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The Viscosity of Five Liquid Hydrocarbons at Pressures Up to 250 MPa

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This paper reports new measurements of the viscosity of toluene, *n*-pentane, *n*-hexane, *n*-octane, and *n*-decane at pressures up to 250 MPa in the temperature range 303 to 348 K. The measurements were performed with a vibrating-wire viscometer and with a relative method of evaluation. Calibration of the instrument was carried out with respect to reference values of the viscosity of the same liquids at their saturation vapour pressure. The viscosity measurements have a precision of $\pm 0.1\%$ but the accuracy is limited by that of the calibration data to be $\pm 0.5\%$. The experimental data have been represented by polynomial functions of pressure for the purposes of interpolation. The data are also used as the most precise test yet applied to a representation of the viscosity of liquids based upon hard-sphere theory.

KEY WORDS: *n*-decane; *n*-hexane; *n*-octane; *n*-pentane; toluene; viscosity.

1. INTRODUCTION

In a series of recent papers [1, 2] the development of a new vibrating-wire viscometer for the measurement of the viscosity of liquids at elevated pressures has been described. The first measurements covered the pressure range up to 80 MPa at temperatures from 303 K to 323 K. In the present work new results are presented for five liquid hydrocarbons, toluene, *n*-pentane, *n*-hexane, *n*-octane, and *n*-decane, that cover a wider pressure range (up to 250 MPa) and temperature range (up to 348 K).

Since the new measurements have a precision of $\pm 0.1\%$ and an accuracy of $\pm 0.5\%$, they provide the most stringent test yet of the ability of representations based upon the hard-sphere model of a liquid to describe experimental viscosity data.

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2. EXPERIMENTAL

The vibrating-wire viscometer employed for the measurements has been described in detail elsewhere [3]. It is therefore sufficient here to record that it was used without change for the present work.

The samples of toluene, *n*-hexane, and *n*-octane were supplied by Merck Ltd. In all cases except for toluene, which was supplied with a

Table I. The Viscosity of Liquid Toluene

Pressure <i>P</i> (MPa)	Frequency ω (rad · s ⁻¹)	Decrement $\Delta \times 10^2$	Density ρ (kg · m ⁻³)	Viscosity μ (mPa · s)
<i>T</i> = 303.15 K				
0.10	5140.26	1.5052	857.8	0.5224
5.22	5135.35	1.5384	861.9	0.5432
10.00	5132.23	1.5732	865.6	0.5636
15.15	5127.78	1.6112	869.4	0.5862
20.38	5124.71	1.6490	873.2	0.6091
25.31	5123.64	1.6858	876.6	0.6321
30.25	5118.12	1.7219	879.9	0.6543
49.07	5112.62	1.8644	891.5	0.7463
74.02	5102.49	2.0419	905.2	0.8748
101.13	5089.50	2.2532	918.3	1.0228
127.73	5081.32	2.4422	929.0	1.1689
151.74	5072.28	2.6469	939.4	1.3352
180.06	5060.07	2.8809	948.7	1.5248
200.63	5053.68	3.0319	956.6	1.6685
225.78	5041.50	3.2377	964.6	1.8564
251.91	5032.09	3.4435	972.5	2.0520
<i>T</i> = 323.15 K				
0.10	5147.31	1.3240	838.9	0.4222
5.24	5146.34	1.3571	843.6	0.4398
10.17	5141.73	1.3904	847.8	0.4576
15.28	5141.97	1.4207	852.0	0.4744
30.31	5135.89	1.5162	863.4	0.5284
50.63	5129.71	1.6501	877.1	0.6085
75.33	5119.62	1.8021	891.5	0.7069
101.50	5108.90	1.9841	905.0	0.8234
125.90	5101.68	2.1444	916.2	0.9402
151.36	5092.83	2.3141	926.9	1.0666
177.22	5088.64	2.4848	936.8	1.2054
200.00	5075.21	2.6412	944.9	1.3360
226.32	5065.73	2.8492	953.6	1.4932
252.04	5059.85	3.0237	961.6	1.6573

Table I. (Continued)

Pressure <i>P</i> (MPa)	Frequency ω (rad · s ⁻¹)	Decrement $\Delta \times 10^2$	Density ρ (kg · m ⁻³)	Viscosity μ (mPa · s)
<i>T</i> = 348.15 K				
0.10	5160.25	1.1501	814.8	0.3386
5.16	5157.73	1.1887	820.1	0.3528
10.95	5158.12	1.2174	825.9	0.3687
16.43	5154.87	1.2500	831.0	0.3831
20.33	5152.10	1.2701	834.5	0.3930
25.07	5149.59	1.3001	838.6	0.4065
31.89	5145.67	1.3388	844.2	0.4267
51.12	5137.26	1.4421	858.4	0.4844
75.20	5130.80	1.5761	873.8	0.5614
100.52	5124.40	1.7181	887.9	0.6482
123.95	5115.81	1.8452	899.6	0.7294
150.71	5106.83	1.9923	911.4	0.8331
179.44	5102.99	2.1725	923.2	0.9527
199.47	5093.25	2.2692	930.7	1.0374
225.64	5087.32	2.4355	939.8	1.1560
251.16	5078.98	2.5957	948.1	1.2842

stated purity of 99.95%, the samples were distilled and degassed before use and the subsequent purity was confirmed as better than 99.99%. The sample of *n*-pentane was supplied by Fluka Chemika with a purity of better than 99.5% and was used without further purification. The sample of *n*-decane was supplied by Rose Chemicals and had a purity of 99.0% after distillation.

As described in our earlier work [3], the viscometer was calibrated with respect to the viscosity data of toluene, *n*-hexane, *n*-heptane, *n*-octane, and *n*-decane reported by Gonçalves *et al.* [4], and Knapstad *et al.* [5] under saturation conditions at the lowest temperature studied. Accounting for the precision of the present viscosity measurements, the uncertainty contributed by the density of the fluid and the estimated error in the calibration data, the accuracy of the present viscosity data is estimated to be $\pm 0.5\%$.

In the evaluation of the viscosity the density of toluene was taken from the work of Kashiwagi *et al.* [6] and has an estimated uncertainty of $\pm 0.1\%$. For *n*-pentane we used the data of Eastcal and Woolf [7], which have an error of $\pm 0.2\%$; for *n*-hexane, *n*-octane, and *n*-decane we used the data of Dymond and his collaborators [8, 9].

