# Excess Molar Volumes and Refractive Indexes of Heptanol + Pentane, + Hexane, + Heptane, + Octane, and + 2,2,4-Trimethylpentane

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Excess molar volumes, $V_m^E$, have been measured using a direct dilatometric technique for binary mixtures of heptanol ($n$-$C_7H_{15}OH$) + hexane ($n$-$C_6H_{14}$), + heptane ($n$-$C_7H_{16}$), + octane ($n$-$C_8H_{18}$), and + 2,2,4-trimethylpentane (2,2,4-TMP) at 308.15 K and for $n$-$C_7H_{15}OH$ + pentane ($n$-$C_5H_{12}$) at 298.15 K. The refractive indexes, $n_D$, have been measured for these mixtures at 303.15 K. $V_m^E$ has been found to be negative throughout the entire range of composition for $xn$-$C_7H_{15}OH + (1 - x)n$-$C_5H_{12}$ and $xn$-$C_7H_{15}OH + (1 - x)n$-$C_6H_{14}$. $V_m^E$ has been found to be positive at lower values of $x$ and negative at higher values of $x$ for $xn$-$C_7H_{15}OH + (1 - x)$2,2,4-TMP, $xn$-$C_7H_{15}OH + (1 - x)n$-$C_7H_{16}$, and $xn$-$C_7H_{15}OH + (1 - x)n$-$C_8H_{18}$, with the inversion of sign from positive to negative values of $V_m^E$ occurring at $x \sim 0.06$, $x \sim 0.20$, and $x \sim 0.45$ respectively for these mixtures. Values of $V_m^E$ and $n_D$ for the various mixtures of $n$-$C_7H_{15}OH$ have been fitted in smoothing equations.

## Introduction

Binary (an alkanol + an alkane) mixtures are of considerable interest from the theoretical viewpoint of the models of the hydrogen-bonded systems. Systematic studies of the thermodynamic excess properties provide important information concerning the deeper understanding of the molecular liquid structure and the intermolecular interactions predominated by the self-association of the alkanol molecules due to hydrogen bond formation. These studies are also important from the viewpoint of the prediction of thermodynamic properties of (alkanol + alkane) mixtures of components having varying numbers of $-CH_2-$ units in the alkyl chain or varying numbers of $-CH_3$ substituents attached to the alkyl chain in the alkanol or alkane. With this viewpoint, Wagner and Heintz (1986) and Heintz et al. (1986) measured excess molar volumes, $V_m^E$, of binary mixtures of nonane and hexane with five different 1-alkanols at various temperatures. Fuente et al. (1992) determined $V_m^E$ of (nonan-1-ol + decane or tetradecane) at the temperatures 298.15 K, 308.15 K, and 318.15 K. Franjo et al. (1994) have measured $V_m^E$ of $xCH_3(CH_2)_5OH + (1 - x)CH_3(CH_2)_4CH_3$ at 298.15 K. Zielkiewicz (1994a,b) has measured total vapor pressures and $V_m^E$ of (heptane + propan-2-ol or butan-1-ol or 2-methylpropan-1-ol or 2-methylpropan-2-ol or pentan-1-ol) at 313.15 K. Although Nath and Pandey (1997) have recently made measurements of $V_m^E$ and refractive indexes, $n_D$, for binary mixtures of butanol with pentane ($n$-$C_5H_{12}$), hexane ($n$-$C_6H_{14}$), heptane ($n$-$C_7H_{16}$), octane ($n$-$C_8H_{18}$), and 2,2,4-trimethylpentane (2,2,4-TMP), a systematic study of mixtures of these alkanes with an alkanol of more complexity has not been made. Hence, in this program, measurements of $V_m^E$ and $n_D$ of heptanol ($n$-$C_7H_{15}OH$) + $n$-$C_5H_{12}$, + $n$-$C_6H_{14}$, + $n$-$C_7H_{16}$, + $n$-$C_8H_{18}$, and + 2,2,4-TMP have been made, and the results of these measurements are reported and discussed in this paper.

## Experimental Section

***Materials.*** Pentane, hexane, heptane, 2,2,4-trimethylpentane, all of HPLC quality and stated purity of the order of 99.8%, and heptanol, of LR quality and stated purity of 99% (GLC), were obtained from S. D. Fine Chemicals Pvt. Ltd., Mumbai, India. Pentane, hexane, heptane, and 2,2,4-TMP were used without any further purification. Heptanol was dried over anhydrous potassium carbonate and then subjected to distillation, and the middle third cut was used for the measurements. Octane (Spectrochem product) of AR quality and stated minimum purity of 99.5% (GLC) was distilled fractionally before use.

