# Densities and Viscosities of Binary Liquid Mixtures of Vinyl Acetate, Diethyl Oxalate, and Dibutyl Phthalate with Normal Alkanols at 303.15 K

**I-Chin Pan, Muoi Tang,† and Yan-Ping Chen***

Department of Chemical Engineering, National Taiwan University, Taipei, Taiwan, Republic of China

Densities and viscosities of binary liquid mixtures of three esters (vinyl acetate, diethyl oxalate, and dibutyl phthalate) with four normal alkanols (ethanol, butan-1-ol, octan-1-ol, and decan-1-ol) are presented over the complete concentration range at 303.15 K and atmospheric pressure. The densities were measured using a vibrating-tube digital densimeter, and the viscosities were measured by a capillary viscometer. The excess molar volumes are positive, except those at low $x_1$ values for dibutyl phthalate (1) + ethanol (2), dibutyl phthalate (1) + butan-1-ol (2), and diethyl oxalate (1) + ethanol (2). The excess molar volumes and viscosity deviations were correlated by the Redlich and Kister type equation. Optimally fitted parameters are presented, and the correlation results are in satisfactory agreement with the experimental data.

## Introduction

Experimental data of the densities and viscosities of binary liquid mixtures containing esters are useful in pharmaceutical and specialty chemicals. These data are not sufficient in the literature.[1] Recently, experimental density and viscosity data have been reported for some esters with *n*-alkanols.[2−6] Vinyl acetate ($C_4H_6O_2$) is a useful specialty chemical for adhesive and lacquer. Diethyl oxalate ($C_6H_{10}O_4$) is used in the manufacturing of plastics or dyestuff. Dibutyl phthalate ($C_{16}H_{22}O_4$) is also used as the plasticizer in organic synthesis. In this study, densities and viscosities for the binary liquid mixtures of these three ester compounds with four alkanols (ethanol, butan-1-ol, octan-1-ol, and decan-1-ol) were measured at 303.15 K and atmospheric pressure. To our knowledge, the data for these systems were not reported previously. Excess molar volumes and deviations of viscosity were then calculated and correlated by polynomial expressions.

## Experimental Section

***Materials***. All chemicals were purchased from Merck Co. The purities of all liquids are better than 99.5 mass % on the basis of GC analyses (Shimadzu 8A with a TCD detector, Japan), and no further purification was made. The densities, viscosities, and refractive indices of all pure chemicals were measured in this study. A comparison of pure fluid properties with literature data is shown in Table 1.

***Measurements***. Liquid mixtures of various compositions were prepared by mass, in a 50 cm$^3$ flask, using a precision Mettler balance (AE 2000, Switzerland) with an accuracy of ±0.1 mg. The corresponding accuracy in the mole fraction calculation was better than ±0.0001. Density and viscosity measurements were carried out using a Neslab RTE220 thermostat. Temperatures were measured with a Hart 1506 thermometer with an accuracy of ±0.01 K. A proportional integral control model was used to maintain the temperature in the thermostat to be within ±0.01 K of the desired value. The refractive indices of pure liquids were measured at 293.15 K with an Abbe refractometer Atago 3T (Japan). The accuracy of the refractive index measurement is ±0.0001.

**Table 1. Comparison of Properties of Pure Liquids with Literature Data**

| | $\rho$/kg·m$^{-3}$ $T$ = 303.15 K | | $\eta$/mPa·s $T$ = 303.15 K | | $n_D$ $T$ = 293.15 K | |
|---|---|---|---|---|---|---|
| solvent | exp | lit. | exp | lit. | exp | lit. |
| dibutyl phthalate | 1038.2 | 1037.9[10] | 13.329 | 13.347[10] | 1.4928 | 1.4926[8] |
| diethyl oxalate | 1066.6 | 1066.9[8] | 1.661 | 1.618[8] | 1.4106 | 1.4102[8] |
| vinyl acetate | 919.2 | 918.2[8] | 0.372 | 0.384[10] | 1.3938 | 1.3934[8] |
| decan-1-ol | 823.1 | 823.1[9] | 9.342 | 9.366[10] | 1.3974 | 1.3971[10] |
| octan-1-ol | 818.2 | 818.1[11] 818.4[9] | 6.100 | 6.010[13] 6.103[10] 6.125[8] | 1.4265 | 1.4261[8] |
| butan-1-ol | 801.9 | 802.0[9] | 2.276 | 2.271[8] 2.280[14] | 1.3990 | 1.3993[8] |
| ethanol | 780.8 | 780.8[9] | 0.995 | 0.994[12] | 1.3611 | 1.3614[9] |

