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Viscosity and Density of the Ternary Mixture Heptane + Methylcyclohexane + 1-Methylnaphthalene

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The dynamic viscosity η and the density ρ of the ternary mixture heptane (mole fraction x_1) + methylcyclohexane (mole fraction x_2) + 1-methylnaphthalene (mole fraction x_3) were measured as a function of temperature T (303.15, 323.15, and 343.15 K) and pressure P ($\leqslant 100$ MPa). The experimental results correspond to 378 values of η and ρ. With reference to the 54 values previously published on pure substances and 378 values for the three associated binaries, the system is globally described by 810 experimental values for various values of P, T, and composition.

KEY WORDS: Density; heptane; hydrocarbons; methylcyclohexane; 1-methylnaphthalene; ternary system; viscosity.

1. INTRODUCTION

As we indicated in a previous article [1], while there is a substantial volume of data describing variations of the dynamic viscosity η of hydrocarbons versus temperature at atmospheric pressure, studies of variations versus pressure are less common, especially for mixtures, particularly for those able to model complex fluids such as petroleum fluids. As for mixtures, while data concerning binaries are now available in which variations as a function of composition, pressure, and temperature are well described. there are practically no systematic studies concerning ternary mixtures.

In a previous paper [1], based on remarks made by Le Roy [2] on the composition of a pure synthetic fluid able to represent a petroleum

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fluid, we selected the heptane + methylcyclohexane + 1-methylnaphthalene mixture. The dynamic viscosity η and density ρ of the three pure substances and the three associated binaries (involving 54 values for the pure substances and 378 for the binaries) have already been published previously [1]. It was then necessary to study the ternary proper under the same pressure and temperature conditions as for the pure substances and the binaries, at enough compositions to cover completely the representative ternary diagram.

2. EXPERIMENTAL PROCEDURE

2.1. Apparatus

The dynamic viscosity η and the density ρ of the samples were determined with the aid of a falling-body viscometer and an Anton-Paar DMA 45 resonance densitometer equipped with an additional DMA 512 cell, the technical details of which are supplied in Ref. 3. Values of ρ between 0.1 and 40 MPa were then extrapolated to 100 MPa with the aid of a procedure described in the same article [3]. It should be indicated that the error in T is estimated as ± 0.5 K for the measurement of η and ± 0.05 K for that of ρ. The error in P is ± 0.05 MPa for ρ and ± 0.1 MPa for η (except at $P = 0.1$ MPa). The error in ρ is $\pm 0.5\ \mathrm{kg \cdot m^{-3}}$, while the relative uncertainty of η is of the order of 2%. As discussed earlier [3, 4], this error is comparable to those obtained by other authors with a similar experimental apparatus. As an example, in the case of pure heptane, Fig. 1 shows η versus pressure at $T = 323.15$ K for our measurements [1] and those of

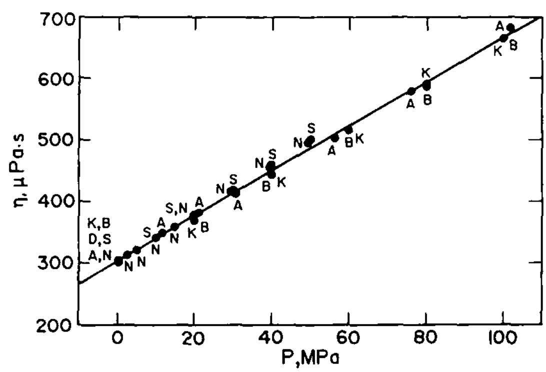

Fig. 1. Dynamic viscosity η of heptane versus pressure at $T = 323.15$ K (——: $\eta = a + bP + cP^2$). A, Ref. 5; K, Ref. 6; N, Ref. 7; S, Ref. 8; D, Ref. 9; B, the present paper.

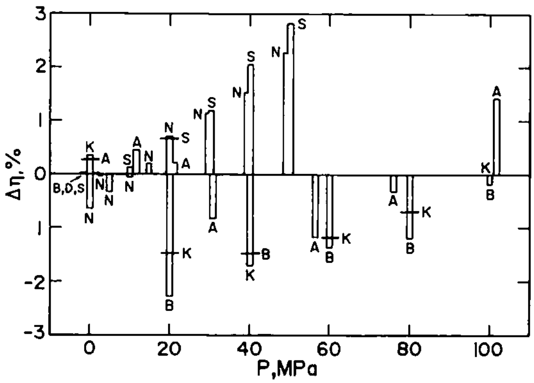

Fig. 2. Deviations $\Delta\eta$ of the literature data for the viscosity of heptane from the correlation of $\eta = a + bP + cP^2$ ($T = 323.15$ K). A, Ref. 5; K, Ref. 6; N, Ref. 7; S, Ref. 8; D, Ref. 9; B, the present paper.

different authors [5–9] and Fig. 2 shows values for the deviation $\Delta\eta = 100(\eta_{exp} - \eta_{cal})/\eta_{exp}$, with $\eta_{cal} = a + bP + cP^2$ ($a = 302.6198$, $b = 3.7064$, $c = -0.00079181$; P in MPa and η_{cal} in μPa · s).

2.2. Characteristics of the Samples

The substances used were commercially available chemicals with the following degrees of purity: heptane (Aldrich: purity, >99%; molecular weight $M = 100.205$ g · mol^{-1}), methylcyclohexane (Aldrich: purity, >99%; $M = 98.189$ g · mol^{-1}), 1-methylnaphthalene (Aldrich: purity, >98%; $M = 142.201$ g · mol^{-1}). Note that at atmospheric pressure $P = 0.1$ MPa and at $T = 303.15$ K the viscosity of heptane is 370 μPa · s, that of methylcyclohexane 638.7 μPa · s, and that of 1-methylnaphthalene 2617 μPa · s. We verified that the viscosity $\eta(P, T)$ of the last of these three components is always much higher than those of the other two. The mixtures were prepared by weight at ambient pressure and temperature so as to obtain the mole fractions x_1, x_2, and x_3 ($x_1 + x_2 + x_3 = 1$ and subscript 1 for heptane, 2 for methylcyclohexane, and 3 for 1-methylnaphthalene) corresponding to the 21 points of the ternary diagram shown in Fig. 3. We recall that the systems associated with the three summits (pure substances) and the three sides (binaries) have already been studied [1].