3. RESULTS

Tables I to V list the viscosity data for each liquid at pressures up to 250 MPa along three isotherms, 303.15, 323.15, and 348.15 K, except for the case of *n*-pentane, where the lack of density data at the highest temperatures confined the study to the lowest two isotherms. The tables include the values of the frequency and decrement of the vibrating wire for each fluid since, should more accurate calibration viscosity data become

Table II. The Viscosity of Liquid *n*-Pentane

Pressure <i>P</i> (MPa)	Frequency ω (rad · s ⁻¹)	Decrement $\Delta \times 10^2$	Density ρ (kg · m ⁻³)	Viscosity μ (mPa · s)
<i>T</i> = 303.15 K				
0.10	5201.62	0.7899	616.3	0.2074
5.55	5198.49	0.8206	623.1	0.2208
10.57	5199.86	0.8458	628.8	0.2317
15.12	5195.05	0.8712	633.8	0.2426
19.72	5194.49	0.8965	638.5	0.2549
30.39	5189.73	0.9531	648.4	0.2810
48.96	5183.61	1.0382	663.3	0.3233
75.93	5175.61	1.1656	681.0	0.3905
102.67	5167.10	1.2823	695.6	0.4566
125.20	5164.64	1.3735	706.2	0.5112
151.39	5154.89	1.4830	717.4	0.5797
179.19	5149.95	1.6000	728.1	0.6558
201.96	5138.32	1.6925	736.3	0.7188
225.43	5135.09	1.8133	744.3	0.8056
251.30	5132.78	2.0036	752.6	0.9534
<i>T</i> = 323.15 K				
5.21	5217.09	0.7270	603.2	0.1811
10.14	5211.63	0.7548	609.9	0.1923
15.09	5208.21	0.7790	616.1	0.2025
20.22	5206.45	0.8042	622.1	0.2137
29.73	5203.41	0.8467	632.3	0.2330
50.06	5195.93	0.9384	650.1	0.2754
74.78	5189.84	1.0410	667.7	0.3272
100.96	5181.39	1.1438	682.9	0.3824
125.27	5175.35	1.2301	695.1	0.4301
151.24	5169.96	1.3254	706.8	0.4872
176.25	5165.63	1.4117	717.0	0.5404
200.73	5154.50	1.4951	726.3	0.5914
226.12	5149.13	1.5760	735.5	0.6441
251.60	5141.56	1.6565	744.2	0.6978

Table III. The Viscosity of Liquid *n*-Hexane

Pressure <i>P</i> (MPa)	Frequency ω (rad · s ⁻¹)	Decrement $\Delta \times 10^2$	Density ρ (kg · m ⁻³)	Viscosity μ (mPa · s)
<i>T</i> = 303.15 K				
0.10	5195.05	0.9529	650.4	0.2809
5.05	5204.75	0.9843	655.6	0.2970
10.22	5198.53	1.0044	660.6	0.3126
19.95	5194.64	1.0777	669.7	0.3443
40.03	5189.27	1.1949	685.2	0.4073
51.29	5181.61	1.2589	691.9	0.4450
60.12	5183.07	1.3069	698.1	0.4732
80.15	5172.35	1.4182	709.4	0.5429
100.01	5169.77	1.5293	719.3	0.6129
125.00	5154.57	1.6776	730.4	0.7133
148.99	5152.11	1.8083	739.9	0.8061
201.09	5130.26	2.1087	757.8	1.0374
249.12	5112.50	2.4082	771.9	1.2882
<i>T</i> = 323.15 K				
0.10	5212.30	0.8509	632.1	0.2338
5.95	5207.35	0.8820	639.4	0.2486
10.09	5204.14	0.9100	644.0	0.2605
20.20	5199.59	0.9709	654.3	0.2877
39.93	5191.87	1.0772	671.5	0.3432
60.06	5182.26	1.1836	685.6	0.4005
80.10	5175.09	1.2803	697.6	0.4584
100.06	5177.88	1.3823	708.2	0.5176
129.07	5167.03	1.5231	721.6	0.6068
181.05	5148.18	1.7817	741.6	0.7845
200.13	5138.61	1.8825	748.0	0.8582
250.02	5128.64	2.1407	763.2	1.0598
<i>T</i> = 348.15 K				
0.10	5231.71	0.7640	607.5	0.1899
5.01	5229.80	0.7748	616.0	0.2017
10.11	5226.66	0.8044	622.9	0.2136
20.03	5216.83	0.8617	635.1	0.2366
39.02	5211.63	0.9598	653.7	0.2824
60.08	5205.20	1.0586	670.2	0.3317
79.93	5192.36	1.1488	683.3	0.3809
100.07	5180.07	1.2369	694.7	0.4294
125.12	5171.84	1.3483	707.1	0.4937
149.03	5174.81	1.4461	717.7	0.5556
177.15	5165.14	1.5683	728.7	0.6346
199.99	5168.01	1.6612	737.0	0.6983
250.02	5142.92	1.8829	752.9	0.8555

available in the future, it is a simple matter to reevaluate the viscosity data given here.

The viscosity of each fluid along each isotherm has been represented as a function of pressure by means of an equation of the form

$$\mu = \sum_{i=0}^n a_i (P/P')^{i/2} \quad (1)$$

Table IV. The Viscosity of Liquid *n*-Octane

Pressure <i>P</i> (MPa)	Frequency ω (rad · s ⁻¹)	Decrement $\Delta \times 10^2$	Density ρ (kg · m ⁻³)	Viscosity μ (mPa · s)
<i>T</i> = 303.15 K				
0.10	5177.10	1.3171	694.7	0.4808
5.62	5174.69	1.3645	699.5	0.5097
10.52	5172.15	1.4071	703.4	0.5363
15.81	5170.12	1.4522	707.5	0.5650
21.11	5166.66	1.4983	711.4	0.5949
26.71	5163.98	1.5461	715.3	0.6265
31.84	5161.28	1.5898	718.8	0.6559
52.36	5149.44	1.7657	731.3	0.7794
75.38	5137.62	1.9656	743.5	0.9293
101.66	5124.87	2.2014	755.7	1.1188
126.10	5113.88	2.4288	765.8	1.3136
152.00	5102.14	2.6826	775.5	1.5443
174.70	5094.24	2.9187	783.2	1.7718
202.71	5077.00	3.2254	792.1	2.0805
226.48	5065.51	3.5027	799.0	2.3757
253.07	5049.59	3.8371	806.3	2.7483
<i>T</i> = 323.15 K				
0.18	5188.76	1.1570	678.4	0.3875
5.12	5185.59	1.1968	683.6	0.4097
10.38	5180.41	1.2386	688.5	0.4333
15.92	5177.31	1.2818	693.3	0.4585
20.86	5175.17	1.3197	697.3	0.4810
25.95	5172.13	1.3582	701.3	0.5043
31.26	5170.42	1.3983	705.2	0.5291
50.75	5161.46	1.5429	718.3	0.6220
76.38	5151.13	1.7323	732.9	0.7525
101.79	5140.08	1.9238	745.4	0.8937
126.51	5129.73	2.1132	756.1	1.0422
152.31	5120.08	2.3198	766.1	1.2140
174.48	5110.79	2.5227	774.7	1.3919
201.53	5096.98	2.7403	782.9	1.5910
226.70	5086.65	2.9691	790.5	1.8117
251.85	5074.30	3.2079	797.6	2.0518

Table IV. (Continued)

