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# High-Pressure Viscosity and Density Behavior of Ternary Mixtures: 1-Methylnaphthalene + *n*-Tridecane + 2,2,4,4,6,8,8-Heptamethylnonane

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The dynamic viscosity $\eta$ and the density $\rho$ of the ternary system, *n*-tridecane + 1-methylnaphthalene + 2,2,4,4,6,8,8-heptamethylnonane, were measured as a function of temperature from 293.15 to 353.15 K in 10 K increments at pressures up to 100 MPa. A falling body viscometer was used for measuring the dynamic viscosity above 0.1 MPa, while at 0.1 MPa the viscosity was obtained with an Ubbelohde viscometer. The overall uncertainty in the reported data is less than 1 $kg \cdot m^{-3}$ for densities and 2% for viscosities, except at 0.1 MPa where the uncertainty is less than 1%. The experimental results correspond to 882 values of viscosity. With reference to the 126 values published previously for the pure compounds and 882 values for the three associated binaries, the system is globally described by 1890 experimental values as a function of pressure, temperature, and composition. The results for the viscosity are discussed in terms of mixing laws and the excess activation energy of viscous flow.

**KEY WORDS:** 1-methylnaphthalene; 2,2,4,4,6,8,8-heptamethylnonane; density; excess activation energy of flow; high pressure; hydrocarbon; mixture; *n*-tridecane; viscosity.

## 1. INTRODUCTION

Our laboratories have been working for several years on the properties of petroleum fluids and synthetic mixtures, more or less representative of

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petroleum fluids, as a function of pressure, temperature, and composition. In petroleum engineering, knowledge of the dynamic viscosity, $\eta$, as a function of the reservoir conditions (temperature and pressure, as well as the composition of the fluid) is of major importance because several models, covering a wide range of aspects such as reservoir simulations or designing of transport equipments, use this coefficient. While there are abundant viscosity data for mixtures versus temperature $T$ at atmospheric pressure, studies of viscosity versus pressure $P$ are less frequent, particularly for mixtures and those likely to be used to model a real petroleum fluid or a real petroleum cut, in other words, a complex fluid. It is important to stress here that viscosity data for some binary systems are available in which the viscosity as a function of composition, pressure, and temperature is well described, but there are only a few systematic studies concerning ternary mixtures [see, e.g., Ref. 1 for the ternary *n*-heptane + methylcyclohexane + 1-methylnaphthalene described by 21 compositions up to 100 MPa (378 experimental data); Ref. 2 for the ternary *n*-pentane + *n*-octane + *n*-decane described by 15 compositions up to 25 MPa (358 experimental data); and Ref. 3 for the ternary water + 2-propanol + diacetone alcohol described by 36 compositions up to 100 MPa (648 experimental data)].

There are many ways to model a real petroleum cut. We decided, some years ago, to choose the ternary system 1-methylnaphthalene + *n*-tridecane + 2,2,4,4,6,8,8-heptamethylnonane, as part of a very simple representation of some petroleum distillation fractions at 510 K. At atmospheric pressure, the boiling temperature is 507.1 K for *n*-tridecane, 514.7 K for 1-methylnaphthalene, and 513.1 K for 2,2,4,4,6,8,8-heptamethylnonane. The dynamic viscosity and density for the three pure compounds and the three associated binary systems have already been studied [4–6] up to 100 MPa and in the temperature range 293.15 to 353.15 K, involving 126 points for the pure compounds and 882 points for the three binary systems. This paper extends the study to cover measurements of the dynamic viscosity and density of ternary mixtures under the same pressure and temperature conditions as for the pure compounds and binary mixtures, with a sufficient range of compositions to provide a complete coverage of the representative ternary diagram.

## 2. EXPERIMENTAL

### 2.1. Apparatus

The dynamic viscosity $\eta$ was determined with the aid of a falling body viscometer, details of which are provided in Ref. 7, of the type designed by Ducoulombier et al. [8] (using *n*-decane and toluene as reference fluids).

Values of the density $\rho$ for pressures between 0.1 and 60 MPa were measured with an Anton–Paar DMA60 resonance densimeter combined with an additional 512P cell. Details of the calibration of this type of apparatus, with vacuum and water as reference fluids, have been described in Ref. 9. The density measurements were extrapolated to 100 MPa according to the procedure described in Ref. 7 using a Tait-like relationship for the variation of the density with pressure:

$$\frac{1}{\rho(P,T)}=\frac{1}{\rho_0(T)}+A(T)\ln\left(1+\frac{P-P_0}{B(T)}\right) \tag{1}$$

where $\rho_0(T)$ is the density at the pressure $P_0$ and at the temperature $T$. Here we chose $P_0 = 0.1$ MPa. For each temperature, the extrapolation of the experimental densities to 80 and 100 MPa was performed by adjusting the $A$ and $B$ parameters. The validity of this method is discussed in Ref. 7 which has been tested with values provided by Dymond et al. [10] for pure alkanes and binary mixtures up to 500 MPa. For example, for isooctane at $T = 348.15$ K, using the parameters $A$ and $B$ adjusted only with density values up to 40 MPa, a small difference of $-0.4\ \mathrm{kg\cdot m^{-3}}$ (i.e., $-0.06\%$) between experimental and calculated values at 105.2 MPa is obtained, which corresponds approximately to our upper limit of viscosity measurements. It should be emphasized here that an error of $1.0\ \mathrm{kg\cdot m^{-3}}$ in the sample density generates an error of 1/8000 in the viscosity [7]. For the viscosity measurements, the uncertainty in the temperature was estimated to be $\pm 0.5$ K, and for the density measurements the uncertainty in the temperature was estimated to be $\pm 0.05$ K. The uncertainty in the pressure was estimated to be $\pm 0.05$ MPa for the density measurements (HBM manometer) and $\pm 0.1$ MPa for the viscosity measurements (SEDEME manometer), except at 0.1 MPa. The overall uncertainty in the reported density values is less than $1\ \mathrm{kg\cdot m^{-3}}$, while the uncertainty in the viscosity is approximately 2%. As discussed previously [1, 7, 11–13], this error is comparable with the error obtained by other authors for similar experimental systems. Comparative curves have been reported for *n*-heptane and methylcyclohexane in Ref. 13, for water and 2-propanol in Ref. 11, and for 2,2,4,4,6,8,8-heptamethylnonane in Ref. 5, which contain plots of our values and of those obtained by other authors. It should be pointed out that at atmospheric pressure the kinematic viscosity $\eta/\rho$ was determined with a classical capillary viscometer. For this purpose several Ubbelohde tubes, connected to an automatic AVS350 Schott Geräte analyzer, were used. In this case, the uncertainty in the temperature was $\pm 0.05$ K. After multiplying by the density, the uncertainty of the dynamic viscosity is less than 1%.

## 2.2. Characteristics of the Samples

The substances used are commercially available chemicals with the following purity levels: *n*-tridecane ($C_{13}H_{28}$; Tokyo Kasei; purity, $>99\%$; molar mass $M = 184.37\ \mathrm{g \cdot mol^{-1}}$), 1-methylnaphthalene ($C_{11}H_{10}$; Fluka; purity, $>97\%$; $M = 142.20\ \mathrm{g \cdot mol^{-1}}$), and 2,2,4,4,6,8,8-heptamethylnonane ($C_{16}H_{34}$; Aldrich; purity, $>98\%$, $M = 226.44\ \mathrm{g \cdot mol^{-1}}$). The mixtures were prepared by very careful weighing (with a Mettler balance) at atmospheric pressure and ambient temperature to obtain the molar fractions $x_i = 0.125$, 0.250, 0.375, 0.500, 0.625, and 0.750 (with $\sum_{i=1}^{i=3} x_i = 1$) corresponding to the 21 points shown in the ternary diagram (Fig. 1). The systems corresponding to the three summits (pure compounds) and the three sides (binaries) have already been studied [4–6]. The pure fluids, not degassed, were stored in hermetically sealed bottles. The samples were in the liquid state within the experimental temperature and pressure domain.

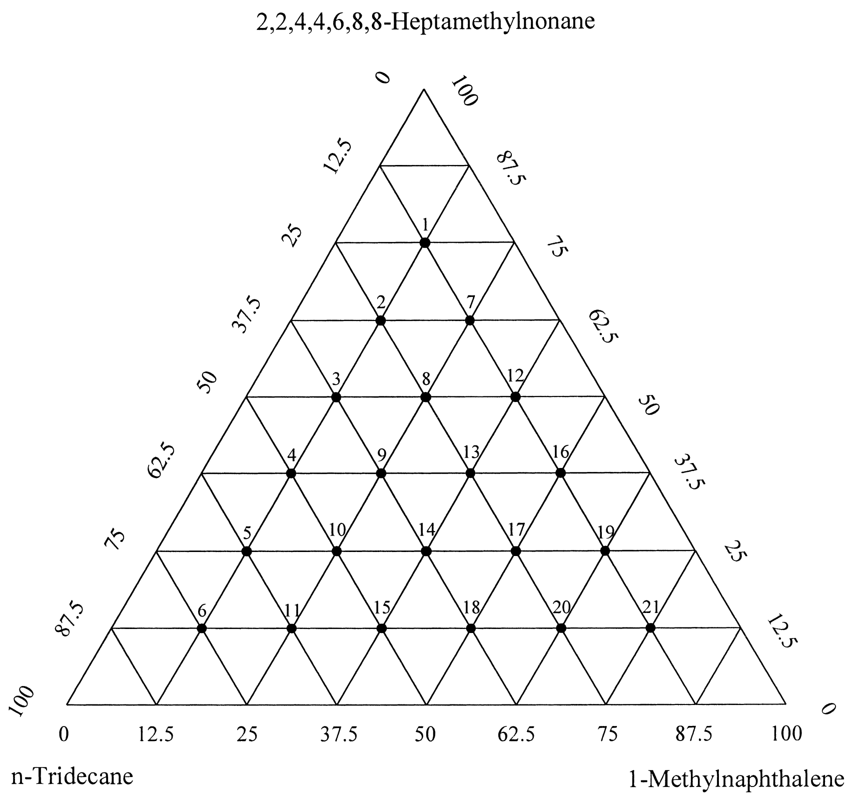


**Fig. 1.** Ternary mixtures studied in this work represented as points 1–21 in the ternary diagram (versus mol%).

## 3. RESULTS

Measurements of the dynamic viscosity $\eta$ were made at seven temperatures (293.15, 303.15, 313.15, 323.15, 333.15, 343.15, and 353.15 K) and at six pressures (0.1, 20, 40, 60, 80, and 100 MPa) for the 21 compositions indicated in Fig. 1. A total of 882 values was obtained for the viscosity. The density measurements were carried out at the same temperatures and compositions at pressures from 0.1 MPa to 60 MPa in steps of 5 MPa (13 pressures), corresponding to 1911 experimental values for the density. These values have been extrapolated (with an error of about 0.1% at $P = 100$ MPa) with the aid [7] of the Tait-type relationship to obtain densities at 80 and 100 MPa (294 values). However, to reduce the amount of tabulated data, we retain only those temperatures and pressures for which the dynamic viscosity has been measured. Additional density data (1029 values) will be published in Refs. 14 and 15. Table I presents the measured dynamic viscosity and density values as a function of temperature $T$, pressure $P$, and mole fraction ($x_m$ for 1-methylnaphthalene, $x_t$ for $n$-tridecane, and $x_h$ for 2,2,4,4,6,8,8-heptamethylnonane; for $x_m$, $x_t$, $x_h \neq 0$ and 1). Figures 2 and 3 show the variations of density with pressure (for different temperatures) and with temperature (for different pressures) in the case corresponding to point 9, which is close to the middle of the ternary diagram in Fig. 1 ($x_m = 0.250$, $x_t = 0.375$, $x_h = 0.375$). Figures 4 and 5 show the dynamic viscosity under the same conditions. Table I and Figs. 2–5 present a general pattern consistent with previous observations made by other authors and by us on either pure hydrocarbons, binary mixtures, or ternary mixtures of hydrocarbons. The group of isothermal and isobaric curves is regular. This is also true for the density, but in the case of isothermal curves, a concavity is observed associated with a second negative derivative. This form is compatible with the logarithmic form proposed by Tait to model the influence of pressure on $1/\rho$, which is the form used for the extrapolation (see, e.g., Ref. 7). It should be noted that the variations of density with $T$ are practically linear due to the small temperature interval (60 K) in this investigation, because the main aim has been to observe the variations of density and viscosity with pressure and composition.

