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Viscosity and Density of Five Hydrocarbon Liquids at Pressures up to 200 MPa and Temperatures up to 473 K[†]

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We report the results of simultaneous measurements of the viscosity and density of five pure hydrocarbon liquids (octane, decane, 1,3-dimethylbenzene, 1,2,3,4-tetrahydronaphthalene, and 1-methylnaphthalene) at temperatures between (298.15 and 473.15) K and at pressures ranging from 0.1 MPa to approximately 200 MPa. The measurements were made with a vibrating-wire instrument, and the estimated expanded relative uncertainties are ± 2 % for viscosity and ± 0.2 % for density with a coverage factor of 2. The densities were correlated by means of a modified Tait equation, while the viscosities were correlated both with the theory of Dymond and Assael and in terms of an empirical function of temperature and pressure. We also present correlations of the viscosity of dodecane and octadecane based on results that we published previously [Caudwell et al. *Int. J. Thermophys.* **2004**, *25*, 1340−1352]. Extensive comparisons with literature data are presented.

Introduction

Despite the importance of viscosity in many physical and chemical processes, there is presently no well-founded theoretical method for the prediction of this property in dense fluids. However, a number of practical prediction schemes do exist that take advantage of the kinetic theory of rigid spheres and the principle of corresponding states.[1−10] The work reported here forms part of a project aimed at developing improved approaches to this problem for both pure fluids and mixtures.[8,9] To establish the accuracy and reliability of predictive methods, it is essential to validate them against a set of primary experimental data of proven accuracy. Furthermore, several predictive schemes for mixtures exploit accurate correlations of experimentally determined pure fluid viscosities, and it is desirable to extend the ranges of fluids and of thermodynamic states for which such correlations are available. The present paper is concerned particularly with reducing gaps in the available viscosity data of pure hydrocarbons under conditions of high temperature and high pressure, thereby permitting more rigorous testing of predictive and correlative methods. Although better understood, there is also a need for improved measurements of compressed-liquid density, and this point was also addressed in the present study. Results are reported for the following five liquids: octane, decane, 1,3-dimethylbenzene (*m*-xylene), 1,2,3,4-tetrahydronaphthalene (tetralin), and 1-methylnaphthalene (1-MNP). The measurements, which were made over extended ranges of temperature [(298.15 to 473.15) K] and pressure [(0.1 to 200) MPa], complement the results reported earlier for dodecane and octadecane.[11] A subsequent paper will deal with binary and multicomponent mixtures containing these and other species.

Experimental

The measurements were made with the steady-state vibrating-wire instrument described earlier.[10,11] This instrument employed a tungsten wire with a nominal radius of 0.5 mm and a nominal length of 70 mm. The wire, located between opposite poles of a pair of permanent magnets, was clamped in a fixed support at its upper end and to an aluminum sinker, with a mass of approximately 0.13 kg, at its lower end. The wire, sinker, and magnet assembly were mounted within a high-pressure vessel which was filled with the liquid under study, and the entire assembly was enclosed within an aluminum-block thermostat. The vibrating wire was driven by a constant sinusoidal current with an amplitude of order of 0.1 mA, and the magnitude and phase of the emf developed across it were measured as a function of frequency in the vicinity of the fundamental transverse resonance frequency. The viscosity η and density ρ of the fluid under study were determined, along with other parameters, in a nonlinear optimization loop that minimized the root-mean-square deviation of the experimental data from the working equation.[11]

The sources and mass-fraction purities w specified by the suppliers of the chemical were as follows: octane ($w \geq 0.995$) and 1-MNP ($w \geq 0.98$) were supplied by Fluka; decane ($w \geq 0.99$), *m*-xylene ($w \geq 0.99$), and tetralin ($w \geq 0.99$) were supplied by Sigma-Aldrich; and methylbenzene ($w \geq 0.999$) was supplied by Riedel-de Haën. The liquids were used without further purification. The purity of 1-MNP was studied by GC-FID, but no impurities were resolved. The other substances were not analyzed for purity, and so it is not possible to make definitive statements about the errors arising from the presence of impurities. However, likely impurities are other hydrocarbons of similar normal boiling temperature and viscosity, leading to relative errors in the measured viscosity of less than 1 %.

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Table 1. Viscosity and Density of Octane

p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)	p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)
	T/K = 298.15			T/K = 373.15	
20.9	715.2	0.631	80.2	709.3	0.503
40.6	728.3	0.753	101.1	721.3	0.576
60.9	740.0	0.889	120.0	730.9	0.645
80.1	749.8	1.030	140.9	740.6	0.726
100.3	759.1	1.190	160.3	748.8	0.804
120.1	767.3	1.360	180.5	756.7	0.890
139.9	774.9	1.550	200.9	764.2	0.981
159.5	782.0	1.750		T/K = 398.15	
179.6	788.7	1.980	21.6	644.4	0.263
197.2	794.2	2.210	42.0	665.9	0.320
139.7	774.8	1.555	80.9	695.8	0.430
80.0	749.6	1.040	120.9	719.0	0.550
0.1	698.6	0.510	160.2	737.2	0.676
	T/K = 323.15		192.3	750.0	0.786
0.1	678.2	0.387	159.5	736.9	0.674
20.1	697.0	0.477	79.4	694.8	0.426
40.0	712.0	0.570	21.6	644.6	0.265
60.6	725.0	0.672		T/K = 423.15	
80.1	735.8	0.778	21.4	626.8	0.226
101.0	746.1	0.897	41.1	650.3	0.276
120.3	754.7	1.010	80.7	683.5	0.372
141.5	763.4	1.150	121.5	708.6	0.476
160.3	770.5	1.280	161.3	728.2	0.587
180.7	777.7	1.440	192.5	741.1	0.676
201.8	784.8	1.610	159.7	727.5	0.581
141.6	763.2	1.150	80.4	683.3	0.372
80.1	735.6	0.781	21.4	626.9	0.226
0.1	678.2	0.387		T/K = 448.15	
	T/K = 348.15		21.0	607.9	0.192
0.1	657.2	0.304	41.2	635.1	0.239
20.4	679.6	0.381	81.4	671.5	0.326
40.8	696.7	0.457	121.5	697.4	0.414
60.7	710.6	0.534	160.3	717.5	0.504
80.1	722.4	0.614	193.0	732.0	0.585
101.0	733.4	0.706	159.4	717.1	0.504
120.4	742.7	0.794	80.5	671.0	0.324
140.3	751.3	0.892	21.0	608.0	0.192
160.7	759.5	0.997		T/K = 473.15	
180.8	767.0	1.110	19.3	586.4	0.163
201.9	774.3	1.230	41.1	620.0	0.210
140.2	751.2	0.893	81.5	659.6	0.289
80.1	722.4	0.614	121.1	686.7	0.367
0.1	657.2	0.304	161.3	708.3	0.449
	T/K = 373.15		193.4	723.0	0.516
0.1	635.3	0.245	160.7	708.0	0.448
20.6	662.0	0.312	81.1	659.3	0.289
39.9	680.4	0.373	20.0	587.7	0.164
60.0	696.0	0.437			