***Methods.*** (i) Excess molar volumes, $V_m^E$, were measured with an imprecision of the order of $\pm 0.002$ cm$^3$·mol$^{-1}$, using a two-limbed Pyrex glass dilatometer which was similar to that used in earlier measurements (Nath and Saini, 1989; Nath and Chaudhary, 1992; Nath and Pandey, 1997). Known amounts of the two liquid components were confined over mercury in the absence of air spaces in the two limbs of the dilatometer. The dilatometer (mounted on a stand) was immersed in a thermostat which was controlled to $\pm 0.01$ K. The mixing of the two liquid components was done by rocking the cell back and forth through a definite angle, and the mercury levels in the capillary of the dilatometer were noted before and after mixing, using a cathetometer which had the accuracy of $\pm 0.001$ cm, as described earlier (Nath and Pandey, 1997).

(ii) The refractive indexes (sodium D line), $n_D$, of the pure samples and the various liquid mixtures were measured with an accuracy of $\pm 0.0002$ at $(303.15 \pm 0.01)$ K, using a thermostated Abbe refractometer. The mole fractions of the components in the various mixtures are accurate to $\pm 0.0001$.

## Results and Discussion

The present experimental values of $n_D$ of $n$-$C_5H_{12}$, $n$-$C_6H_{14}$, $n$-$C_7H_{16}$, $n$-$C_8H_{18}$, 2,2,4-TMP, and $n$-$C_7H_{15}OH$ are 1.3520, 1.3695, 1.3825, 1.3925, 1.3865, and 1.4202, respectively. The values of $n_D$ of $n$-$C_5H_{12}$, $n$-$C_6H_{14}$, $n$-$C_7H_{16}$, $n$-$C_8H_{18}$, and 2,2,4-TMP at $T = 303.15$ K, listed in Riddick and Bunger (1970) are 1.352 07, 1.369 55, 1.382 57, 1.392 64, and 1.386 60, respectively. The experimental values of $V_m^E$ for the mixtures of $n$-$C_7H_{15}OH$ + $n$-$C_5H_{12}$, + $n$-$C_6H_{14}$,

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**Table 1. Experimental Values of the Excess Molar Volumes, $V_m^E$, for $n$-$C_7H_{15}OH$ + $n$-$C_6H_{14}$, + $n$-$C_7H_{16}$, + $n$-$C_8H_{18}$, + 2,2,4-TMP at 308.15 K and for $n$-$C_7H_{15}OH$ + $n$-$C_5H_{12}$ at 298.15 K**

| $x$ | $V_m^E$/cm$^3$·mol$^{-1}$ | $x$ | $V_m^E$/cm$^3$·mol$^{-1}$ | $x$ | $V_m^E$/cm$^3$·mol$^{-1}$ | $x$ | $V_m^E$/cm$^3$·mol$^{-1}$ |
|---|---|---|---|---|---|---|---|
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_5H_{12}$ | | | | |
| 0.0248 | −0.077 | 0.2090 | −0.439 | 0.5042 | −0.620 | 0.7713 | −0.493 |
| 0.0693 | −0.190 | 0.2705 | −0.508 | 0.6017 | −0.614 | 0.8338 | −0.412 |
| 0.1213 | −0.303 | 0.3588 | −0.574 | 0.6381 | −0.602 | 0.8882 | −0.303 |
| 0.1550 | −0.359 | 0.3967 | −0.595 | 0.7296 | −0.540 | | |
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_6H_{14}$ | | | | |
| 0.0369 | −0.013 | 0.2047 | −0.181 | 0.5173 | −0.368 | 0.8427 | −0.230 |
| 0.0843 | −0.052 | 0.2945 | −0.261 | 0.6417 | −0.363 | 0.8710 | −0.198 |
| 0.1134 | −0.084 | 0.3722 | −0.316 | 0.6956 | −0.343 | 0.9306 | −0.115 |
| 0.1667 | −0.143 | 0.4148 | −0.338 | 0.7203 | −0.331 | | |
| 0.1882 | −0.166 | 0.4655 | −0.361 | 0.8136 | −0.257 | | |
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_7H_{16}$ | | | | |
| 0.0225 | 0.042 | 0.2895 | −0.049 | 0.5634 | −0.183 | 0.8026 | −0.168 |
| 0.1153 | 0.048 | 0.3323 | −0.076 | 0.5891 | −0.195 | 0.8325 | −0.148 |
| 0.1283 | 0.036 | 0.3825 | −0.100 | 0.6379 | −0.200 | 0.8545 | −0.136 |
| 0.1629 | 0.020 | 0.4323 | −0.131 | 0.6879 | −0.196 | 0.8886 | −0.117 |
| 0.2276 | −0.016 | 0.4837 | −0.154 | 0.7350 | −0.190 | 0.9400 | −0.063 |
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_8H_{18}$ | | | | |
| 0.0411 | 0.043 | 0.1833 | 0.108 | 0.4637 | −0.012 | 0.7984 | −0.098 |
| 0.0698 | 0.073 | 0.2310 | 0.095 | 0.4899 | −0.028 | 0.8244 | −0.091 |
| 0.0720 | 0.072 | 0.2849 | 0.077 | 0.5882 | −0.072 | 0.8288 | −0.090 |
| 0.1275 | 0.103 | 0.3750 | 0.034 | 0.6679 | −0.099 | 0.8775 | −0.070 |
| 0.1303 | 0.104 | 0.4343 | 0.006 | 0.7385 | −0.103 | 0.9391 | −0.035 |
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)$2,2,4-TMP | | | | |
| 0.0255 | 0.025 | 0.3512 | −0.273 | 0.6713 | −0.348 | 0.9112 | −0.149 |
| 0.0419 | 0.017 | 0.3806 | −0.294 | 0.7590 | −0.306 | 0.9439 | −0.105 |
| 0.1202 | −0.052 | 0.4440 | −0.323 | 0.7958 | −0.286 | 0.9605 | −0.073 |
| 0.1851 | −0.121 | 0.5059 | −0.347 | 0.8217 | −0.256 | | |
| 0.2700 | −0.207 | 0.5791 | −0.360 | 0.8777 | −0.199 | | |