Densities of pure liquids and binary liquid mixtures were measured at 303.15 K with an Anton Parr digital vibrating tube densimeter (model 60/602, Anton Parr, Austria). The densimeter was calibrated with degassed water and dehumidized air at atmospheric pressure. More than eight readings were taken for each density measurement. The precision in the density measurement is estimated to be better than ±0.01 kg·m$^{-3}$. The corresponding uncertainty in the excess molar volume calculation is $\pm 2 \times 10^{-9}$ m$^3$·mol$^{-1}$.

Kinematic viscosities of binary liquid mixtures were measured by factory-calibrated Ubbelohde capillary viscometers (Schott Gerate, FRG). A Lauda Viscoboy 2 timer (Lauda, FRG), which measured automatically the flow time of a liquid, was used with an accuracy of time measurement of 0.01 s. The kinematic viscosities of liquids were calculated with the measured flow time and the calibration

* Corresponding author. Fax: +886-2-2362-3040. E-mail: ypchen@ccms.ntu.edu.tw.
† Present address: Department of Chemical Engineering, Chinese Culture University, Taipei, Taiwan.

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**Table 2. Densities, ρ, and Viscosities, η, for Vinyl Acetate (1) with Various Alkanols (2) at 303.15 K**

| $x_1$ | $\rho$/kg·m$^{-3}$ | $\eta$/mPa·s | $x_1$ | $\rho$/kg·m$^{-3}$ | $\eta$/mPa·s |
|---|---|---|---|---|---|
| | (2) = Ethanol | | | (2) = Butan-1-ol | |
| 0.0000 | 780.79 | 0.995 | 0.0000 | 801.89 | 2.276 |
| 0.0998 | 801.05 | 0.787 | 0.1008 | 812.57 | 1.622 |
| 0.2001 | 819.23 | 0.651 | 0.1999 | 823.38 | 1.211 |
| 0.2995 | 835.45 | 0.564 | 0.3008 | 834.66 | 0.944 |
| 0.4007 | 850.54 | 0.501 | 0.3993 | 845.87 | 0.768 |
| 0.5002 | 864.17 | 0.457 | 0.5055 | 858.21 | 0.634 |
| 0.5990 | 876.57 | 0.425 | 0.6002 | 869.40 | 0.547 |
| 0.7013 | 888.46 | 0.402 | 0.6998 | 881.39 | 0.482 |
| 0.8005 | 899.24 | 0.386 | 0.7997 | 893.66 | 0.433 |
| 0.9002 | 909.49 | 0.378 | 0.8749 | 903.09 | 0.405 |
| 1.0000 | 919.24 | 0.372 | 1.0000 | 919.24 | 0.372 |
| | (2) = Octan-1-ol | | | (2) = Decan-1-ol | |
| 0.0000 | 818.17 | 6.100 | 0.0000 | 823.14 | 9.342 |
| 0.1001 | 823.12 | 4.448 | 0.1013 | 826.79 | 6.774 |
| 0.2008 | 828.92 | 3.155 | 0.1998 | 831.25 | 4.804 |
| 0.3002 | 835.48 | 2.302 | 0.3007 | 836.58 | 3.437 |
| 0.4017 | 843.11 | 1.705 | 0.3991 | 842.76 | 2.488 |
| 0.4997 | 851.49 | 1.279 | 0.5006 | 850.26 | 1.793 |
| 0.6008 | 861.38 | 0.965 | 0.6004 | 859.01 | 1.310 |
| 0.6993 | 872.42 | 0.743 | 0.6957 | 869.01 | 0.952 |
| 0.7981 | 885.29 | 0.577 | 0.8003 | 882.45 | 0.682 |
| 0.8965 | 900.30 | 0.459 | 0.9040 | 899.25 | 0.489 |
| 1.0000 | 919.24 | 0.372 | 1.0000 | 919.24 | 0.372 |