Table 2. Viscosity and Density of Decane

p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)	p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)
	T/K = 298.15			T/K = 348.15	
0.1	726.6	0.849	0.1	687.7	0.458
50.1	758.5	1.416	51.0	729.2	0.759
102.8	782.8	2.199	101.5	757.0	1.102
150.5	800.6	3.146	150.9	777.9	1.504
190.9	813.3	4.185	190.8	792.2	1.893
150.4	801.0	3.136	149.7	777.8	1.494
50.3	759.2	1.414	51.1	730.1	0.758
0.1	726.8	0.849	0.1	687.9	0.459
	T/K = 323.15			T/K = 373.15	
0.1	707.4	0.607	0.1	667.6	0.360
52.0	744.6	1.017	51.1	714.7	0.601
100.7	769.2	1.481	101.7	744.7	0.866
150.8	789.1	2.080	150.6	766.8	1.159
191.7	803.0	2.684	190.4	782.2	1.433
150.9	789.5	2.084	149.1	766.7	1.152
52.4	745.7	1.016	51.5	715.7	0.605
0.1	707.4	0.607	0.1	667.5	0.360

Table 3. Viscosity and Density of *m*-Xylene

p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)	p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)
	T/K = 298.15			T/K = 373.15	
0.1	860.5	0.584	191.3	910.2	0.838
40.1	885.9	0.783	158.6	897.5	0.731
80.4	906.5	1.010	80.8	859.3	0.504
120.2	923.5	1.280	0.1	793.4	0.291
161.3	938.5	1.610		T/K = 398.15	
198.5	950.9	1.960	0.1	768.4	0.241
160.0	938.3	1.600	39.7	811.9	0.331
80.3	906.9	1.010	79.7	841.6	0.423
0.1	860.6	0.585	119.9	865.0	0.526
	T/K = 298.15		160.7	884.8	0.624
0.1	860.8	0.584	197.1	899.9	0.721
41.0	886.7	0.789	159.7	884.4	0.622
79.7	906.3	1.010	80.3	841.9	0.423
120.4	923.7	1.280	0.1	768.4	0.241
159.4	938.2	1.590		T/K = 423.15	
191.3	948.8	1.890	21.3	774.2	0.249
159.1	938.3	1.590	40.5	794.6	0.287
80.8	907.5	1.020	80.6	826.8	0.366
0.1	861.0	0.584	122.2	852.4	0.450
	T/K = 323.15		161.6	872.4	0.534
0.1	838.9	0.447	195.7	887.5	0.611
41.7	868.8	0.607	160.3	871.9	0.531
80.5	890.3	0.765	80.4	826.8	0.366
121.7	909.5	0.957	21.8	774.8	0.250
160.0	924.6	1.160		T/K = 448.15	
189.6	935.3	1.340	19.9	751.9	0.213
159.2	924.9	1.160	40.5	776.8	0.251
81.4	891.8	0.771	79.6	811.4	0.320
0.1	838.9	0.448	120.5	838.5	0.393
	T/K = 348.15		162.3	861.2	0.471
0.1	816.4	0.355	190.7	874.9	0.528
41.4	850.2	0.481	161.0	861.1	0.470
80.9	874.3	0.610	81.0	812.7	0.323
121.2	894.6	0.748	20.9	753.3	0.215
160.8	911.2	0.906		T/K = 473.15	
192.3	923.0	1.040	20.1	731.1	0.187
158.9	910.9	0.899	40.0	758.5	0.221
80.9	875.3	0.609	80.2	797.5	0.287
0.1	816.4	0.355	120.8	826.9	0.353
	T/K = 373.15		163.8	852.1	0.426
0.1	793.4	0.291	196.4	868.0	0.483
41.8	832.4	0.398	163.0	851.9	0.425
81.4	858.7	0.504	79.9	798.0	0.287
121.1	880.1	0.616	19.9	730.9	0.187
160.8	897.9	0.737			

Prior to charging the viscometer with a new liquid sample, the entire system was flushed thoroughly with hexane, drained, and dried under a vacuum. The sample was then drawn into the previously evacuated system and pressurized to approximately 10 MPa by means of a hand pump to ensure complete filling of the instrument. The desired initial pressure was then established before starting a sequence of measurements along an isotherm with pressure increasing in approximately even increments. At each state point, two or three measurements were usually made, and the mean results obtained were ascribed to the mean temperature and the mean pressure for that group.