Figure 6 shows, for $P = 40$ MPa and $T = 323.15$ K, the variations of density with $x_t$ ($n$-tridecane) for a constant $x_m$ (1-methylnaphthalene). Points M, T, and H correspond to 1-methylnaphthalene, $n$-tridecane, and 2,2,4,4,6,8,8-heptamethylnonane, respectively, and the sides MT, MH, and TH to the associated binaries. To complete this figure and the following figures, the reported data for the pure compounds and the binaries given in Refs. 4–6 have been used. Within the experimental uncertainty the variations are practically linear, which correspond to very low excess volumes.

**Table I.** Density $\rho$ and Dynamic Viscosity $\eta$ Versus Temperature $T$ and Pressure $P$ for Ternary Mixtures of 1-Methylnaphthalene (m) + $n$-Tridecane (t) + 2,2,4,4,6,8,8-Heptamethylnonane (h)

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.125$, $x_t = 0.125$, $x_h = 0.750$ | | |
| 293.15 | 0.1 | 0.7964 | 3.130 |
| 293.15 | 20 | 0.8089 | 4.380 |
| 293.15 | 40 | 0.8197 | 6.070 |
| 293.15 | 60 | 0.8292 | 8.153 |
| 293.15 | 80 | 0.8377 | 11.055 |
| 293.15 | 100 | 0.8454 | 14.696 |
| 303.15 | 0.1 | 0.7895 | 2.504 |
| 303.15 | 20 | 0.8027 | 3.481 |
| 303.15 | 40 | 0.8139 | 4.661 |
| 303.15 | 60 | 0.8238 | 6.221 |
| 303.15 | 80 | 0.8325 | 8.258 |
| 303.15 | 100 | 0.8403 | 10.896 |
| 313.15 | 0.1 | 0.7827 | 2.049 |
| 313.15 | 20 | 0.7965 | 2.801 |
| 313.15 | 40 | 0.8083 | 3.718 |
| 313.15 | 60 | 0.8184 | 4.903 |
| 313.15 | 80 | 0.8272 | 6.411 |
| 313.15 | 100 | 0.8351 | 8.314 |
| 323.15 | 0.1 | 0.7759 | 1.708 |
| 323.15 | 20 | 0.7905 | 2.386 |
| 323.15 | 40 | 0.8027 | 3.127 |
| 323.15 | 60 | 0.8133 | 4.020 |
| 323.15 | 80 | 0.8226 | 5.132 |
| 323.15 | 100 | 0.8310 | 6.588 |
| 333.15 | 0.1 | 0.7691 | 1.447 |
| 333.15 | 20 | 0.7844 | 2.002 |
| 333.15 | 40 | 0.7972 | 2.585 |
| 333.15 | 60 | 0.8081 | 3.287 |
| 333.15 | 80 | 0.8177 | 4.127 |
| 333.15 | 100 | 0.8263 | 5.179 |
| 343.15 | 0.1 | 0.7621 | 1.248 |
| 343.15 | 20 | 0.7783 | 1.740 |
| 343.15 | 40 | 0.7915 | 2.250 |
| 343.15 | 60 | 0.8029 | 2.856 |
| 343.15 | 80 | 0.8127 | 3.567 |
| 343.15 | 100 | 0.8216 | 4.389 |
| 353.15 | 0.1 | 0.7552 | 1.083 |
| 353.15 | 20 | 0.7721 | 1.500 |
| 353.15 | 40 | 0.7860 | 1.920 |
| 353.15 | 60 | 0.7977 | 2.434 |
| 353.15 | 80 | 0.8079 | 3.054 |
| 353.15 | 100 | 0.8169 | 3.721 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.125$, $x_t = 0.250$, $x_h = 0.625$ | | |
| 293.15 | 0.1 | 0.7936 | 2.775 |
| 293.15 | 20 | 0.8061 | 3.894 |
| 293.15 | 40 | 0.8169 | 5.210 |
| 293.15 | 60 | 0.8264 | 6.900 |
| 293.15 | 80 | 0.8349 | 9.042 |
| 293.15 | 100 | 0.8427 | 11.735 |
| 303.15 | 0.1 | 0.7870 | 2.241 |
| 303.15 | 20 | 0.8002 | 3.113 |
| 303.15 | 40 | 0.8114 | 4.120 |
| 303.15 | 60 | 0.8213 | 5.376 |
| 303.15 | 80 | 0.8301 | 6.920 |
| 303.15 | 100 | 0.8381 | 8.799 |
| 313.15 | 0.1 | 0.7802 | 1.846 |
| 313.15 | 20 | 0.7940 | 2.550 |
| 313.15 | 40 | 0.8057 | 3.364 |
| 313.15 | 60 | 0.8159 | 4.354 |
| 313.15 | 80 | 0.8249 | 5.540 |
| 313.15 | 100 | 0.8330 | 6.944 |
| 323.15 | 0.1 | 0.7732 | 1.550 |
| 323.15 | 20 | 0.7877 | 2.128 |
| 323.15 | 40 | 0.7999 | 2.781 |
| 323.15 | 60 | 0.8105 | 3.568 |
| 323.15 | 80 | 0.8199 | 4.501 |
| 323.15 | 100 | 0.8283 | 5.594 |
| 333.15 | 0.1 | 0.7663 | 1.326 |
| 333.15 | 20 | 0.7816 | 1.815 |
| 333.15 | 40 | 0.7943 | 2.331 |
| 333.15 | 60 | 0.8053 | 2.945 |
| 333.15 | 80 | 0.8148 | 3.662 |
| 333.15 | 100 | 0.8233 | 4.493 |
| 343.15 | 0.1 | 0.7593 | 1.144 |
| 343.15 | 20 | 0.7755 | 1.561 |
| 343.15 | 40 | 0.7887 | 1.983 |
| 343.15 | 60 | 0.8000 | 2.489 |
| 343.15 | 80 | 0.8099 | 3.083 |
| 343.15 | 100 | 0.8187 | 3.774 |
| 353.15 | 0.1 | 0.7523 | 0.999 |
| 353.15 | 20 | 0.7692 | 1.375 |
| 353.15 | 40 | 0.7830 | 1.740 |
| 353.15 | 60 | 0.7948 | 2.167 |
| 353.15 | 80 | 0.8050 | 2.658 |
| 353.15 | 100 | 0.8140 | 3.216 |

Table I. *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g\cdot cm^{-3}$) | $\eta$ ($mPa\cdot s$) |
|---|---|---|---|
| $x_m = 0.125$, $x_t = 0.375$, $x_h = 0.500$ | | | |
| 293.15 | 0.1 | 0.7903 | 2.524 |
| 293.15 | 20 | 0.8027 | 3.436 |
| 293.15 | 40 | 0.8135 | 4.526 |
| 293.15 | 60 | 0.8230 | 5.871 |
| 293.15 | 80 | 0.8315 | 7.506 |
| 293.15 | 100 | 0.8392 | 9.519 |
| 303.15 | 0.1 | 0.7833 | 2.052 |
| 303.15 | 20 | 0.7965 | 2.765 |
| 303.15 | 40 | 0.8076 | 3.614 |
| 303.15 | 60 | 0.8176 | 4.644 |
| 303.15 | 80 | 0.8263 | 5.875 |
| 303.15 | 100 | 0.8343 | 7.284 |
| 313.15 | 0.1 | 0.7764 | 1.702 |
| 313.15 | 20 | 0.7902 | 2.306 |
| 313.15 | 40 | 0.8019 | 2.990 |
| 313.15 | 60 | 0.8121 | 3.801 |
| 313.15 | 80 | 0.8211 | 4.749 |
| 313.15 | 100 | 0.8293 | 5.819 |
| 323.15 | 0.1 | 0.7695 | 1.434 |
| 323.15 | 20 | 0.7840 | 1.930 |
| 323.15 | 40 | 0.7961 | 2.475 |
| 323.15 | 60 | 0.8068 | 3.114 |
| 323.15 | 80 | 0.8161 | 3.854 |
| 323.15 | 100 | 0.8246 | 4.704 |
| 333.15 | 0.1 | 0.7625 | 1.229 |
| 333.15 | 20 | 0.7778 | 1.650 |
| 333.15 | 40 | 0.7905 | 2.097 |
| 333.15 | 60 | 0.8014 | 2.613 |
| 333.15 | 80 | 0.8110 | 3.199 |
| 333.15 | 100 | 0.8196 | 3.877 |
| 343.15 | 0.1 | 0.7554 | 1.066 |
| 343.15 | 20 | 0.7716 | 1.414 |
| 343.15 | 40 | 0.7848 | 1.783 |
| 343.15 | 60 | 0.7961 | 2.213 |
| 343.15 | 80 | 0.8060 | 2.706 |
| 343.15 | 100 | 0.8148 | 3.279 |
| 353.15 | 0.1 | 0.7484 | 0.934 |
| 353.15 | 20 | 0.7654 | 1.245 |
| 353.15 | 40 | 0.7792 | 1.568 |
| 353.15 | 60 | 0.7910 | 1.944 |
| 353.15 | 80 | 0.8011 | 2.376 |
| 353.15 | 100 | 0.8101 | 2.859 |

Table I. *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ (mPa·s) |
|---|---|---|---|
| | $x_m = 0.125$, $x_t = 0.500$, $x_h = 0.375$ | | |
| 293.15 | 0.1 | 0.7872 | 2.319 |
| 293.15 | 20 | 0.7997 | 3.083 |
| 293.15 | 40 | 0.8105 | 3.978 |
| 293.15 | 60 | 0.8201 | 5.073 |
| 293.15 | 80 | 0.8286 | 6.390 |
| 293.15 | 100 | 0.8363 | 7.962 |
| 303.15 | 0.1 | 0.7805 | 1.889 |
| 303.15 | 20 | 0.7936 | 2.551 |
| 303.15 | 40 | 0.8048 | 3.259 |
| 303.15 | 60 | 0.8148 | 4.092 |
| 303.15 | 80 | 0.8236 | 5.056 |
| 303.15 | 100 | 0.8316 | 6.158 |
| 313.15 | 0.1 | 0.7736 | 1.573 |
| 313.15 | 20 | 0.7874 | 2.129 |
| 313.15 | 40 | 0.7990 | 2.715 |
| 313.15 | 60 | 0.8093 | 3.396 |
| 313.15 | 80 | 0.8183 | 4.173 |
| 313.15 | 100 | 0.8265 | 5.050 |
| 323.15 | 0.1 | 0.7666 | 1.333 |
| 323.15 | 20 | 0.7811 | 1.814 |
| 323.15 | 40 | 0.7932 | 2.304 |
| 323.15 | 60 | 0.8038 | 2.859 |
| 323.15 | 80 | 0.8131 | 3.477 |
| 323.15 | 100 | 0.8215 | 4.153 |
| 333.15 | 0.1 | 0.7595 | 1.149 |
| 333.15 | 20 | 0.7748 | 1.544 |
| 333.15 | 40 | 0.7875 | 1.936 |
| 333.15 | 60 | 0.7985 | 2.387 |
| 333.15 | 80 | 0.8080 | 2.896 |
| 333.15 | 100 | 0.8166 | 3.465 |
| 343.15 | 0.1 | 0.7524 | 0.998 |
| 343.15 | 20 | 0.7685 | 1.340 |
| 343.15 | 40 | 0.7817 | 1.666 |
| 343.15 | 60 | 0.7930 | 2.034 |
| 343.15 | 80 | 0.8030 | 2.440 |
| 343.15 | 100 | 0.8119 | 2.884 |
| 353.15 | 0.1 | 0.7453 | 0.882 |
| 353.15 | 20 | 0.7622 | 1.177 |
| 353.15 | 40 | 0.7759 | 1.461 |
| 353.15 | 60 | 0.7877 | 1.777 |
| 353.15 | 80 | 0.7979 | 2.123 |
| 353.15 | 100 | 0.8070 | 2.497 |

