Speed of Sound, Density, and Compressibility of Alkyl-Benzenes as a Function of Pressure and Temperature: Heptadecylbenzene and Octadecylbenzene

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Measurements of speed of sound were carried out in liquid heptadecylbenzene and octadecylbenzene at pressures from atmospheric up to 150 MPa in the temperature range 303 to 383 K using a pulse technique operating at 3 MHz. Additional density measurements were performed up to 60 MPa. From these measurements, the density was evaluated up to 150 MPa and the isentropic and isothermal compressibilities were determined in the same *P* and *T* domain. The results were fitted to a Tait-like equation.

1. Introduction

As a part of a broad project on thermophysical properties of heavy components belonging to the families of chemicals found essentially in crude oils (paraffins, naphthenes, and aromatics), a measurement program on the heavy alkylbenzene was initiated[1] in order to acquire a substantial body of experimental data on different thermophysical properties over a wide range of pressure (0.1 to 150) MPa. As the speed of sound is a complex thermodynamic property which can be determined experimentally with a high degree of accuracy including at high pressures and which presents the advantage of giving access to various derived properties, we have focused our measurements on this property.

In this paper, which follows on other investigations into tridecylbenzene and pentadecylbenzene, ultrasonic measurements were carried out under high pressure in heavier components, namely, heptadecylbenzene (23 carbon atoms) and octadecylbenzene (24 carbon atoms). The speed of sound data linked with complementary density measurements performed in a narrow pressure range were then used to determine the density as well as the isentropic and isothermal compressibilities of these high molecular weight alkyl-benzenes up to 150 Mpa.

2. Experimental Section

Ultrasonic speed measurements were carried out using a pulse−echo technique operating at 3 MHz. The apparatus is essentially made up of a high-pressure cell closed at both ends by two identical transducers. The details of this apparatus have been extensively described in a previous paper.[2,3] To ensure satisfactory thermal uniformity within the fluid, the vessel was immersed in a bath of heat-carrying fluid agitated and thermoregulated by a Bioblock thermostat with a stability of 0.02 K. The temperature was recorded by means of a platinum probe (Pt100) placed inside the experimental vessel, whereas the pressure was measured by an HBM P3M gauge, which is frequently checked against a dead-weight tester to an accuracy better than 0.02%.

Table 1. Speed of Sound c ($m \cdot s^{-1}$) in the Liquids Heptadecylbenzene and Octadecylbenzene as a Function of Pressure and Temperature

	T/K							
P/MPa	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
				Heptadecylbenzene				
0.1	1422.6	1387.1	1352.6	1319.9	1287.0	1255.2	1223.6	1193.0
10	1467.5	1433.6	1402.3	1370.2	1338.7	1308.6	1278.7	1249.5
20	1510.9	1479.2	1447.9	1417.3	1387.3	1359.3	1330.3	1302.6
30	1552.0	1520.4	1490.8	1461.4	1432.3	1405.3	1378.0	1351.4
40	1590.5	1559.7	1531.1	1502.9	1475.5	1448.5	1422.2	1396.8
50	1627.2	1597.9	1569.2	1542.1	1515.3	1489.5	1464.0	1439.2
60	1661.9	1633.6	1605.8	1579.2	1553.3	1528.3	1503.3	1479.7
70	1695.6	1667.3	1640.7	1614.6	1589.5	1564.8	1541.1	1517.7
80		1700.0	1673.9	1648.7	1624.1	1600.1	1576.4	1553.8
90		1731.8	1705.8	1681.3	1657.2	1633.4	1610.7	1588.6
100		1762.0	1736.6	1712.2	1688.0	1665.5	1643.4	1621.8
110		1790.2	1766.3	1742.3	1719.2	1696.9	1674.6	1653.6
120		1818.7	1794.9	1771.7	1748.7	1726.9	1705.0	1684.3
130		1847.1	1822.8	1799.8	1777.4	1755.8	1734.4	1713.8
140			1849.3	1826.9	1805.0	1783.8	1763.3	1742.7
150			1875.5	1853.6	1832.0	1811.1	1790.0	1770.5
				Octadecylbenzene				
0.1		1392.9	1357.8	1325.5	1292.7	1260.7	1229.5	1198.7
10		1439.6	1406.8	1374.8	1344.1	1313.4	1284.8	1255.2
20		1484.1	1452.4	1422.6	1392.8	1364.1	1334.9	1308.3
30		1525.8	1495.5	1466.1	1437.7	1410.0	1382.7	1356.6
40		1564.8	1535.6	1507.5	1480.2	1454.0	1427.1	1401.7
50		1602.4	1574.0	1546.7	1519.9	1494.4	1469.1	1444.5
60		1638.1	1610.3	1583.5	1558.1	1532.6	1508.3	1484.7
70		1672.1	1645.1	1619.1	1594.2	1569.2	1545.7	1522.0
80		1704.7	1678.7	1652.8	1628.6	1604.3	1581.7	1558.6
90			1710.4	1685.7	1661.5	1638.2	1615.7	1592.9
100			1741.3	1717.0	1693.1	1670.3	1648.3	1626.3
110			1771.2	1747.3	1723.6	1701.4	1679.8	1657.9
120			1799.2	1775.7	1753.5	1731.3	1710.0	1688.5
130				1803.9	1781.9	1759.9	1739.2	1718.7
140				1830.9	1809.4	1788.8	1767.5	1746.6
150				1857.5	1836.3	1815.1	1795.5	1774.6