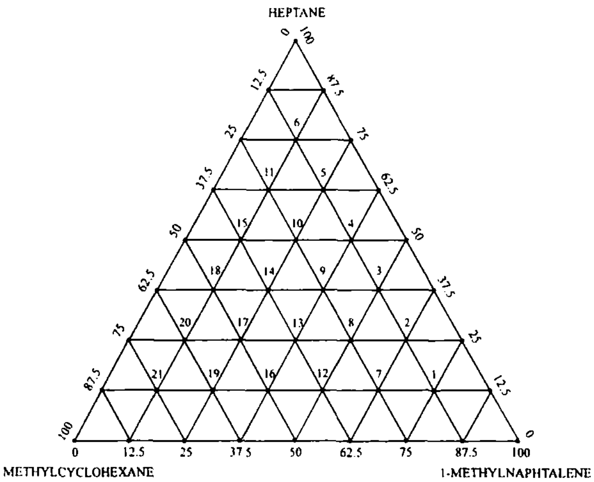

Fig. 3. Points representative of the three pure substances, the three binaries, and the ternary, in the ternary diagram (as mole fraction).

3. RESULTS

Measurements of dynamic viscosity η were made at 303.15, 323.15, and 343.15 K and at 0.1, 20, 40, 60, 80, and 100 MPa. As we have already indicated, the values of density ρ were extrapolated between 40 and 100 MPa. Twenty-one compositions were considered; we thus obtained 378 values for η and ρ. The values are presented in Table I as a function of pressure P, temperature T, and composition ($x_1, x_2, x_3 \neq 0$ and 1). Figures 4 and 5 represent variations of ρ versus P (for different T) and versus T (for different P) in the case corresponding to point 14 on the ternary

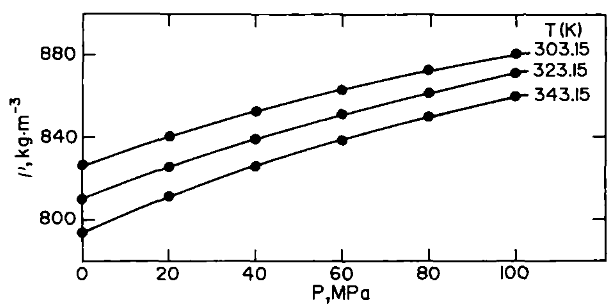

Fig. 4. Density ρ versus pressure P at various temperatures T, for composition $x_1 = 0375$, $x_2 = 0.375$, and $x_3 = 0.25$.

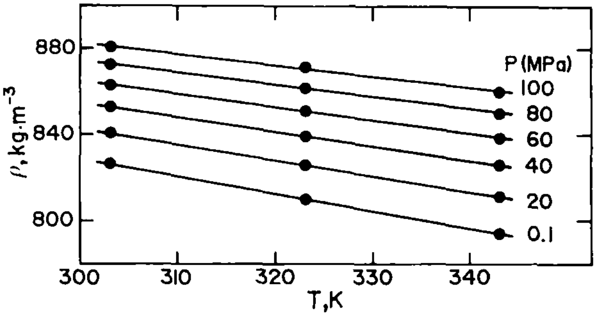

Fig. 5. Density ρ versus temperature T at various pressures P, for composition $x_1 = 0.375$, $x_2 = 0.375$, and $x_3 = 0.25$.

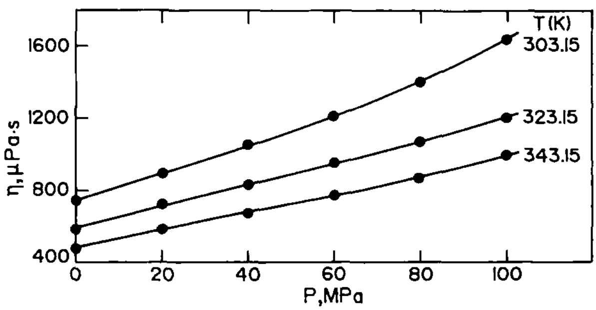

Fig. 6. Dynamic viscosity η versus pressure P at various temperatures T, for composition $x_1 = 0.375$, $x_2 = 0.375$, and $x_3 = 0.25$.

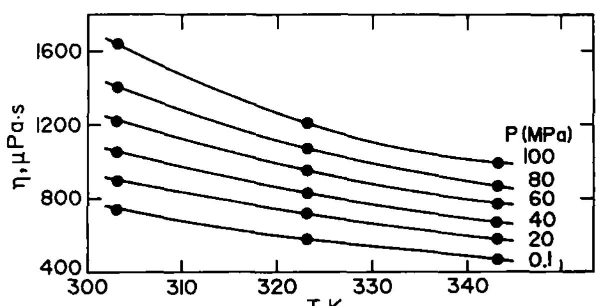

Fig. 7. Dynamic viscosity η versus temperature T at various pressures P, for composition $x_1 = 0.375$, $x_2 = 0.375$, and $x_3 = 0.25$.

