519 | 5.001 | 4.563 |
| 0.0238 | 0.923 | 0.869 | 0.2186 | 1.704 | 1.586 | 0.4779 | 3.121 | 2.872 | 0.8090 | 5.427 | 4.945 |
| 0.0517 | 1.015 | 0.954 | 0.2347 | 1.780 | 1.655 | 0.5084 | 3.312 | 3.044 | 0.8539 | 5.773 | 5.255 |
| 0.0770 | 1.104 | 1.036 | 0.2630 | 1.923 | 1.785 | 0.5648 | 3.684 | 3.380 | 0.8891 | 6.037 | 5.491 |
| 0.1095 | 1.227 | 1.149 | 0.3234 | 2.238 | 2.072 | 0.6213 | 4.069 | 3.726 | 0.9729 | 6.677 | 6.063 |
| 0.1430 | 1.363 | 1.274 | 0.3962 | 2.640 | 2.436 | 0.6516 | 4.279 | 3.915 | | | |
| Dipropylene Glycol *tert*-Butyl Ether (1) + *N*,*N*-Dimethylacetamide (2) | | | | | | | | | | | |
| 0.0063 | 1.026 | 0.971 | 0.1621 | 1.678 | 1.570 | 0.3584 | 2.674 | 2.477 | 0.7498 | 5.228 | 4.775 |
| 0.0128 | 1.055 | 0.998 | 0.1771 | 1.745 | 1.631 | 0.4371 | 3.152 | 2.909 | 0.8172 | 5.693 | 5.190 |
| 0.0260 | 1.114 | 1.052 | 0.2060 | 1.875 | 1.750 | 0.4932 | 3.506 | 3.229 | 0.8646 | 6.015 | 5.477 |
| 0.0385 | 1.167 | 1.101 | 0.2369 | 2.023 | 1.885 | 0.5540 | 3.906 | 3.590 | 0.9159 | 6.360 | 5.783 |
| 0.0673 | 1.286 | 1.211 | 0.2538 | 2.108 | 1.962 | 0.6263 | 4.386 | 4.021 | 0.9518 | 6.591 | 5.988 |
| 0.1023 | 1.428 | 1.341 | 0.3141 | 2.426 | 2.252 | 0.6666 | 4.663 | 4.269 | 0.9758 | 6.739 | 6.119 |
| Dipropylene Glycol *tert*-Butyl Ether (1) + *N*-Methyl-2-pyrrolidinone (2) | | | | | | | | | | | |
| 0.0144 | 1.680 | 1.722 | 0.1758 | 2.493 | 2.476 | 0.4191 | 3.997 | 3.830 | 0.7111 | 5.706 | 5.300 |
| 0.0220 | 1.710 | 1.749 | 0.2097 | 2.692 | 2.658 | 0.4785 | 4.368 | 4.155 | 0.7883 | 6.094 | 5.622 |
| 0.0332 | 1.756 | 1.792 | 0.2267 | 2.795 | 2.752 | 0.5483 | 4.793 | 4.524 | 0.8341 | 6.300 | 5.790 |
| 0.0611 | 1.880 | 1.907 | 0.2653 | 3.033 | 2.968 | 0.5908 | 5.045 | 4.741 | 0.8904 | 6.526 | 5.971 |
| 0.0867 | 2.008 | 2.027 | 0.3460 | 3.533 | 3.417 | 0.6698 | 5.485 | 5.114 | 0.9603 | 6.786 | 6.176 |
| 0.1331 | 2.256 | 2.257 | 0.3861 | 3.786 | 3.643 | | | | | | |
| Dipropylene Glycol *tert*-Butyl Ether (1) + 2-Pyrrolidinone (2) | | | | | | | | | | | |
| 0.0045 | 11.943 | 13.193 | 0.2060 | 12.217 | 12.593 | 0.4717 | 11.038 | 10.723 | 0.8100 | 8.584 | 7.946 |
| 0.0200 | 12.011 | 13.185 | 0.2399 | 12.154 | 12.414 | 0.5334 | 10.628 | 10.217 | 0.9026 | 7.817 | 7.163 |
| 0.0569 | 12.133 | 13.129 | 0.2685 | 12.075 | 12.243 | 0.6368 | 9.901 | 9.370 | 0.9475 | 7.428 | 6.776 |
| 0.1050 | 12.232 | 13.010 | 0.3451 | 11.765 | 11.718 | 0.6843 | 9.557 | 8.986 | 0.9766 | 7.155 | 6.507 |
| 0.1529 | 12.261 | 12.838 | 0.4028 | 11.466 | 11.284 | 0.7549 | 9.021 | 8.406 | | | |