Pressure <i>P</i> (MPa)	Frequency ω (rad · s ⁻¹)	Decrement $\Delta \times 10^2$	Density ρ (kg · m ⁻³)	Viscosity μ (mPa · s)
<i>T</i> = 348.15 K				
0.23	5203.23	1.0009	658.0	0.3051
5.11	5198.87	1.0377	663.8	0.3235
10.55	5193.82	1.0775	669.7	0.3440
15.60	5192.79	1.1134	674.8	0.3630
20.67	5189.23	1.1489	679.6	0.3821
25.85	5185.92	1.1840	684.3	0.4014
30.68	5183.95	1.2861	688.3	0.4193
53.30	5173.79	1.3639	705.1	0.5062
76.42	5165.21	1.5105	719.4	0.5986
101.81	5156.10	1.6824	732.7	0.7101
126.31	5148.16	1.8258	744.0	0.8170
152.08	5138.67	1.9928	754.6	0.9429
175.28	5131.30	2.1469	763.2	1.0652
200.64	5121.20	2.3196	771.8	1.2082
225.83	5113.07	2.4978	779.8	1.3632
247.41	5103.87	2.6560	786.1	1.5057

Table V. The Viscosity of Liquid *n*-Decane

Pressure <i>P</i> (MPa)	Frequency ω (rad · s ⁻¹)	Decrement $\Delta \times 10^2$	Density ρ (kg · m ⁻³)	Viscosity μ (mPa · s)
<i>T</i> = 303.15 K				
0.10	5148.37	1.7733	722.5	0.7913
4.67	5145.05	1.8300	726.1	0.8333
10.24	5141.04	1.8983	730.3	0.8848
15.60	5138.02	1.9664	734.1	0.9376
20.53	5133.73	2.0285	737.5	0.9864
25.43	5131.32	2.0912	740.7	1.0374
30.66	5126.84	2.1574	743.9	1.0915
51.34	5114.88	2.4249	755.6	1.3207
75.49	5098.34	2.7534	767.4	1.6235
101.64	5082.63	3.1334	778.7	2.0016
125.82	5066.95	3.5083	788.1	2.4004
152.44	5047.77	3.9478	797.7	2.8972
175.19	5032.80	4.4417	805.6	3.4985
199.87	5010.12	4.9011	813.9	4.0786
238.79	4976.77	5.8090	827.0	5.3165

Table IV. (Continued)

Pressure P (MPa)	Frequency ω (rad \cdot s $^{-1}$)	Decrement $\Delta \times 10^2$	Density ρ (kg \cdot m $^{-3}$)	Viscosity μ (mPa \cdot s)
$T = 323.15$ K				
0.38	5166.17	1.5223	707.3	0.6141
5.11	5162.93	1.5915	711.5	0.6447
10.22	5160.74	1.6286	715.7	0.6863
15.30	5156.04	1.6832	719.7	0.7246
20.31	5151.96	1.7357	723.5	0.7620
25.71	5146.86	1.7940	727.3	0.8038
30.80	5146.53	1.8457	730.8	0.8424
52.02	5134.77	2.0729	743.8	1.0189
76.20	5122.85	2.3389	756.5	1.2419
101.94	5106.22	2.6320	768.2	1.5036
126.26	5090.72	2.9297	778.0	1.7882
151.39	5077.31	3.2599	787.4	2.1249
171.74	5067.46	3.5651	794.5	2.4554
200.56	5050.02	4.0606	804.1	2.9911
231.80	5029.95	4.5841	814.1	3.6645
$T = 348.15$ K				
0.24	5181.44	1.2847	687.9	0.4628
5.85	5174.20	1.3383	693.6	0.4948
10.41	5171.92	1.3811	698.0	0.5211
15.44	5171.17	1.4271	702.6	0.5501
20.71	5165.59	1.4759	707.1	0.5809
25.30	5165.57	1.5160	710.9	0.6073
30.55	5161.96	1.5621	715.0	0.6377
50.80	5152.25	1.7441	729.2	0.7640
67.31	5144.42	1.8861	739.1	0.8682
101.39	5128.63	2.1919	756.4	1.1099
125.99	5118.57	2.4186	766.9	1.3035
151.00	5106.57	2.6612	776.3	1.5226
169.93	5099.42	2.8643	782.9	1.7167
200.80	5083.04	3.1768	792.8	2.0296
224.76	5071.70	3.4357	799.9	2.2994
254.37	5052.34	3.7990	808.3	2.6990

The coefficients a_i and the scaling parameter P' for each isotherm and each fluid are listed in Table VI. The fitting was performed with the algorithm SEEQ [10] so as to select only those terms in Eq. (1) that are statistically significant.

The representation of the present experimental data secured by Eq. (1) and the coefficients in Table VI are compared with both the present data and the results of earlier measurements in Figures 1 to 5.

Table VI. Coefficients of the Correlation for the Pressure Dependence of the Viscosity with Eq. (1)

T (K)	P' (MPa)	a_0 (MPa \cdot s)	a_1	a_2	a_3	a_4
Toluene						
303.15	126.0	0.5201	—	0.5358	—	0.1162
323.15	126.1	0.4227	—	0.4170	—	0.1001
348.15	125.6	0.3394	—	0.3241	—	0.0735
<i>n</i> -Pentane						
303.15	125.7	0.2079	—	0.2076	—	0.0785
323.15	128.4	0.1707	—	0.2155	—	—
<i>n</i> -Hexane						
303.15	124.6	0.2795	—	0.4477	−0.1661	0.1456
323.15	125.1	0.2315	—	0.4033	−0.1573	0.1166
348.15	125.1	0.1888	—	0.3363	−0.1049	0.0726
<i>n</i> -Octane						
303.15	126.6	0.4699	—	0.9436	−0.6082	0.5690
323.15	126.0	0.3890	—	0.8452	−0.5192	0.3803
348.15	123.8	0.3027	—	0.4927	−0.1975	0.1584
<i>n</i> -Decane						
303.15	119.4	0.8034	−0.3515	2.7486	−2.7059	1.7910
323.15	116.1	0.6285	−0.2660	2.0309	−1.9644	1.2308
348.15	127.3	0.4658	−0.0920	1.1656	−0.7885	0.5647

For toluene, Gonçalves *et al.* [4] have made measurements under saturation conditions within the lower temperature range studied here using a capillary viscometer. Krall [11] has made measurements over the entire temperature range at pressures up to 20 MPa with an oscillating-disk instrument, Kashiwagi and Makita [12] have made measurements at pressures up to 100 MPa with a torsional crystal viscometer and have presented a correlation of their data, Assael *et al.* [13] have employed a vibrating-wire viscometer for measurements to 80 MPa, while Dymond *et al.* [14] have made measurements to 500 MPa using a falling-body viscometer.