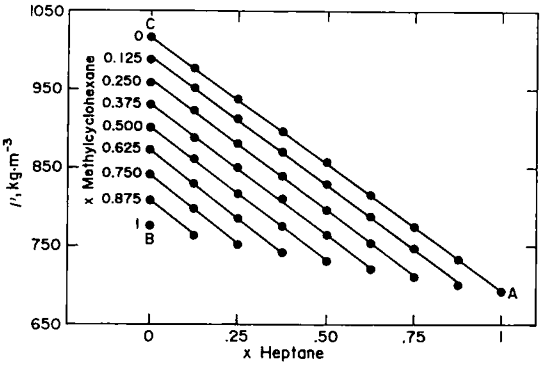

Fig. 8. Density ρ versus x_1 (heptane content) for various x_2 (methylcyclohexane content) at $P = 40$ MPa and $T = 323.15$ K.

diagram in Fig. 3 ($x_1 = 0.375$, $x_2 = 0.375$, $x_3 = 0.25$). Figures 6 and 7 show the dynamic viscosity η under the same conditions. Table I and the figures present a general pattern consistent with previous observations made by other authors and by us on pure hydrocarbons or binary mixtures of hydrocarbons. The pressure coefficient of the viscosity variation $(\partial\eta/\partial P)_T$ is positive for all the compositions and the shape of $\eta(P)$ variations shows a sharp increase, while on the contrary, the temperature variation coefficient $(\partial\eta/\partial T)_P$ is always negative. The group of isotherm and isobar curves is regular. This is also true for density, but in the case of the isotherm 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 $1/\rho$; this logarithmic form is the one used for

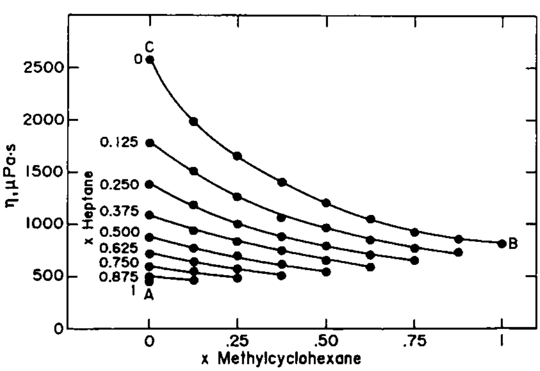

Fig. 9. Dynamic viscosity η versus x_2 (methylcyclohexane content) for various x_1 (heptane content) at $P = 40$ MPa and $T = 323.15$ K.

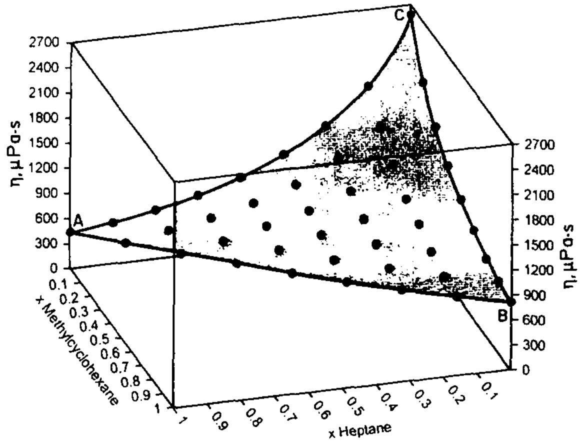

Fig. 10. Dynamic viscosity η versus x_1 (heptane content) and x_2 (methylcyclohexane content) at $P=40$ MPa and $T=323.15$ K.

extrapolation (see, e.g., Ref. 3). It should be noted that variations of ρ versus T are practically linear, but it should be recalled that in the present investigation the temperature variation is relatively small, the main aim being to observe variations of ρ and η as a function of pressure and the mole fractions of the components.

Figure 8 shows, at $P=40$ MPa and $T=323.15$ K, ρ as a function of x_1 (heptane) for a constant x_2 (methylcyclohexane). Within experimental

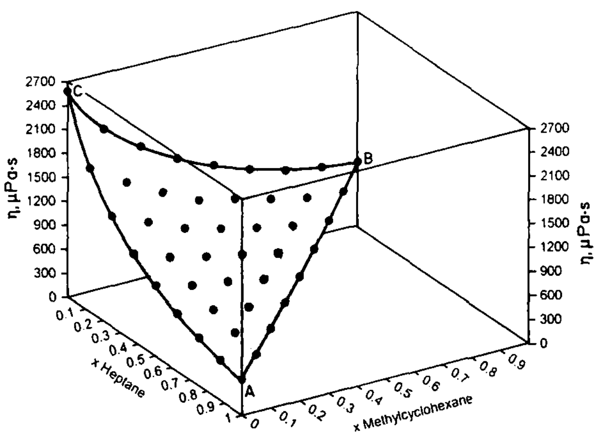

Fig. 11. Dynamic viscosity η versus x_1 (heptane content) and x_2 (methylcyclohexane content) at $P=40$ MPa and $T=323.15$ K (different viewing angle).

accuracy the variations are practically linear, which corresponds to very low excess volumes. Figure 9 shows η as a function of x_2 at constant x_1 at $P = 40$ MPa and $T = 323.15$ K. Points A, B, and C correspond to the pure substances and the sides AB, BC, and AC to the binaries (to complete the figures we used data on the pure substances and binaries given in Ref. 1). Figures 10 and 11 represent the surface $\eta(x_1, x_2)$ at $P = 40$ MPa and $T = 323.15$ K, seen from two angles. Finally, Fig. 12 represents the surface $\rho(x_1, x_2)$ under the same pressure and temperature conditions. We note that the plane $\rho = a + bx_1 + cx_2$ represents the experimental values of ρ with a maximum error of 0.6%.