than with *N*-methyl-2-pyrrolidinone. The large, more-negative values of $V_m^E$ suggest a strong chemical or specific interaction between the components, which is maximized in the case of dipropylene glycol *tert*-butyl ether-2-pyrrolidinone. That is, the more-negative excess volumes for the mixtures of 2-pyrrolidinone indicate that substitution of a proton for the methyl group of *N*-methyl-2 -pyrrolidinone results in a less-negative contribution to the excess volume. Again, the dissociation of self-associated species of 2-pyrrolidinone by dipropylene glycol *tert*-butyl ether and/or because of simultaneous interaction, mainly due to hydrogen bonding in addition to interstitial accommodation, leads to more negative $V_m^E$ for 2-pyrrolidinone + dipropylene glycol *tert*-butyl ether compared with that of dipropylene glycol monobutyl ether + 2-pyrrolidinone mixtures.

The more-negative $V_m^E$ values for dimethylformamide might be due to the somewhat better proton-accepting ability of dimethylformamide than that of dimethylacetamide but also to the more effective association of the oxygen atom of dimethylformamide to the hydrogen atom of alkoxypropanol than to the oxygen atom of dimethylacetamide. Furthermore, because of the steric hindrance of the two methyl groups of dimethylformamide, the strength of interactions due to the formation of O−H- - -O as compared to O−H- - -N bonds is expected to decrease with straight-

**Table 4. Values of the Parameters $A_i$ and Standard Deviation $\sigma$ Obtained with Equation 3 for Various Mixtures at 298.15 K**

| | | $A_0$ | $A_1$ | $A_2$ | $A_3$ | $A_4$ | $\sigma$ |
|---|---|---|---|---|---|---|---|
| dipropylene glycol monobutyl ether (1) + *N*,*N*-dimethyl formamide (2) | $V_m^E$/cm$^3$ mol$^{-1}$ | −1.222 | 0.219 | −0.215 | 0.354 | | 0.003 |
| | $\Delta\eta$/mPa s | −0.041 | 0.639 | −0.110 | 0.200 | | 0.003 |
| *N*,*N*-dimethyl acetamide (2) | $V_m^E$/cm$^3$ mol$^{-1}$ | −1.016 | −0.238 | 0.074 | 0.308 | | 0.002 |
| | $\Delta\eta$/mPa s | 0.596 | 0.694 | 0.023 | −0.457 | | 0.003 |
| *N*-methyl-2-pyrrolidinone (2) | $V_m^E$/cm$^3$ mol$^{-1}$ | −2.053 | 0.269 | 0.058 | 0.018 | −0.427 | 0.003 |
| | $\Delta\eta$/mPa s | 2.039 | 0.494 | −0.477 | 0.273 | | 0.003 |
| 2-pyrrolidinone (2) | $V_m^E$/cm$^3$ mol$^{-1}$ | −3.124 | 0.265 | −0.169 | 0.508 | | 0.003 |
| | $\Delta\eta$/mPa s | −6.385 | 2.035 | −0.657 | 1.477 | | 0.004 |
| dipropylene glycol *tert*-butyl ether (1) + *N*,*N*-dimethylformamide (2) | $V_m^E$/cm$^3$ mol$^{-1}$ | −1.333 | 0.484 | −0.069 | 0.092 | −0.390 | 0.002 |
| | $\Delta\eta$/mPa s | −2.121 | 0.502 | 0.244 | 0.178 | −0.339 | 0.003 |
| *N*,*N*-dimethylacetamide (2) | $V_m^E$/cm$^3$ mol$^{-1}$ | −0.919 | 0.104 | −0.058 | 0.003 | −0.826 | 0.003 |
| | $\Delta\eta$/mPa s | −1.321 | 1.082 | −0.012 | −0.538 | | 0.002 |
| *N*-methyl-2-pyrrolidinone (2) | $V_m^E$/cm$^3$ mol$^{-1}$ | −2.301 | 0.274 | 0.069 | 0.257 | −0.405 | 0.003 |
| | $\Delta\eta$/mPa s | 1.226 | 1.460 | −0.441 | 0.435 | | 0.004 |
| 2-pyrrolidinone (2) | $V_m^E$/cm$^3$ mol$^{-1}$ | −3.411 | 0.607 | −0.425 | 0.375 | −0.645 | 0.002 |
| | $\Delta\eta$/mPa s | 3.104 | −2.595 | 1.893 | 1.021 | | 0.008 |

