122 *J. Chem. Eng. Data* **1990**, *35*, 122-124

# Density and Viscosity of Ethanol + 1,2-Dichloroethane, Ethanol + 1,1,1-Trichloroethane, and Ethanol + 1,1,2,2-Tetrachloroethane Binary Mixtures

**G. Sivaramprasad and M. Venkateshwara Rao**

*College of Technology, Osmania University, Hyderabad 500 007, India*

**D. H. L. Prasad***

*Indian Institute of Chemical Technology, Hyderabad 500 007, India*

**Density and viscosity measurements on the binary mixtures of ethanol + 1,2-dichloroethane, ethanol + 1,1,1-trichloroethane, and ethanol + 1,1,2,2-tetrachloroethane in the temperature range of 303.15-333.15 K are reported. The data are found to be satisfactorily represented by Arrhenius and Kendall and Monroe mixing rules. The Lobe equation fails, predicting lower values of mixture viscosity at all the data points.**

## Introduction

In continuation of the investigative work on the properties of binary mixtures such as phase equilibria (*1-3*), enthalpy of mixing (*4*), and viscosity and surface tension (*5*), this study on the density and viscosity of the binary mixtures of ethanol + 1,2-dichloroethane, ethanol + 1,1,1-trichloroethane, and ethanol + 1,1,2,2-tetrachloroethane is carried out, as a part of our studies on the properties of binary mixtures having an aliphatic alcohol as one component and a chloroethane as the second component.

## Experimental Section

***Materials.*** Anhydrous ethanol ($T_C$ = 516.2 K and $P_C$ = 63.0 atm) procured from Aldrich Chemical Co., Gillingham, England, has been dried over silica gel and distilled twice. The middle fraction of the second distillation (boiling at 350.3 K, corresponding to the local atmospheric pressure) has been collected and stored in an amber-colored bottle for use in the experimental work.

Analar Grade 1,2-dichloroethane ($T_C$ = 561.0 K and $P_C$ = 53.0 atm) supplied by BDH Chemicals, Poole, England, is washed with a dilute solution of potassium hydroxide and then with water. The sample is then dried over phosphorus pentoxide and distilled twice. The middle fraciton of the second distillation (collected at a constant temperature of 355.5 K, corresponding to the boiling point at the local atmospheric pressure) is collected in an amber-colored bottle for use in the experimentation.

Analar Grade 1,1,1-trichloroethane ($T_C$ = 584.5 K and $P_C$ = 44.36 atm) suppled by SDS Fine Chemicals, Boisar, India, is washed with concentrated hydrochloric acid and then with 10% potassium carbonate solution and finally with 10% sodium chloride solution. The compound is distilled twice after drying over calcium chloride. The middle fraction of the second distillation (boiling at a constant temperature of 345.8 K, corresponding to the local atmospheric pressure) is collected and stored in an amber-colored bottle for use in the preparation of the mixtures for the study.

Laboratory Grade 1,1,2,2-tetrachloroethane ($T_C$ = 661.0 K and $P_C$ = 39.5 atm) supplied by BDH is stirred with 25 mL of concentrated sulphuric acid at 353 K for 10 min. The discolored acid is removed, and the procedure is repeated until the discoloration of the acid stops. The sample is then washed with water and dried over potassium carbonate. The dry sample is distilled twice, and the middle fraction of the second distillation (boiling at constant temperature of 417.0 K, corresponding to the local atmospheric pressure) is collected into amber-colored bottles, for use in the experimental work.

**Table I. Comparison of the Physical Properties of the Substances Used with Literature Data at 293.15 K**

| | refractive index | | density, g/mL | |
|---|---|---|---|---|
| substance | this work | lit. | this work | lit. |
| ethanol | 1.3614 | 1.36143[a] | 0.7894 | 0.7893[a] |
| 1,2-dichloroethane | 1.4450 | 1.4451[b] | 1.2530 | 1.2529[b] |
| 1,1,1-trichloroethane | 1.4379 | 1.4377[c] | 1.3247 | 1.3249[c] |
| 1,1,2,2-tetrachloroethane | 1.4494 | 1.4492[d] | 1.5932 | 1.5930[d] |

[a] Source of literature data *Kirk-Othmer Encyclopedia of Chemical Technology*, 3rd Ed.; Wiley: New York, 1980; Vol. 9, p 339. [b] 1978; Vol. 5, p 725. [c] 1979; Vol. 5, p 729. [d] 1979; Vol. 5, p 735.