The temperature of the fluid was inferred from the reading of a platinum resistance thermometer that was mounted in a well in the cap of the pressure vessel. This thermometer was calibrated by comparison with a standard platinum resistance thermometer that had itself been calibrated on ITS-90 at the UK National Physical Laboratory. The expanded uncertainty of the temperature measurements was ± 0.02 K at a coverage factor, k, of 2. The pressure was measured in the external pipework by means of a Paroscientific transducer (model 40K-110) with a full scale reading of 276 MPa. Prior to the present

Table 4. Viscosity and Density of Tetralin

p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)	p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)
	T/K = 298.15			T/K = 373.15	
0.1	965.8	1.980	160.2	991.4	2.090
40.3	986.3	2.920	190.6	1003	2.500
78.6	1003	4.153	123.0	977.2	1.680
119.8	1019	6.000	81.1	957.5	1.280
158.9	1032	8.490	0.1	904.8	0.696
190.2	1042	11.200		T/K = 398.15	
120.6	1020	6.040	0.1	883.9	0.541
80.9	1005	4.240	40.5	916.9	0.752
0.1	965.7	1.980	80.6	941.5	0.989
	T/K = 323.15		120.8	961.5	1.270
0.1	945.5	1.290	159.9	978.1	1.587
39.9	968.5	1.830	200.6	993.3	1.970
80.3	987.7	2.550	161.0	978.5	1.590
121.0	1004	3.480	81.8	942.0	1.000
161.2	1019	4.703	0.1	883.9	0.542
190.6	1028	5.833		T/K = 423.15	
120.9	1005	3.470	0.1	863.1	0.438
80.6	988.7	2.550	40.5	900.0	0.610
0.1	945.6	1.290	81.0	926.8	0.800
	T/K = 348.15		120.8	948.0	1.010
0.1	925.1	0.920	161.3	966.1	1.250
41.0	951.4	1.290	201.0	981.7	1.530
79.7	971.3	1.730	160.8	965.9	1.250
119.5	988.9	2.270	82.2	927.4	0.807
163.0	1006	3.027	0.1	863.0	0.438
190.8	1015	3.620		T/K = 448.15	
121.7	990.9	2.310	0.1	841.6	0.362
80.6	972.7	1.740	40.4	883.2	0.509
0.1	925.3	0.921	80.4	912.0	0.664
	T/K = 373.15		120.2	934.5	0.833
0.1	904.6	0.696	161.2	954.1	1.030
40.5	933.9	0.963	200.1	970.1	1.240
80.3	956.3	1.277	160.6	953.7	1.030
122.5	976.1	1.670	81.2	912.4	0.667
			0.1	841.6	0.362

Table 5. Viscosity and Density of 1-Methylnaphthalene

p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)	p/MPa	ρ/(kg·m^{-3})	η/(mPa·s)
	T/K = 298.15			T/K = 398.15	
0.1	1020	2.913	0.1	942.6	0.664
20.5	1030	3.620	40.5	972	0.905
40.1	1038	4.430	79.7	994.3	1.180
81.4	1056	6.780	120.5	1014	1.510
100.1	1062	8.350	162.9	1031	1.920
121.3	1070	10.300	202.4	1045	2.390
140.8	1077	12.700	160.5	1030	1.900
161.5	1083	16.000	82.7	995.8	1.200
121.3	1071	10.300	0.1	942.7	0.665
80.2	1056	6.720			
0.1	1020	2.920			
	T/K = 323.15			T/K = 423.15	
0.1	1001	1.740	0.1	923.1	0.530
39.6	1022	2.490	40.5	955.8	0.722
80.4	1040	3.540	81.0	980.6	0.937
120.8	1056	4.960	120.5	1000	1.170
160.5	1068	7.020	160.0	1017	1.450
193.0	1080	9.180	199.7	1032	1.780
123.3	1057	5.120	161.0	1018	1.460
79.6	1040	3.510	81.9	981	0.941
0.1	1001	1.730	0.1	923	0.530
	T/K = 348.15			T/K = 448.15	
0.1	981.3	1.190	0.1	903.1	0.435
40.8	1005	1.660	40.2	939.4	0.592
79.3	1024	2.240	80.1	965.8	0.762
121.9	1042	3.050	120.7	987.5	0.957
162.2	1057	4.050	160.8	1006	1.170
189.9	1066	4.930	200.8	1022	1.410
121.4	1043	3.030	160.1	1005	1.160
78.9	1025	2.220	81.8	966.7	0.769
0.1	981.3	1.190	0.1	903.1	0.435
	T/K = 373.15			T/K = 473.15	
0.1	962.0	0.869	0.1	882.8	0.364
41.5	989.0	1.200	40.6	923.7	0.501
79.8	1009	1.570	81.0	952.6	0.645
120.3	1027	2.060	121.6	975.5	0.802
159.7	1043	2.630	160.8	994.2	0.972
189.5	1054	3.140	200.2	1011	1.160
121.7	1029	2.070	160.9	994.3	0.970
80.1	1010	1.580	81.5	952.9	0.646
0.1	962.0	0.869	0.1	882.7	0.364

work, the pressure transducer was calibrated against a pressure balance (piston gauge) in the range (0.1 to 200) MPa. The expanded uncertainty of pressure was ± 0.02 MPa ($k = 2$).