chain rather than with branched-chain alkoxypropanols. In other words, the inductive effect of the alkyl group in the branched-chain alkoxypropanol increases the electron density in the oxygen atom, enhancing the formation of O−H- - -O bonding. This behavior is inconsistent with that of the $V_m^E$ for the 2-alkoxyethanols + *N*,*N*-dialkyl amides:[10] $V_m^E$ becomes more negative with dimethylacetamide than with dimethylformamide. However, the values of $V_m^E$ are found to decrease with dimethylacetamide + dipropylene glycol monobutyl ether more than with dimethylacetamide + dipropylene glycol *tert*-butyl ether, which is attributable to the increased interaction involving hydrogen bonding/dipole−dipole interaction between dipropylene glycol monobutyl ether and dimethylacetamide molecules in the mixture.

We have determined $\eta$ and calculated $\Delta\eta$ at 298.15 K. Figure 3 reveals that $\eta$ decreases with 2-pyrrolidinone and increases for all other mixtures with increasing mole fraction $x_1$ of alkoxypropanol. Figure 4 shows that the viscosity deviations ($\Delta\eta$) are positive with dipropylene glycol monobutyl ether + *N*-methyl-2-pyrrolidinone, and + *N*,*N*-dimethylacetamide, and dipropylene glycol *tert*-butyl ether + 2-pyrrolidinone, negative with dipropylene glycol monobutyl ether + 2-pyrrolidinone, dipropylene glycol *tert*-butyl ether + *N*,*N*-dimethylacetamide, and + *N*,*N*-dimethylformamide, and change sign from negative to positive with dipropylene glycol monobutyl ether + *N*,*N*-dimethylformamide and dipropylene glycol *tert*-butyl ether + *N*-methyl-2-pyrrolidinone. In fact, a large positive deviation over the entire range of composition for dipropylene glycol *tert*-butyl ether + 2-pyrrolidinone and a large nega-

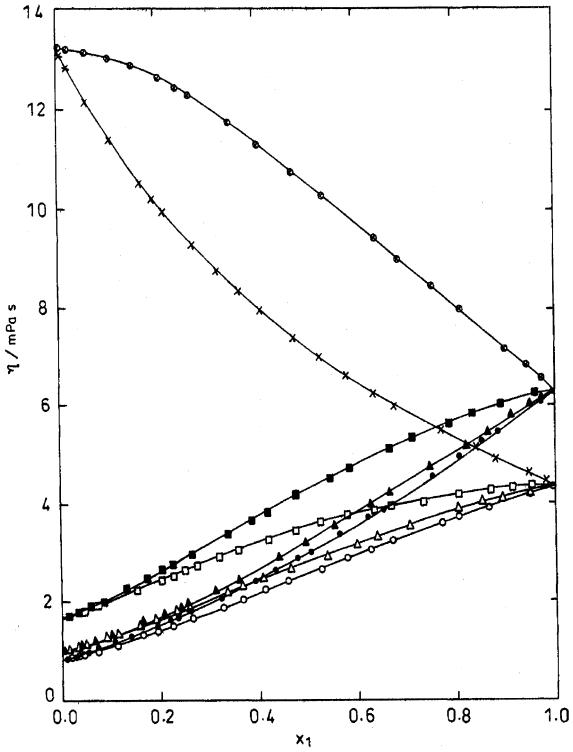


**Figure 3.** Dynamic viscosity $\eta$ at 298.15 K for dipropylene glycol monobutyl ether (1) + *N*,*N*-dimethylformamide (2) ○, + *N*,*N*-dimethylacetamide (2) △, + *N*-methyl-2-pyrrolidinone (2) □, + 2-pyrrolidinone (2) × and dipropylene glycol *tert*-butyl ether (1) + *N*,*N*-dimethylformamide (2) ●, + *N*,*N*-dimethylacetamide (2) ▲, + *N*-methyl-2-pyrrolidinone (2) ■, + 2-pyrrolidinone (2) ⊗. The solid curves have been drawn through experimental points.