The data obtained on the ternary mixture in the course of this investigation, combined with those obtained previously [1] on the three pure substances and the three associated binaries, represent a total of 810 experimental 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 with very different models. In the case of the Grunberg and Nissan mixing rule [10],

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_3 \ln \eta_3 \quad (1)$$

we obtained an absolute average deviation (AAD) of 16.2% and a maximum deviation (MD) of 39.11%. The method is very simple since there are no adjustable parameters and only viscosity data for the pure substances are required. The above relationship can be modified by introducing adjustable parameters believed to be in some way representative of the

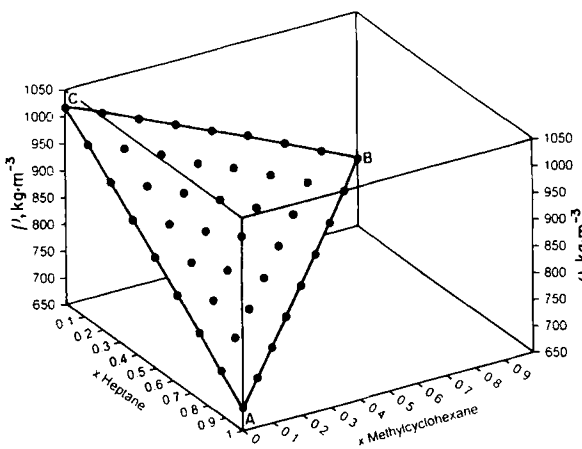

Fig. 12. Density ρ versus x_1 (heptane content) and x_2 (methylcyclohexane content) at $P = 40$ MPa and $T = 323.15$ K.

Table I. Dynamic Viscosity η and Density ρ for the Ternary Mixture[a]

x_1	x_2	x_3	T (K)	P (MPa)	ρ ($kg \cdot m^{-3}$)	η ($\mu Pa \cdot s$)
0.125	0.125	0.75	303.15	0.1	940.7	1500
0.125	0.125	0.75	303.15	20	952.5	1760
0.125	0.125	0.75	303.15	40	963.1	2090
0.125	0.125	0.75	303.15	60	972.8	2470
0.125	0.125	0.75	303.15	80	981.7	2880
0.125	0.125	0.75	303.15	100	989.9	3380
0.125	0.125	0.75	323.15	0.1	927.2	1100
0.125	0.125	0.75	323.15	20	940.2	1300
0.125	0.125	0.75	323.15	40	951.8	1510
0.125	0.125	0.75	323.15	60	962.2	1730
0.125	0.125	0.75	323.15	80	971.7	2000
0.125	0.125	0.75	323.15	100	980.4	2290
0.125	0.125	0.75	343.15	0.1	913.4	869
0.125	0.125	0.75	343.15	20	927.7	966
0.125	0.125	0.75	343.15	40	940.2	1160
0.125	0.125	0.75	343.15	60	951.2	1310
0.125	0.125	0.75	343.15	80	961.2	1490
0.125	0.125	0.75	343.15	100	970.3	1710
0.125	0.25	0.625	303.15	0.1	913.5	1220
0.125	0.25	0.625	303.15	20	925.8	1460
0.125	0.25	0.625	303.15	40	936.6	1700
0.125	0.25	0.625	303.15	60	946.2	2000
0.125	0.25	0.625	303.15	80	954.8	2330
0.125	0.25	0.625	303.15	100	962.7	2770
0.125	0.25	0.625	323.15	0.1	897.6	911
0.125	0.25	0.625	323.15	20	911.2	1100
0.125	0.25	0.625	323.15	40	923	1270
0.125	0.25	0.625	323.15	60	933.6	1460
0.125	0.25	0.625	323.15	80	943.1	1650
0.125	0.25	0.625	323.15	100	951.7	1880
0.125	0.25	0.625	343.15	0.1	881.7	715
0.125	0.25	0.625	343.15	20	896.9	869
0.125	0.25	0.625	343.15	40	909.7	994
0.125	0.25	0.625	343.15	60	920.7	1140
0.125	0.25	0.625	343.15	80	930.5	1280
0.125	0.25	0.625	343.15	100	939.3	1480
0.125	0.375	0.5	303.15	0.1	877.2	991
0.125	0.375	0.5	303.15	20	890	1190
0.125	0.375	0.5	303.15	40	901.4	1390
0.125	0.375	0.5	303.15	60	911.6	1630
0.125	0.375	0.5	303.15	80	921	1900

[a] x_1, mole fraction of heptane; x_2, methylcyclohexane; x_3, 1-methylnaphthalene.

Table I. *(Continued)*

x_1	x_2	x_3	T (K)	P (MPa)	ρ (kg · m^{-3})	η (μPa · s)
0.125	0.375	0.5	303.15	100	929.5	2210
0.125	0.375	0.5	323.15	0.1	860.8	758
0.125	0.375	0.5	323.15	20	875.1	939
0.125	0.375	0.5	323.15	40	887.8	1060
0.125	0.375	0.5	323.15	60	899.1	1230
0.125	0.375	0.5	323.15	80	909.4	1390
0.125	0.375	0.5	323.15	100	918.8	1600
0.125	0.375	0.5	343.15	0.1	844.5	602
0.125	0.375	0.5	343.15	20	860.6	739
0.125	0.375	0.5	343.15	40	874.4	861
0.125	0.375	0.5	343.15	60	886.5	981
0.125	0.375	0.5	343.15	80	897.3	1120
0.125	0.375	0.5	343.5	100	907.1	1250
0.125	0.5	0.375	303.15	0.1	849.1	870
0.125	0.5	0.375	303.15	20	862.8	1050
0.125	0.5	0.375	303.15	40	874.7	1240
0.125	0.5	0.375	303.15	60	885.1	1460
0.125	0.5	0.375	303.15	80	894.5	1670
0.125	0.5	0.375	303.15	100	902.9	1940
0.125	0.5	0.375	323.15	0.1	832.6	672
0.125	0.5	0.375	323.15	20	848	868
0.125	0.5	0.375	323.15	40	861.1	970
0.125	0.5	0.375	323.15	80	872.4	1110
0.125	0.5	0.375	323.15	80	882.5	1260
0.125	0.5	0.375	323.15	100	891.5	1440
0.125	0.5	0.375	343.15	0.1	816.4	543
0.125	0.5	0.375	343.15	20	833.4	663
0.125	0.5	0.375	343.15	40	847.6	790
0.125	0.5	0.375	343.15	60	859.7	911
0.125	0.5	0.375	343.15	80	870.3	1030
0.125	0.5	0.375	343.15	100	879.8	1160
0.125	0.625	0.25	303.15	0.1	816.2	748
0.125	0.625	0.25	303.15	20	830.8	891
0.125	0.625	0.25	303.15	40	843.4	1070
0.125	0.625	0.25	303.15	60	854.5	1250
0.125	0.625	0.25	303.15	80	864.5	1460
0.125	0.625	0.25	303.15	100	873.5	1710
0.125	0.625	0.25	323.15	0.1	799.3	585
0.125	0.625	0.25	323.15	20	815.7	742
0.125	0.625	0.25	323.15	40	829.5	851
0.125	0.625	0.25	323.15	60	841.4	975
0.125	0.625	0.25	323.15	80	851.9	1100