The working equations of the instrument contain four quantities that we choose to determine by calibration.[11] The resonance frequency and logarithmic decrement of the wire under vacuum were inferred from measurements in ambient air, while the radius of the wire and the volume of the tensioning weight were obtained from a measurement in methylbenzene at $T = 298.15$ K and $p = 0.1$ MPa. The viscosity of methylbenzene under those conditions was taken to be $\eta = (0.553 \pm 0.003)$ mPa·s, which is the average value from three sources[12−14] that were selected as primary data in the correlation of Assael et al.,[15] while the density was taken to be $\rho = 862.3$ kg·m^{-3}.[16] Other quantities in the working equations were either directly measured or were material properties with values taken from the literature.[10]

The repeatability of results was investigated both by returning to the initial state point after a sequence of measurements on a single sample and by refilling and repeating measurements with a fresh fluid charge. The relative repeatability of the viscosity was not worse than ± 1 %, while that of the density was not worse than ± 0.1 %. The overall relative uncertainties are ± 2 % for viscosity and ± 0.2 % for density with $k = 2$.

Results and Correlation

The experimental viscosities and densities of the five liquids are reported in Tables 1 to 5. Since the actual experimental temperatures differed from the nominal isotherm temperatures by ± 0.1 K, small fractional adjustments (never more than 0.2 % in viscosity and 0.01 % in density) have been applied to reduce all tabulated results to the stated temperatures.

Density. The density was correlated by the modified Tait equation

$$\rho = \rho_0\left[1 - C\log_{10}\left\{\frac{p+B}{p_0+B}\right\}\right]^{-1} \tag{1}$$

where $p_0 = 0.1$ MPa; ρ_0 is the density at $p = p_0$; C is a constant; and B is a temperature-dependent parameter. In this work, we correlated ρ_0 and B as follows

$$\rho_0/(\mathrm{kg\cdot m^{-3}}) = \sum_{i=0}^{2} a_i(T/\mathrm{K})^i \tag{2}$$

$$B/\mathrm{MPa} = \sum_{i=0}^{2} b_i(T/\mathrm{K})^i \tag{3}$$

To establish the parameters of this correlation, eq 1 was first applied to individual isotherms, and ρ_0, B, and C were adjusted to minimize the absolute differences between the measured and correlated values. Here and below, goodness of fit was measured by the absolute average relative deviation, $\Delta_{\mathrm{AAD},X}$, given by

Table 6. Parameters of Equations 1 to 3 for the Density

fluid	a_0	a_1	a_2	b_0	b_1	b_2	C
octane	814.34	−0.03758	$-1.186 \cdot 10^{-3}$	314.88	−1.13918	$1.046 \cdot 10^{-3}$	−0.2042
decane	918.45	−0.52524	$-3.949 \cdot 10^{-4}$	398.06	−1.49775	$1.488 \cdot 10^{-3}$	−0.2070
m-xylene	1048.90	−0.41812	$-7.150 \cdot 10^{-4}$	428.70	−1.44109	$1.240 \cdot 10^{-3}$	−0.2127
tetralin	1191.20	−0.70936	$-1.570 \cdot 10^{-4}$	512.20	−1.63032	$1.376 \cdot 10^{-3}$	−0.2043
1-MNP	1233.45	−0.67681	$-1.350 \cdot 10^{-4}$	541.82	−1.65868	$1.361 \cdot 10^{-3}$	−0.2008

Table 7. Parameters of Equation 10 for the Correlation of Viscosity at $p = 0.1$ MPa

fluid	A_η	B_η	C_η
octane	−5.87065	2495.86	183.07
decane	−4.07477	1045.98	−30.80
dodecane	−4.71516	1562.59	14.23
octadecane	−3.81885	1187.42	−71.54
m-xylene	−4.63386	1545.59	79.66
tetralin	−3.68755	1055.47	−55.99
1-MNP[a]	−3.37631	895.753	−96.00

[a] Results from individual isotherm results.

$$\Delta_{\mathrm{AAD},X} = \frac{1}{N}\sum_{i=1}^{N}(|X_i - X_{i,\mathrm{fit}}|/X_i) \qquad (4)$$

where X_i is an experimental datum; $X_{i,\mathrm{fit}}$ is calculated from the correlation applied at the same state point; and N is the total number of points. We also calculated the maximum absolute relative deviation, $\Delta_{\mathrm{MAD},X}$, defined by

$$\Delta_{\mathrm{MAD},X} = \mathrm{Max}(|X_i - X_{i,\mathrm{fit}}|/X_i) \qquad (5)$$

Once values of ρ_0, B, and C were obtained at each temperature, the parameters a_i and b_i were estimated by linear regressions with eqs 2 and 3, and a mean value of C was calculated. Finally, all seven parameters (a_0, a_1, a_2, b_0, b_1, b_2, and C) were adjusted in a simultaneous nonlinear optimization to minimize $\Delta_{\mathrm{AAD},\rho}$ for the entire data set pertaining to a given fluid. The parameters obtained are given in Table 6.

With our choice of reference pressure $p_0 = 0.1$ MPa, $\rho_0(T)$ represents hypothetical liquid states when the temperature exceeds the boiling temperature at $p = 0.1$ MPa.

Viscosity. The viscosity was correlated in two ways. The first approach sought to represent η as a function of T, and p and made use of the equation

$$\eta = A_\eta \exp\left(\frac{B_\eta}{T - C_\eta}\right)\left(\frac{p + E}{p_0 + E}\right)^D \qquad (6)$$

where A_η, B_η, and C_η are constants, $p_0 = 0.1$ MPa as before, and D and E are functions of temperature given by

$$D = \sum_{i=0}^{2} d_i(\mathrm{K}/T)^i \qquad (7)$$

$$E/\mathrm{MPa} = \sum_{i=0}^{2} e_i(T/\mathrm{K})^i \qquad (8)$$

This correlation method has been found both useful and accurate for a number of pure fluids and mixtures over wide ranges of temperature and pressure.[17,18] The correlating procedure was as follows. First, individual isotherms were fitted by the equation

$$\eta = \eta_0\left(\frac{p + E}{p_0 + E}\right)^D \qquad (9)$$

to obtain η_0 (the viscosity at $p = p_0$), E, and D at each temperature. Second, we estimated A_η, B_η, and C_η by linear regression with the equation

$$\ln \eta_0 = \ln A_\eta + \left(\frac{B_\eta}{T - C_\eta}\right) \qquad (10)$$

using trial values of C_η that were adjusted to achieve a good fit. Next, we estimated the parameters d_i and e_i by linear regressions with eqs 5 and 6. Finally, all nine parameters (A_η, B_η, C_η, d_0, d_1, d_2, e_0, e_1, and e_2) were adjusted in a simultaneous nonlinear optimization to minimize $\Delta_{\mathrm{AAD},\eta}$ for the entire data set.