Figure 1 shows that at the lowest temperature the available data are all consistent within $\pm 1\%$, which represents the limit of the mutual uncertainty. For the data of Assael *et al.* [13] and Kashiwagi and Makita [12] there seems to be a systematic trend away from the present results but it is still quite small. A similar pattern emerges at higher temperatures, and

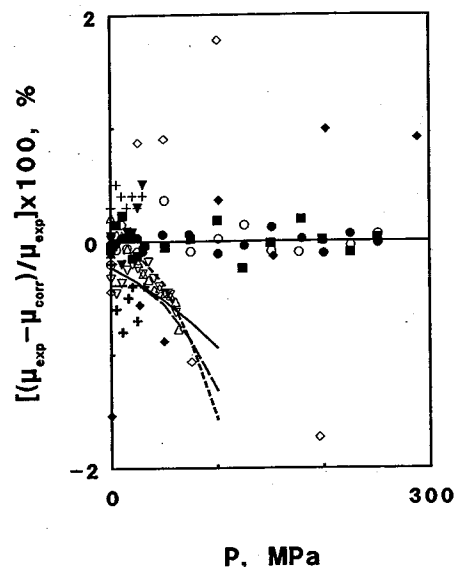


Fig. 1. Deviations of viscosity data for toluene from the optimum representation of the present results as a function of pressure. Present work—●, 303.15 K; ○, 313.15 K; ■, 348.15 K. Earlier work—303.15 K: — [12], + [11], △ [13], and △ [4]; 323.15 K: — [12], ▼ [11], ▽ [13], ◇ [14], and □ [4]; 348.15 K: --- [12], + [11], and ◆ [14].

although the data of Dymond *et al.* [14] are rather scattered, the data are consistent with the present results within the mutual uncertainty of $\pm 2.5\%$. The agreement with the precise work of Gonçalves *et al.* [4] and Krall *et al.* [11] (within $\pm 0.5\%$) is particularly gratifying given that three different instruments were employed for the measurements.

For *n*-pentane (for which the deviations are shown in Fig. 2), only Collings and McLaughlin [15], with a torsional crystal viscometer, and Brazier and Freeman [16], with a rolling-ball instrument, seem to have reported viscosity measurements on this fluid. Figure 2 shows that the results of these earlier studies depart markedly (by up to 5%) from those reported here. The present data are to be preferred owing to their higher accuracy.

For *n*-hexane, Fig. 3 shows the deviations of earlier results from the representation of the present data. At the lowest two temperatures, the

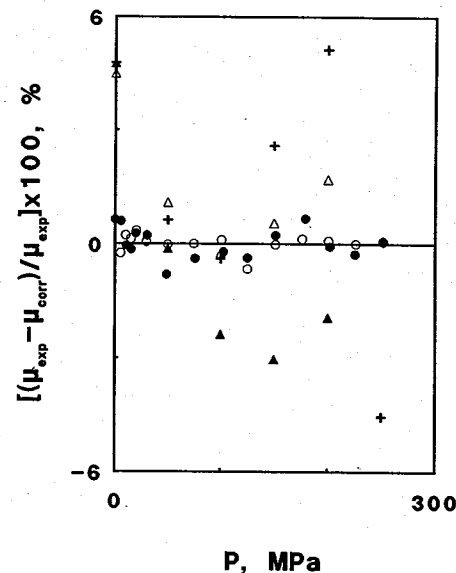


Fig. 2. Deviations of viscosity data for *n*-pentane from the optimum representation of the present results as a function of pressure. Present work—●, 303.15 K; ○, 323.15 K. Earlier work—303.15 K: △ [15] and + [16]; 323.15 K: ▲ [15].

results of Kashiwagi and Makita [12] show a systematic deviation from the present work, similar to that observed for toluene. However, the magnitude of the deviation is still comparable with the mutual uncertainty in the data. The precise measurements of Knapstad *et al.* [5] at saturation are in satisfactory agreement ($\pm 0.5\%$) with the present data at all three temperatures. The results of Isdale *et al.* [17] with the falling-body viscometer lie up to 5% below the present data.

The deviations of the earlier work on *n*-octane from the present results are shown in Fig. 4. Once again, the correlation of the experimental data given by Kashiwagi and Makita [12] shows a systematic pressure dependence different than observed here but the magnitude of the deviation does not exceed $\pm 2\%$. The data of Dymond *et al.* [9] show an opposing systematic trend with pressure in this case but are consistent with the present results within their estimated error. The data of Knapstad *et al.* [5] at saturation are consistent with the present values within $\pm 0.5\%$.

Finally, Fig. 5 shows the results for *n*-decane. Here the results of

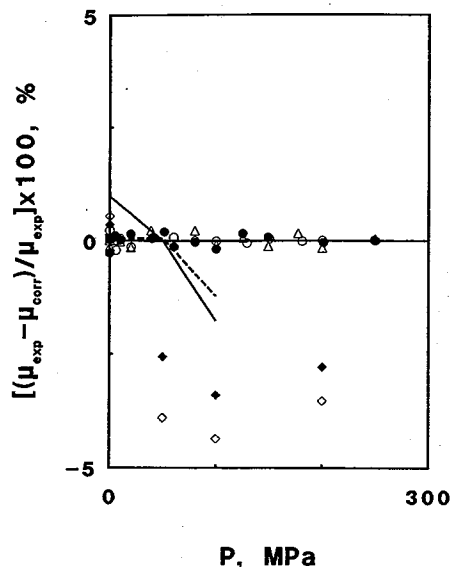


Fig. 3. Deviations of viscosity data for *n*-hexane from the optimum representation of the present results as a function of pressure. Present work—●, 303.15 K; ○, 323.15 K; △, 348.15 K. Earlier work—303.15 K: — [12] and □ [5]; 323.15 K: — [12], ■ [5], and ◇ [17]; 348.15 K: ▼ [5] and ♦ [9].

Kashiwagi and Makita [12] show a different trend and differ by more than 2% from the present results at the lower temperatures. On the other hand, the results of Dymond *et al.* [8] and Knapstad *et al.* [5] at saturation are consistent with the present values at each temperature.

These comparisons, particularly those with the most accurate earlier measurements near saturation, support the contention that the accuracy of the present results is $\pm 0.5\%$ over the complete range of conditions studied.

4. DENSITY DEPENDENCE OF THE VISCOSITY

The most valuable means of representing the viscosity of liquid hydrocarbons has proved to be based upon a rigid-sphere model of the fluid behavior. First, it has been shown on a number of occasions [18, 19] that this procedure allows the viscosity of a hydrocarbon to be estimated

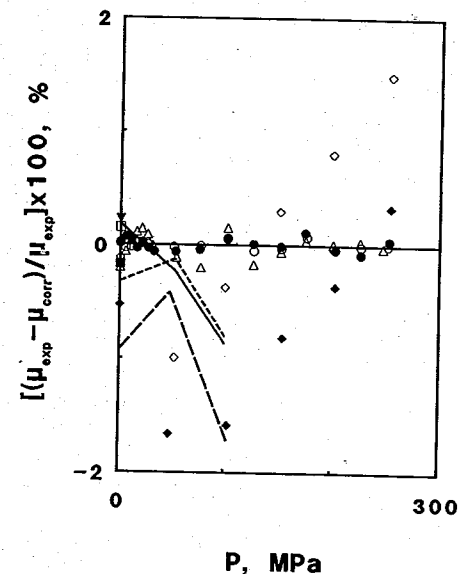


Fig. 4. Deviations of viscosity data for *n*-octane from the optimum representation of the present results as a function of pressure. Present work—●, 303.15 K; ○, 323.15 K; △, 348.15 K. Earlier work—303.15 K: — [12] and □ [5]; 323.15 K: — [12], ▼ [5], and ◇ [9]; 348.15 K: — [12], ■ [5], and ♦ [9].

in a range of states beyond that in which it was measured experimentally. Second, and more recently, a correlation scheme based on the same ideas has made it possible to estimate values of one transport property from measurements of another [20] and for one fluid from those of another.