Table I. *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.125$, $x_t = 0.625$, $x_h = 0.250$ | | |
| 293.15 | 0.1 | 0.7839 | 2.141 |
| 293.15 | 20 | 0.7963 | 2.841 |
| 293.15 | 40 | 0.8071 | 3.606 |
| 293.15 | 60 | 0.8166 | 4.518 |
| 293.15 | 80 | 0.8252 | 5.587 |
| 293.15 | 100 | 0.8329 | 6.829 |
| 303.15 | 0.1 | 0.7769 | 1.758 |
| 303.15 | 20 | 0.7901 | 2.326 |
| 303.15 | 40 | 0.8013 | 2.939 |
| 303.15 | 60 | 0.8112 | 3.649 |
| 303.15 | 80 | 0.8200 | 4.458 |
| 303.15 | 100 | 0.8280 | 5.368 |
| 313.15 | 0.1 | 0.7700 | 1.469 |
| 313.15 | 20 | 0.7838 | 1.942 |
| 313.15 | 40 | 0.7954 | 2.445 |
| 313.15 | 60 | 0.8057 | 3.028 |
| 313.15 | 80 | 0.8147 | 3.618 |
| 313.15 | 100 | 0.8229 | 4.347 |
| 323.15 | 0.1 | 0.7629 | 1.249 |
| 323.15 | 20 | 0.7774 | 1.668 |
| 323.15 | 40 | 0.7896 | 2.099 |
| 323.15 | 60 | 0.8002 | 2.590 |
| 323.15 | 80 | 0.8097 | 3.077 |
| 323.15 | 100 | 0.8182 | 3.600 |
| 333.15 | 0.1 | 0.7557 | 1.077 |
| 333.15 | 20 | 0.7710 | 1.437 |
| 333.15 | 40 | 0.7837 | 1.789 |
| 333.15 | 60 | 0.7947 | 2.186 |
| 333.15 | 80 | 0.8043 | 2.622 |
| 333.15 | 100 | 0.8129 | 3.092 |
| 343.15 | 0.1 | 0.7486 | 0.942 |
| 343.15 | 20 | 0.7646 | 1.237 |
| 343.15 | 40 | 0.7779 | 1.527 |
| 343.15 | 60 | 0.7893 | 1.857 |
| 343.15 | 80 | 0.7993 | 2.226 |
| 343.15 | 100 | 0.8082 | 2.633 |
| 353.15 | 0.1 | 0.7414 | 0.837 |
| 353.15 | 20 | 0.7583 | 1.096 |
| 353.15 | 40 | 0.7721 | 1.353 |
| 353.15 | 60 | 0.7840 | 1.642 |
| 353.15 | 80 | 0.7943 | 1.961 |
| 353.15 | 100 | 0.8035 | 2.309 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.125$, $x_t = 0.750$, $x_h = 0.125$ | | |
| 293.15 | 0.1 | 0.7800 | 1.990 |
| 293.15 | 20 | 0.7924 | 2.582 |
| 293.15 | 40 | 0.8032 | 3.245 |
| 293.15 | 60 | 0.8127 | 4.014 |
| 293.15 | 80 | 0.8213 | 4.988 |
| 293.15 | 100 | 0.8291 | 6.017 |
| 303.15 | 0.1 | 0.7728 | 1.642 |
| 303.15 | 20 | 0.7859 | 2.159 |
| 303.15 | 40 | 0.7971 | 2.685 |
| 303.15 | 60 | 0.8070 | 3.288 |
| 303.15 | 80 | 0.8158 | 3.966 |
| 303.15 | 100 | 0.8237 | 4.690 |
| 313.15 | 0.1 | 0.7658 | 1.381 |
| 313.15 | 20 | 0.7795 | 1.805 |
| 313.15 | 40 | 0.7912 | 2.227 |
| 313.15 | 60 | 0.8014 | 2.711 |
| 313.15 | 80 | 0.8105 | 3.257 |
| 313.15 | 100 | 0.8187 | 3.848 |
| 323.15 | 0.1 | 0.7586 | 1.178 |
| 323.15 | 20 | 0.7730 | 1.544 |
| 323.15 | 40 | 0.7852 | 1.902 |
| 323.15 | 60 | 0.7958 | 2.307 |
| 323.15 | 80 | 0.8053 | 2.757 |
| 323.15 | 100 | 0.8138 | 3.252 |
| 333.15 | 0.1 | 0.7514 | 1.017 |
| 333.15 | 20 | 0.7666 | 1.311 |
| 333.15 | 40 | 0.7794 | 1.617 |
| 333.15 | 60 | 0.7903 | 1.966 |
| 333.15 | 80 | 0.8000 | 2.356 |
| 333.15 | 100 | 0.8087 | 2.801 |
| 343.15 | 0.1 | 0.7442 | 0.893 |
| 343.15 | 20 | 0.7602 | 1.153 |
| 343.15 | 40 | 0.7735 | 1.415 |
| 343.15 | 60 | 0.7849 | 1.715 |
| 343.15 | 80 | 0.7949 | 2.052 |
| 343.15 | 100 | 0.8039 | 2.437 |
| 353.15 | 0.1 | 0.7370 | 0.789 |
| 353.15 | 20 | 0.7538 | 0.999 |
| 353.15 | 40 | 0.7677 | 1.221 |
| 353.15 | 60 | 0.7795 | 1.478 |
| 353.15 | 80 | 0.7899 | 1.769 |
| 353.15 | 100 | 0.7991 | 2.088 |

Table I. *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ (g·cm$^{-3}$) | $\eta$ (mPa·s) |
|---|---|---|---|
| $x_m = 0.250$, $x_t = 0.125$, $x_h = 0.625$ | | | |
| 293.15 | 0.1 | 0.8146 | 2.903 |
| 293.15 | 20 | 0.8269 | 4.016 |
| 293.15 | 40 | 0.8376 | 5.412 |
| 293.15 | 60 | 0.8470 | 7.194 |
| 293.15 | 80 | 0.8555 | 9.440 |
| 293.15 | 100 | 0.8631 | 12.299 |
| 303.15 | 0.1 | 0.8078 | 2.333 |
| 303.15 | 20 | 0.8207 | 3.197 |
| 303.15 | 40 | 0.8318 | 4.254 |
| 303.15 | 60 | 0.8416 | 5.608 |
| 303.15 | 80 | 0.8503 | 7.320 |
| 303.15 | 100 | 0.8581 | 9.405 |
| 313.15 | 0.1 | 0.8009 | 1.925 |
| 313.15 | 20 | 0.8145 | 2.606 |
| 313.15 | 40 | 0.8260 | 3.423 |
| 313.15 | 60 | 0.8362 | 4.444 |
| 313.15 | 80 | 0.8452 | 5.702 |
| 313.15 | 100 | 0.8533 | 7.207 |
| 323.15 | 0.1 | 0.7940 | 1.612 |
| 323.15 | 20 | 0.8083 | 2.161 |
| 323.15 | 40 | 0.8203 | 2.821 |
| 323.15 | 60 | 0.8308 | 3.627 |
| 323.15 | 80 | 0.8401 | 4.598 |
| 323.15 | 100 | 0.8485 | 5.764 |
| 333.15 | 0.1 | 0.7871 | 1.376 |
| 333.15 | 20 | 0.8021 | 1.861 |
| 333.15 | 40 | 0.8146 | 2.388 |
| 333.15 | 60 | 0.8254 | 3.028 |
| 333.15 | 80 | 0.8349 | 3.795 |
| 333.15 | 100 | 0.8435 | 4.728 |
| 343.15 | 0.1 | 0.7803 | 1.187 |
| 343.15 | 20 | 0.7959 | 1.594 |
| 343.15 | 40 | 0.8089 | 2.042 |
| 343.15 | 60 | 0.8202 | 2.579 |
| 343.15 | 80 | 0.8301 | 3.215 |
| 343.15 | 100 | 0.8390 | 3.975 |
| 353.15 | 0.1 | 0.7732 | 1.037 |
| 353.15 | 20 | 0.7897 | 1.398 |
| 353.15 | 40 | 0.8033 | 1.787 |
| 353.15 | 60 | 0.8150 | 2.245 |
| 353.15 | 80 | 0.8252 | 2.777 |
| 353.15 | 100 | 0.8343 | 3.380 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.250$, $x_t = 0.250$, $x_h = 0.500$ | | |
| 293.15 | 0.1 | 0.8110 | 2.621 |
| 293.15 | 20 | 0.8233 | 3.545 |
| 293.15 | 40 | 0.8340 | 4.728 |
| 293.15 | 60 | 0.8435 | 6.130 |
| 293.15 | 80 | 0.8520 | 7.920 |
| 293.15 | 100 | 0.8597 | 10.100 |
| 303.15 | 0.1 | 0.8043 | 2.122 |
| 303.15 | 20 | 0.8172 | 2.867 |
| 303.15 | 40 | 0.8283 | 3.730 |
| 303.15 | 60 | 0.8381 | 4.799 |
| 303.15 | 80 | 0.8468 | 6.102 |
| 303.15 | 100 | 0.8546 | 7.674 |
| 313.15 | 0.1 | 0.7974 | 1.757 |
| 313.15 | 20 | 0.8110 | 2.362 |
| 313.15 | 40 | 0.8225 | 3.082 |
| 313.15 | 60 | 0.8327 | 3.922 |
| 313.15 | 80 | 0.8417 | 4.882 |
| 313.15 | 100 | 0.8498 | 5.966 |
| 323.15 | 0.1 | 0.7904 | 1.486 |
| 323.15 | 20 | 0.8047 | 1.989 |
| 323.15 | 40 | 0.8168 | 2.535 |
| 323.15 | 60 | 0.8273 | 3.182 |
| 323.15 | 80 | 0.8365 | 3.935 |
| 323.15 | 100 | 0.8449 | 4.800 |
| 333.15 | 0.1 | 0.7834 | 1.272 |
| 333.15 | 20 | 0.7984 | 1.701 |
| 333.15 | 40 | 0.8110 | 2.161 |
| 333.15 | 60 | 0.8218 | 2.703 |
| 333.15 | 80 | 0.8313 | 3.331 |
| 333.15 | 100 | 0.8399 | 4.051 |
| 343.15 | 0.1 | 0.7764 | 1.109 |
| 343.15 | 20 | 0.7922 | 1.473 |
| 343.15 | 40 | 0.8052 | 1.859 |
| 343.15 | 60 | 0.8165 | 2.274 |
| 343.15 | 80 | 0.8262 | 2.777 |
| 343.15 | 100 | 0.8349 | 3.390 |
| 353.15 | 0.1 | 0.7693 | 0.969 |
| 353.15 | 20 | 0.7858 | 1.304 |
| 353.15 | 40 | 0.7995 | 1.622 |
| 353.15 | 60 | 0.8112 | 1.996 |
| 353.15 | 80 | 0.8213 | 2.427 |
| 353.15 | 100 | 0.8303 | 2.918 |