**Table 3. Densities, ρ, and Viscosities, η, for Diethyl Oxalate (1) with Various Alkanols (2) at 303.15 K**

| $x_1$ | $\rho$/kg·m$^{-3}$ | $\eta$/mPa·s | $x_1$ | $\rho$/kg·m$^{-3}$ | $\eta$/mPa·s |
|---|---|---|---|---|---|
| | (2) = Ethanol | | | (2) = Butan-1-ol | |
| 0.0000 | 780.79 | 0.995 | 0.0000 | 801.89 | 2.276 |
| 0.1001 | 839.78 | 0.948 | 0.1064 | 840.12 | 1.803 |
| 0.2004 | 886.25 | 0.968 | 0.1924 | 869.35 | 1.613 |
| 0.2897 | 920.05 | 1.009 | 0.2954 | 900.63 | 1.496 |
| 0.3980 | 953.91 | 1.069 | 0.3832 | 925.89 | 1.438 |
| 0.5098 | 982.72 | 1.146 | 0.5033 | 957.88 | 1.405 |
| 0.5982 | 1002.17 | 1.213 | 0.6097 | 984.06 | 1.405 |
| 0.6828 | 1018.58 | 1.283 | 0.6734 | 998.89 | 1.419 |
| 0.7700 | 1033.64 | 1.371 | 0.7804 | 1022.46 | 1.461 |
| 0.8931 | 1052.35 | 1.515 | 0.9074 | 1048.60 | 1.556 |
| 1.0000 | 1066.63 | 1.661 | 1.0000 | 1066.63 | 1.661 |
| | (2) = Octan-1-ol | | | (2) = Decan-1-ol | |
| 0.0000 | 818.17 | 6.100 | 0.0000 | 823.14 | 9.342 |
| 0.0949 | 837.13 | 4.702 | 0.1042 | 839.98 | 6.782 |
| 0.2003 | 859.25 | 3.667 | 0.1986 | 856.64 | 5.372 |
| 0.2883 | 878.56 | 3.152 | 0.2934 | 874.72 | 4.324 |
| 0.4000 | 904.18 | 2.636 | 0.4010 | 897.00 | 3.516 |
| 0.5026 | 928.89 | 2.296 | 0.5111 | 921.83 | 2.851 |
| 0.6064 | 955.02 | 2.027 | 0.5985 | 943.15 | 2.488 |
| 0.6984 | 979.10 | 1.860 | 0.7108 | 973.00 | 2.078 |
| 0.7809 | 1001.70 | 1.740 | 0.8081 | 999.30 | 1.837 |
| 0.8763 | 1029.00 | 1.638 | 0.8863 | 1026.38 | 1.691 |
| 1.0000 | 1066.63 | 1.661 | 1.0000 | 1066.63 | 1.661 |

constants. Dynamic viscosities were determined from these data and the corresponding densities of the mixtures at the same compositions. The reported dynamic viscosities were the averaged values of at least five to eight runs. The accuracy of the dynamic viscosity measurement was estimated to be ±0.003 mPa·s.

## Results and Discussion

Density data of various binary mixtures are listed in Tables 2−4, respectively. The excess molar volumes, $V^E$, were calculated from the density data by the relationship

$$V^E/(\mathrm{cm^3{\cdot}mol^{-1}}) = x_1M_1(1/\rho_m - 1/\rho_1) + x_2M_2(1/\rho_m - 1/\rho_2) \quad (1)$$

where $x$, $M$, and $\rho$ are the mole fraction, molecular weight, and density, respectively. The subscript m represents the mixture property. Esters and alkanols are designated as components 1 and 2, respectively. Dynamic viscosities $\eta$ of the binary mixtures of three ester compounds with various alkanols are calculated from the measured density and