In addition to the five liquids studied experimentally in this work, we have applied the correlation method for the first time to the viscosity data reported previously for dodecane and octadecane.[11] The results of the correlations are reported in Tables 7 and 8.

In all cases, eq 9 was found to represent accurately the viscosity along an isotherm, and the parameters D and E were found to increase with decreasing temperature. In most cases, this variation was accounted for in a satisfactory way by eqs 7 and 8. However, in the case of 1-MNP at the lowest temperature of 298.15 K, E was found to increase rapidly, apparently without bound, during the isothermal parameter optimization loop but such that $D' = D/E$ approached a constant value. This corresponds to a situation in which eq 9 reduces to a simple exponential dependence upon pressure

$$\eta = \eta_0 \exp\{D'(p - p_0)\} \qquad (11)$$

We do not have an explanation for this behavior. Thus, in the case of 1-MNP, a satisfactory representation of the whole experimental $\eta(T,p)$ surface was not obtained with eqs 6 to 8, and only the parameters A_η, B_η, and C_η are reported. As with the correlation of density, $\eta_0(T)$ represents hypothetical liquid states when the temperature exceeds the boiling temperature at $p = p_0$.

The second approach to the correlation of the viscosity was based on a substance-specific version of the hard-sphere theory of Dymond and Assael.[19,2] In this approach, a dimensionless reduced viscosity η^* is defined such that

$$\eta^* = \frac{16}{5}(2N_\mathrm{A})^{1/3}(\pi/MRT)^{1/2}V_\mathrm{m}^{2/3}\eta \qquad (12)$$

where N_A is Avogadro's constant, $V_\mathrm{m} = M/\rho$ is the molar volume, and M is the molar mass. The reduced viscosity is then correlated as a function of (V_m/V_0) as follows

$$1/\eta^* = \sum_{i=0}^{3} f_i(V_\mathrm{m}/V_0)^i \qquad (13)$$

where V_0 is a temperature-dependent molar core volume given by

$$V_0/(\mathrm{cm}^3 \cdot \mathrm{mol}^{-1}) = \sum_{i=0}^{3} g_i(T/\mathrm{K})^i \qquad (14)$$

The values of the eight parameters so determined for each fluid are given in Tables 9 and 10. To apply the correlation at a given temperature and pressure, eqs 1 to 3 may be used to obtain $\rho(T,p)$ and then eqs 12 to 14 to obtain $\eta(T,p)$.

Table 8. Parameters of Equation 7 to 8 for the Correlation of Viscosity

fluid	d_0	d_1	d_2	e_0	e_1	e_2
octane	2.99480	−2174.86	$5.89895 \cdot 10^{5}$	1108.58	−4.27394	$4.38223 \cdot 10^{-3}$
decane	11.6438	−8608.68	$1.79954 \cdot 10^{6}$	2670.84	−12.8767	$1.62362 \cdot 10^{-2}$
dodecane	3.62843	−3184.60	$9.28430 \cdot 10^{5}$	1412.44	−5.36373	$5.31960 \cdot 10^{-3}$
octadecane	4.21920	−3747.83	$1.06532 \cdot 10^{6}$	1219.99	−4.56228	$4.50438 \cdot 10^{-3}$
m-xylene	6.89403	−5393.26	$1.29136 \cdot 10^{6}$	2546.45	−10.3921	$1.12223 \cdot 10^{-2}$
tetralin	75.4440	−66400.5	$1.49943 \cdot 10^{7}$	19134.7	−85.4852	$9.66338 \cdot 10^{-2}$

Table 9. Parameters of Equation 13 for the Fluid-Specific Reduced-Viscosity Correlation

fluid	f_0	f_1	f_2	f_3
octane	0.398074	−0.7252300	0.410386	−0.0676790
decane	0.412930	−0.6786820	0.349327	−0.0534100
xylene	0.368771	−0.6745920	0.377256	−0.0602089
tetralin	0.658061	−1.0511615	0.531687	−0.0821090
1-MNP	0.689529	−1.0699490	0.525441	−0.0787304

Table 10. Parameters of Equation 14 for the Molar Core Volume

fluid	g_0	g_1	g_2	g_3
octane	161.917	−0.439890	$9.70200 \cdot 10^{-4}$	$-7.6170 \cdot 10^{-7}$
octadecane	285.892	−0.403541	$6.10544 \cdot 10^{-4}$	$-3.7920 \cdot 10^{-7}$
m-xylene	93.602	−0.126664	$2.25422 \cdot 10^{-4}$	$-1.6159 \cdot 10^{-7}$
tetralin	97.978	−0.095833	$1.31890 \cdot 10^{-4}$	$-7.4350 \cdot 10^{-8}$
1-MNP	100.018	−0.095826	$1.11058 \cdot 10^{-4}$	$-4.6500 \cdot 10^{-8}$

Comparison with Literature: Viscosity

The present results for viscosity are plotted in Figures 1 to 5, along with data from the literature, as deviations from eqs 12 to 14. In cases where the original authors did not report the corresponding densities, eqs 1 to 3 were employed to obtain ρ at the specified temperature and pressure.