Table I compares the density and refractive index of the pure substances, used in this study on mixtures, with literature data.

***Methods.*** Measurements on the density of the pure liquids as well as the mixtures are carried out by means of pycnometers carefully calibrated by using double-distilled water. The weights required in the entire work are recorded with a Mettler balance accurate to ±0.0001 g. The temperatures are maintained to within ±0.05 K of the desired value by keeping the pycnometers immersed in an electronically controlled water bath for a sufficient length of time (usually 1 h).

An Oswald-type glass viscometer carefully maintained to within ±0.05 K of the desired temperature (by keeping it immersed in an electronically controlled thermostart for about 1 h) is used for the measurements on the viscosity of pure liquids and the mixtures. While the experiments are conducted, care is taken to avoid the entrainment of any air bubbles or particulate matter into the apparatus. The apparatus is filled exactly to the same level in all the experiments, to meet as closely as possible equal pressure and the same amount of volumetric flow requirements. The time taken by the liquid (or liquid mixture) to flow between the two fixed marks is noted by starting the stopwatch when the lower miniscus of the liquid is at the top mark and stopping the watch when the lower miniscus of the liquid is at the bottom mark. The accuracy of the time measurement is ±0.05 s. The experiments are first conducted at each temperature using water (disilled twice). The data of the experiments are used in conjunction with those on the samples (pure liquids and mixtures studied) to calculate the viscosity of the samples by

$$\eta_s/\eta_w = \rho_s t_s/\rho_w t_w \qquad (1)$$

Comparison of the viscosity measurements on some pure liquids—benzene, toluene, *p*-xylene, chlorobenzene—with the

0021-9568/90/1735-0122$02.50/0

**Table II. Comparison of the Pure Liquid Viscosity of Some Test Liquids with Literature Data at 298.15 K**

| substance | viscosity, cP<br>this work | lit. |
|---|---|---|
| benzene | 0.644 | 0.6462[a] |
| toluene | 0.590 | 0.5876[b] |
| *p*-xylene | 0.642 | 0.6441[c] |
| chlorobenzene | 0.802 | 0.7997[d] |

[a] Source of literature data: Dreisbach, R. R. *Physical Properties of Chemical Compounds*; American Chemical Society: Washington, DC, 1955; p 11. [b] p 12. [c] p 16. [d] p 134.

literature values given in Table II shows that the method and apparatus used in this work are capable of producing viscosities within ±0.5%.

Mixture samples are prepared from weighed quantities of pure liquids, and the constancy of the compositions is checked after each experiment by density measurement at 303.15 K. Randomly carried out duplicate measurements on mixture samples gave results within ±0.2% of the values reported in Tables III–V.

## Results and Discussion

The results are presented in Tables III–V. All the mixture density data agree with the values predicted from the law of additive volumes rearranged for convenience as

$$\rho_m = \frac{\rho_1\rho_2}{\rho_1 x_1 + \rho_2 x_2} \tag{2}$$

with an average absolute deviation of 0.11%.

The Arrhenius equation

$$\log \eta_m = x_1 \log \eta_1 + x_2 \log \eta_2 \tag{3}$$

The Kendall and Monroe equation

$$\eta^{1/3} = x_1\eta_1^{1/3} + x_2\eta_2^{1/3} \tag{4}$$

and Lobe's equation

$$\eta_m/\rho_m = \phi_1(\eta_1/\rho_1)e^{\phi_2\beta} + \phi_2(\eta_2/\rho_2)e^{\phi_1\alpha} \tag{5}$$

with

$$\alpha = -1.7 \ln \frac{(\eta_2/\rho_2)}{(\eta_1/\rho_1)} \tag{6}$$

$$\beta = 0.27 \ln \frac{(\eta_2/\rho_2)}{(\eta_1/\rho_1)} + \left[1.3 \ln \frac{(\eta_2/\rho_2)}{(\eta_1/\rho_1)}\right]^{0.5} \tag{7}$$

given by Bretsznajder (*6*) and Reid et al. (*7*) have been used to calculate mixture viscosities from pure liquid viscosities. The type of agreement of the present experimental mixture viscosity data with those calculated with the three methods requiring