The ultrasonic speed was determined from the measurement, by direct chronometry,[4] of the travelling time of the wave through the sample by means of a numerical oscilloscope with memory storage. The length of the sample path was calibrated with degassed water by using the data of Del Grosso et al.,[5] Wilson,[6] and Petitet et al.[7] The

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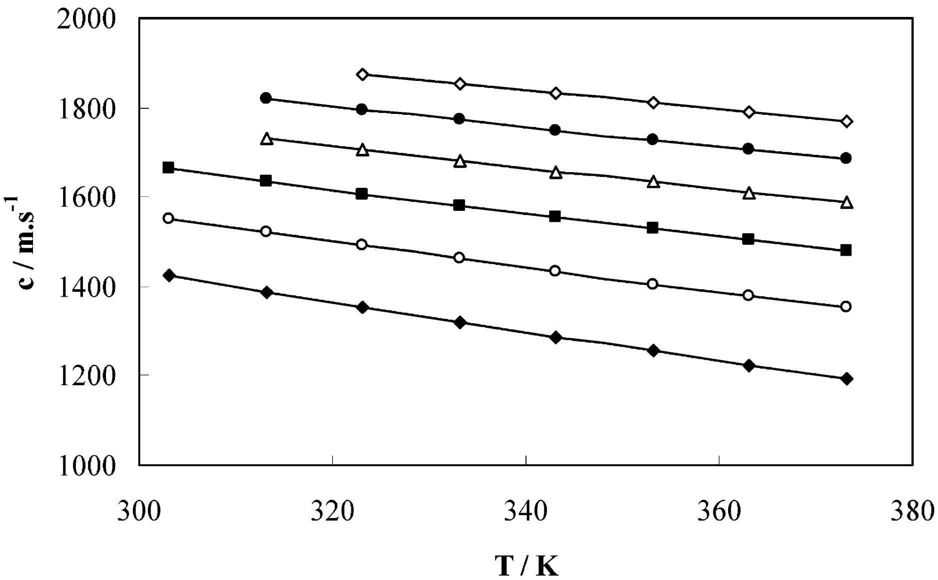

Figure 1. Speed of sound *c* in liquid heptadecylbenzene as a function of temperature along various isobars. ◆, 0.1 MPa; ○, 30 MPa; ■, 60 MPa; △, 90 MPa; ●, 120 Mpa; ◇, 150 MPa.