**Table 2. Experimental Values of the Refractive Indexes, $n_D$, for $n$-$C_7H_{15}OH$ + $n$-$C_5H_{12}$, + $n$-$C_6H_{14}$, + $n$-$C_7H_{16}$, + $n$-$C_8H_{18}$, and + 2,2,4-TMP at 303.15 K**

| $x$ | $n_D$ | $x$ | $n_D$ | $x$ | $n_D$ | $x$ | $n_D$ |
|---|---|---|---|---|---|---|---|
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_5H_{12}$ | | | | |
| 0.0257 | 1.3535 | 0.3003 | 1.3728 | 0.5686 | 1.3910 | 0.8345 | 1.4090 |
| 0.1311 | 1.3610 | 0.3511 | 1.3762 | 0.5836 | 1.3915 | 0.8828 | 1.4120 |
| 0.1429 | 1.3620 | 0.4156 | 1.3805 | 0.6490 | 1.3965 | 0.9442 | 1.4165 |
| 0.2810 | 1.3710 | 0.4314 | 1.3818 | 0.7313 | 1.4015 | 0.9776 | 1.4190 |
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_6H_{14}$ | | | | |
| 0.0204 | 1.3705 | 0.2852 | 1.3835 | 0.5414 | 1.3972 | 0.8810 | 1.4145 |
| 0.1107 | 1.3748 | 0.3322 | 1.3862 | 0.5859 | 1.3995 | 0.9321 | 1.4168 |
| 0.1364 | 1.3765 | 0.3973 | 1.3898 | 0.6189 | 1.4008 | 0.9665 | 1.4182 |
| 0.2186 | 1.3802 | 0.4276 | 1.3910 | 0.6939 | 1.4045 | | |
| 0.2466 | 1.3822 | 0.5017 | 1.3950 | 0.7302 | 1.4068 | | |
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_7H_{16}$ | | | | |
| 0.0198 | 1.3830 | 0.3040 | 1.3940 | 0.5547 | 1.4032 | 0.8104 | 1.4132 |
| 0.0949 | 1.3858 | 0.3308 | 1.3950 | 0.6109 | 1.4055 | 0.8235 | 1.4135 |
| 0.1339 | 1.3875 | 0.4022 | 1.3980 | 0.6457 | 1.4072 | 0.9216 | 1.4170 |
| 0.1728 | 1.3890 | 0.4493 | 1.3995 | 0.7117 | 1.4095 | 0.9701 | 1.4192 |
| 0.2509 | 1.3915 | 0.4797 | 1.4008 | 0.7482 | 1.4110 | | |
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_8H_{18}$ | | | | |
| 0.0365 | 1.3935 | 0.4359 | 1.4045 | 0.6842 | 1.4115 | 0.8581 | 1.4160 |
| 0.1560 | 1.3965 | 0.4505 | 1.4048 | 0.6918 | 1.4115 | 0.9386 | 1.4182 |
| 0.2789 | 1.4000 | 0.5374 | 1.4070 | 0.7547 | 1.4135 | | |
| 0.4070 | 1.4035 | 0.6000 | 1.4088 | 0.7950 | 1.4148 | | |
| | | | $xn$-$C_7H_{15}OH$ + $(1 - x)$2,2,4-TMP | | | | |
| 0.0331 | 1.3875 | 0.3256 | 1.3975 | 0.5914 | 1.4062 | 0.8277 | 1.4145 |
| 0.1280 | 1.3908 | 0.3527 | 1.3985 | 0.6269 | 1.4075 | 0.8947 | 1.4165 |
| 0.1693 | 1.3925 | 0.4308 | 1.4012 | 0.6717 | 1.4090 | 0.9296 | 1.4178 |
| 0.1960 | 1.3932 | 0.4922 | 1.4030 | 0.7424 | 1.4115 | 0.9479 | 1.4185 |
| 0.2694 | 1.3955 | 0.5115 | 1.4040 | 0.7903 | 1.4130 | | |