Table I. *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ (g·cm$^{-3}$) | $\eta$ (mPa·s) |
|---|---|---|---|
| $x_m = 0.250$, $x_t = 0.375$, and $x_h = 0.375$ | | | |
| 293.15 | 0.1 | 0.8089 | 2.396 |
| 293.15 | 20 | 0.8212 | 3.161 |
| 293.15 | 40 | 0.8319 | 4.063 |
| 293.15 | 60 | 0.8414 | 5.159 |
| 293.15 | 80 | 0.8498 | 6.470 |
| 293.15 | 100 | 0.8575 | 8.102 |
| 303.15 | 0.1 | 0.8019 | 1.959 |
| 303.15 | 20 | 0.8148 | 2.622 |
| 303.15 | 40 | 0.8259 | 3.358 |
| 303.15 | 60 | 0.8358 | 4.239 |
| 303.15 | 80 | 0.8445 | 5.277 |
| 303.15 | 100 | 0.8525 | 6.487 |
| 313.15 | 0.1 | 0.7950 | 1.625 |
| 313.15 | 20 | 0.8085 | 2.154 |
| 313.15 | 40 | 0.8200 | 2.756 |
| 313.15 | 60 | 0.8302 | 3.471 |
| 313.15 | 80 | 0.8392 | 4.305 |
| 313.15 | 100 | 0.8474 | 5.266 |
| 323.15 | 0.1 | 0.7879 | 1.376 |
| 323.15 | 20 | 0.8021 | 1.842 |
| 323.15 | 40 | 0.8142 | 2.341 |
| 323.15 | 60 | 0.8247 | 2.915 |
| 323.15 | 80 | 0.8339 | 3.566 |
| 323.15 | 100 | 0.8422 | 4.291 |
| 333.15 | 0.1 | 0.7808 | 1.182 |
| 333.15 | 20 | 0.7958 | 1.560 |
| 333.15 | 40 | 0.8083 | 1.967 |
| 333.15 | 60 | 0.8192 | 2.433 |
| 333.15 | 80 | 0.8288 | 2.958 |
| 333.15 | 100 | 0.8373 | 3.542 |
| 343.15 | 0.1 | 0.7738 | 1.029 |
| 343.15 | 20 | 0.7895 | 1.352 |
| 343.15 | 40 | 0.8026 | 1.683 |
| 343.15 | 60 | 0.8139 | 2.063 |
| 343.15 | 80 | 0.8236 | 2.491 |
| 343.15 | 100 | 0.8325 | 2.968 |
| 353.15 | 0.1 | 0.7666 | 0.905 |
| 353.15 | 20 | 0.7831 | 1.199 |
| 353.15 | 40 | 0.7968 | 1.480 |
| 353.15 | 60 | 0.8085 | 1.809 |
| 353.15 | 80 | 0.8187 | 2.186 |
| 353.15 | 100 | 0.8278 | 2.612 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ (g·cm$^{-3}$) | $\eta$ (mPa·s) |
|---|---|---|---|
| $x_m = 0.250$, $x_t = 0.500$, $x_h = 0.250$ | | | |
| 293.15 | 0.1 | 0.8059 | 2.203 |
| 293.15 | 20 | 0.8182 | 2.877 |
| 293.15 | 40 | 0.8289 | 3.666 |
| 293.15 | 60 | 0.8384 | 4.610 |
| 293.15 | 80 | 0.8468 | 5.720 |
| 293.15 | 100 | 0.8545 | 7.013 |
| 303.15 | 0.1 | 0.7990 | 1.805 |
| 303.15 | 20 | 0.8118 | 2.357 |
| 303.15 | 40 | 0.8229 | 2.980 |
| 303.15 | 60 | 0.8328 | 3.710 |
| 303.15 | 80 | 0.8415 | 4.550 |
| 303.15 | 100 | 0.8495 | 5.505 |
| 313.15 | 0.1 | 0.7918 | 1.506 |
| 313.15 | 20 | 0.8054 | 1.988 |
| 313.15 | 40 | 0.8169 | 2.499 |
| 313.15 | 60 | 0.8270 | 3.075 |
| 313.15 | 80 | 0.8360 | 3.712 |
| 313.15 | 100 | 0.8442 | 4.404 |
| 323.15 | 0.1 | 0.7847 | 1.281 |
| 323.15 | 20 | 0.7989 | 1.686 |
| 323.15 | 40 | 0.8110 | 2.092 |
| 323.15 | 60 | 0.8216 | 2.591 |
| 323.15 | 80 | 0.8309 | 3.126 |
| 323.15 | 100 | 0.8394 | 3.678 |
| 333.15 | 0.1 | 0.7775 | 1.107 |
| 333.15 | 20 | 0.7925 | 1.449 |
| 333.15 | 40 | 0.8050 | 1.783 |
| 333.15 | 60 | 0.8159 | 2.167 |
| 333.15 | 80 | 0.8255 | 2.600 |
| 333.15 | 100 | 0.8342 | 3.082 |
| 343.15 | 0.1 | 0.7704 | 0.967 |
| 343.15 | 20 | 0.7861 | 1.268 |
| 343.15 | 40 | 0.7992 | 1.561 |
| 343.15 | 60 | 0.8104 | 1.897 |
| 343.15 | 80 | 0.8203 | 2.275 |
| 343.15 | 100 | 0.8292 | 2.696 |
| 353.15 | 0.1 | 0.7631 | 0.849 |
| 353.15 | 20 | 0.7797 | 1.126 |
| 353.15 | 40 | 0.7933 | 1.373 |
| 353.15 | 60 | 0.8051 | 1.656 |
| 353.15 | 80 | 0.8152 | 1.973 |
| 353.15 | 100 | 0.8243 | 2.325 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ (mPa $\cdot$ s) |
|---|---|---|---|
| $x_m = 0.250$, $x_t = 0.625$, $x_h = 0.125$ | | | |
| 293.15 | 0.1 | 0.8033 | 2.068 |
| 293.15 | 20 | 0.8155 | 2.627 |
| 293.15 | 40 | 0.8261 | 3.295 |
| 293.15 | 60 | 0.8356 | 4.083 |
| 293.15 | 80 | 0.8441 | 4.998 |
| 293.15 | 100 | 0.8519 | 6.076 |
| 303.15 | 0.1 | 0.7961 | 1.728 |
| 303.15 | 20 | 0.8088 | 2.150 |
| 303.15 | 40 | 0.8200 | 2.668 |
| 303.15 | 60 | 0.8298 | 3.267 |
| 303.15 | 80 | 0.8386 | 3.949 |
| 303.15 | 100 | 0.8467 | 4.685 |
| 313.15 | 0.1 | 0.7887 | 1.442 |
| 313.15 | 20 | 0.8022 | 1.807 |
| 313.15 | 40 | 0.8138 | 2.227 |
| 313.15 | 60 | 0.8239 | 2.713 |
| 313.15 | 80 | 0.8330 | 3.266 |
| 313.15 | 100 | 0.8412 | 3.871 |
| 323.15 | 0.1 | 0.7814 | 1.224 |
| 323.15 | 20 | 0.7956 | 1.558 |
| 323.15 | 40 | 0.8077 | 1.915 |
| 323.15 | 60 | 0.8183 | 2.323 |
| 323.15 | 80 | 0.8276 | 2.780 |
| 323.15 | 100 | 0.8360 | 3.289 |
| 333.15 | 0.1 | 0.7743 | 1.052 |
| 333.15 | 20 | 0.7891 | 1.346 |
| 333.15 | 40 | 0.8017 | 1.651 |
| 333.15 | 60 | 0.8126 | 1.998 |
| 333.15 | 80 | 0.8223 | 2.385 |
| 333.15 | 100 | 0.8310 | 2.825 |
| 343.15 | 0.1 | 0.7670 | 0.911 |
| 343.15 | 20 | 0.7827 | 1.168 |
| 343.15 | 40 | 0.7957 | 1.421 |
| 343.15 | 60 | 0.8071 | 1.725 |
| 343.15 | 80 | 0.8171 | 2.062 |
| 343.15 | 100 | 0.8261 | 2.437 |
| 353.15 | 0.1 | 0.7597 | 0.806 |
| 353.15 | 20 | 0.7762 | 1.025 |
| 353.15 | 40 | 0.7899 | 1.248 |
| 353.15 | 60 | 0.8016 | 1.492 |
| 353.15 | 80 | 0.8118 | 1.773 |
| 353.15 | 100 | 0.8210 | 2.070 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.375$, $x_t = 0.125$, $x_h = 0.500$ | | |
| 293.15 | 0.1 | 0.8353 | 2.770 |
| 293.15 | 20 | 0.8473 | 3.725 |
| 293.15 | 40 | 0.8579 | 4.939 |
| 293.15 | 60 | 0.8672 | 6.481 |
| 293.15 | 80 | 0.8756 | 8.410 |
| 293.15 | 100 | 0.8832 | 10.852 |
| 303.15 | 0.1 | 0.8284 | 2.232 |
| 303.15 | 20 | 0.8410 | 2.995 |
| 303.15 | 40 | 0.8518 | 3.892 |
| 303.15 | 60 | 0.8616 | 5.019 |
| 303.15 | 80 | 0.8702 | 6.416 |
| 303.15 | 100 | 0.8781 | 8.077 |
| 313.15 | 0.1 | 0.8213 | 1.841 |
| 313.15 | 20 | 0.8346 | 2.453 |
| 313.15 | 40 | 0.8460 | 3.171 |
| 313.15 | 60 | 0.8560 | 4.053 |
| 313.15 | 80 | 0.8649 | 5.120 |
| 313.15 | 100 | 0.8730 | 6.371 |
| 323.15 | 0.1 | 0.8143 | 1.550 |
| 323.15 | 20 | 0.8283 | 2.078 |
| 323.15 | 40 | 0.8402 | 2.657 |
| 323.15 | 60 | 0.8506 | 3.347 |
| 323.15 | 80 | 0.8598 | 4.155 |
| 323.15 | 100 | 0.8681 | 5.097 |
| 333.15 | 0.1 | 0.8074 | 1.325 |
| 333.15 | 20 | 0.8220 | 1.745 |
| 333.15 | 40 | 0.8344 | 2.218 |
| 333.15 | 60 | 0.8451 | 2.771 |
| 333.15 | 80 | 0.8547 | 3.408 |
| 333.15 | 100 | 0.8633 | 4.155 |
| 343.15 | 0.1 | 0.8003 | 1.146 |
| 343.15 | 20 | 0.8157 | 1.508 |
| 343.15 | 40 | 0.8286 | 1.886 |
| 343.15 | 60 | 0.8397 | 2.335 |
| 343.15 | 80 | 0.8495 | 2.887 |
| 343.15 | 100 | 0.8583 | 3.512 |
| 353.15 | 0.1 | 0.7933 | 1.004 |
| 353.15 | 20 | 0.8095 | 1.324 |
| 353.15 | 40 | 0.8229 | 1.649 |
| 353.15 | 60 | 0.8344 | 2.038 |
| 353.15 | 80 | 0.8444 | 2.496 |
| 353.15 | 100 | 0.8534 | 3.021 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| $x_m = 0.375,\ x_t = 0.250,\ x_h = 0.375$ | | | |
| 293.15 | 0.1 | 0.8331 | 2.495 |
| 293.15 | 20 | 0.8451 | 3.308 |
| 293.15 | 40 | 0.8557 | 4.227 |
| 293.15 | 60 | 0.8650 | 5.446 |
| 293.15 | 80 | 0.8734 | 6.867 |
| 293.15 | 100 | 0.8811 | 8.567 |
| 303.15 | 0.1 | 0.8261 | 2.028 |
| 303.15 | 20 | 0.8387 | 2.680 |
| 303.15 | 40 | 0.8496 | 3.436 |
| 303.15 | 60 | 0.8593 | 4.336 |
| 303.15 | 80 | 0.8680 | 5.391 |
| 303.15 | 100 | 0.8760 | 6.612 |
| 313.15 | 0.1 | 0.8190 | 1.684 |
| 313.15 | 20 | 0.8322 | 2.231 |
| 313.15 | 40 | 0.8436 | 2.823 |
| 313.15 | 60 | 0.8537 | 3.529 |
| 313.15 | 80 | 0.8626 | 4.354 |
| 313.15 | 100 | 0.8707 | 5.308 |
| 323.15 | 0.1 | 0.8119 | 1.423 |
| 323.15 | 20 | 0.8258 | 1.869 |
| 323.15 | 40 | 0.8377 | 2.388 |
| 323.15 | 60 | 0.8480 | 2.952 |
| 323.15 | 80 | 0.8573 | 3.634 |
| 323.15 | 100 | 0.8656 | 4.374 |
| 333.15 | 0.1 | 0.8048 | 1.221 |
| 333.15 | 20 | 0.8194 | 1.597 |
| 333.15 | 40 | 0.8318 | 1.986 |
| 333.15 | 60 | 0.8426 | 2.440 |
| 333.15 | 80 | 0.8520 | 2.961 |
| 333.15 | 100 | 0.8606 | 3.552 |
| 343.15 | 0.1 | 0.7977 | 1.065 |
| 343.15 | 20 | 0.8130 | 1.381 |
| 343.15 | 40 | 0.8259 | 1.723 |
| 343.15 | 60 | 0.8371 | 2.122 |
| 343.15 | 80 | 0.8468 | 2.579 |
| 343.15 | 100 | 0.8556 | 3.096 |
| 353.15 | 0.1 | 0.7905 | 0.938 |
| 353.15 | 20 | 0.8066 | 1.214 |
| 353.15 | 40 | 0.8200 | 1.496 |
| 353.15 | 60 | 0.8316 | 1.820 |
| 353.15 | 80 | 0.8418 | 2.187 |
| 353.15 | 100 | 0.8508 | 2.597 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.375$, $x_t = 0.375$, $x_h = 0.250$ | | |
| 293.15 | 0.1 | 0.8307 | 2.314 |
| 293.15 | 20 | 0.8427 | 3.032 |
| 293.15 | 40 | 0.8532 | 3.853 |
| 293.15 | 60 | 0.8626 | 4.838 |
| 293.15 | 80 | 0.8710 | 6.002 |
| 293.15 | 100 | 0.8786 | 7.363 |
| 303.15 | 0.1 | 0.8236 | 1.886 |
| 303.15 | 20 | 0.8362 | 2.477 |
| 303.15 | 40 | 0.8471 | 3.110 |
| 303.15 | 60 | 0.8569 | 3.866 |
| 303.15 | 80 | 0.8656 | 4.752 |
| 303.15 | 100 | 0.8736 | 5.780 |
| 313.15 | 0.1 | 0.8165 | 1.574 |
| 313.15 | 20 | 0.8298 | 2.051 |
| 313.15 | 40 | 0.8411 | 2.578 |
| 313.15 | 60 | 0.8512 | 3.192 |
| 313.15 | 80 | 0.8602 | 3.814 |
| 313.15 | 100 | 0.8684 | 4.589 |
| 323.15 | 0.1 | 0.8093 | 1.332 |
| 323.15 | 20 | 0.8232 | 1.747 |
| 323.15 | 40 | 0.8351 | 2.165 |
| 323.15 | 60 | 0.8456 | 2.648 |
| 323.15 | 80 | 0.8548 | 3.198 |
| 323.15 | 100 | 0.8632 | 3.813 |
| 333.15 | 0.1 | 0.8021 | 1.149 |
| 333.15 | 20 | 0.8168 | 1.490 |
| 333.15 | 40 | 0.8292 | 1.836 |
| 333.15 | 60 | 0.8399 | 2.238 |
| 333.15 | 80 | 0.8495 | 2.696 |
| 333.15 | 100 | 0.8580 | 3.211 |
| 343.15 | 0.1 | 0.7949 | 1.002 |
| 343.15 | 20 | 0.8103 | 1.295 |
| 343.15 | 40 | 0.8232 | 1.583 |
| 343.15 | 60 | 0.8344 | 1.916 |
| 343.15 | 80 | 0.8441 | 2.294 |
| 343.15 | 100 | 0.8528 | 2.717 |
| 353.15 | 0.1 | 0.7878 | 0.884 |
| 353.15 | 20 | 0.8039 | 1.142 |
| 353.15 | 40 | 0.8173 | 1.382 |
| 353.15 | 60 | 0.8289 | 1.659 |
| 353.15 | 80 | 0.8391 | 1.972 |