**Table 4. Densities, ρ, and Viscosities, η, for Dibutyl Phthalate (1) with Various Alkanols (2) at 303.15 K**

| $x_1$ | $\rho$/kg·m$^{-3}$ | $\eta$/mPa·s | $x_1$ | $\rho$/kg·m$^{-3}$ | $\eta$/mPa·s |
|---|---|---|---|---|---|
| | (2) = Ethanol | | | (2) = Butan-1-ol | |
| 0.0000 | 780.79 | 0.995 | 0.0000 | 801.89 | 2.276 |
| 0.0982 | 867.97 | 1.555 | 0.1016 | 860.34 | 2.585 |
| 0.1991 | 919.40 | 2.302 | 0.1991 | 900.90 | 3.095 |
| 0.4046 | 976.37 | 4.211 | 0.2997 | 932.69 | 3.757 |
| 0.5046 | 993.12 | 5.310 | 0.4016 | 957.76 | 4.553 |
| 0.5634 | 1001.14 | 6.002 | 0.5004 | 977.33 | 5.431 |
| 0.6940 | 1015.58 | 7.761 | 0.5969 | 993.13 | 6.463 |
| 0.8070 | 1025.35 | 9.476 | 0.6966 | 1006.90 | 7.645 |
| 0.8877 | 1031.20 | 10.910 | 0.8044 | 1019.53 | 9.216 |
| 0.9316 | 1034.08 | 11.678 | 0.8662 | 1025.93 | 10.270 |
| 1.0000 | 1038.18 | 13.329 | 1.0000 | 1038.18 | 13.329 |
| | (2) = Octan-1-ol | | | (2) = Decan-1-ol | |
| 0.0000 | 818.17 | 6.100 | 0.0000 | 823.14 | 9.342 |
| 0.1003 | 852.22 | 5.900 | 0.1004 | 851.02 | 8.572 |
| 0.2011 | 882.55 | 5.876 | 0.1995 | 876.97 | 8.061 |
| 0.3001 | 909.13 | 6.078 | 0.3035 | 902.47 | 7.851 |
| 0.3996 | 933.03 | 6.444 | 0.4004 | 924.80 | 7.910 |
| 0.5000 | 954.84 | 6.901 | 0.4902 | 944.33 | 8.075 |
| 0.5999 | 974.50 | 7.600 | 0.5977 | 966.34 | 8.479 |
| 0.6958 | 991.75 | 8.396 | 0.6985 | 985.78 | 9.095 |
| 0.7975 | 1008.54 | 9.530 | 0.7997 | 1004.25 | 9.985 |
| 0.8980 | 1023.82 | 11.052 | 0.8948 | 1020.73 | 11.176 |
| 1.0000 | 1038.18 | 13.329 | 1.0000 | 1038.18 | 13.329 |

**Table 5. Regression Results for the Excess Molar Volumes of Various Binary Mixtures at $T$ = 303.15 K**

| binary systems | $a_0$ | $a_1$ | $a_2$ | $10^9\sigma(V^E)$ m$^3$·mol$^{-1}$ |
|---|---|---|---|---|
| diethyl oxalate + ethanol | 0.0891 | 0.3512 | −0.0185 | 0.99 |
| diethyl oxalate + butan-1-ol | 1.4048 | −0.2081 | 0.3511 | 1.59 |
| diethyl oxalate + octan-1-ol | 3.1027 | −0.1455 | 0.5198 | 3.86 |
| diethyl oxalate + decan-1-ol | 3.7474 | 0.0238 | 0.8950 | 1.58 |
| dibutyl phthalate + ethanol | −0.4645 | 1.1744 | −0.7935 | 5.97 |
| dibutyl phthalate + butan-1-ol | 0.2534 | 0.4772 | 0.0068 | 2.21 |
| dibutyl phthalate + octan-1-ol | 1.2361 | −0.1587 | 0.3042 | 4.54 |
| dibutyl phthalate + decan-1-ol | 1.9133 | −0.4530 | 0.6764 | 5.81 |
| vinyl acetate + ethanol | 0.5731 | 0.2080 | 0.0555 | 3.42 |
| vinyl acetate + butan-1-ol | 1.4710 | −0.0381 | 0.1884 | 0.54 |
| vinyl acetate + octan-1-ol | 2.4274 | 0.1474 | 0.4457 | 0.91 |
| vinyl acetate + decan-1-ol | 2.9618 | 0.3056 | 0.7369 | 3.94 |