Table I. *(Continued)*

x_1	x_2	x_3	T (K)	P (MPa)	ρ ($kg \cdot m^{-3}$)	η ($\mu Pa \cdot s$)
0.125	0.625	0.25	323.15	100	861.3	1240
0.125	0.625	0.25	343.15	0.1	783	474
0.125	0.625	0.25	343.15	20	801	582
0.125	0.625	0.25	343.15	40	816.1	701
0.125	0.625	0.25	343.15	60	829	809
0.125	0.625	0.25	343.15	80	840.4	913
0.125	0.625	0.25	343.15	100	850.6	1010
0.125	0.75	0.125	303.15	0.1	782.4	684
0.125	0.75	0.125	303.15	20	797.8	796
0.125	0.75	0.125	303.15	40	810.8	956
0.125	0.75	0.125	303.15	60	821.9	1110
0.125	0.75	0.125	303.15	80	831.8	1290
0.125	0.75	0.125	303.15	100	840.6	1490
0.125	0.75	0.125	323.15	0.1	765.3	551
0.125	0.75	0.125	323.15	20	782.8	659
0.125	0.75	0.125	323.15	40	797.2	763
0.125	0.75	0.125	323.15	60	809.5	886
0.125	0.75	0.125	323.15	80	820.2	1010
0.125	0.75	0.125	323.15	100	829.8	1160
0.125	0.75	0.125	343.15	0.1	748.3	455
0.125	0.75	0.125	343.15	20	768	526
0.125	0.75	0.125	343.15	40	783.5	631
0.125	0.75	0.125	343.15	60	796.3	736
0.125	0.75	0.125	343.15	80	807.3	838
0.125	0.75	0.125	343.15	100	817	952
0.25	0.125	0.625	303.15	0.1	898.1	1120
0.25	0.125	0.625	303.15	20	910.3	1320
0.25	0.125	0.625	303.15	40	921.4	1540
0.25	0.125	0.625	303.15	60	931.8	1840
0.25	0.125	0.625	303.15	80	941.4	2100
0.25	0.125	0.625	303.15	100	950.3	2450
0.25	0.125	0.625	323.15	0.1	885.2	848
0.25	0.125	0.625	323.15	20	899.6	1020
0.25	0.125	0.625	323.15	40	912	1190
0.25	0.125	0.625	323.15	60	923	1350
0.25	0.125	0.625	323.15	80	932.9	1540
0.25	0.125	0.625	323.15	100	941.9	1750
0.25	0.125	0.625	343.15	0.1	871.6	671
0.25	0.125	0.625	343.15	20	887.1	793
0.25	0.125	0.625	343.15	40	900.6	920
0.25	0.125	0.625	343.15	60	912.7	1050
0.25	0.125	0.625	343.15	80	923.5	1190

Table I. *(Continued)*

x_1	x_2	x_3	T (K)	P (MPa)	ρ (kg · m^{-3})	η (μPa · s)
0.25	0.125	0.625	343.15	100	933.4	1360
0.25	0.25	0.5	303.15	0.1	869.7	934
0.25	0.25	0.5	303.15	20	882.6	1130
0.25	0.25	0.5	303.15	40	894.2	1340
0.25	0.25	0.5	303.15	60	904.7	1550
0.25	0.25	0.5	303.15	80	914.3	1810
0.25	0.25	0.5	303.15	100	923.2	2090
0.25	0.25	0.5	323.15	0.1	853.4	718
0.25	0.25	0.5	323.15	20	867.8	900
0.25	0.25	0.5	323.15	40	880.6	1000
0.25	0.25	0.5	323.15	60	891.9	1150
0.25	0.25	0.5	323.15	80	902.2	1290
0.25	0.25	0.5	323.15	100	911.6	1450
0.25	0.25	0.5	343.15	0.1	837.1	575
0.25	0.25	0.5	343.15	20	853.2	701
0.25	0.25	0.5	343.15	40	867.1	825
0.25	0.25	0.5	343.15	60	879.3	935
0.25	0.25	0.5	343.15	80	890.1	1060
0.25	0.25	0.5	343.15	100	899.9	1200
0.25	0.375	0.375	303.15	0.1	837.7	797
0.25	0.375	0.375	303.15	20	851.2	966
0.25	0.375	0.375	303.15	40	863.5	1130
0.25	0.375	0.375	303.15	60	874.8	1320
0.25	0.375	0.375	303.15	80	885.2	1500
0.25	0.375	0.375	303.15	100	894.9	1750
0.25	0.375	0.375	323.15	0.1	821.3	622
0.25	0.375	0.375	323.15	20	836.3	779
0.25	0.375	0.375	323.15	40	850	883
0.25	0.375	0.375	323.15	60	862.5	1010
0.25	0.375	0.375	323.15	80	874.1	1140
0.25	0.375	0.375	323.15	100	884.9	1290
0.25	0.375	0.375	343.15	0.1	804.5	503
0.25	0.375	0.375	343.15	20	821.9	612
0.25	0.375	0.375	343.15	40	836.1	735
0.25	0.375	0.375	343.15	60	848.2	825
0.25	0.375	0.375	343.15	80	858.8	929
0.25	0.375	0.375	343.15	100	868.2	1040
0.25	0.5	0.25	303.15	0.1	802.9	684
0.25	0.5	0.25	303.15	20	817.7	839
0.25	0.5	0.25	303.15	40	830.3	982
0.25	0.5	0.25	303.15	60	841.2	1120
0.25	0.5	0.25	303.15	80	850.9	1300