**Table III. Density and Viscosity Data on Ethanol + 1,2-Dichloroethane Mixtures**

| mole fraction of ethanol | temp = 303.15 K<br>density, g/mL | viscosity, cP | temp = 313.15 K<br>density, g/mL | viscosity, cP | temp = 323.15 K<br>density, g/mL | viscosity, cP | temp = 333.15 K<br>density, g/mL | viscosity, cP |
|---|---|---|---|---|---|---|---|---|
| 1.0000 | 0.7810 | 1.004 | 0.7729 | 0.835 | 0.7632 | 0.702 | 0.7522 | 0.592 |
| 0.9241 | 0.8036 | 0.987 | 0.7957 | 0.819 | 0.7849 | 0.691 | 0.7740 | 0.584 |
| 0.8441 | 0.8289 | 0.962 | 0.8202 | 0.803 | 0.8091 | 0.683 | 0.7984 | 0.583 |
| 0.8061 | 0.8414 | 0.945 | 0.8319 | 0.793 | 0.8211 | 0.679 | 0.8105 | 0.571 |
| 0.6700 | 0.8898 | 0.914 | 0.8792 | 0.764 | 0.8670 | 0.666 | 0.8571 | 0.563 |
| 0.5301 | 0.9457 | 0.868 | 0.9330 | 0.735 | 0.9207 | 0.652 | 0.9110 | 0.558 |
| 0.4744 | 0.9670 | 0.852 | 0.9564 | 0.728 | 0.9438 | 0.646 | 0.9345 | 0.552 |
| 0.3672 | 1.0203 | 0.819 | 1.0048 | 0.701 | 0.9911 | 0.637 | 0.9815 | 0.547 |
| 0.2528 | 1.0713 | 0.784 | 1.0620 | 0.671 | 1.0483 | 0.624 | 1.0409 | 0.540 |
| 0.1307 | 1.1522 | 0.767 | 1.1310 | 0.650 | 1.1165 | 0.613 | 1.100 | 0.537 |
| 0.0000 | 1.2400 | 0.727 | 1.2296 | 0.623 | 1.2132 | 0.601 | 1.1961 | 0.531 |

**Table IV. Density and Viscosity Data on Ethanol + 1,1,1-Trichloroethane Mixtures**

| mole fraction of ethanol | temp = 303.15 K<br>density, g/mL | viscosity, cP | temp = 313.15 K<br>density, g/mL | viscosity, cP | temp = 323.15 K<br>density, g/mL | viscosity, cP | temp = 333.15 K<br>density, g/mL | viscosity, cP |
|---|---|---|---|---|---|---|---|---|
| 1.0000 | 0.7810 | 1.004 | 0.7738 | 0.835 | 0.7632 | 0.702 | 0.7522 | 0.592 |
| 0.9392 | 0.7991 | 0.984 | 0.7937 | 0.819 | 0.7823 | 0.692 | 0.7717 | 0.589 |
| 0.8728 | 0.8220 | 0.942 | 0.8613 | 0.807 | 0.8054 | 0.684 | 0.7942 | 0.572 |
| 0.8001 | 0.8484 | 0.924 | 0.8426 | 0.784 | 0.8316 | 0.664 | 0.8203 | 0.563 |
| 0.7202 | 0.8797 | 0.902 | 0.8738 | 0.761 | 0.8626 | 0.655 | 0.8512 | 0.558 |
| 0.6318 | 0.9175 | 0.870 | 0.9109 | 0.745 | 0.8996 | 0.639 | 0.8882 | 0.544 |
| 0.5336 | 0.9625 | 0.839 | 0.9561 | 0.710 | 0.9446 | 0.614 | 0.9332 | 0.528 |
| 0.4237 | 1.0189 | 0.787 | 1.0122 | 0.683 | 1.0007 | 0.604 | 0.9893 | 0.517 |
| 0.3002 | 1.0907 | 0.745 | 1.0846 | 0.669 | 1.0722 | 0.597 | 1.0611 | 0.511 |
| 0.1602 | 1.1856 | 0.705 | 1.1783 | 0.614 | 1.1668 | 0.560 | 1.1560 | 0.496 |
| 0.0000 | 1.3207 | 0.642 | 1.3088 | 0.586 | 1.2977 | 0.535 | 1.2881 | 0.483 |