**Table 6. Regression Results for the Viscosities of Various Binary Mixtures at $T$ = 303.15 K**

| binary systems | $a_0$ | $a_1$ | $a_2$ | $a_3$ | $10^3\sigma[\delta(\ln \eta)]$/ mPa·s |
|---|---|---|---|---|---|
| diethyl oxalate + ethanol | −0.4811 | 0.2026 | -0.3761 | 0.3710 | 1.90 |
| diethyl oxalate + butan-1-ol | −1.2950 | 0.4423 | −0.4962 | 0.2786 | 1.35 |
| diethyl oxalate + octan-1-ol | −1.2900 | 0.1172 | −0.5186 | −0.1867 | 5.65 |
| diethyl oxalate + decan-1-ol | −1.1827 | −0.0512 | −0.7010 | 0.2490 | 5.40 |
| dibutyl phthalate + ethanol | 1.4742 | −0.7788 | 0.1039 | −0.0942 | 4.61 |
| dibutyl phthalate + butan-1-ol | −0.0417 | 0.0050 | −0.5104 | 0.3165 | 3.60 |
| dibutyl phthalate + octan-1-ol | −1.0558 | 0.1104 | −0.2544 | −0.0892 | 2.83 |
| dibutyl phthalate + decan-1-ol | −1.2801 | 0.0568 | −0.2236 | −0.1893 | 2.35 |
| vinyl acetate + ethanol | −1.1431 | 0.3204 | −0.1354 | 0.0913 | 1.17 |
| vinyl acetate + butan-1-ol | −1.4563 | 0.2981 | −0.0923 | −0.0126 | 1.85 |
| vinyl acetate + octan-1-ol | −0.6603 | 0.0429 | −0.0271 | −0.3426 | 5.07 |
| vinyl acetate + decan-1-ol | −0.1430 | 0.0656 | −0.0881 | −0.5236 | 4.48 |

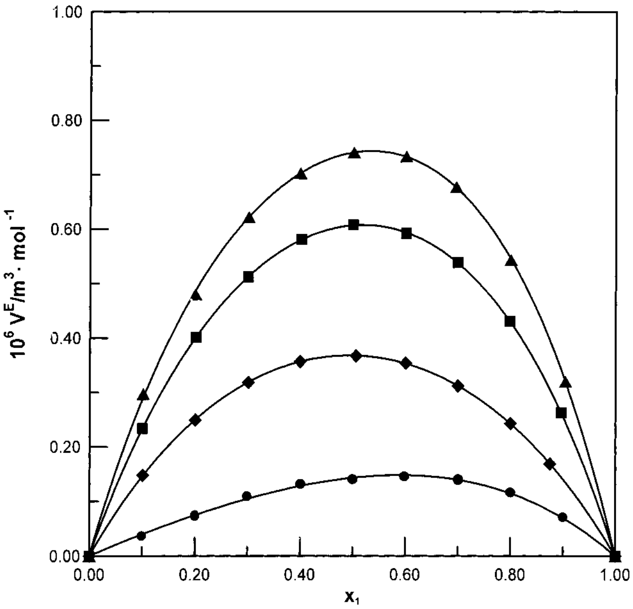


**Figure 1.** Excess molar volumes of vinyl acetate (1) with various alkanols (2) at 303.15 K: ●, vinyl acetate (1) + ethanol (2); ◆, vinyl acetate (1) + butan-1-ol (2); ■, vinyl acetate (1) + octan-1-ol (2); ▲, vinyl acetate (1) + decan-1-ol (2); —, correlated results.

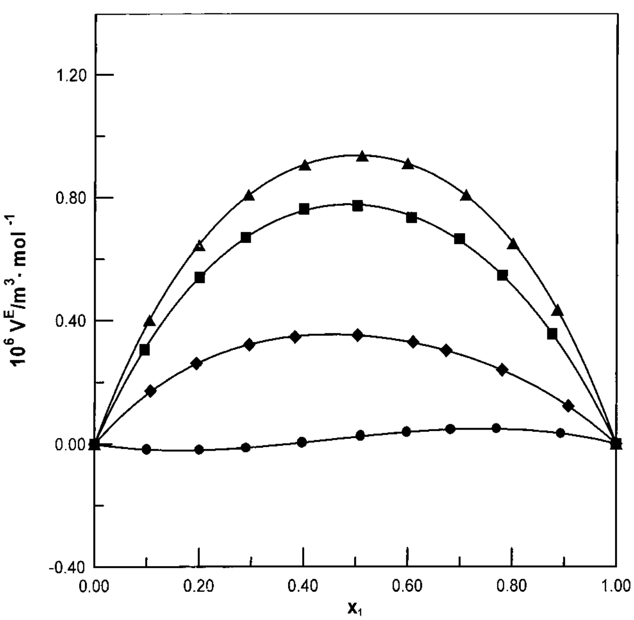


**Figure 2.** Excess molar volumes of diethyl oxalate (1) with various alkanols (2) at 303.15 K: ●, diethyl oxalate (1) + ethanol (2); ◆, diethyl oxalate (1) + butan-1-ol (2); ■, diethyl oxalate (1) + octan-1-ol (2); ▲, diethyl oxalate (1) + decan-1-ol (2); —, correlated results.