Table I. *(Continued)*

x_1	x_2	x_3	T (K)	P (MPa)	ρ ($kg \cdot m^{-3}$)	η ($\mu Pa \cdot s$)
0.25	0.5	0.25	303.15	100	859.6	1480
0.25	0.5	0.25	323.15	0.1	786.3	540
0.25	0.5	0.25	323.15	20	802.8	684
0.25	0.5	0.25	323.15	40	816.6	790
0.25	0.5	0.25	323.15	60	828.6	897
0.25	0.5	0.25	323.15	80	839.2	1020
0.25	p.5	0.25	323.15	00	848.7	1150
0.25	0.5	0.25	343.15	0.1	769.5	439
0.25	0.5	0.25	343.15	20	788.1	540
0.25	0.5	0.25	343.15	40	803.1	633
0.25	0.5	0.25	343.15	60	815.8	753
0.25	0.5	0.25	343.15	80	826.7	847
0.25	0.5	0.25	343.15	100	836.4	940
0.25	0.625	0.125	303.15	0.1	770.7	600
0.25	0.625	0.125	303.15	20	786.1	728
0.25	0.625	0.125	303.15	40	799.1	870
0.25	0.625	0.125	303.15	60	810.3	1020
0.25	0.625	0.125	303.15	80	820.3	1180
0.25	0.625	0.125	303.15	100	829.2	1340
0.25	0.625	0.125	323.15	0.1	753.4	477
0.25	0.625	0.125	323.15	20	771.3	606
0.25	0.625	0.125	323.15	40	785.6	703
0.25	0.625	0.125	323.15	60	797.7	811
0.25	0.625	0.125	323.15	80	808.1	924
0.25	0.625	0.125	323.15	100	817.4	1030
0.25	0.625	0.125	343.15	0.1	736.4	391
0.25	0.625	0.125	343.15	20	756.4	476
0.25	0.625	0.125	343.15	40	772.1	568
0.25	0.625	0.125	343.15	60	785	673
0.25	0.625	0.125	343.15	80	796.1	764
0.25	0.625	0.125	343.15	100	805.8	864
0.375	0.125	0.5	303.15	0.1	856.3	870
0.375	0.125	0.5	303.15	20	869.4	1040
0.375	0.125	0.5	303.15	40	881.1	1200
0.375	0.125	0.5	303.15	60	891.7	1400
0.375	0.125	0.5	303.15	80	901.4	1620
0.375	0.125	0.5	303.15	100	910.4	1840
0.375	0.125	0.5	323.15	0.1	842.6	677
0.375	0125	0.5	323.15	20	857.5	822
0.375	0.125	0.5	323.15	40	870.6	934
0.375	0.125	0.5	323.15	60	882.3	1070
0.375	0.125	0.5	323.15	80	892.9	1210

Table I. *(Continued)*

x_1	x_2	x_3	T (K)	P (MPa)	ρ ($kg \cdot m^{-3}$)	η ($\mu Pa \cdot s$)
0.375	0.125	0.5	323.15	100	902.6	1380
0.375	0.125	0.5	343.15	0.1	828	545
0.375	0.125	0.5	343.15	20	845.2	647
0.375	0.125	0.5	343.15	40	859.6	756
0.375	0.125	0.5	343.15	60	872	862
0.375	0.125	0.5	343.15	80	883	972
0.375	0.125	0.5	343.15	100	892.8	1100
0.375	0.25	0.375	303.15	0.1	826.3	737
0.375	0.25	0.375	303.15	20	840.5	892
0.375	0.25	0.375	303.15	40	852.6	1050
0.375	0.25	0.375	303.15	60	863.1	1220
0.375	0.25	0.375	303.15	80	872.4	1400
0.375	0.25	0.375	303.15	100	880.8	1640
0.375	0.25	0.375	323.15	0.1	809.9	580
0.375	0.25	0.375	323.15	20	825.6	730
0.375	0.25	0.375	323.15	40	839.1	829
0.375	0.25	0.375	323.15	60	851	950
0.375	0.25	0.375	323.15	80	861.7	1070
0.375	0.25	0.375	323.15	100	871.4	1210
0.375	0.25	0.375	343.15	0.1	793.6	471
0.375	0.25	0.375	343.15	20	811.1	579
0.375	0.25	0.375	343.15	40	825.8	671
0.375	0.25	0.375	343.15	60	838.5	773
0.375	0.25	0.375	343.15	80	849.7	867
0.375	0.25	0.375	343.15	100	859.8	993
0.375	0.375	0.25	303.15	0.1	797.3	652
0.375	0.375	0.25	303.15	20	812.4	793
0.375	0.375	0.25	303.15	40	825	919
0.375	0.375	0.25	303.15	60	835.8	1050
0.375	0.375	0.25	303.15	80	845.3	1210
0.375	0.375	0.25	303.15	100	853.8	1390
0.375	0.375	0.25	323.15	0.1	780.8	518
0.375	0.375	0.25	323.15	20	797.4	647
0.375	0.375	0.25	323.15	40	811.3	750
0.375	0.375	0.25	323.15	60	823.3	851
0.375	0.375	0.25	323.15	80	833.9	956
0.375	0.375	0.25	323.15	100	843.4	1080
0.375	0.375	0.25	343.15	0.1	764	423
0.375	0.375	0.25	343.15	20	782.7	513
0.375	0.375	0.25	343.15	40	797.7	607
0.375	0.375	0.25	343.15	60	810.3	701
0.375	0.375	0.25	343.15	80	821.3	800