**Table V. Density and Viscosity Data on Ethanol + 1,1,2,2-Tetrachloroethane Mixtures**

| mole fraction of ethanol | temp = 303.15 K<br>density, g/mL | viscosity, cP | temp = 313.15 K<br>density, g/mL | viscosity, cP | temp = 323.15 K<br>density, g/mL | viscosity, cP | temp = 333.15 K<br>density, g/mL | viscosity, cP |
|---|---|---|---|---|---|---|---|---|
| 1.0000 | 0.7810 | 1.004 | 0.7738 | 0.835 | 0.7632 | 0.702 | 0.7522 | 0.592 |
| 0.9420 | 0.8046 | 1.026 | 0.7973 | 0.868 | 0.7865 | 0.732 | 0.7753 | 0.622 |
| 0.8783 | 0.8322 | 1.047 | 0.8247 | 0.903 | 0.8137 | 0.787 | 0.8024 | 0.640 |
| 0.8081 | 0.8649 | 1.095 | 0.8572 | 0.934 | 0.8461 | 0.795 | 0.8345 | 0.673 |
| 0.7303 | 0.9042 | 1.120 | 0.8964 | 0.965 | 0.8875 | 0.819 | 0.8733 | 0.687 |
| 0.6435 | 0.9526 | 1.166 | 0.9495 | 1.007 | 0.9330 | 0.869 | 0.9210 | 0.725 |
| 0.5461 | 1.0134 | 1.210 | 1.0050 | 1.049 | 0.9933 | 0.910 | 0.9811 | 0.764 |
| 0.4362 | 1.0921 | 1.257 | 1.0836 | 1.107 | 1.0715 | 0.961 | 1.0591 | 0.820 |
| 0.3109 | 1.1982 | 1.324 | 1.1894 | 1.160 | 1.1772 | 1.068 | 1.1647 | 0.886 |
| 0.1670 | 1.3486 | 1.369 | 1.3396 | 1.240 | 1.3272 | 1.121 | 1.3154 | 0.955 |
| 0.0000 | 1.5787 | 1.450 | 1.5698 | 1.367 | 1.5588 | 1.180 | 1.5478 | 1.028 |