The present experimental data, being both more precise and more accurate than earlier results, provide an opportunity to test these ideas in a more stringent way. The basis of the representation based upon the hard-sphere model was developed by Dymond and Brawn [21] and makes use of the experimental quantity

$$\mu' = 9.118 \times 10^7 \mu V^{2/3} / (MRT)^{1/2} \quad (2)$$

Here, $\mu'(V, T)$, the viscosity at temperature T and molar volume V , is measured in cP, V is the molar volume in $\text{cm}^3 \cdot \text{g}^{-1}$, M the molar mass of the substance, and R the universal gas constant.

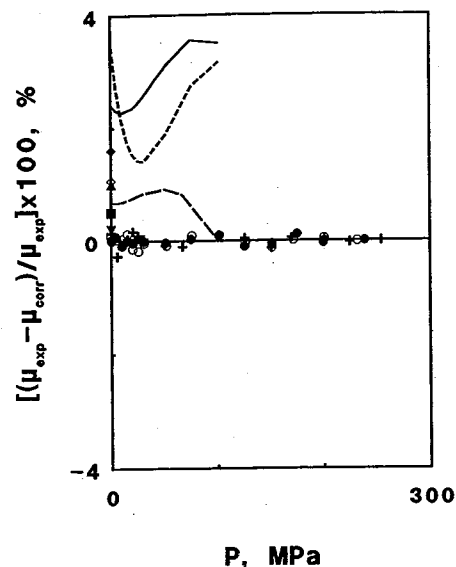


Fig. 5. Deviations of viscosity data for *n*-octane from the optimum representation of the present results as a function of pressure. Present work—●, 303.15 K; ○, 323.15 K; +, 348.15 K. Earlier work—303.15 K: — [12], ◇ [8], and □ [5]; 323.15 K: — [12], ◆ [8], and [5]; 348.15 K: — [12], ▲ [8], and ▼ [5].

According to the hypothesis of Dymond and Bawn's representation, μ' is a function only of (V/V_0) for a particular liquid, where V_0 is a weakly temperature-dependent characteristic volume. We have tested this hypothesis with the aid of the present data on the viscosity of normal alkanes and toluene as well as the results for *n*-heptane reported previously. For this purpose, we have represented the viscosity along all isotherms for a particular fluid by the equation

$$\ln \mu' = \sum_{i=0}^n b_i \left[\frac{V_0}{V - V_0} \right]^i \quad (3)$$

The parameter V_0 was determined for each isotherm by superposition relative to a reference value at 303.15 K so that the value of the ratio $V_0(T)/V_0(303.15 \text{ K})$ is determined for each fluid. The optimum values of

Table VII. Coefficients in the Representation of the Density Dependence of the Viscosity with the Aid of Eq. (3)

Fluid	b_0	b_1	b_2	b_3	b_4
Toluene	-0.208	0.645	-0.054	0.002	-0.00004
<i>n</i> -Pentane	-1.696	3.181	-0.437	—	—
<i>n</i> -Hexane	-1.053	2.120	-0.007	—	—
<i>n</i> -Octane	-0.842	1.861	-0.067	—	—
<i>n</i> -Decane	-0.510	1.473	-0.012	—	—

Fluid	$10^6 V_0 (\text{m}^3 \cdot \text{mol}^{-1})$ at T (K)		
	303.15 ^a	323.15	348.15
Toluene	89.80	88.81	88.09
<i>n</i> -Pentane	62.76	61.94	—
<i>n</i> -Hexane	72.92	72.41	71.75
<i>n</i> -Octane	97.88	97.00	96.02
<i>n</i> -Decane	133.86	132.52	130.78

^a Reference values taken from Assael *et al.* [22].

the coefficients b_i as well as the values of V_0 for the fluids studied are listed in Table VII.

Figures 6a to e show the deviations of the present data from these fits to individual fluids. In examining these plots it is necessary to emphasize that whereas the precision of the viscosity itself determined in the present work is $\pm 0.2\%$, the projected error in the viscosity arising from an error of $\pm 0.2\%$ in the density is up to $\pm 2.4\%$ at the highest viscosities. Since the figure of $\pm 0.2\%$ is typical of the uncertainties in the density values, it follows that only deviations of viscosity data from universality as a function of density outside of that range can be considered to disprove the original hypothesis. Figures 6a to e show that none of the fluids reveal discrepancies exceeding $\pm 1.5\%$ and that they are frequently much less. There is therefore substantial evidence that the hypothesis of a temperature-independent behavior of $\mu'(V/V_0)$ is valid.

5. CORRELATION

Assael *et al.* [22] have made use of observations similar to those reported above to develop a correlation/prediction scheme for all of the transport properties of liquid *n*-alkanes. Necessarily, because their development was more general and based upon data of poorer accuracy in some

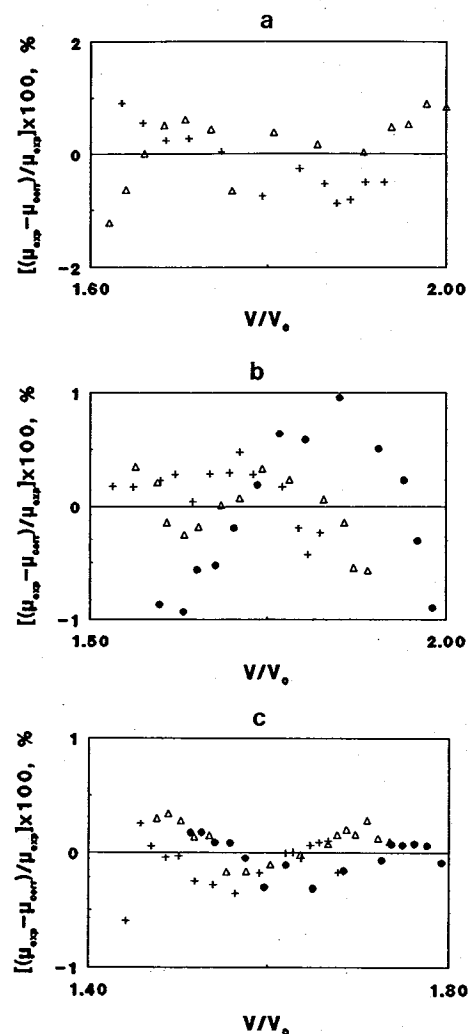


Fig. 6. Deviations of the viscosity data for the five fluids from the temperature-independent representation of the density dependence according to Eq. (3). (a) *n*-Pentane: +, 303.15 K; Δ, 323.15 K. (b) *n*-Hexane: +, 303.15 K; Δ, 323.15 K; ●, 348.15 K. (c) *n*-Octane: +, 303.15 K; Δ, 323.15 K; ●, 348.15 K. (d) *n*-Decane: +, 303.15 K; Δ, 323.15 K; ●, 348.15 K. (e) Toluene: +, 303.15 K; Δ, 323.15 K; ●, 348.15 K.