Table I. *(Continued)*

x_1	x_2	x_3	T (K)	P (MPa)	ρ (kg·m^{-3})	η (μPa·s)
0.375	0.375	0.25	343.15	100	831	898
0.375	0.5	0.125	303.15	0.1	759.4	559
0.375	0.5	0.125	303.15	20	775.2	691
0.375	0.5	0.125	303.15	40	788.3	807
0.375	0.5	0.125	303.15	60	799.5	938
0.375	0.5	0.125	303.15	80	809.2	1090
0.375	0.5	0.125	303.15	100	817.9	1260
0.375	0.5	0.125	323.15	0.1	742.5	447
0.375	0.5	0.125	323.15	20	760.2	563
0.375	0.5	0.125	323.15	40	774.8	652
0.375	0.5	0.125	323.15	60	787.2	756
0.375	0.5	0.125	323.15	80	798.1	852
0.375	0.5	0.125	323.15	100	807.8	964
0.375	0.5	0.125	343.15	0.1	725.6	368
0.375	0.5	0.125	343.15	20	745.4	453
0.375	0.5	0.125	343.15	40	761.6	541
0.375	0.5	0.125	343.15	60	775.4	623
0.375	0.5	0.125	343.15	80	787.4	718
0.375	0.5	0.125	343.15	100	798.2	817
0.5	0.125	0.375	303.15	0.1	813.9	693
0.5	0.125	0.375	303.15	20	828.2	826
0.5	0.125	0.375	303.15	40	840.7	970
0.5	0.125	0.375	303.15	60	851.8	1120
0.5	0.125	0.375	303.15	80	861.7	1290
0.5	0.125	0.375	303.15	100	870.8	1470
0.5	0.125	0.375	323.15	0.1	799.6	547
0.5	0.125	0.375	323.15	20	815.9	644
0.5	0.125	0.375	323.15	40	829.6	769
0.5	0.125	0.375	323.15	60	841.5	895
0.5	0.125	0.375	323.15	80	852	1000
0.5	0.125	0.375	323.15	100	861.5	1130
0.5	0.125	0.375	343.15	0.1	784.9	449
0.5	0.125	0.375	343.15	20	803.4	529
0.5	0.125	0.375	343.15	40	818.3	615
0.5	0.125	0.375	343.15	60	830.9	726
0.5	0.125	0.375	343.15	80	841.7	823
0.5	0.125	0.375	343.15	100	851.3	927
0.5	0.25	0.25	303.15	0.1	782.1	594
0.5	0.25	0.25	303.15	20	797	728
0.5	0.25	0.25	303.15	40	809.6	848
0.5	0.25	0.25	303.15	60	820.6	985
0.5	0.25	0.25	303.15	80	830.4	1140

Table I. *(Continued)*

x_1	x_2	x_3	T (K)	P (MPa)	ρ (kg·m^{-3})	η (μPa·s)
0.5	0.25	0.25	303.15	100	839.2	1300
0.5	0.25	0.25	323.15	0.1	765	476
0.5	0.25	0.25	323.15	20	782.1	610
0.5	0.25	0.25	323.15	40	796.1	697
0.5	0.25	0.25	323.15	60	808	800
0.5	0.25	0.25	323.15	80	818.3	898
0.5	0.25	0.25	323.15	100	827.5	1030
0.5	0.25	0.25	343.15	0.1	748.3	393
0.5	0.25	0.25	343.15	20	767.3	492
0.5	0.25	0.25	343.15	40	782.6	586
0.5	0.25	0.25	343.15	60	795.4	662
0.5	0.25	0.25	343.15	80	806.5	758
0.5	0.25	0.25	343.15	100	816.3	855
0.5	0.375	0.125	303.15	0.1	748.7	521
0.5	0.375	0.125	303.15	20	764.4	639
0.5	0.375	0.125	303.15	40	777.6	762
0.5	0.375	0.125	303.15	60	789	889
0.5	0.375	0.125	303.15	80	799	1010
0.5	0.375	0.125	303.15	100	808	1160
0.5	0.375	0.125	323.15	0.1	731.5	421
0.5	0.375	0.125	323.15	20	749.5	532
0.5	0.375	0.125	323.15	40	764.1	619
0.5	0.375	0.125	323.15	60	776.4	717
0.5	0.375	0.125	323.15	80	787.1	813
0.5	0.375	0.125	323.15	100	796.7	922
0.5	0.375	0.125	343.15	0.1	714.4	347
0.5	0.375	0.125	343.15	20	734.8	420
0.5	0.375	0.125	343.15	40	750.8	511
0.5	0.375	0.125	343.15	60	764	590
0.5	0.375	0.125	343.15	80	775.4	679
0.5	0.375	0.125	343.15	100	785.4	776
0.625	0.125	0.25	303.15	0.1	771	560
0.625	0.125	0.25	303.15	20	786.4	683
0.625	0.125	0.25	303.15	40	799.1	806
0.625	0.125	0.25	303.15	60	809.9	928
0.625	0.125	0.25	303.15	80	819.4	1070
0.625	0.125	0.25	303.15	100	827.8	1210
0.625	0.125	0.25	323.15	0.1	756.6	451
0.625	0.125	0.25	323.15	20	774	551
0.625	0.125	0.25	323.15	40	788.3	644
0.625	0.125	0.25	323.15	60	800.6	737
0.625	0.125	0.25	323.15	80	811.4	847