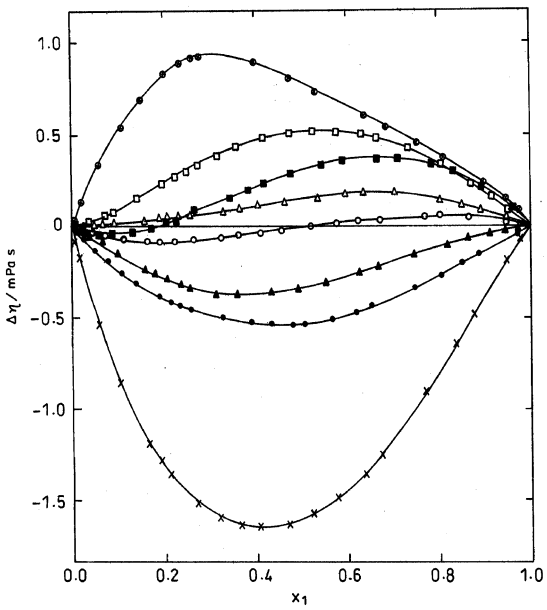


**Figure 4.** Viscosity deviations $\Delta\eta$ at 298.15 K for dipropylene glycol monobutyl ether (1) + *N*,*N*-dimethylformamide (2) ○, + *N*,*N*-dimethylacetamide (2) △, + *N*-methyl-2-pyrrolidinone (2) □, + 2-pyrrolidinone (2) × and dipropylene glycol *tert*-butyl ether (1) + *N*,*N*-dimethylformamide (2) ●, + *N*,*N*-dimethylacetamide (2) ▲, + *N*-methyl-2-pyrrolidinone (2) ■, + 2-pyrrolidinone (2) ⊗. The solid curves have been drawn from eq 3.

**Table 5. Values of the Parameters and Standard Percentage Deviation for the (Alkoxypropanol + Amide) Systems Represented by Equations 4−6**

| | eq 4 | | | | eq 5 | | | | eq 6 | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | $Z_{1112}$ | $Z_{1122}$ | $Z_{2221}$ | $\sigma$ | $A$ | $B$ | $C$ | $\sigma$ | $B_{12}$ | $B_{21}$ | $A_{21}$ | $\sigma$ |
| dipropylene glycol monobutyl ether (1) + *N*,*N*-dimethylformamide (2) | 4.07 | 4.51 | 2.13 | 0.02 | 1.74 | −0.49 | 0.10 | 0.15 | 0.49 | 1.31 | 0.66 | 0.17 |
| *N*,*N*-dimethylacetamide (2) | 4.34 | 4.25 | 2.59 | 0.27 | 1.60 | −0.56 | 0.34 | 0.52 | 0.87 | 0.72 | 0.89 | 0.41 |
| *N*-methyl-2-pyrrolidinone (2) | 6.14 | 10.14 | 9.74 | 0.05 | 0.24 | −0.04 | 0.09 | 0.23 | −2.07 | −0.49 | −1.23 | 0.56 |
| 2-pyrrolidinone (2) | 4.51 | 5.67 | 2.99 | 0.10 | 1.36 | −0.28 | −0.10 | 0.32 | 0.53 | 0.23 | 0.46 | 0.19 |
| dipropylene glycol *tert*-butyl ether (1) + *N*,*N*-dimethylformamide (2) | 5.25 | 5.28 | 2.23 | 0.07 | 1.64 | −0.43 | 0.08 | 0.27 | 0.43 | 1.51 | 0.86 | 0.27 |
| *N*,*N*-dimethylacetamide (2) | 5.78 | 4.92 | 2.62 | 0.71 | 1.49 | −0.38 | 0.31 | 0.86 | 0.66 | 1.00 | 1.08 | 0.98 |
| *N*-methyl-2-pyrrolidinone (2) | 9.90 | 15.11 | 13.91 | 0.05 | 1.04 | −0.16 | 0.24 | 0.21 | −0.43 | 6.14 | 0.30 | 0.34 |
| 2-pyrrolidinone (2) | 6.12 | 7.00 | 3.17 | 0.18 | 1.40 | −0.14 | −0.15 | 0.43 | 0.55 | 0.82 | 0.71 | 0.34 |