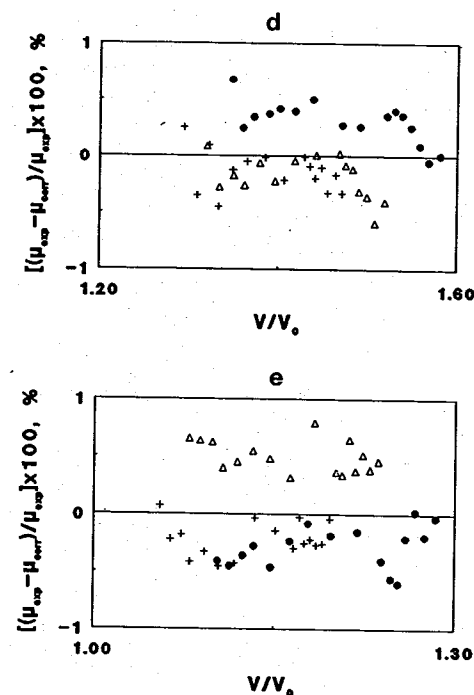


Fig. 6. (Continued)

cases, the accuracy of their procedure is estimated to be no better than $\pm 5\%$. We have tested their prediction scheme against the present viscosity data. Generally, the agreement is within the claimed uncertainty of the representation of Assael *et al.* However, for *n*-pentane the deviations amount to $\pm 15\%$. This is because at the time that the representation was developed, there were no reliable viscosity data for *n*-pentane available. The present data therefore provide the information necessary to improve the representation of Assael *et al.* [22].

6. CONCLUSIONS

New viscosity data for five hydrocarbons over the pressure range up to 250 MPa and temperatures in the range 303 to 348 K have been determined. The precision of the viscosity data is estimated to be $\pm 0.1\%$, although the accuracy of the results is limited to $\pm 0.5\%$. The results have allowed confirmation of the temperature-independent representation of the density dependence of the viscosity with a higher degree of accuracy than

has been possible hitherto. As a means of reliably extending the range of thermodynamic states over which the viscosity of the hydrocarbons can be evaluated, the procedure is remarkably accurate.

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Measurements of the Viscosity of Toluene + Mesitylene Mixtures at Pressures Up to 55 MPa

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New absolute measurements of the viscosity of mesitylene and binary mixtures of toluene + mesitylene are presented. The measurements were performed in a vibrating-wire instrument and cover a temperature range of 295–330 K and pressures up to 55 MPa. The concentrations studied were 40 and 70%, by weight, of toluene. The overall uncertainty in the reported data is estimated to be $\pm 0.5\%$. A recently extended semiempirical scheme for the prediction of the viscosity of mixtures from the pure components is used to predict successfully the viscosity of these mixtures, as a function of composition, temperature, and pressure.

KEY WORDS: high pressure; mesitylene; mixtures; toluene; viscosity.

1. INTRODUCTION

In a series of recent papers, a semiempirical scheme based on considerations of the hard-sphere theory of transport properties has been applied for the correlation and prediction of the liquid-phase transport properties of simple molecular fluids [1], *n*-alkanes [2, 3], *n*-alkane mixtures [4], and aromatic hydrocarbons [5]. For the pure liquids it has been shown that the thermal conductivity, viscosity, and diffusion coefficients can, simultaneously, be successfully correlated over a temperature range of 100–400 K and pressures up to 600 MPa, with an uncertainty of $\pm 6\%$. In the case of *n*-alkane mixtures it has further been demonstrated that, by adopting a simple mixing rule, the thermal conductivity and viscosity can

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Measurements of the Viscosity of Benzene, Toluene, and *m*-Xylene at Pressure up to 80 MPa

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New absolute measurements of the viscosity of benzene, toluene, and *m*-xylene are presented. The measurements were performed in a recently developed vibrating-wire instrument, at temperatures of 303.15 and 323.15 K and pressures up to 80 MPa. The overall uncertainty in the reported viscosity data is estimated to be $\pm 0.5\%$.

KEY WORDS: benzene; high pressure; toluene; viscosity; vibrating wire; *m*-xylene.

1. INTRODUCTION

In recent years semiempirical schemes, based on the exact hard-sphere theory of transport properties, have been applied to the correlation and prediction of these properties. In a recent paper [1], the thermal conductivity, the viscosity, and the self-diffusion coefficient of liquid *n*-alkanes were satisfactorily simultaneously correlated with an uncertainty of $\pm 5\%$, and universal curves were developed for the prediction of these properties in a wide range of temperatures and pressures. In order to extend the applicability of this scheme to other groups of liquids, accurate high-pressure measurements of the viscosity are essential.

In a previous paper [2], a new vibrating-wire instrument for the measurement of the viscosity of liquids at high pressures was described. The instrument, based on a complete theory, permits the absolute measurements of the viscosity with an estimated uncertainty less than

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$\pm 0.5\%$. In this paper, absolute measurements of the viscosity of benzene, toluene, and *m*-xylene at 303.15 and 323.15 K and up to 80 MPa pressure are presented.

2. EXPERIMENTAL

According to the ideal model of the technique, a thin vertical wire, constrained not to move at its two ends, is placed in the liquid. If an oscillation is initiated by the displacement of the wire from its equilibrium position then, following a short-lived initial transient, the motion will conform to a damped, simple harmonic motion [2, 3],

$$y = y_0 e^{(i-\Delta)\omega t} \quad (1)$$

with an angular frequency ω and a logarithmic decrement Δ . In circumstances when the length of the wire is much larger than its radius, and provided that the amplitude of the motion is sufficiently small, it has been shown that it is possible to linearize the Navier-Stokes equations and relate the viscosity, μ , and density, ρ , of the fluid to the values of ω and Δ and to their corresponding values *in vacuo*, ω_0 and Δ_0 . In these circumstances, coupled with a distinct set of design criteria, the working equation for the viscometer, can be written [2, 3] as

$$\Delta = \frac{(\rho/\rho_s)k' + 2\Delta_0}{2[1 + (\rho/\rho_s)k]} \quad (2)$$

In this equation ρ_s denotes the density of the wire material, while the quantities k and k' are given by

$$k = -1 + 2 \operatorname{Im}(A) \quad (3)$$

and

$$k' = 2 \operatorname{Re}(A) + 2\Delta \operatorname{Im}(A) \quad (4)$$

where

$$A = (i - \Delta) \left[1 + \frac{2K_1[(i - \Delta)\Omega]^{1/2}}{[(i - \Delta)\Omega]^{1/2} K_0[(i - \Delta)\Omega]^{1/2}} \right] \quad (5)$$

and

$$\Omega = \frac{\rho\omega R^2}{\mu} \quad (6)$$

In the above equations, K_0 and K_1 are modified Bessel functions. Equations (1)–(6) represent a consistent set from which the viscosity of a liquid can be calculated from the frequency of oscillation, ω , the logarithmic decrement, Δ , in the liquid, and the logarithmic decrement, Δ_0 , *in vacuo*.