| 353.15 | 100 | 0.8482 | 2.323 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.375$, $x_t = 0.500$, $x_h = 0.125$ | | |
| 293.15 | 0.1 | 0.8287 | 2.120 |
| 293.15 | 20 | 0.8408 | 2.717 |
| 293.15 | 40 | 0.8512 | 3.406 |
| 293.15 | 60 | 0.8606 | 4.217 |
| 293.15 | 80 | 0.8691 | 5.158 |
| 293.15 | 100 | 0.8769 | 6.263 |
| 303.15 | 0.1 | 0.8215 | 1.741 |
| 303.15 | 20 | 0.8340 | 2.212 |
| 303.15 | 40 | 0.8451 | 2.740 |
| 303.15 | 60 | 0.8548 | 3.348 |
| 303.15 | 80 | 0.8636 | 4.035 |
| 303.15 | 100 | 0.8716 | 4.771 |
| 313.15 | 0.1 | 0.8142 | 1.459 |
| 313.15 | 20 | 0.8274 | 1.859 |
| 313.15 | 40 | 0.8388 | 2.287 |
| 313.15 | 60 | 0.8489 | 2.768 |
| 313.15 | 80 | 0.8580 | 3.297 |
| 313.15 | 100 | 0.8662 | 3.856 |
| 323.15 | 0.1 | 0.8068 | 1.243 |
| 323.15 | 20 | 0.8207 | 1.578 |
| 323.15 | 40 | 0.8326 | 1.926 |
| 323.15 | 60 | 0.8431 | 2.325 |
| 323.15 | 80 | 0.8525 | 2.772 |
| 323.15 | 100 | 0.8609 | 3.271 |
| 333.15 | 0.1 | 0.7996 | 1.078 |
| 333.15 | 20 | 0.8141 | 1.374 |
| 333.15 | 40 | 0.8266 | 1.676 |
| 333.15 | 60 | 0.8374 | 2.024 |
| 333.15 | 80 | 0.8470 | 2.416 |
| 333.15 | 100 | 0.8557 | 2.867 |
| 343.15 | 0.1 | 0.7924 | 0.947 |
| 343.15 | 20 | 0.8076 | 1.197 |
| 343.15 | 40 | 0.8204 | 1.454 |
| 343.15 | 60 | 0.8318 | 1.746 |
| 343.15 | 80 | 0.8418 | 2.071 |
| 343.15 | 100 | 0.8509 | 2.437 |
| 353.15 | 0.1 | 0.7849 | 0.834 |
| 353.15 | 20 | 0.8011 | 1.040 |
| 353.15 | 40 | 0.8145 | 1.268 |
| 353.15 | 60 | 0.8262 | 1.517 |
| 353.15 | 80 | 0.8363 | 1.784 |
| 353.15 | 100 | 0.8454 | 2.061 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ (mPa·s) |
|---|---|---|---|
| $x_m = 0.500$, $x_t = 0.125$, $x_h = 0.375$ | | | |
| 293.15 | 0.1 | 0.8602 | 2.677 |
| 293.15 | 20 | 0.8719 | 3.539 |
| 293.15 | 40 | 0.8823 | 4.577 |
| 293.15 | 60 | 0.8915 | 5.890 |
| 293.15 | 80 | 0.8999 | 7.526 |
| 293.15 | 100 | 0.9075 | 9.590 |
| 303.15 | 0.1 | 0.8531 | 2.161 |
| 303.15 | 20 | 0.8654 | 2.840 |
| 303.15 | 40 | 0.8761 | 3.637 |
| 303.15 | 60 | 0.8857 | 4.618 |
| 303.15 | 80 | 0.8943 | 5.807 |
| 303.15 | 100 | 0.9022 | 7.187 |
| 313.15 | 0.1 | 0.8460 | 1.786 |
| 313.15 | 20 | 0.8589 | 2.333 |
| 313.15 | 40 | 0.8701 | 2.949 |
| 313.15 | 60 | 0.8800 | 3.709 |
| 313.15 | 80 | 0.8890 | 4.633 |
| 313.15 | 100 | 0.8971 | 5.719 |
| 323.15 | 0.1 | 0.8389 | 1.510 |
| 323.15 | 20 | 0.8525 | 1.969 |
| 323.15 | 40 | 0.8641 | 2.474 |
| 323.15 | 60 | 0.8744 | 3.077 |
| 323.15 | 80 | 0.8836 | 3.786 |
| 323.15 | 100 | 0.8920 | 4.616 |
| 333.15 | 0.1 | 0.8319 | 1.294 |
| 333.15 | 20 | 0.8461 | 1.673 |
| 333.15 | 40 | 0.8582 | 2.081 |
| 333.15 | 60 | 0.8688 | 2.561 |
| 333.15 | 80 | 0.8783 | 3.115 |
| 333.15 | 100 | 0.8868 | 3.767 |
| 343.15 | 0.1 | 0.8247 | 1.120 |
| 343.15 | 20 | 0.8397 | 1.449 |
| 343.15 | 40 | 0.8523 | 1.800 |
| 343.15 | 60 | 0.8633 | 2.211 |
| 343.15 | 80 | 0.8731 | 2.681 |
| 343.15 | 100 | 0.8818 | 3.227 |
| 353.15 | 0.1 | 0.8176 | 0.983 |
| 353.15 | 20 | 0.8332 | 1.261 |
| 353.15 | 40 | 0.8464 | 1.553 |
| 353.15 | 60 | 0.8577 | 1.890 |
| 353.15 | 80 | 0.8677 | 2.272 |
| 353.15 | 100 | 0.8767 | 2.694 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.500,\ x_t = 0.250,\ x_h = 0.250$ | | |
| 293.15 | 0.1 | 0.8581 | 2.421 |
| 293.15 | 20 | 0.8699 | 3.200 |
| 293.15 | 40 | 0.8802 | 4.050 |
| 293.15 | 60 | 0.8895 | 5.081 |
| 293.15 | 80 | 0.8978 | 6.310 |
| 293.15 | 100 | 0.9055 | 7.761 |
| 303.15 | 0.1 | 0.8511 | 1.977 |
| 303.15 | 20 | 0.8633 | 2.596 |
| 303.15 | 40 | 0.8740 | 3.261 |
| 303.15 | 60 | 0.8837 | 4.047 |
| 303.15 | 80 | 0.8924 | 4.959 |
| 303.15 | 100 | 0.9004 | 6.005 |
| 313.15 | 0.1 | 0.8439 | 1.639 |
| 313.15 | 20 | 0.8568 | 2.131 |
| 313.15 | 40 | 0.8680 | 2.676 |
| 313.15 | 60 | 0.8780 | 3.303 |
| 313.15 | 80 | 0.8868 | 4.010 |
| 313.15 | 100 | 0.8948 | 4.798 |
| 323.15 | 0.1 | 0.8368 | 1.392 |
| 323.15 | 20 | 0.8503 | 1.820 |
| 323.15 | 40 | 0.8619 | 2.253 |
| 323.15 | 60 | 0.8723 | 2.748 |
| 323.15 | 80 | 0.8815 | 3.305 |
| 323.15 | 100 | 0.8899 | 3.922 |
| 333.15 | 0.1 | 0.8296 | 1.195 |
| 333.15 | 20 | 0.8437 | 1.545 |
| 333.15 | 40 | 0.8559 | 1.897 |
| 333.15 | 60 | 0.8666 | 2.298 |
| 333.15 | 80 | 0.8761 | 2.747 |
| 333.15 | 100 | 0.8847 | 3.243 |
| 343.15 | 0.1 | 0.8223 | 1.043 |
| 343.15 | 20 | 0.8373 | 1.341 |
| 343.15 | 40 | 0.8499 | 1.632 |
| 343.15 | 60 | 0.8609 | 1.967 |
| 343.15 | 80 | 0.8707 | 2.344 |
| 343.15 | 100 | 0.8794 | 2.764 |
| 353.15 | 0.1 | 0.8152 | 0.918 |
| 353.15 | 20 | 0.8307 | 1.185 |
| 353.15 | 40 | 0.8439 | 1.438 |
| 353.15 | 60 | 0.8554 | 1.725 |
| 353.15 | 80 | 0.8655 | 2.045 |
| 353.15 | 100 | 0.8746 | 2.397 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.500,\ x_t = 0.375,\ x_h = 0.125$ | | |
| 293.15 | 0.1 | 0.8565 | 2.222 |
| 293.15 | 20 | 0.8682 | 2.838 |
| 293.15 | 40 | 0.8786 | 3.538 |
| 293.15 | 60 | 0.8879 | 4.382 |
| 293.15 | 80 | 0.8963 | 5.382 |
| 293.15 | 100 | 0.9041 | 6.584 |
| 303.15 | 0.1 | 0.8493 | 1.818 |
| 303.15 | 20 | 0.8616 | 2.285 |
| 303.15 | 40 | 0.8724 | 2.826 |
| 303.15 | 60 | 0.8821 | 3.456 |
| 303.15 | 80 | 0.8908 | 4.176 |
| 303.15 | 100 | 0.8989 | 4.958 |
| 313.15 | 0.1 | 0.8421 | 1.519 |
| 313.15 | 20 | 0.8548 | 1.889 |
| 313.15 | 40 | 0.8660 | 2.344 |
| 313.15 | 60 | 0.8760 | 2.864 |
| 313.15 | 80 | 0.8850 | 3.423 |
| 313.15 | 100 | 0.8933 | 3.988 |
| 323.15 | 0.1 | 0.8347 | 1.293 |
| 323.15 | 20 | 0.8481 | 1.626 |
| 323.15 | 40 | 0.8598 | 1.989 |
| 323.15 | 60 | 0.8702 | 2.398 |
| 323.15 | 80 | 0.8794 | 2.850 |
| 323.15 | 100 | 0.8879 | 3.347 |
| 333.15 | 0.1 | 0.8273 | 1.115 |
| 333.15 | 20 | 0.8415 | 1.415 |
| 333.15 | 40 | 0.8536 | 1.721 |
| 333.15 | 60 | 0.8643 | 2.068 |
| 333.15 | 80 | 0.8739 | 2.455 |
| 333.15 | 100 | 0.8826 | 2.895 |
| 343.15 | 0.1 | 0.8200 | 0.979 |
| 343.15 | 20 | 0.8348 | 1.224 |
| 343.15 | 40 | 0.8474 | 1.480 |
| 343.15 | 60 | 0.8585 | 1.771 |
| 343.15 | 80 | 0.8683 | 2.095 |
| 343.15 | 100 | 0.8772 | 2.460 |
| 353.15 | 0.1 | 0.8125 | 0.865 |
| 353.15 | 20 | 0.8283 | 1.074 |
| 353.15 | 40 | 0.8414 | 1.304 |
| 353.15 | 60 | 0.8528 | 1.559 |
| 353.15 | 80 | 0.8629 | 1.836 |
| 353.15 | 100 | 0.8720 | 2.127 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ (mPa $\cdot$ s) |
|---|---|---|---|
| $x_m = 0.625,\ x_t = 0.125,\ x_h = 0.250$ | | | |
| 293.15 | 0.1 | 0.8873 | 2.615 |
| 293.15 | 20 | 0.8986 | 3.392 |
| 293.15 | 40 | 0.9088 | 4.301 |
| 293.15 | 60 | 0.9178 | 5.438 |
| 293.15 | 80 | 0.9261 | 6.839 |
| 293.15 | 100 | 0.9337 | 8.588 |
| 303.15 | 0.1 | 0.8801 | 2.113 |
| 303.15 | 20 | 0.8920 | 2.724 |
| 303.15 | 40 | 0.9024 | 3.434 |
| 303.15 | 60 | 0.9119 | 4.285 |
| 303.15 | 80 | 0.9205 | 5.286 |
| 303.15 | 100 | 0.9284 | 6.410 |
| 313.15 | 0.1 | 0.8729 | 1.751 |
| 313.15 | 20 | 0.8854 | 2.247 |
| 313.15 | 40 | 0.8963 | 2.794 |
| 313.15 | 60 | 0.9061 | 3.449 |
| 313.15 | 80 | 0.9149 | 4.219 |
| 313.15 | 100 | 0.9230 | 5.095 |
| 323.15 | 0.1 | 0.8657 | 1.482 |
| 323.15 | 20 | 0.8788 | 1.890 |
| 323.15 | 40 | 0.8902 | 2.331 |
| 323.15 | 60 | 0.9003 | 2.848 |
| 323.15 | 80 | 0.9094 | 3.446 |
| 323.15 | 100 | 0.9177 | 4.134 |
| 333.15 | 0.1 | 0.8585 | 1.268 |
| 333.15 | 20 | 0.8723 | 1.622 |
| 333.15 | 40 | 0.8841 | 1.990 |
| 333.15 | 60 | 0.8945 | 2.417 |
| 333.15 | 80 | 0.9039 | 2.903 |
| 333.15 | 100 | 0.9123 | 3.466 |
| 343.15 | 0.1 | 0.8516 | 1.100 |
| 343.15 | 20 | 0.8659 | 1.389 |
| 343.15 | 40 | 0.8783 | 1.693 |
| 343.15 | 60 | 0.8891 | 2.042 |
| 343.15 | 80 | 0.8988 | 2.435 |
| 343.15 | 100 | 0.9076 | 2.885 |
| 353.15 | 0.1 | 0.8443 | 0.967 |
| 353.15 | 20 | 0.8595 | 1.216 |
| 353.15 | 40 | 0.8723 | 1.483 |
| 353.15 | 60 | 0.8835 | 1.775 |
| 353.15 | 80 | 0.8934 | 2.088 |
| 353.15 | 100 | 0.9024 | 2.412 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ ($mPa \cdot s$) |
|---|---|---|---|
| | $x_m = 0.625$, $x_t = 0.250$, $x_h = 0.125$ | | |
| 293.15 | 0.1 | 0.8862 | 2.381 |
| 293.15 | 20 | 0.8976 | 3.027 |
| 293.15 | 40 | 0.9077 | 3.766 |
| 293.15 | 60 | 0.9168 | 4.638 |
| 293.15 | 80 | 0.9251 | 5.647 |
| 293.15 | 100 | 0.9328 | 6.971 |
| 303.15 | 0.1 | 0.8789 | 1.938 |
| 303.15 | 20 | 0.8908 | 2.440 |
| 303.15 | 40 | 0.9012 | 3.002 |
| 303.15 | 60 | 0.9107 | 3.668 |
| 303.15 | 80 | 0.9193 | 4.442 |
| 303.15 | 100 | 0.9272 | 5.298 |
| 313.15 | 0.1 | 0.8716 | 1.646 |
| 313.15 | 20 | 0.8841 | 2.029 |
| 313.15 | 40 | 0.8950 | 2.485 |
| 313.15 | 60 | 0.9047 | 3.016 |
| 313.15 | 80 | 0.9136 | 3.621 |
| 313.15 | 100 | 0.9216 | 4.284 |
| 323.15 | 0.1 | 0.8643 | 1.374 |
| 323.15 | 20 | 0.8774 | 1.712 |
| 323.15 | 40 | 0.8887 | 2.082 |
| 323.15 | 60 | 0.8989 | 2.512 |
| 323.15 | 80 | 0.9080 | 3.005 |
| 323.15 | 100 | 0.9164 | 3.566 |
| 333.15 | 0.1 | 0.8569 | 1.178 |
| 333.15 | 20 | 0.8706 | 1.466 |
| 333.15 | 40 | 0.8825 | 1.774 |
| 333.15 | 60 | 0.8930 | 2.131 |
| 333.15 | 80 | 0.9023 | 2.537 |
| 333.15 | 100 | 0.9108 | 3.008 |
| 343.15 | 0.1 | 0.8496 | 1.029 |
| 343.15 | 20 | 0.8640 | 1.256 |
| 343.15 | 40 | 0.8763 | 1.509 |
| 343.15 | 60 | 0.8872 | 1.799 |
| 343.15 | 80 | 0.8968 | 2.123 |
| 343.15 | 100 | 0.9056 | 2.490 |
| 353.15 | 0.1 | 0.8422 | 0.907 |
| 353.15 | 20 | 0.8573 | 1.109 |
| 353.15 | 40 | 0.8702 | 1.339 |
| 353.15 | 60 | 0.8814 | 1.594 |
| 353.15 | 80 | 0.8915 | 1.871 |
| 353.15 | 100 | 0.9005 | 2.161 |