Table I. *(Continued)*

x_1	x_2	x_3	T (K)	P (MPa)	ρ (kg·m^{-3})	η (μPa·s)
0.625	0.125	0.25	323.15	100	821.1	943
0.625	0.125	0.25	343.15	0.1	741.6	372
0.625	0.125	0.25	343.15	20	761.1	451
0.625	0.125	0.25	343.15	40	776.9	529
0.625	0.125	0.25	343.15	60	790.3	624
0.625	0.125	0.25	343.15	80	802	710
0.625	0.125	0.25	343.15	100	812.4	806
0.625	0.25	0.125	303.15	0.1	738	490
0.625	0.25	0.125	303.15	20	754	600
0.625	0.25	0.125	303.15	40	767.3	710
0.625	0.25	0.125	303.15	60	778.8	814
0.625	0.25	0.125	303.15	80	788.9	925
0.625	0.25	0.125	303.15	100	797.9	1060
0.625	0.25	0.125	323.15	0.1	721.2	396
0.625	0.25	0.125	323.15	20	739	496
0.625	0.25	0.125	323.15	40	753.9	573
0.625	0.25	0.125	323.15	60	766.7	665
0.625	0.25	0.125	323.15	80	778	755
0.625	0.25	0.125	323.15	100	788.2	847
0.625	0.25	0.125	343.15	0.1	703.5	327
0.625	0.25	0.125	343.15	20	724.3	406
0.625	0.25	0.125	343.15	40	740.6	472
0.625	0.25	0.125	343.15	60	754.1	551
0.625	0.25	0.125	343.15	80	765.6	630
0.625	0.25	0.125	343.15	100	775.8	710
0.75	0.125	0.125	303.15	0.1	728.6	462
0.75	0.125	0.125	303.15	20	745	570
0.75	0.125	0.125	303.15	40	758.6	673
0.75	0.125	0.125	303.15	60	770.2	772
0.75	0.125	0.125	303.15	80	780.4	884
0.75	0.125	0.125	303.15	100	789.6	1010
0.75	0.125	0.125	323.15	0.1	713.8	377
0.75	0.125	0.125	323.15	20	732.4	468
0.75	0.125	0.125	323.15	40	747.5	549
0.75	0.125	0.125	323.15	60	760.3	625
0.75	0.125	0.125	323.15	80	771.3	725
0.75	0.125	0.125	323.15	100	781.2	807
0.75	0.125	0.125	343.15	0.1	697.7	312
0.75	0.125	0.125	343.15	20	719.7	407
0.75	0.125	0.125	343.15	40	736.4	479
0.75	0.125	0.125	343.15	60	750.1	548
0.75	0.125	0.125	343.15	80	761.7	622
0.75	0.125	0.125	343.15	100	771.8	704

interactions of the system studied. We obtained the following relationship in which a corrective term was added:

$$\ln \eta = x_1 \ln \eta_1 + x_2 \ln \eta_2 + x_3 \ln \eta_3 + (x_1 x_2 + x_1 x_3 + x_2 x_3) \times \left(\frac{a + bP}{T}\right) \frac{1}{1 + c(x_1 M_1 + x_2 M_2 + x_3 M_3)} \tag{2}$$

which, by a procedure of minimization on the AAD, provides the following results: AAD = 2.7% and MD = 9.0%, which is a very satisfactory result, since the AAD is of the order of magnitude of the experimental error.

A completely different model is the self-referencing model (see, e.g., Refs. 11 and 12), which can be applied both to pure substances and to mixtures and which requires only viscosity data at 0.1 MPa and at a reference temperature T_0. For our system we used 45 reference points and we obtained, for the 765 remaining points ($810 - 45 = 765$), AAD = 6.1% and MD = 20.7% with $T_0 = 323.15$ K. Readjusting the initial coefficients of Kanti et al. [11], we obtained AAD = 2.4% and MD = 17.9% for the same value of T_0. It is possible to combine the self-referencing model with a mixing rule. In that case it is sufficient to know the viscosity of each of the three pure substances at atmospheric pressure and at the reference temperature T_0. Using the ideal Grunberg and Nissan mixing rule [Eq. (1)] and the initial coefficients of the self-referencing model, i.e., without introducing any new numerical adjustment, we obtained, for the 807 points concerned, AAD = 17.4% and MD = 58.6% at $T_0 = 323.15$ K. But if the modified mixing rule [Eq. (2)] is used, and if the coefficients of the self-referencing model are adjusted, then AAD = 2.5% and MD = 13.9%, once again for $T_0 = 323.15$ K.

The models used to account for the viscous behavior versus pressure, temperature, and, in some cases, composition are very varied in their approaches to the problem. Interested readers will find information in Ref. 13. However, it is interesting to mention here the method recently developed by Dymond and Awan [14] and Assael et al. [15] based on the theory of hard spheres, which entails developing viscosity as a function of density. If the coefficients supplied by those authors are used, the results obtained are poor because their adjustment base concerns linear alkanes, but if a new adjustment is performed on the basis of our 810 values, one obtains AAD = 3.6% and MD = 18.7%, which represents a very good performance. The three models mentioned in this investigation, although they are conceptually different, yield comparable results.

It is to be hoped that the experimental data supplied in this paper and in the earlier publication [1], which provide a substantial description of variations of η and ρ versus pressure, temperature, and composition, will be included in databases and used to test other models of viscous behavior more sophisticated than those discussed here.

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