tive deviation for dipropylene glycol monobutyl ether + 2-pyrrolidinone are observed. This reveals that the strength of specific interaction is not the only factor influencing the viscosity deviation of liquid mixtures. The molecular size and shape of the components also play an equally important role.

Assuming a four-body interaction model, McAllister[33] proposed the following relation for the viscosity of mixtures:

$$\ln \nu = x_1^4 \ln \nu_1 + 4x_1^3 x_2 \ln Z_{1112} + 6x_1^2 x_2^2 \ln Z_{1122} + 4x_1 x_2^3 \ln Z_{2221} + x_2^4 \ln \nu_2 - \ln\left[x_1 + x_2\left(\frac{M_2}{M_1}\right)\right] + 4x_1^3 x_2 \ln\left[\frac{3 + (M_2/M_1)}{4}\right] + 6x_1^2 2x_2^2 \ln\left[\frac{1 + (M_2/M_1)}{2}\right] + 4x_1 x_2^3 \ln\left[\frac{1 + (3M_2/M_1)}{4}\right] + x_2^4 \ln\left(\frac{M_2}{M_1}\right) \quad (4)$$

where $\nu$ refers to the kinematic viscosity of the mixture, $\nu_1$ and $\nu_2$ are the kinematic viscosities of pure components 1 and 2, respectively, and $Z_{1112}$, $Z_{1122}$, and $Z_{2221}$ are interaction parameters that are characteristic of the binary system and can be evaluated by the least-squares procedure.

Heric[34] suggested the following relation for correlating the kinematic viscosities of the binary liquid mixtures:

$$\ln \nu = x_1 \ln \nu_1 + x_2 \ln \nu_2 + x_1 \ln M_1 + x_2 \ln M_2 - \ln(x_1 M_1 + x_2 M_2) + x_1 x_2 [a + b(x_1 - x_2) + c(x_1 - x_2)^2] \quad (5)$$

where $a$, $b$, and $c$ are the best-fit coefficients of the Heric equation and represent the interaction between unlike molecules.

Auslaender[35] developed the following expression for kinematic viscosities of binary mixtures

$$x_1(x_1 + B_{12}x_2)(\nu - \nu_1) + A_{21}x_2(B_{21}x_2 + x_2)(\nu - \nu_2) = 0 \quad (6)$$

where $B_{12}$, $A_{21}$, and $B_{21}$ are adjustable parameters representing binary 12 interactions. The correlating capibility of eqs 4−6 was tested by calculating the standard percentage deviation $\sigma$% between the experimental and calculated viscosities using the following relation

$$\sigma\% = \left[\frac{1}{(p-k)}\sum\left\{\frac{100(\nu_{\text{exptl}} - \nu_{\text{calcd}})}{\nu_{\text{exptl}}}\right\}^2\right]^{1/2} \quad (7)$$

where $p$ represents the number of experimental points and $k$ represents the number of numerical coefficients in the respective equations.

The values of the different fitting parameters in eqs 4−6 and the standard percentage deviation are listed in Table 5. All three correlative models are capable of representing with a higher or lesser degree of accuracy the viscometric behavior of the studied mixtures. The values of the parameters for most of the models vary regularly while going from alkylamide to cyclic amides. It is observed that the McAllister and the Heric relations fit the experimental results better as compared to the Auslaender because the $\sigma$% values for the latter equation are larger than the others in all of the systems except with *N*-methyl-2-pyrrolidinone, as evidenced by small $\sigma$% values.

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