**Table I.** *(Continued)*

| $T$ (K) | $P$ (MPa) | $\rho$ ($g \cdot cm^{-3}$) | $\eta$ (mPa · s) |
|---|---|---|---|
| | $x_m = 0.750,\ x_t = 0.125,\ x_h = 0.125$ | | |
| 293.15 | 0.1 | 0.9222 | 2.624 |
| 293.15 | 20 | 0.9332 | 3.321 |
| 293.15 | 40 | 0.9431 | 4.127 |
| 293.15 | 60 | 0.9520 | 5.108 |
| 293.15 | 80 | 0.9601 | 6.283 |
| 293.15 | 100 | 0.9677 | 7.774 |
| 303.15 | 0.1 | 0.9149 | 2.135 |
| 303.15 | 20 | 0.9264 | 2.663 |
| 303.15 | 40 | 0.9366 | 3.254 |
| 303.15 | 60 | 0.9459 | 3.980 |
| 303.15 | 80 | 0.9543 | 4.855 |
| 303.15 | 100 | 0.9621 | 5.899 |
| 313.15 | 0.1 | 0.9076 | 1.765 |
| 313.15 | 20 | 0.9196 | 2.192 |
| 313.15 | 40 | 0.9302 | 2.667 |
| 313.15 | 60 | 0.9398 | 3.228 |
| 313.15 | 80 | 0.9485 | 3.876 |
| 313.15 | 100 | 0.9564 | 4.618 |
| 323.15 | 0.1 | 0.9003 | 1.490 |
| 323.15 | 20 | 0.9128 | 1.864 |
| 323.15 | 40 | 0.9239 | 2.248 |
| 323.15 | 60 | 0.9339 | 2.700 |
| 323.15 | 80 | 0.9428 | 3.186 |
| 323.15 | 100 | 0.9511 | 3.768 |
| 333.15 | 0.1 | 0.8930 | 1.273 |
| 333.15 | 20 | 0.9061 | 1.569 |
| 333.15 | 40 | 0.9176 | 1.901 |
| 333.15 | 60 | 0.9279 | 2.254 |
| 333.15 | 80 | 0.9371 | 2.705 |
| 333.15 | 100 | 0.9456 | 3.200 |
| 343.15 | 0.1 | 0.8856 | 1.106 |
| 343.15 | 20 | 0.8994 | 1.383 |
| 343.15 | 40 | 0.9113 | 1.668 |
| 343.15 | 60 | 0.9220 | 1.971 |
| 343.15 | 80 | 0.9315 | 2.284 |
| 343.15 | 100 | 0.9402 | 2.629 |
| 353.15 | 0.1 | 0.8782 | 0.965 |
| 353.15 | 20 | 0.8926 | 1.220 |
| 353.15 | 40 | 0.9051 | 1.447 |
| 353.15 | 60 | 0.9161 | 1.696 |
| 353.15 | 80 | 0.9259 | 1.964 |
| 353.15 | 100 | 0.9349 | 2.248 |

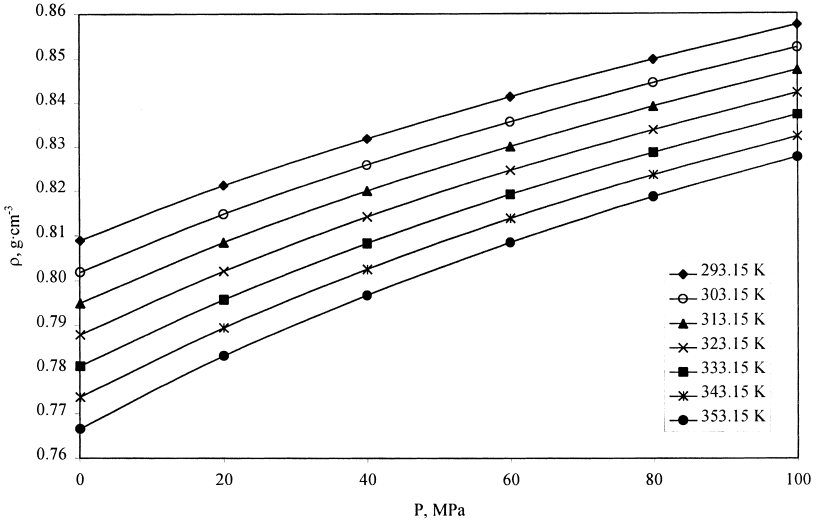


**Fig. 2.** Density $\rho$ versus pressure $P$ at various temperatures $T$ for the composition $x_m = 0.250$, $x_t = 0.375$, $x_h = 0.375$.