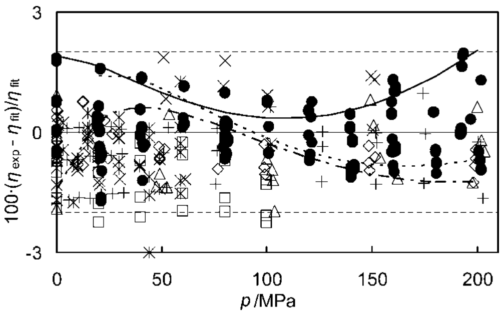

Figure 1. Deviations of viscosity of octane from the substance-specific hard-sphere correlation: ●, this work; Δ, Dymond et al.;[24] ◇, Harris et al.;[22] □, Kashiwagi et al.;[25] *, Badalyan and Rodchenko;[23] +, Oliveira et al.;[21] ×, Tanaka et al.[26] −, Huber et al. $T = 298.15$ K;[20] —•—•—, Huber et al. $T = 373.15$ K;[20] ••••••••, Huber et al. $T = 473.15$ K;[20] and - - - - -, uncertainty of the substance-specific hard-sphere correlation.

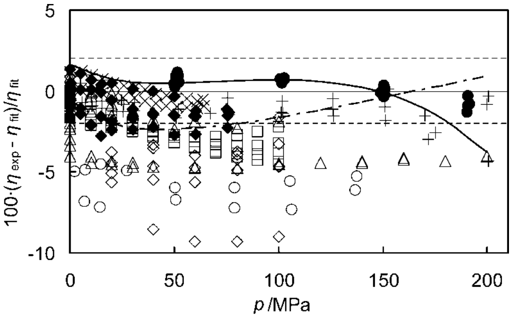

Figure 2. Deviations of viscosity of decane from the substance-specific hard-sphere correlation: ●, this work; □, Kashiwagi et al.;[25] +, Oliveira et al.;[21] ×, Assael et al.;[27] ♦, Audonnet and Paduá;[28] Δ, Naake et al.;[29] ○, Tohidi et al.;[30] ◇, Ducoulombier et al.[31] −, Huber et al. $T = 298.15$ K;[20] —•—•—, Huber et al. $T = 373.15$ K;[20] and - - - - -, uncertainty of the substance-specific hard-sphere correlation.

Octane. The available viscosity data for octane have been critically reviewed by Huber et al.,[20] and in Figure 1, we compare our experimental results with the primary[21−23] and secondary data[24−26] that they selected in the compressed-liquid region. The present viscosities all lie within ± 2 % of the correlation and agree well with most of the available data. In comparison with the correlation reported by Huber et al.,[20] which is partially based on the present data, our results exhibit an absolute average relative deviation of 0.6 % and a maximum absolute relative deviation of 2.7 %.

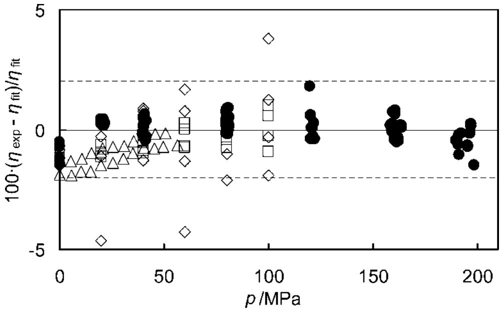

Figure 3. Deviations of viscosity of *m*-xylene from the substance-specific hard-sphere correlation: ●, this work; □, Kashiwagi et al.;[25] Δ, Assael et al.;[32] ◇, Et-Tahir et al.;[33] and - - - - -, uncertainty of the substance-specific hard-sphere correlation.

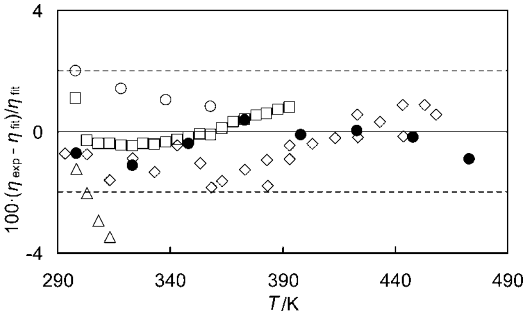

Figure 4. Deviations of viscosity of tetralin at $p = 0.1$ MPa from eq 10: ●, this work; □, Goncalves et al.;[34] ◇, Byers et al.;[35] ○, Oshmyansky et al.;[36] Δ, Aminabhavi et al.;[37] and - - - - -, uncertainty of eq 10.

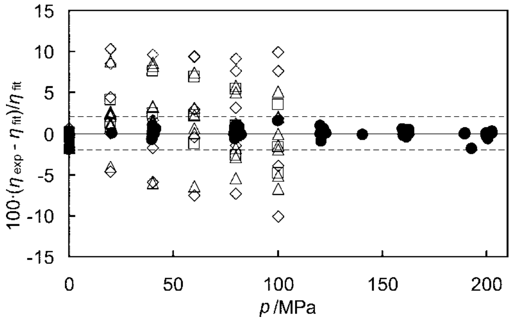

Figure 5. Deviations of viscosity of 1-methylnaphthalene from the substance-specific hard-sphere correlation: ●, this work; ◇, Et-Tahir;[33] □, Baylaucq et al.;[40] Δ, Canet et al.;[41] and - - - - -, uncertainty of the substance-specific hard-sphere correlation.