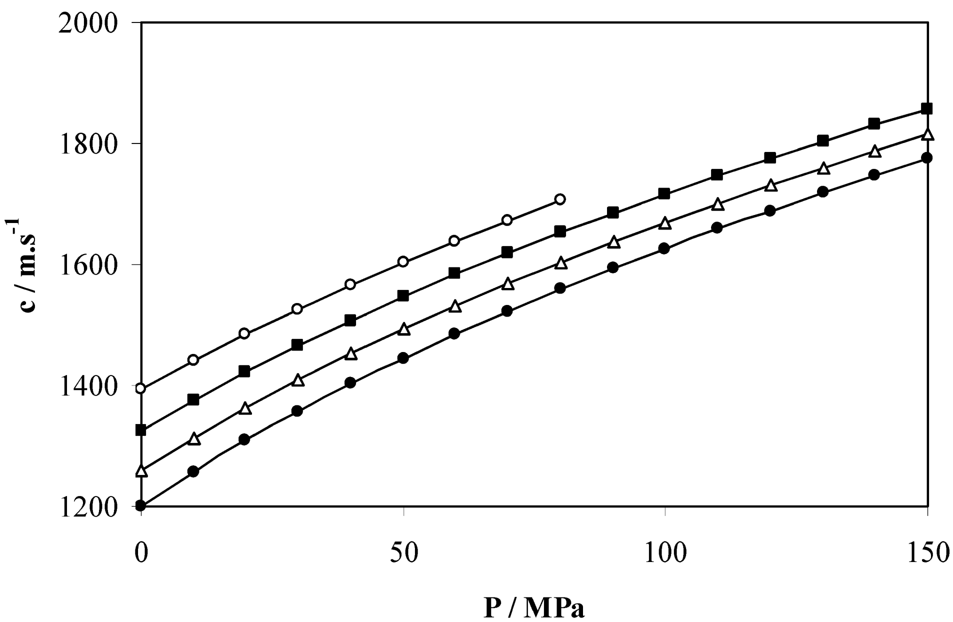

Figure 2. Speed of sound *c* in liquid octadecylbenzene as a function of pressure along various isotherms. ○, 313.15 K; ■, 333.15 K; △, 353.15 K; ●, 373.15 K.

experimental uncertainty of the speed of sound measurements has been estimated to be less than 0.2% over the entire pressure range (0.1 to 150) MPa, an estimation confirmed after tests performed with various hydrocarbons.[3−4,8]

The complementary density measurements were carried out by means of an ANTON-PAAR densimeter (DMA 60 model) equipped with a high-pressure cell (DMA 512 P) with an operating range of (0.1 to 70) MPa. The principle of this apparatus is to measure the period of oscillation of a U-shaped tube and to deduce the density which is related to the square of the period by a linear law whose parameters are calibrated by the method proposed by Lagourette et al.[9] using water reference data.[10] The overall accuracy obtained by this apparatus is estimated to be better than 0.1 kg/m^3.

Both compounds were supplied by Fluka with a purity higher than 98% and used without further purification. The heptadecylbenzene, also called 1-phenylheptadecane, has the chemical formula $C_{23}H_{40}$ and a molar mass of 316.56 $g\cdot mol^{-1}$, whereas the octadecylbenzene (1-phenyloctadecane) has the chemical formula $C_{24}H_{42}$ and a molar mass of 330.59 $g\cdot mol^{-1}$.

3. Results and Discussion

The speed of sound *c* of liquid heptadecylbenzene and octadecylbenzene was measured at 10-K intervals from (303.15 to 373.15) K in the pressure range from atmospheric to the pressure of solidification. The pressure step adopted during the experiments was fixed at 10 MPa in order to have a sufficient number of data (16) to allow a good fit by correlation functions on each isotherm. The values of the speed of sound obtained for these two alkylbenzenes are given in Table 1 and are plotted as a function of temperature and pressure in Figures 1 and 2. The results were fitted to a rational function which correlates c^2 with 9 adjustable parameters

$$c^2 = \frac{E + FP}{A + BP + CP^2 + DP^3} \tag{1}$$

where

$$A = A_0 + A_1T + A_2T^2 + A_3T^3 \tag{2}$$

and

$$E = 1 + E_1T \tag{3}$$

The parameters obtained by a least-squares method are listed in Table 2 along with the average deviation (AD%), the average absolute deviation (AAD%), and the maximum deviation MD% for both components.