+ $n$-$C_7H_{16}$, + $n$-$C_8H_{18}$, and + 2,2,4-TMP are given in Table 1, whereas the values of $n_D$ for these mixtures at 303.15 K are given in Table 2, where $x$ refers to the mole fraction of $n$-$C_7H_{15}OH$ in the mixture. $x$ has an uncertainty of ±0.0001. Values of $V_m^E$ for the present mixtures have been plotted against $x$ in Figure 1 and have been fitted by the method of least squares to the equation

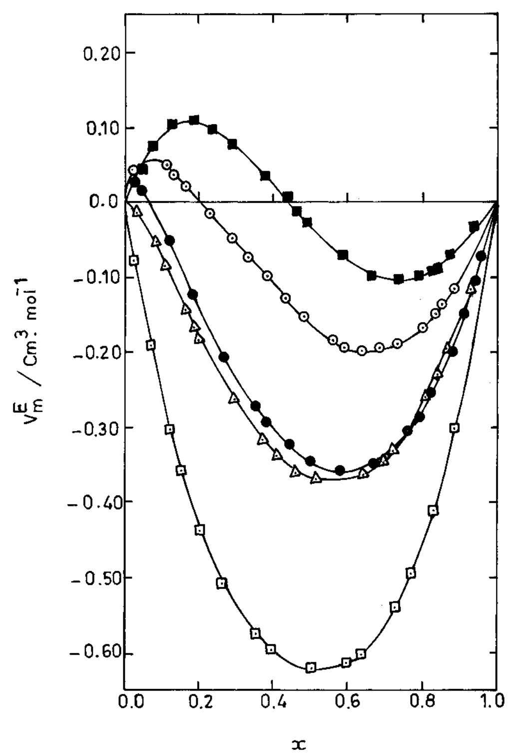


**Figure 1.** $V_m^E$ plotted against $x$ for the following systems: (⊡) $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_5H_{12}$, $T$ = 298.15 K; (△) $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_6H_{14}$, $T$ = 308.15 K; (⊙) $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_7H_{16}$, $T$ = 308.15 K; (■) $xn$-$C_7H_{15}OH$ + $(1 - x)n$-$C_8H_{18}$, $T$ = 308.15 K; (●) $xn$-$C_7H_{15}OH$ + $(1 - x)$2,2,4-TMP, $T$ = 308.15 K.

$$V_m^E/x(1 - x) = \sum_{j=1}^{m} A_j Y_j \qquad (1)$$

where $Y_j = [x - 1/(1 + Dx)]^{j-1}$ and $A_j$ are the coefficients characteristic of a system at a given temperature. As

**Table 3. Values of the Coefficients $A_j$ of Eq 1 and the Standard Deviations, $\delta(V_m^E)$, for the Various Mixtures**

| mixture | $T$/K | $A_1$ (cm$^3$·mol$^{-1}$) | $A_2$ | $A_3$ | $A_4$ | $\delta(V_m^E)$ |
|---|---|---|---|---|---|---|
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_5H_{12}$ | 298.15 | −2.6871 | 0.9160 | −0.5902 | −1.2403 | 0.003 |
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_6H_{14}$ | 308.15 | −1.0462 | −1.2040 | 0.4920 | −0.0670 | 0.003 |
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_7H_{16}$ | 308.15 | 0.0962 | −2.1882 | 1.0706 | −0.2254 | 0.003 |
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_8H_{18}$ | 308.15 | 0.7271 | −1.8080 | −1.3265 | 1.8399 | 0.003 |
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)2,2,4-TMP | 308.15 | −0.7922 | −1.8824 | 1.5101 | −0.9027 | 0.003 |