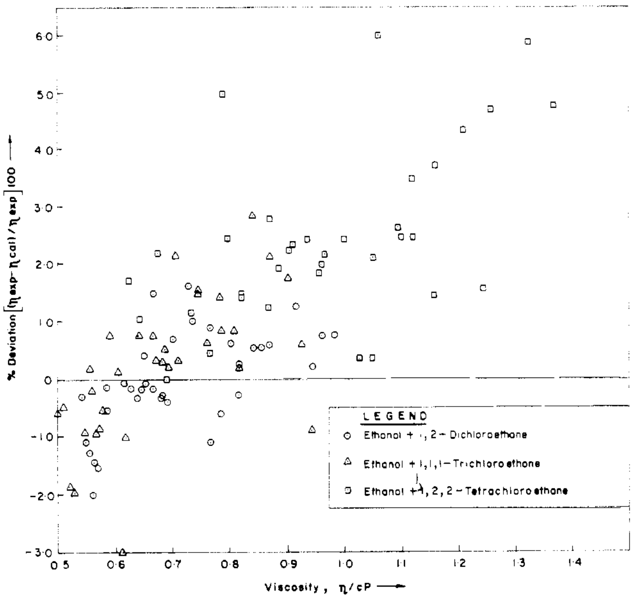

**Figure 1.** Representation of mixture viscosity data by the Arrhenius equation.

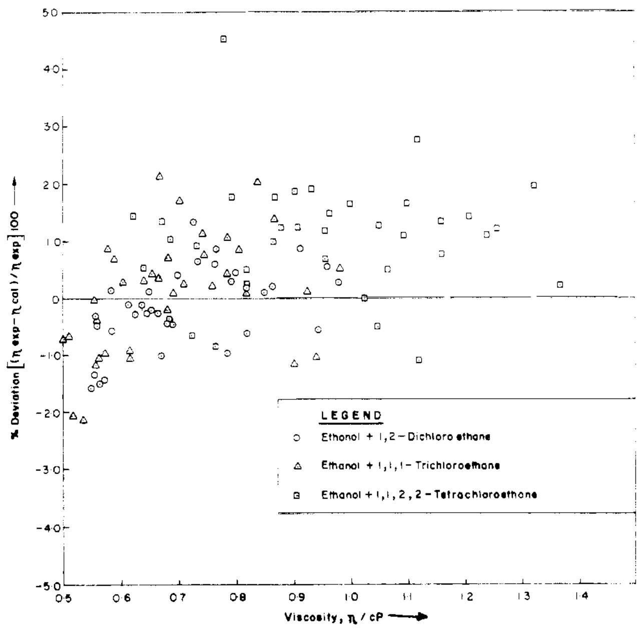

**Figure 2.** Representation of mixture viscosity data by the Kendall and Monroe equation.

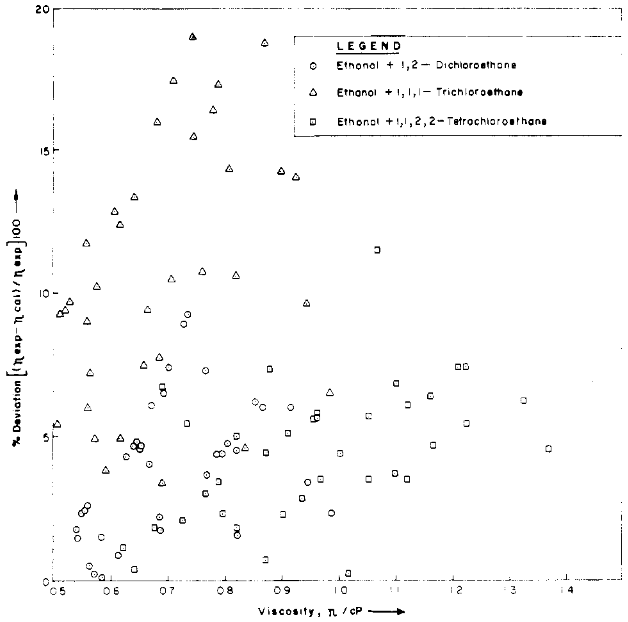

**Figure 3.** Representation of mixture viscosity data by Lobe's equation.

pure-component density, viscosity, and composition as the only input data is summarized in Figures 1–3. It can be seen from the figures that the Kendall and Monroe equation gives better agreement with a percent average absolute deviation of 0.85 compared to 0.98 for the Arrhenius equation and 6.32 for Lobe's equation. Lobe's equation predicts lower values of the liquid mixture viscosity at all the data points.

## Glossary

| | |
|---|---|
| $T$ | temperature, K |
| $t$ | time, s |
| $P$ | pressure, atm |
| $x$ | mole fraction |

### *Subscripts*

| | |
|---|---|
| C | critical |
| m | mixture |
| s | sample |
| w | water |
| 1 | component 1 |
| 2 | component 2 |

### *Greek Letters*

| | |
|---|---|
| $\alpha$ | characteristic viscosity parameter defined by eq 6 |
| $\beta$ | characteristic viscosity parameter defined by eq 7 |
| $\rho$ | density, g/mL |
| $\eta$ | viscosity, cP |
| $\phi$ | volume fraction |

**Registry No.** Ethanol, 64-17-5; 1,2-dichloroethane, 107-06-2; 1,1,1-trichloroethane, 71-55-6; 1,1,2,2-tetrachloroethane, 79-34-5.

## Literature Cited

(1) Venkateshwara Rao, M.; Viswanath, D. S. *J. Chem. Eng. Data* **1982**, *27*, 41.
(2) Viswanath, D. S.; Venkateshwara Rao, M. *J. Chem. Eng. Data* **1982**, *27*, 414.
(3) Venkateshwara Rao, M.; Viswanath, D. S. *J. Chem. Eng. Data* **1982**, *27*, 417.
(4) Rao, M. V.; Viswanath, D. S. *J. Chem. Thermodyn.* **1983**, *15*, 89.
(5) Prabhakar, J.; Ashok Kumar, K.; Narayana Swamy, V.; Venkateshwara Rao, M.; Prasad, D. H. L. *J. Ind. Inst. Sci.* **1986**, *66*, 449.
(6) Bretsznajder, S. *Prediction of Transport and Other Physical Properties of Fluids*; Pergamon: Oxford, 1971; p 232.
(7) Reid, R. C.; Prausnitz, J. M.; Sherwood, T. K. *The Properties of Gases and Liquids*; McGraw-Hill: New York, 1977, p 461.

Received for review September 9, 1988. Revised July 14, 1989. Accepted October 25, 1989.