The actual instrument used for these measurements has been described in detail elsewhere [2] and is presented only briefly here. A 100- μm -diameter, 50-mm-length, tungsten wire was used as the vibrating wire. Constant tensioning of the wire was accomplished by a two-weight assembly at the bottom of the wire. The volumes of the two weights (the inner one is made from tungsten and the outer one from stainless steel) are chosen so that the net effect of changing the density of the surrounding liquid results in a negligible effect upon the frequency of oscillation. Two permanent magnets, made from samarium-cobalt and placed around the wire, are used for the electromagnetic initiation of the oscillations of the wire and their detection. Following initiation of the motion, the signal induced in the vibrating wire is observed with a bridge in which the wire forms one arm. The out-of-balance signal, amplified by 30,000 times, is then observed with an A/D converter coupled to a microcomputer. This configuration enables sampling of the oscillation signal at a rate of 50 kHz with a resolution of 12 bits. The values of the logarithmic decrement and the frequency of oscillation are then obtained from the recorded data with the aid of a nonlinear least-squares fitting procedure.

The samples of benzene and *m*-xylene used for these measurements were supplied by B.D.H. Ltd. with nominal purities of 99.8 and 99.0%, respectively, while the sample of toluene was supplied by Merck with a nominal purity of 99.5%.

3. RESULTS

As described in a previous paper [2], the uncertainty of the instrument was found to be $\pm 0.5\%$. In order to check the continuing good operation of the instrument, the viscosity of toluene was measured at atmospheric pressure as a function of temperature (Table I), before and after each set of measurements. The values obtained were found to agree with those obtained by Gonçalves et al. [4] (in an absolute Ubbelohde capillary viscometer with an uncertainty of $\pm 0.3\%$) within $\pm 0.1\%$, which is well within the mutual uncertainty of both instruments.

In Tables II, III, and IV the present measurements of the viscosity of benzene, toluene, and *m*-xylene at 303.15 and 323.15 K as a function of pressure, are presented. The density data used for the measurements of benzene were obtained from Li [5] and Dymond et al. [6], with an

Table I. Viscosity of Toluene as a Function of Temperature at Atmospheric Pressure

Temperature (K)	Viscosity ($\mu\text{Pa}\cdot\text{s}$)
303.15	522.7
304.72	513.8
312.52	470.6
322.24	426.0
323.15	421.6
334.69	376.9

uncertainty less than $\pm 0.1\%$. The density data used for the measurements of toluene were obtained by Kashiwagi et al. [7], while the density data for *m*-xylene are those used by Taxis et al. [8], both with an uncertainty less than $\pm 0.1\%$. Since the density data for *m*-xylene extended only up to 55-MPa pressure, the measurements of the viscosity of *m*-xylene were restricted to this pressure range.

Table II. Viscosity of Benzene at 303.15 and 323.15 K, as a Function of Pressure

303.15 K			323.15 K		
Pressure (MPa)	Density ($\text{kg}\cdot\text{m}^{-3}$)	Viscosity ($\mu\text{Pa}\cdot\text{s}$)	Pressure (MPa)	Density ($\text{kg}\cdot\text{m}^{-3}$)	Viscosity ($\mu\text{Pa}\cdot\text{s}$)
0.10	868.3	562.7	0.10	846.8	438.1
5.37	872.8	588.5	5.37	851.8	456.5
10.44	876.8	614.5	10.34	856.3	476.0
15.50	880.8	640.5	15.40	860.7	496.2
20.57	884.5	667.5	20.27	864.7	516.6
25.74	888.2	695.4	25.64	869.0	538.4
30.90	891.8	724.5	30.90	873.1	560.6
35.87	895.1	752.6	35.26	876.3	579.4
41.14	898.5	783.8	39.72	879.5	598.8
46.10	901.6	814.3	45.90	883.8	626.4
50.97	904.5	844.1	50.26	886.8	645.8
55.02	906.9	870.1	55.02	889.9	668.5
59.38	909.4	897.3	62.11	894.3	702.0
			62.92	894.8	706.3
			69.41	898.7	735.9

Table III. Viscosity of Toluene at 303.15 and 323.15 K, as a Function of Pressure

303.15 K			323.15 K		
Pressure (MPa)	Density ($\text{kg}\cdot\text{m}^{-3}$)	Viscosity ($\mu\text{Pa}\cdot\text{s}$)	Pressure (MPa)	Density ($\text{kg}\cdot\text{m}^{-3}$)	Viscosity ($\mu\text{Pa}\cdot\text{s}$)
0.10	857.8	522.7	0.10	839.0	421.6
5.78	862.4	546.0	5.17	843.6	437.9
10.54	866.0	566.5	10.44	848.1	456.2
15.50	869.7	587.6	15.40	852.2	473.9
20.87	873.5	611.3	20.27	856.0	491.3
26.04	877.1	634.3	25.53	860.0	510.0
30.70	880.2	655.2	30.50	863.6	527.9
35.77	883.4	678.6	35.57	867.2	547.3
40.94	886.6	702.7	40.63	870.7	565.6
45.90	889.6	726.7	45.80	874.1	585.0
50.87	892.5	750.8	50.76	877.2	602.8
55.83	895.4	775.4	54.01	879.2	615.6
60.90	898.2	800.6	55.83	880.3	624.0
63.53	899.6	812.1	61.10	883.5	644.2
			66.17	886.5	663.7
			71.33	889.4	684.0

Table IV. Viscosity of *m*-Xylene at 303.15 and 323.15 K, as a Function of Pressure

303.15 K			323.15 K		
Pressure (MPa)	Density ($\text{kg}\cdot\text{m}^{-3}$)	Viscosity ($\mu\text{Pa}\cdot\text{s}$)	Pressure (MPa)	Density ($\text{kg}\cdot\text{m}^{-3}$)	Viscosity ($\mu\text{Pa}\cdot\text{s}$)
0.10	856.1	549.4	0.10	840.0	444.4
5.17	860.0	570.4	5.57	844.6	463.0
10.64	864.0	594.3	10.44	848.5	480.8
15.40	867.3	616.4	15.00	852.0	496.9
20.57	870.9	640.5	19.76	855.6	515.2
25.74	874.3	664.3	25.48	859.7	536.4
30.80	877.5	687.8	30.50	863.2	555.6
35.57	880.5	711.5	35.26	866.3	574.7
40.63	883.5	737.5	40.63	869.7	596.2
45.80	886.4	762.9	45.50	872.6	614.6
50.87	889.2	788.3	49.45	874.9	630.5
			56.34	878.6	658.1

For comparison purposes, we have used a Tait-like equation to correlate each isotherm of the present measurements as

$$\ln \left[\frac{\mu}{\mu_0} \right] = E \ln \left[\frac{D+P}{D+0.1} \right] \quad (7)$$

where μ_0 represents the experimental viscosity at atmospheric pressure. The values of the constants for each isotherm for the three liquids are shown in Table V. In the same table, the standard deviation of each fit is shown. It can be seen that the maximum standard deviation is $\pm 0.1\%$.