Measurements of density were undertaken along the same isotherms but were restricted in pressure to 60 MPa. To extend the density data to higher pressures than those accessible from the densimeter, the change in density ρ with pressure was evaluated from speed of sound integration[11] up to 150 MPa

$$\rho(P,T) - \rho(P_{atm},T) = \int_{P_{atm}}^{P} 1/c^2\, dP + T\int_{P_{atm}}^{P} (\alpha_P{}^2/C_p)\, dP \tag{4}$$

In this relation α_P is the isobaric expansion coefficient and C_p the heat capacity at constant pressure. The first integral is evaluated by analytical integration of eq 1 using the fitted parameters listed in Table 2. The last contribution is calculated numerically using a predictor−corrector method[2] in which the initialization procedure proposed by Denielou et al.[12] was used. The values of the isobaric expansion coefficient are evaluated at each pressure by numerical derivation of density vs temperature, whereas the heat capacity C_p required for the evaluation of the integral is estimated at each pressure step by the following thermodynamic relation

$$C_p(P,T) = C_p(P_{atm},T) - \int_{P_{atm}}^{P} T[\alpha_P{}^2 + (\partial\alpha_P/\partial T)_p]/\rho\, dP$$

Unfortunately, the heat capacity at atmospheric pressure is not available for these compounds. To overcome this lack, the moderate pressure density data were used to initiate the numerical calculation of the integral by an inverse technique.[13] The densities were then evaluated up to 150 MPa. The measurements of density as well as the data deduced from speed of sound are listed in Table 3. The accuracy of these data has been estimated to 0.1% on the basis of several tests performed on pure hexane.[8] These density data were mathematically correlated as a function of pressure by a Tait-type equation which reproduces the density data within the experimental uncertainty

$$\frac{1}{\rho} = \frac{1}{\rho_{atm}} + a\ln\left(\left[\frac{P + b}{P_{atm} + b}\right]\right) \tag{5}$$

in which a, b, and ρ_{atm} are correlated with temperature by means of polynomial functions

$$\rho_{atm} = \rho_0 + \rho_1T + \rho_2T^2 + \rho_3T^3 \tag{6}$$

$$a = a_0 + a_1T + a_2T^2 \tag{7}$$

$$b = b_0 + b_1T + b_2T^2 \tag{8}$$

Parameters of eq 6 were first evaluated alone by fitting atmospheric density data. The other parameters were then evaluated by an unweighted least-squares procedure. The parameters ρ_i, a_i, and b_i for each compound are listed in Table 4 together with the related deviations.

As the determination of density from speed of sound is based on the relationships which link the isentropic compressibility κ_S to the speed of sound c and the isother-

Table 2. Parameters of Equations 1−3 with T in K, P in MPa, and c in m·s^{-1}

parameters	heptadecylbenzene	octadecylbenzene
A_0	5.39276×10^{-8}	-2.03590×10^{-7}
A_1	7.61871×10^{-10}	2.99536×10^{-9}
A_2	-4.11400×10^{-14}	-6.48960×10^{-12}
A_3	-9.78950×10^{-16}	5.31208×10^{-15}
B	9.26883×10^{-10}	1.00769×10^{-9}
C	-2.19690×10^{-12}	-2.62030×10^{-12}
D	3.98678×10^{-15}	5.19506×10^{-15}
E_1	-1.60736×10^{-3}	-1.57796×10^{-3}
F	5.37290×10^{-3}	5.57940×10^{-3}
deviations of c		
AD%	-4.6×10^{-4}	-7.2×10^{-4}
AAD%	1.6×10^{-2}	1.6×10^{-2}
MD%	6.4×10^{-2}	6.2×10^{-2}

Table 3. Density ρ (kg·m^{-3}) of Heptadecylbenzene and Octadecylbenzene as a Function of Pressure and Temperature