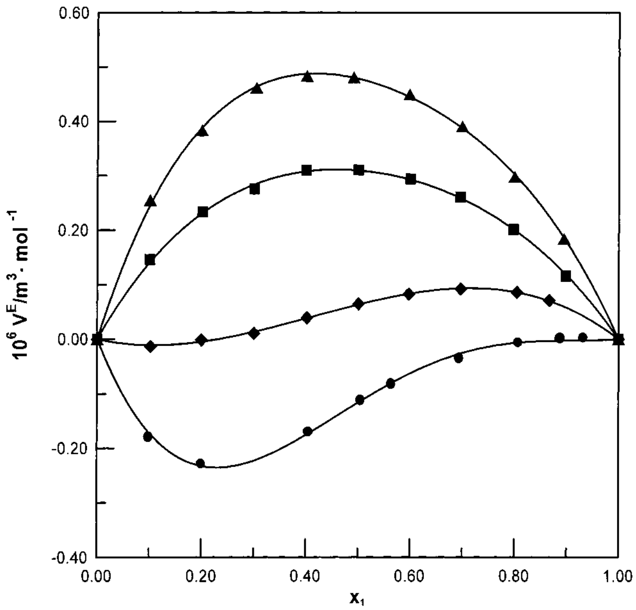


**Figure 3.** Excess molar volumes of dibutyl phthalate (1) with various alkanols (2) at 303.15 K: ●, dibutyl phthalate (1) + ethanol (2); ◆, dibutyl phthalate (1) + butan-1-ol (2); ■, dibutyl phthalate (1) + octan-1-ol (2); ▲, dibutyl phthalate (1) + decan-1-ol (2); —, correlated results.

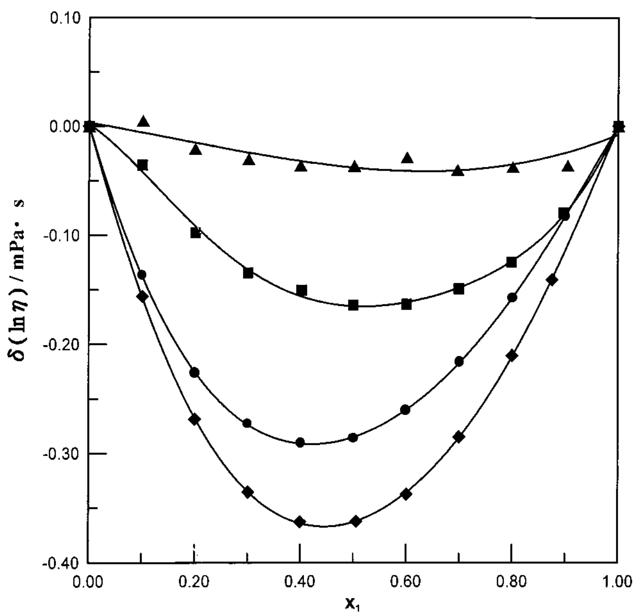


**Figure 4.** Deviation of viscosities of vinyl acetate (1) with various alkanols (2) at 303.15 K: ●, vinyl acetate (1) + ethanol (2); ◆, vinyl acetate (1) + butan-1-ol (2); ■, vinyl acetate (1) + octan-1-ol (2); ▲, vinyl acetate (1) + decan-1-ol (2); —, correlated results.

kinematic viscosity data. The results are also listed in Tables 2−4, respectively. The deviation of viscosity was calculated by

$$\delta(\ln \eta) = \ln \eta_m - \sum_{i=1}^{2} x_i \ln \eta_i \qquad (2)$$

The excess molar volumes and the deviations of viscosity were then correlated by a Redlich and Kister type equation[7]

$$V^E(\text{or } \delta(\ln \eta)) = x_1 x_2 \sum_{i=1}^{3 \text{ or } 4} a_{i-1}(x_1 - x_2)^{i-1} \qquad (3)$$

The coefficients in eq 3 were determined by the least-squares method, where the square of the difference between the experimental and calculated results is minimized. The optimally fitted coefficients in eq 3 for the excess molar volumes and the deviations of viscosity are listed in Tables 5 and 6, respectively. The standard deviations were calculated by

$$\sigma(Y) = [\sum (Y^{exp} - Y^{cal})^2/(m - n)]^{0.5} \qquad (4)$$

where $Y$ represents the excess molar volume or the deviation of viscosity, $m$ is the number of data points, and $n$ is the number of parameters. It is observed that the average