In Fig. 1 the deviations of the present experimental measurements of the viscosity of benzene, from those correlated by Eq. (7), are presented. It can be seen that the maximum deviation is less than $\pm 0.2\%$. In the same figure measurements of other investigators are also included. The only other absolute measurements are those performed by Knapstad et al. [9] at atmospheric pressure in an oscillating cylinder viscometer, with an uncertainty of $\pm 0.5\%$. The deviation of these measurements from those correlated by Eq. (7) is less than 0.25% , which is well within the mutual uncertainty of the two instruments. The other measurements included in Fig. 1 were all performed on a relative basis. The fitted measurements of Kashiwagi and Makita [10], performed in a torsionally vibrating crystal instrument with an uncertainty of $\pm 2\%$, show a maximum deviation of 0.5% from the present measurements. The measurements of Dymond et al. [6], performed in a self-centering falling-body viscometer with an uncertainty of $\pm 2\%$, show a maximum deviation of 0.6% from the present measurements. Also, the measurements of Parkhurst and Jonas [11] performed in a rolling-ball viscometer and those of Collings and McLaughlin [12], performed in a torsionally vibrating quartz-crystal viscometer, both show a deviation of less than 1% from the present

Table V. Coefficients of Eq. (7)

Liquid	Temperature (K)	μ_0 ($\mu\text{Pa} \cdot \text{s}$)	E	D (MPa)	σ (%)
Toluene	303.15	522.66	1.871	237.9	± 0.06
	323.15	421.63	1.586	199.4	± 0.09
Benzene	303.15	562.72	2.718	316.1	± 0.03
	323.15	438.10	2.179	257.0	± 0.10
<i>m</i> -Xylene	303.15	549.40	2.326	301.6	± 0.08
	323.15	444.40	1.757	223.9	± 0.09

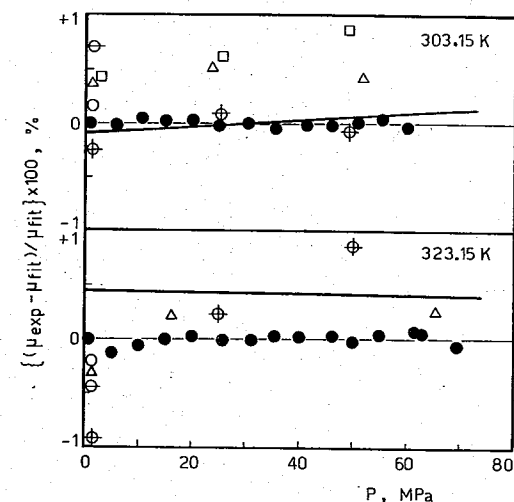


Fig. 1. The deviations of the experimental measurements of the viscosity of benzene from Eq. (7). (●) Present work; (Δ) Ref. 6; (○) Ref. 9; (—) Ref. 10; (□) Ref. 11; (+) Ref. 12; (○) Ref. 13.

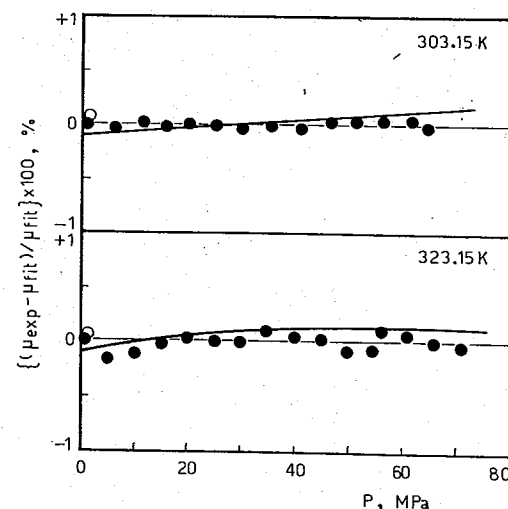


Fig. 2. The deviations of the experimental measurements of the viscosity of toluene from Eq. (7). (●) Present work; (○) Ref. 4; (—) Ref. 10.

measurements, which is well within the uncertainty of the instruments. At atmospheric pressure the measurements of Howard and Pike [13], performed in a Cannon-Ubbelohde viscometer, calibrated with water, show a 0.7% deviation from the present measurements.

In Fig. 2 the deviations of the present experimental measurements of the viscosity of toluene from those correlated by Eq. (7) are presented. It can be seen that the maximum deviation is less than $\pm 0.2\%$. In the same figure measurements of other investigators are also included. The atmospheric-pressure measurements of Gonçalves et al. [4] were performed in an Ubbelohde-type viscometer on an absolute basis with an uncertainty of $\pm 0.3\%$. The maximum deviation of these measurements from the correlation of Eq. (7) is less than 0.1%. The other set of measurements included in Fig. 2 are the high-pressure measurements of Kashiwagi and Makita [10], performed in a torsionally vibrating quartz crystal on a relative basis, with an uncertainty of $\pm 2\%$. The maximum deviation of these measurements from the correlation of Eq. (7) is less than 0.2%, which is well within the mutual uncertainty of the two instruments.

In Fig. 3 the deviations of the present experimental measurements of the viscosity of *m*-xylene from those correlated by Eq. (7) are presented. It can be seen that the maximum deviation is less than $\pm 0.2\%$. To our knowledge no other absolute measurements of the viscosity of *m*-xylene

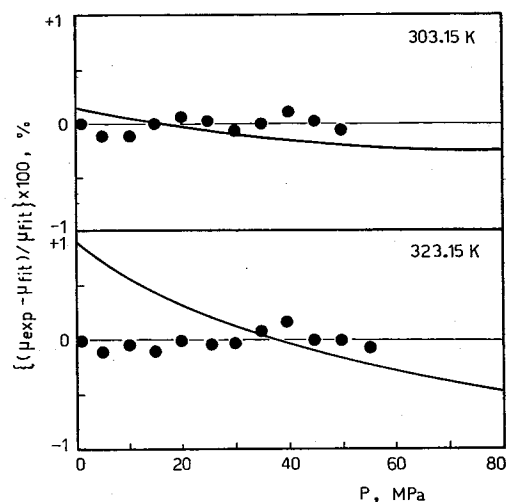


Fig. 3. The deviations of the experimental measurements of the viscosity of *m*-xylene from Eq. (7). (●) Present work; (—) Ref. 10.

exist. The only other measurements included in Fig. 3 are those of Kashiwagi and Makita [10], performed in a torsionally vibrating quartz-crystal viscometer on a relative basis, with an uncertainty of $\pm 2\%$. The maximum deviation of these measurements from the correlation of Eq. (7) is less than 1%, which is well within the mutual uncertainty of the two instruments.

4. CONCLUSIONS

New absolute measurements of the viscosity of benzene, toluene, and *m*-xylene at 303.15 and 323.15 K and at pressures up to 80 MPa were presented. The uncertainty of the measurements is estimated to be $\pm 0.5\%$.

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