	T/K							
P/MPa	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
Heptadecylbenzene								
0.1[a]	847.62	840.87	834.27	827.59	820.95	814.43	807.80	801.11
5[a]	850.37	843.75	837.23	830.84	824.32	817.93	811.30	805.07
10[a]	853.14	846.65	840.29	833.94	827.71	821.35	815.14	808.86
15[a]	855.85	849.47	843.15	837.13	830.75	824.69	818.57	812.48
20[a]	858.47	852.17	845.93	839.97	833.93	827.90	821.81	815.93
25[a]	861.00	854.73	848.70	842.84	836.75	830.86	824.97	819.17
30[a]	863.37	857.38	851.34	845.52	839.62	833.85	828.00	822.40
35[a]	865.81	859.90	853.89	848.14	842.40	836.90	830.94	825.42
40[a]	868.19	862.24	856.43	850.62	845.11	839.55	833.81	828.43
45[a]	870.43	864.56	858.84	853.30	847.75	842.10	836.50	831.28
50[a]	872.65	866.80	861.35	855.69	850.31	844.78	839.26	834.03
55[a]	874.70	869.08	863.55	858.00	852.73	847.34	841.95	836.72
60[a]	876.93	870.60	865.80	860.35	855.09	849.77	844.41	839.37
70[b]	880.88	875.35	869.96	864.68	859.51	854.43	849.41	844.44
80[b]		879.39	874.10	868.95	863.90	858.94	854.05	849.22
90[b]		883.27	878.09	873.04	868.10	863.25	858.49	853.78
100[b]		887.01	881.92	876.97	872.13	867.39	862.73	858.13
110[b]		890.62	885.62	880.76	876.01	871.36	866.80	862.31
120[b]		894.12	889.19	884.41	879.75	875.19	870.72	866.32
130[b]		897.50	892.65	887.95	883.36	878.89	874.50	870.19
140[b]			896.01	891.37	886.86	882.46	878.15	873.92
150[b]			899.26	894.69	890.25	885.92	881.68	877.53
Octadecylbenzene								
0.1[a]		840.55	833.94	827.32	820.78	814.10	807.70	800.89
5[a]		843.37	836.91	830.40	824.10	817.72	811.19	804.86
10[a]		846.27	840.02	833.56	827.33	821.14	814.87	808.59
15[a]		849.15	842.88	836.59	830.48	824.47	818.14	812.16
25[a]		854.46	848.32	842.30	836.54	830.64	824.70	818.84
30[a]		856.89	850.96	845.04	839.35	833.58	827.83	821.97
35[a]		859.36	853.46	847.71	841.97	836.52	830.62	825.09
40[a]		861.48	856.00	850.30	844.62	839.12	833.38	828.00
45[a]		864.12	858.41	852.76	847.32	841.72	836.17	830.80
50[a]		866.37	860.91	855.26	849.82	844.35	838.78	833.60
55[a]		868.59	863.12	857.46	852.25	846.91	841.42	836.29
60[a]		870.16	865.36	859.86	854.60	849.39	843.92	838.88
70[b]		874.62	869.43	864.26	859.12	854.01	848.95	843.93
80[b]		878.61	873.54	868.50	863.49	858.51	853.58	848.70
90[b]			877.49	872.57	867.67	862.82	858.00	853.24
100[b]			881.28	876.47	871.69	866.94	862.23	857.58
110[b]			884.94	880.24	875.55	870.91	866.30	861.75
120[b]			888.48	883.87	879.28	874.73	870.21	865.75
130[b]				887.37	882.88	878.41	873.99	869.61
140[b]				890.77	886.36	881.98	877.63	873.33
150[b]				894.07	889.73	885.43	881.16	876.94

[a] U-shaped tube densimeter measurements. [b] Determined from speed of sound.