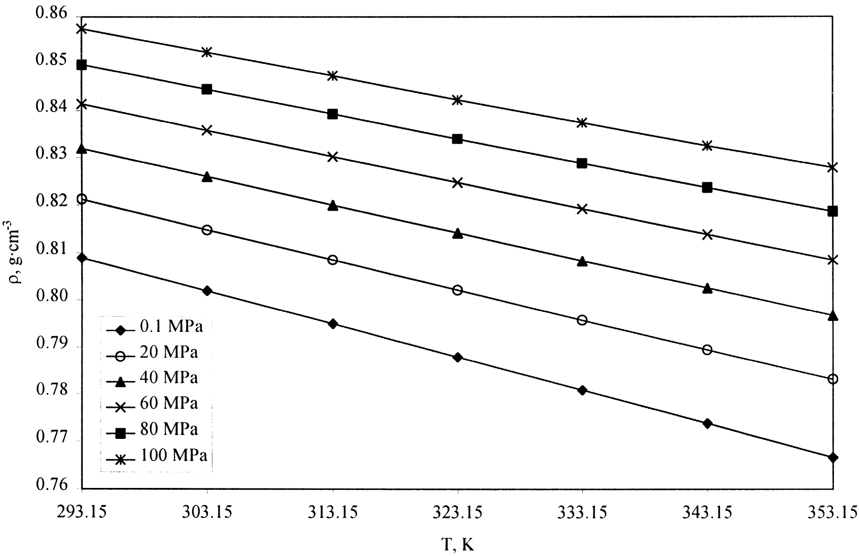


**Fig. 3.** Density $\rho$ versus temperature $T$ at various pressures $P$ for the composition $x_m = 0.250$, $x_t = 0.375$, $x_h = 0.375$.

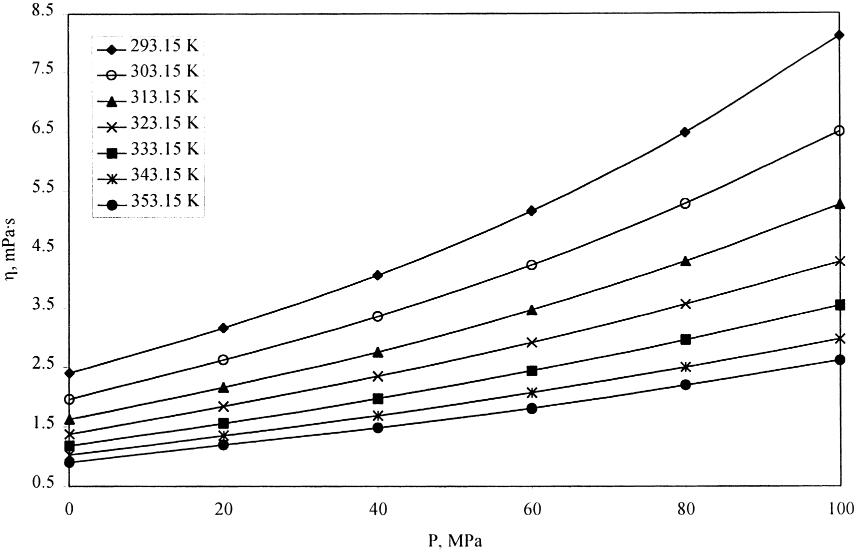


**Fig. 4.** Dynamic viscosity $\eta$ versus pressure $P$ at various temperatures $T$ for the composition $x_m = 0.250$, $x_t = 0.375$, $x_h = 0.375$.

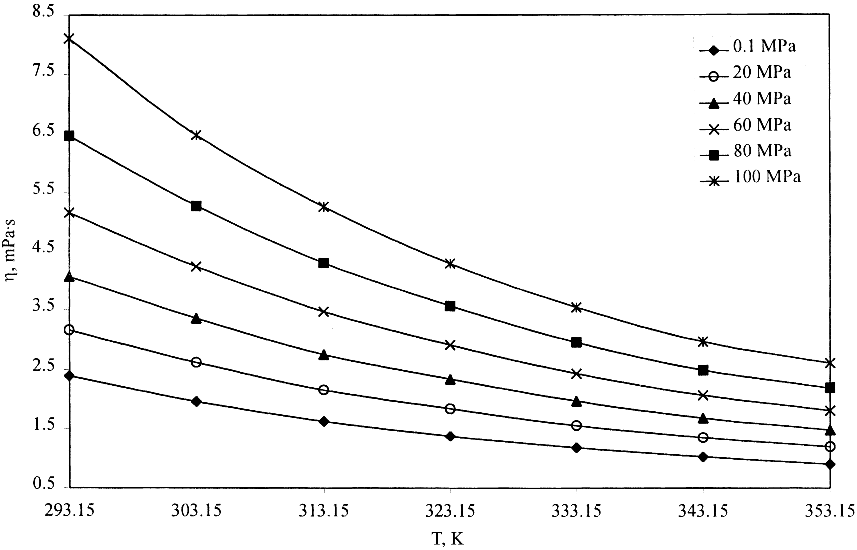


**Fig. 5.** Dynamic viscosity $\eta$ versus temperature $T$ at various pressures $P$ for the composition $x_m = 0.250$, $x_t = 0.375$, $x_h = 0.375$.

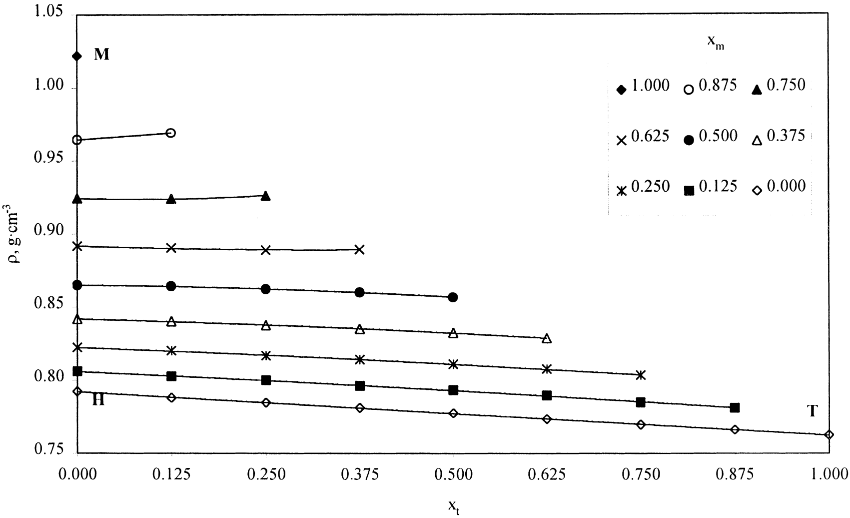


**Fig. 6.** Density $\rho$ versus $x_t$ ($n$-tridecane content) for various $x_m$ (1-methylnaphthalene content) at $P = 40$ MPa and $T = 323.15$ K.

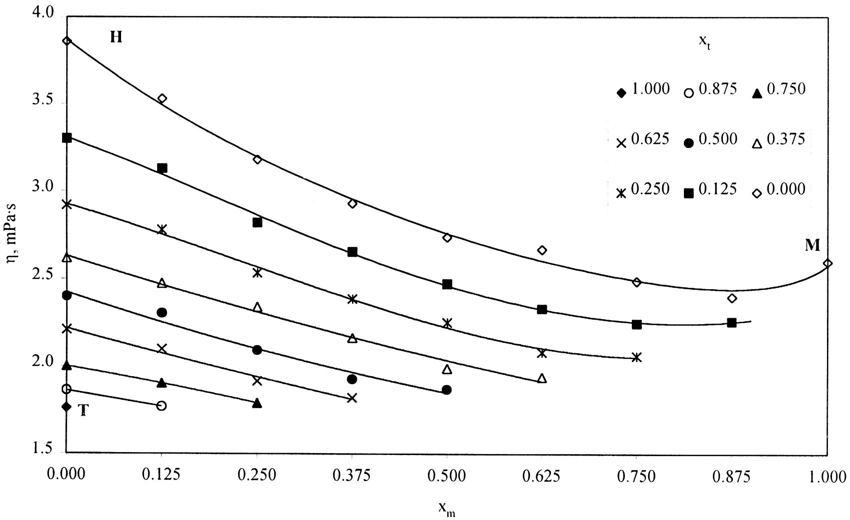


**Fig. 7.** Dynamic viscosity $\eta$ versus $x_m$ (1-methylnaphthalene content) for various $x_t$ ($n$-tridecane content) at $P = 40$ MPa and $T = 323.15$ K.

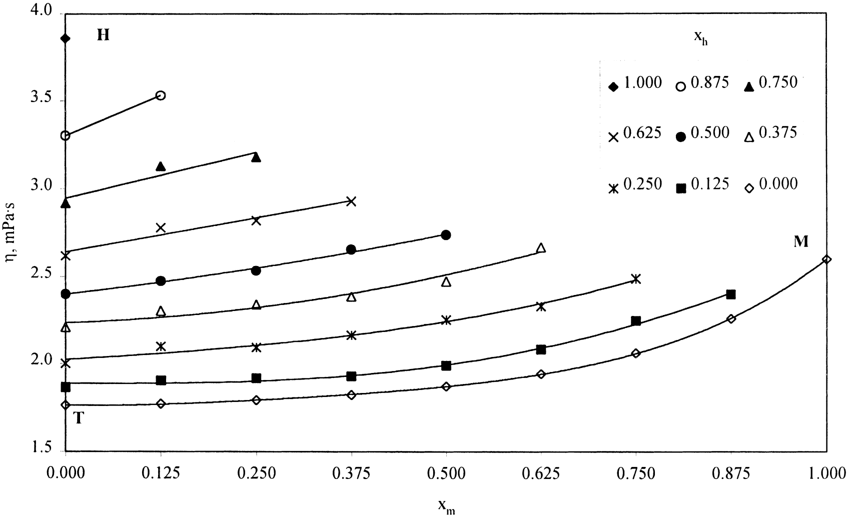


**Fig. 8.** Dynamic viscosity $\eta$ versus $x_m$ (1-methylnaphthalene content) for various $x_h$ (2,2,4,4,6,8,8-heptamethylnonane content) at $P = 40$ MPa and $T = 323.15$ K.

Figure 7 shows the variations of viscosity with $x_m$ for constant $x_t$ at $P = 40$ MPa and $T = 323.15$ K, and Fig. 8 shows another plot as it corresponds to $\eta$ as a function of $x_m$ for constant $x_h$ at $P = 40$ MPa and $T = 323.15$ K. Figures 9 and 10 present the viscosity surface and density surface in a ternary representation for $P = 40$ MPa and $T = 323.15$ K. It should be noted that near the 1-methylnaphthalene + 2,2,4,4,6,8,8-heptamethylnonane side at a given $P, T$, the curves reveal a nonmonotonic behavior with respect to the composition which may be the effect of repulsive interactions (as for the binary; see Ref. 6). The minimum disappears when the amount of $x_t$ increases. At a given composition Table I shows that it also disappears when the pressure increases. Furthermore, by keeping the concentration of 2,2,4,4,6,8,8-heptamethylnonane constant for $x_h \leqslant 0.375$ and plotting the viscosity as a function of the concentration of 1-methylnaphthalene for $0 < x_m < 0.500$, a very slow increase in the viscosity is observed: see Fig. 8. Overall, for the viscosity of this ternary system, there is a negative deviation from an ideal mixture, indicating that the viscosity is probably influenced by "repulsive interactions."