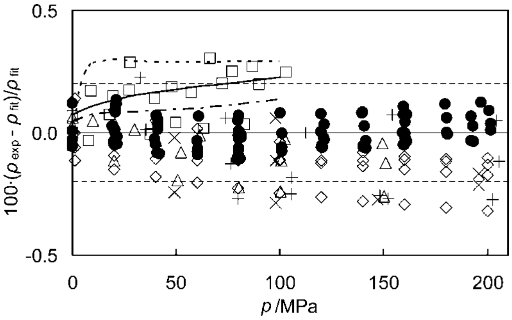

Figure 6. Deviations of density of octane from the modified Tait equation: ●, this work; □, Dix et al.;[42] Δ, Tanaka et al.;[43] ◇, Benson et al.;[44] +, Dymond et al.;[45] ×, Bridgman;[46] −, Span and Wagner $T = 298.15$ K;[50] —•—•—, Span and Wagner $T = 373.15$ K;[50] •••••••, Span and Wagner $T = 473.15$ K;[50] and - - - - -, uncertainty of the modified Tait equation.

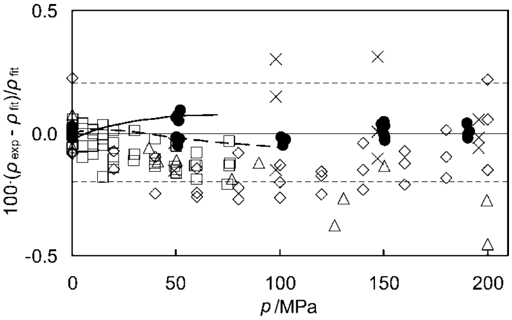

Figure 7. Deviations of density of decane from the modified Tait equation: ●, this work; □, Audonnet and Paduá;[28] Δ, Dymond et al.;[45] ×, Bridgman;[46] ◇, Gehrig and Lentz;[51] −, Lemmon and Span $T = 298.15$ K;[52] —•—•—, Lemmon and Span $T = 373.15$ K;[52] and - - - - -, uncertainty of the modified Tait equation.

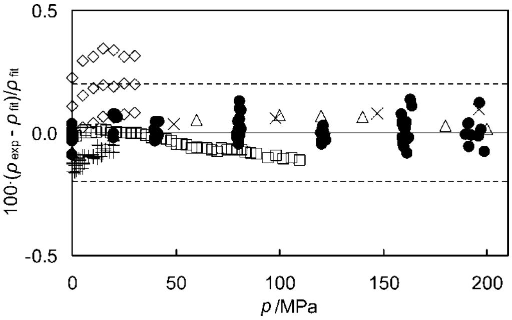

Figure 8. Deviations of density of *m*-xylene from the modified Tait equation: ●, this work; ×, Bridgman;[46] □, Taravillo et al.;[53] +, Chang and Lee;[54] Δ, Takagi;[55] ◇, Chang et al.;[56] and - - - - -, uncertainty of the modified Tait equation.

Decane. The available data for decane have also been critically assessed by Huber et al.,[20] and as shown in Figure 2, the present results are in good agreement with the primary data of Oliveira[21] and Assael[27] and also with the secondary data of Audonnet and Paduá,[28] with relative differences mostly within ± 3 %. The correlation of Huber et al.,[20] again based in part on our data, is also shown. Other workers report results that are systematically lower. The data of Kaskiwagi[25] and Naake[29] exhibit relative deviations of up to −5 %, while those of Tohidi[30] are systematically below our correlation by about 0.06η, and the viscosities reported by Ducoulombier[31] have relative deviations approaching −10 %.

m-Xylene. There are few results in the literature for the viscosity of *m*-xylene at pressures above atmospheric. As shown in Figure 3, the present results agree well with those of Assael[32] and Kashiwagi,[25] while the data of Et-Tahir[33] are more scattered with relative deviation of ± 5 %.

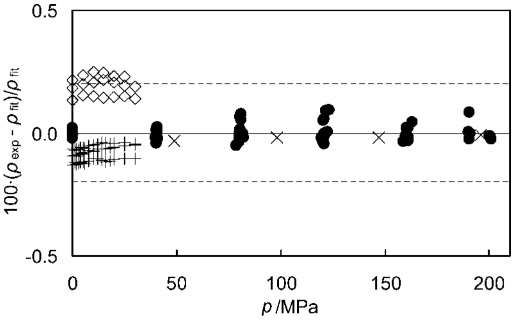

Figure 9. Deviations of density of tetralin from the modified Tait equation: ●, this work; ×, Bridgman;[46] +, Chang and Lee;[54] ◇, Chang et al.;[56] and - - - - -, uncertainty of the modified Tait equation.

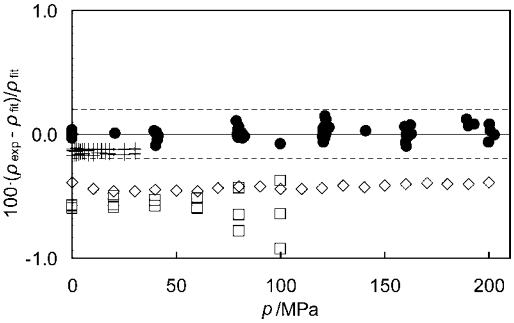

Figure 10. Deviations of density of 1-methylnaphthalene from the modified Tait equation: ●, this work; +, Chang and Lee;[58] ◇, Yokoyama et al.;[59] □, Baylaucq et al.;[40] and - - - - -, uncertainty of the modified Tait equation.

Table 11. Absolute Average Relative Deviations and Maximum Absolute Relative Deviations for Density and Viscosity Correlations

fluid	$10^2\,\Delta_{AAD,\rho}$	$10^2\,\Delta_{MAD,\rho}$	$10^2\,\Delta_{AAD,\eta}$	$10^2\,\Delta_{MAD,\eta}$	$10^2\,\Delta_{AAD,\eta}$	$10^2\,\Delta_{MAD,\eta}$
	eq 1		eq 6		eq 12	
octane	0.05	0.12	0.7	2.3	0.6	2.0
decane	0.03	0.08	0.5	1.0	0.6	1.7
dodecane[a]	0.03	0.12	0.8	2.0	0.5	1.4
octadecane[a]	0.03	0.09	0.3	0.8	0.2	0.8
m-xylene	0.03	0.13	1.1	2.6	0.4	1.8
tetralin	0.02	0.10	1.4	4.1	0.4	1.2
MNP	0.03	0.13	-	-	0.3	2.0

[a] Parameters of eqs 1 and 12 from ref 11.