Table 4. Parameters of the Tait Equation (Eqs 7 to 10) with *T* in K, *P* in MPa, and *ρ* in kg·m^{-3}

parameters	heptadecylbenzene	octadecylbenzene
ρ_0	1.21923×10^{3}	1.04436×10^{3}
ρ_1	-2.16408	-6.43666×10^{-1}
ρ_2	4.38678×10^{-3}	-2.30940×10^{-5}
ρ_3	-4.26090×10^{-6}	
a_0	-1.20970×10^{-4}	5.60785×10^{-5}
a_1	1.84126×10^{-7}	-7.92570×10^{-7}
a_2	-3.93230×10^{-10}	9.47530×10^{-10}
b_0	4.60775×10^{2}	2.58094×10^{2}
b_1	-1.52877	-4.10940×10^{-1}
b_2	1.40000×10^{-3}	-1.29430×10^{-4}
deviations of *c*		
AD%	3.3×10^{-4}	4.5×10^{-4}
AAD%	7.7×10^{-3}	7.7×10^{-3}
MD%	7.2×10^{-2}	4.2×10^{-2}

Table 5. Isentropic Compressibility κ_S (GPa^{-1}) of Heptadecylbenzene and Octadecylbenzene as a Function of Pressure and Temperature

	T/K							
P/MPa	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
	Heptadecylbenzene							
0.1	0.5830	0.6181	0.6552	0.6936	0.7354	0.7793	0.8268	0.8770
10	0.5443	0.5747	0.6052	0.6387	0.6741	0.7110	0.7503	0.7918
20	0.5102	0.5363	0.5639	0.5927	0.6231	0.6537	0.6876	0.7223
30	0.4808	0.5046	0.5285	0.5538	0.5806	0.6073	0.6361	0.6658
40	0.4553	0.4767	0.4981	0.5205	0.5435	0.5677	0.5929	0.6187
50	0.4328	0.4518	0.4715	0.4914	0.5122	0.5335	0.5559	0.5789
60	0.4129	0.4304	0.4479	0.4660	0.4847	0.5039	0.5240	0.5441
70	0.3949	0.4110	0.4270	0.4436	0.4605	0.4779	0.4957	0.5141
80		0.3935	0.4083	0.4234	0.4389	0.4547	0.4712	0.4877
90		0.3775	0.3914	0.4052	0.4195	0.4342	0.4490	0.4641
100		0.3631	0.3760	0.3890	0.4024	0.4156	0.4292	0.4431
110		0.3503	0.3619	0.3740	0.3862	0.3986	0.4114	0.4241
120		0.3381	0.3491	0.3602	0.3717	0.3832	0.3951	0.4069
130		0.3266	0.3372	0.3477	0.3583	0.3691	0.3801	0.3912
140			0.3263	0.3361	0.3461	0.3561	0.3662	0.3768
150			0.3162	0.3253	0.3347	0.3441	0.3540	0.3635
	Octadecylbenzene							
0.1		0.6132	0.6504	0.6880	0.7291	0.7729	0.8190	0.8689
10		0.5702	0.6015	0.6347	0.6690	0.7060	0.7435	0.7849
20		0.5330	0.5607	0.5886	0.6184	0.6493	0.6832	0.7163
30		0.5013	0.5254	0.5505	0.5764	0.6035	0.6318	0.6610
40		0.4741	0.4954	0.5175	0.5404	0.5637	0.5892	0.6147
50		0.4495	0.4688	0.4887	0.5094	0.5303	0.5524	0.5749
60		0.4283	0.4457	0.4638	0.4820	0.5012	0.5208	0.5408
70		0.4090	0.4250	0.4414	0.4580	0.4755	0.4930	0.5115
80		0.3917	0.4062	0.4215	0.4366	0.4525	0.4683	0.4850
90			0.3895	0.4033	0.4175	0.4319	0.4465	0.4619
100			0.3742	0.3870	0.4002	0.4135	0.4269	0.4409
110			0.3602	0.3721	0.3845	0.3967	0.4091	0.4222
120			0.3477	0.3588	0.3699	0.3814	0.3930	0.4051
130				0.3463	0.3567	0.3675	0.3783	0.3893
140				0.3349	0.3446	0.3543	0.3647	0.3753
150				0.3242	0.3333	0.3428	0.3520	0.3621

mal compressibility κ_T to κ_S

$$\kappa_S = \frac{1}{\rho c^2} \tag{9}$$

$$\kappa_T = \kappa_S + \frac{T{\alpha_p}^2}{\rho C_p} \tag{10}$$

the procedure leads also to the evaluation of the isentropic and isothermal compressibilities with an accuracy of 0.3% and 2% respectively. These data are summarized in Tables 5 and 6. The isothermal compressibility can also be derived from the fitted Tait equation

$$\kappa_T = -\rho \frac{a}{P + b} \tag{11}$$

Table 6. Isothermal Compressibility κ_T (GPa^{-1}) of Heptadecylbenzene and Octadecylbenzene as a Function of Pressure and Temperature