## 4. DISCUSSION

The data obtained for the ternary system in the course of this investigation, combined with those obtained previously [4–6] on the three pure

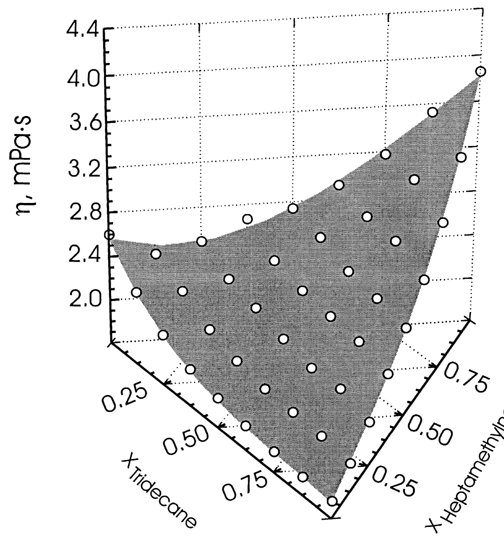


**Fig. 9.** Surface of the dynamic viscosity $\eta(x_m, x_t, x_h)$ in the ternary diagram at $P = 40$ MPa and $T = 323.15$ K.

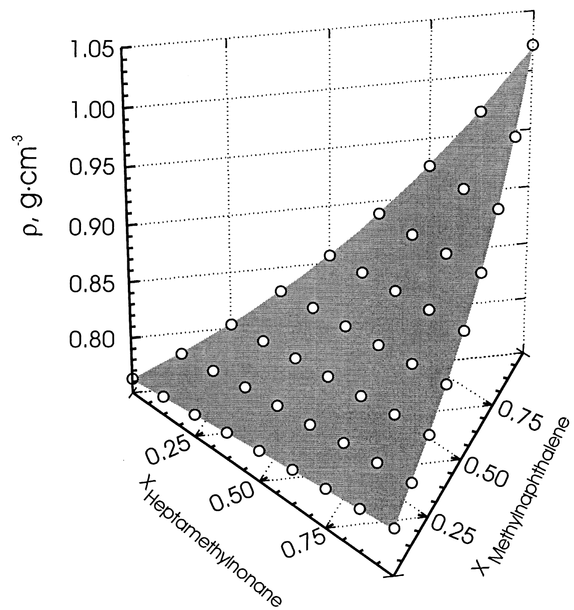


**Fig. 10.** Surface of the density $\rho(x_m, x_t, x_h)$ in the ternary diagram at $P = 40$ MPa and $T = 323.15$ K.

compounds and the three binaries, represent the most comprehensive set of experimental data for the dynamic viscosity (a total of 1890 data points), which can be used to test different representative models incorporating the effects of temperature, pressure, and composition. A preliminary study has been carried out focusing on mixing laws. In the case of the Grunberg and Nissan mixing law [16],

$$\ln(\eta) = x_{\mathrm{m}} \ln(\eta_{\mathrm{m}}) + x_{\mathrm{t}} \ln(\eta_{\mathrm{t}}) + x_{\mathrm{h}} \ln(\eta_{\mathrm{h}}) \tag{2}$$

an absolute average deviation (AAD) of 13.0% and a absolute maximum deviation (MD) of 34.2% are obtained. This model is very simple since no adjustable parameters are required, only the viscosity of the pure compounds and the composition. But with this relationship, the variation of the viscosity with composition becomes monotonic and any interaction between the pure compounds influencing the total viscosity is not taken into account. The Grunberg–Nissan mixing law can be modified by introducing adjustable parameters believed to be representative in some way of the interactions within the system studied. Using the following relationship, in which a corrective term has been added,

$$\ln(\eta) = x_{\mathrm{m}} \ln(\eta_{\mathrm{m}}) + x_{\mathrm{t}} \ln(\eta_{\mathrm{t}}) + x_{\mathrm{h}} \ln(\eta_{\mathrm{h}}) + (x_{\mathrm{m}} x_{\mathrm{t}} + x_{\mathrm{t}} x_{\mathrm{h}} + x_{\mathrm{m}} x_{\mathrm{h}})\, d \tag{3}$$

each binary pair is involved in the same manner. By minimizing the least squares, the adjustable parameter ($d = -0.41484$) in Eq. (3) has been obtained, resulting in an AAD = 2.92% and an MD = 19.0%. Equation (2) can be made less symmetrical by introducing three parameters instead of one:

$$\ln(\eta) = x_{\mathrm{m}} \ln(\eta_{\mathrm{m}}) + x_{\mathrm{t}} \ln(\eta_{\mathrm{t}}) + x_{\mathrm{h}} \ln(\eta_{\mathrm{h}}) + (x_{\mathrm{m}} x_{\mathrm{t}} d_{\mathrm{mt}} + x_{\mathrm{t}} x_{\mathrm{h}} d_{\mathrm{th}} + x_{\mathrm{m}} x_{\mathrm{h}} d_{\mathrm{mh}}) \tag{4}$$

In Eq. (4) $d_{ij}$ is characteristic of the intermolecular interactions between component $i$ and component $j$. The $d_{ij}$ parameter can be evaluated using viscosity data for only the binary $i+j$ system. Using the three parameters $d_{\mathrm{mt}} = -0.58110$, $d_{\mathrm{mh}} = -0.50034$, and $d_{\mathrm{th}} = -0.28387$ evaluated from the binary systems and reported in Ref. 17, an AAD = 2.45% and a MD = 15.7% are obtained for the ternary. The result is satisfactory, in the sense that the AAD is of the same order of magnitude as the experimental error. Furthermore, it can be seen from the values of the $d_{ij}$ parameters that the largest binary interactions are obtained between 1-methylnaphthalene and *n*-tridecane or 2,2,4,4,6,8,8-heptamethylnonane (aromatic hydrocarbon + alkane) (see Figs. 7 and 8), rather than between *n*-tridecane and 2,2,4,4,6,8,8-heptamethylnonane (alkane + alkane).

Another interesting property, which can be obtained from the measured values of the viscosity and density, is the excess activation energy of viscous flow $\Delta G^{E}$, which appears in

$$\ln(\eta V) = x_t \ln(\eta_t V_t) + x_m \ln(\eta_m V_m) + x_h \ln(\eta_h V_h) + \Delta G^{E}/RT \quad (5)$$

where $R$ is the gas constant and $V_i = M_i/\rho_i$ is the molar volume of component $i$. $M_i$ is the molecular weight of component $i$, and for a mixture the molecular weight $M = \sum x_i M_i$. This relation is a modified form of the equation of Katti and Chaudhri [18] and is theoretically justified by Eyring's representation of the dynamic viscosity of a pure fluid [19]. It is important to note here that the quantity $\eta V$ is also obtained from the time-correlation expression for shear viscosity [20]. Thus, the quantities $\eta V$ and $\Delta G^{E}$ have a theoretical background, while the corrective terms in Eqs. (3) and (4) do not. The excess activation energy of viscous flow $\Delta G^{E}$ can be calculated from the results in Table I and in Refs. 4–6. Figure 11 shows the surface $\Delta G^{E}(x_m, x_t, x_h)$ in a ternary representation for $P = 40$ MPa and $T = 323.15$ K. We note that $\Delta G^{E}$ is generally negative and $|\Delta G^{E}|$ increases with pressure. For some authors [21, 22] the fact that the excess activation energy of viscous flow $\Delta G^{E}$ is negative means that the predominant effect in the mixture is the breaking-up of the ordered structure present in the pure liquids. Other authors [23, 24] interpret the negative values of $\Delta G^{E}$ by the fact that the repulsive forces of interaction are the forces which predominate, corresponding to the breaking of bonds within the ordered structure. For the very associative system water + alcohol (see, e.g., Ref. 11) $|\Delta G^{E}|$ can reach 5000 $J \cdot mol^{-1}$, whereas in this work the maximum value of $|\Delta G^{E}|$ is about 600 $J \cdot mol^{-1}$, which corresponds to weak interactions and consequently to a weakly interactive system. The estimation of the excess activation energy of viscous flow is based on Eq. (5), which can be thought to model the viscosity of an ideal mixture by assuming $\Delta G^{E} = 0$ and, therefore, using only the properties of the pure components. In this case an AAD = 8.65% and a MD = 27.7% are obtained. This relationship can also be used to model the viscosity of real mixtures. We tried $\Delta G^{E} = (x_m x_t + x_m x_h + x_t x_h) W$ in which $W$ is an adjustable parameter representative of the interactions. By least-squares minimization, we obtained $W = -788.13$ $J \cdot mol^{-1}$ resulting in an AAD = 2.12% and a MD = 16.8%. We also tried $\Delta G^{E} = (x_m x_t W_{mt} + x_m x_h W_{mh} + x_t x_h W_{th})$, where $W_{ij}$ is characteristic of intermolecular interactions between component $i$ and component $j$ responsible for the excess energy of activation for viscous flow. The $W_{ij}$ parameters can be evaluated using viscosity and density data concerning only the binary $i + j$ system. For each of the three binary $i + j$ systems we found $W_{mt} = -1173.6$ $J \cdot mol^{-1}$, $W_{mh} = -611.55$ $J \cdot mol^{-1}$, and $W_{th} = -726.83$ $J \cdot mol^{-1}$ by

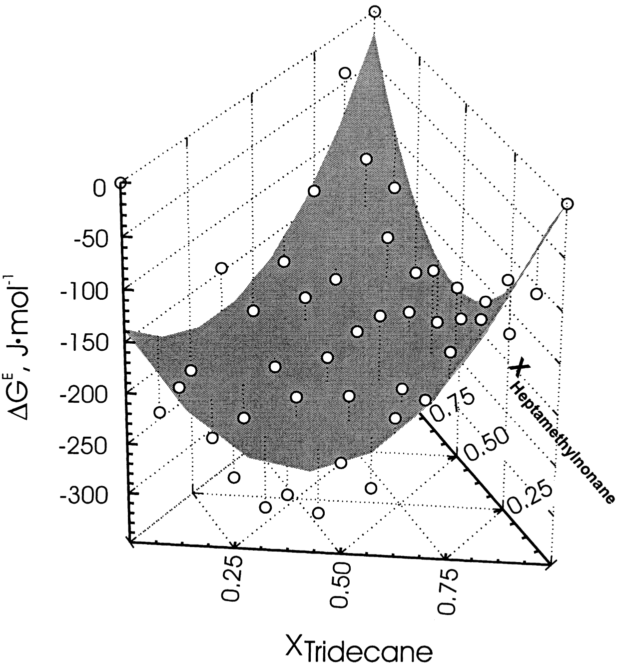


**Fig. 11.** Surface of excess activation energy of viscous flow $\Delta G^E(x_m, x_t, x_h)$ in the ternary diagram at $P = 40$ MPa and $T = 323.15$ K.

least squares. Using these three $W_{ij}$ parameters results in an AAD = 2.12% and a MD = 14.6% for the ternary mixtures. The $W_{ij}$ parameters are related to the excess activation energy of viscous flow and they are all negative, indicating that repulsive forces dominate between component $i$ and component $j$.

## 5. CONCLUSION

The viscosity and density measurements reported in this work together with the viscosity and density measurements for the pure components and binary mixtures [4–6] represent the most comprehensive study for the ternary system 1-methylnaphthalene + $n$-tridecane + 2,2,4,4,6,8,8-heptamethylnonane. This study is a part of a more general study concerning various systems (associative and nonassociative mixtures, various binary mixtures with different compounds, ternary systems, and even systems with more than three components). We intend to undertake a modeling study of these measurements concerning three recently proposed viscosity models:

the hard-sphere viscosity scheme [25, 26], a free-volume viscosity model [27–29], and the viscosity model based on the friction theory [30, 31]. Each of these three models has a strong physical background and they are conceptually very different.

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