Tetralin. As far as we are aware, the present results for tetralin at elevated pressures are the first to be reported. Accordingly, we show in Figure 4 a comparison with literature data at atmospheric pressure only. The results reported by Goncalves,[34] which have a claimed relative uncertainty of ± 0.3 %, agree especially well with our data, except at $T = 298.15$ K where there is a relative deviation of 1 %. The results of Byers[35] and Oshmyansky[36] are also in good agreement with our data, but the data of Aminabhavi[37] exhibit relative deviations of as much as −4 %.

1-Methylnaphthalene. The literature data found for the viscosity of 1-MNP at pressures above atmospheric are very limited and were all measured with the falling-body apparatus described by Ducoulombier[38] and Zéberg-Mikkelsen.[39] As shown in Figure 5, these data[33,40,41] exhibit relative deviation of ± 10 % from the present results, although these reduce to ± 2 % at atmospheric pressure. We note that viscosities measured with the same falling-body apparatus for decane and octadecane[31] and for *m*-xylene[33] also show significant relative deviations of 5 % to 10 % in comparison with our present and previous work.[10,11]

Comparison with Literature: Density

The present results for density are plotted in Figures 6 to 10, along with data from the literature, as deviations from eqs 1 to 3.

Octane. Figure 6 shows the results for octane in comparison with selected data from the literature,[42−47] all of which are in relative agreement to within about ± 0.3 %. Here and below, the relative volumes of Bridgman[46,47] were combined with our measured density at $p = 0.1$ MPa to obtain ρ for the compressed liquid. We also consider the recent equation of state of Span and Wagner,[48] and this equation is valid at pressures of up to 100 MPa and shows small positive relative deviations from our results that reach a maximum of about 0.3 % at $T = 473.15$ K and $p = 20$ MPa. Cibulka and Hnědkovský[49] have also carried out a critical evaluation of experimental compressed-liquid densities for *n*-alkanes from C_5 to C_{16}. These authors present a correlation in terms of a modified Tait equation which may be used with Cibulka's[50] correlation of saturated liquid densities to obtain $\rho(T,p)$. For octane, these correlations agree with the present results to within relative deviation of ± 0.13 % at temperatures up to 323.15 K but, at higher temperatures, show larger relative deviations in the range −0.9 % to 0.5 %.

Decane. Densities of decane are compared with the literature[28,45,46,51] in Figure 7, and the relative deviations are generally within ± 0.3 % with most other workers reporting values slightly lower than ours. The recent results of Audonnet and Paduá,[28] which extend in pressure up to 76 MPa, agree closely with ours and show a mean relative deviation (bias) of −0.08 %. The equation of state of Lemmon and Span,[52] which is valid at pressures of up to 100 MPa, agrees very closely with our results and exhibits an absolute average relative deviation of 0.03 % and a maximum absolute relative deviation of 0.08 %. The correlation of Cibulka and Hnědkovský[48] again used with that of Cibulka[49] shows negative relative deviations which generally increase in magnitude with both temperature and pressure reaching about −0.5 % at $T = 373.15$ K and $p = 200$ MPa.

m-Xylene. Densities of compressed liquid *m*-xylene are compared with literature data in Figure 8. The present results are in excellent agreement with those of Bridgman,[46] Travillo,[53] Chang and Lee,[54] and Takagi,[55] the latter estimated from ultrasonic measurements. The data of Chang, Lee, and Lin[56] show significant positive relative deviations reaching a maximum of 0.34 % at $T = 413.15$ K and $p = 20$ MPa.

Tetralin. Figure 9 compared the present results with the few available data from the literature. As with *m*-xylene, our results are in excellent agreement with Bridgman[46] at $T = 298.15$ K. We also observe good agreement with Chang and Lee,[54] while the data of Chang, Lee, and Lin[56] show significant positive deviations. Siddiqi and Teja[57] report densities measured on three isotherms in the range (298 to 338) K at $p < 34.5$ MPa. Their result (not shown in Figure 9) shows the same dependence upon pressure as ours; however, they have relative deviations of −1.1 % at $T = 298$ K and −0.75 % at $T = 318$ K.

1-Methylnaphthalene. The available data for 1-MNP are compared in Figure 10. The results of Chang and Lee[58] agree well with the present data, while those of Yokoyama[59] show a mean relative deviation of −0.43 %. The results of Baylaucq et al.[40] also show large negative relative deviations with a mean of about −0.6 %.

Conclusions

The simultaneous measurements of viscosity and density reported here significantly extend and improve the data situation for these properties of the liquids investigated. We present wide-ranging correlations of both properties in the ranges $298.15 \le T/\mathrm{K} \le 473.15$ K and $p < 200$ MPa. The performance of these correlation schemes is summarized statistically in Table 11. The Tait equation provides an excellent correlation of the experimental densities with maximum absolute deviations that are well within the expanded uncertainty of the measurements. The equation of Comuñas et al.[17] was found to provide a convenient and reasonably accurate representation of viscosity as a function of temperature and pressure for all but one fluid. The hard-sphere scheme[2] is always reliable, more accurate for all but one fluid, and probably more robust in extrapolation. This approach has the further merit of being theoretically based, although it does require the equation of state for practical application. Comparisons between the available literature data are broadly consistent with the claimed mutual uncertainties, but there are some notable exceptions in the case of viscosity.

Acknowledgment

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