	T/K							
P/MPa	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
	Heptadecylbenzene							
0.1	0.6820	0.7201	0.7609	0.8039	0.8512	0.9018	0.9573	1.0171
10	0.6347	0.6672	0.7004	0.7373	0.7769	0.8188	0.8643	0.9131
20	0.5934	0.6209	0.6505	0.6818	0.7154	0.7500	0.7886	0.8290
30	0.5578	0.5826	0.6080	0.6351	0.6644	0.6941	0.7267	0.7611
40	0.5271	0.5491	0.5715	0.5954	0.6203	0.6469	0.6752	0.7046
50	0.5000	0.5195	0.5397	0.5607	0.5830	0.6063	0.6311	0.6571
60	0.4762	0.4939	0.5117	0.5306	0.5504	0.5711	0.5933	0.6159
70	0.4547	0.4707	0.4870	0.5041	0.5218	0.5405	0.5599	0.5804
80		0.4501	0.4649	0.4802	0.4964	0.5132	0.5310	0.5493
90		0.4312	0.4449	0.4589	0.4736	0.4890	0.5050	0.5216
100		0.4142	0.4268	0.4398	0.4535	0.4673	0.4817	0.4969
110		0.3991	0.4104	0.4223	0.4347	0.4474	0.4610	0.4748
120		0.3848	0.3953	0.4062	0.4178	0.4295	0.4420	0.4547
130		0.3713	0.3814	0.3916	0.4022	0.4131	0.4246	0.4365
140			0.3687	0.3782	0.3880	0.3981	0.4086	0.4198
150			0.3569	0.3656	0.3748	0.3842	0.3944	0.4044
	Octadecylbenzene							
0.1		0.7132	0.7560	0.7993	0.8464	0.8965	0.9491	1.0058
10		0.6604	0.6963	0.7343	0.7734	0.8152	0.8577	0.9042
20		0.6150	0.6466	0.6785	0.7122	0.7471	0.7849	0.8219
30		0.5764	0.6040	0.6325	0.6616	0.6919	0.7235	0.7557
40		0.5434	0.5677	0.5927	0.6185	0.6445	0.6726	0.7005
50		0.5137	0.5358	0.5583	0.5814	0.6047	0.6290	0.6534
60		0.4881	0.5079	0.5284	0.5488	0.5701	0.5915	0.6132
70		0.4649	0.4832	0.5017	0.5203	0.5396	0.5587	0.5786
80		0.4442	0.4608	0.4780	0.4949	0.5125	0.5296	0.5476
90			0.4409	0.4565	0.4723	0.4881	0.5040	0.5205
100			0.4227	0.4372	0.4519	0.4664	0.4810	0.4959
110			0.4061	0.4196	0.4333	0.4467	0.4602	0.4741
120			0.3912	0.4038	0.4162	0.4288	0.4414	0.4543
130				0.3891	0.4007	0.4126	0.4242	0.4359
140				0.3756	0.3865	0.3973	0.4084	0.4197
150				0.3630	0.3733	0.3838	0.3937	0.4044

The comparison reveals excellent agreement between the two sets of compressibility data, those resulting from eq 10 on one hand and those resulting from the derivative of the Tait equation (eq 11) on the other hand. The two data sets deviate by 0.2% on average with an average absolute deviation of 0.4% for both compounds.

4. Conclusion

A program of measurement of ultrasonic velocity in heavy alkyl-benzenes has been previously initiated[1] in order to evaluate the density as well as its derivatives with respect to pressure κ_S and κ_T under high pressure. The results of this work complete these earlier measurements and help to characterize these compounds which have an importance in the petroleum industry due to their presence in significant amount in heavy fractions of crude oils.

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