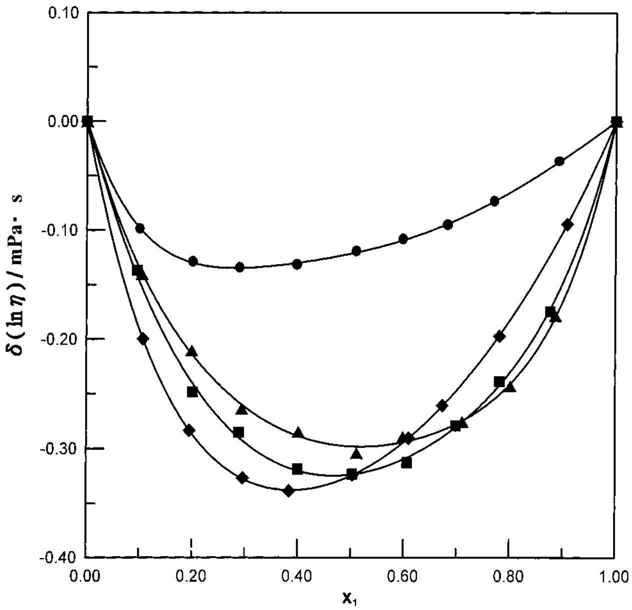


**Figure 5.** Deviation of viscosities of diethyl oxalate (1) with various alkanols (2) at 303.15 K: ●, diethyl oxalate (1) + ethanol (2); ◆, diethyl oxalate (1) + butan-1-ol (2); ■, diethyl oxalate (1) + octan-1-ol (2); ▲, diethyl oxalate (1) + decan-1-ol (2); —, correlated results.

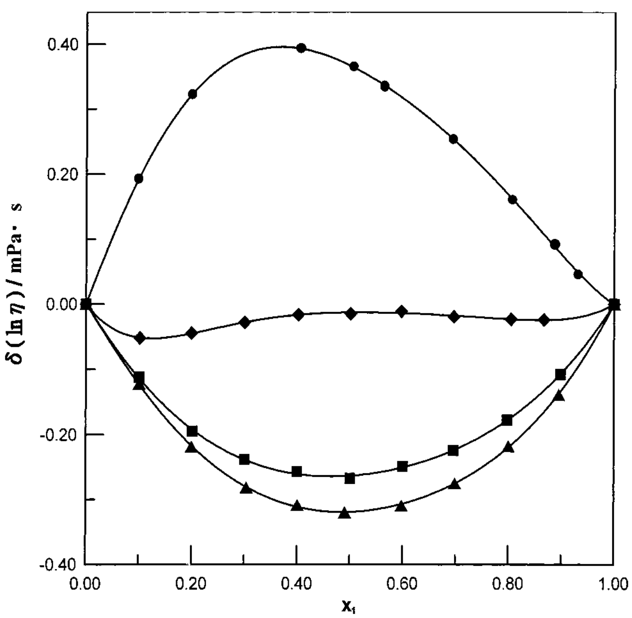


**Figure 6.** Deviation of viscosities of dibutyl phthalate (1) with various alkanols (2) at 303.15 K: ●, dibutyl phthalate (1) + ethanol (2); ◆, dibutyl phthalate (1) + butan-1-ol (2); ■, dibutyl phthalate (1) + octan-1-ol (2); ▲, dibutyl phthalate (1) + decan-1-ol (2); —, correlated results.

value of the standard deviations of the excess molar volumes for all binary systems is less than $3 \times 10^{-9}$ $m^3{\cdot}mol^{-1}$. The average standard deviation value of the deviations of viscosity for all systems is 0.0034 mPa·s.

Graphical presentations of the excess molar volumes are shown in Figures 1−3, respectively. The binary mixtures of vinyl acetate with four alkanols show positive deviations in the complete concentration range. Maximum values of the excess molar volumes occur at the mole fractions of vinyl acetate between 0.5 and 0.6 for these binary mixtures. The positive values of $V^E$ indicate the declustering of alkanols. The $V^E$ values are negative at some concentration ranges for the binary mixtures of diethyl oxalate (1) + ethanol (2), dibutyl phthalate (1) + ethanol (2), and dibutyl phthalate (1) + butan-1-ol (2). Graphical presentations of the deviations of viscosities of various esters with four alkanols are shown in Figures 4−6, respectively. Good agreement between the correlated results and the experimental data is also observed.

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