**Table 4. Values of the Coefficients $B_j$ of Eq 2 and the Standard Deviations, $\delta(n_D)$, for the Various Mixtures at 303.15 K**

| mixture | $B_1$ | $B_2$ | $B_3$ | $B_4$ | $\delta(n_D)$ |
|---|---|---|---|---|---|
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_5H_{12}$ | 1.351 84 | 0.071 21 | −0.007 32 | 0.004 63 | 0.0002 |
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_6H_{14}$ | 1.369 49 | 0.048 96 | 0.005 65 | −0.004 01 | 0.0002 |
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_7H_{16}$ | 1.382 34 | 0.037 79 | 0.001 61 | −0.001 61 | 0.0002 |
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_8H_{18}$ | 1.392 52 | 0.025 35 | 0.005 47 | −0.003 28 | 0.0002 |
| $x$$n$-$C_7H_{15}OH$ + (1 − $x$)2,2,4-TMP | 1.386 42 | 0.035 46 | −0.004 87 | 0.003 33 | 0.0001 |

pointed out by Berro and Péneloux (1984), the calculations were performed for all these systems with $D$ = 25. The values of the coefficients $A_j$ of eq 1, along with the standard deviations, $\delta(V_m^E)$, for the various mixtures are given in Table 3.

The refractive indexes, $n_D$, of the present mixtures at 303.15 K have been fitted by the method of least squares to the equation

$$n_D = \sum_{j=1}^{m} B_j x^{j-1} \qquad (2)$$

The values of the coefficients $B_j$ of eq 2, along with the standard deviations, $\delta(n_D)$, are given in Table 4.

$V_m^E$ is positive at lower mole fractions of $n$-$C_7H_{15}OH$ for $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_7H_{16}$, $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_8H_{18}$, and $x$$n$-$C_7H_{15}OH$ + (1 − $x$)2,2,4-TMP and is negative at higher mole fractions of $n$-$C_7H_{15}OH$ for these mixtures, with inversion of sign of $V_m^E$ from positive to negative values occurring at $x \sim 0.20$, $x \sim 0.45$, and $x \sim 0.06$, respectively, for the above mixtures at $T$ = 308.15 K. $V_m^E$ is found to be negative throughout the entire range of composition for $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_5H_{12}$ at $T$ = 298.15 K, and for $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_6H_{14}$ at $T$ = 308.15 K. At $x$ = 0.5, $V_m^E$ for the various present mixtures of $n$-$C_7H_{15}OH$ with alkanes has the sequence

$$n\text{-}C_8H_{18} > n\text{-}C_7H_{16} > 2,2,4\text{-TMP} > n\text{-}C_6H_{14} > n\text{-}C_5H_{12}$$

It is quite interesting to note that the above (same) sequence is also found (Nath and Pandey, 1997) in the values of $V_m^E$ at $x$ = 0.5, and at $T$ = 298.15 K, for binary mixtures of butanol ($n$-$C_4H_9OH$) with $n$-$C_5H_{12}$, $n$-$C_6H_{14}$, $n$-$C_7H_{16}$, $n$-$C_8H_{18}$, and 2,2,4-TMP.

The values of $V_m^E$ of the (alkanol + alkane) mixtures may be interpreted as the result of the contributions of the various types of intermolecular interactions operating between the alkane and alkanol molecules. Mainly three type of the contributions, (i) physical, due to nonspecific interactions; (ii) chemical, due to hydrogen bonding, and (iii) structural, due to changes of interstitial accommodation and free volume, are important in determining the thermodynamic excess properties of the alkanol + alkane mixtures. The chemical contribution is important at low concentrations of alkanol, where the breaking of self-association of the alkanol molecules due to H bonds makes a positive contribution to $V_m^E$. The positive values of $V_m^E$ for $x$$n$-$C_7H_{15}OH$ + (1 − $x$)$n$-$C_7H_{16}$, $x$$n$-$C_7H_{15}OH$ + (1 − $x$)-$n$-$C_8H_{18}$, and $x$$n$-$C_7H_{15}OH$ + (1 − $x$)2,2,4-TMP at low $x$ values may thus be attributed to the predominance of the contributions to the values of $V_m^E$ due to the breaking of self-association due to H bonds in the heptanol molecules in these mixtures.

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