Speed of Sound, Density, and Compressibility of Alkylbenzenes as a Function of Pressure and Temperature: Tridecylbenzene and Pentadecylbenzene

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Speed of sound measurements were carried out in liquid tridecylbenzene and pentadecylbenzene at pressures from atmospheric up to 150 MPa in the temperature range from (293 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 compressibility was determined in the same $P-T$ domain.

Introduction

The current trend toward the production of crude petroleum oils of much greater densities than those exploited a few years ago makes the thermodynamic properties of organic compounds with high molecular weights (and especially those found in reservoir fluids) all the more important. To make the best possible use of these fossil fuels, down to the residual fuels, it is necessary to be able to characterize the behavior of petroleum fluids as a whole, their so-called heavy fractions, and liquid mixtures such as distillation cuts with high boiling points.

Although a number of correlation functions have been developed—each with its own limitations—to generate predictions of such properties, and particularly volumetric properties and phase equilibrium, there is no real substitute for direct experimental data. This is especially true for heavy hydrocarbons, which have so far not been widely studied, especially at pressures other than atmospheric pressure, and for which the predictive models such as group contributions often give disappointing results.

A thorough review of the literature for data as a function of pressure reveals no trace of information on the volumetric and thermoelastic properties of aromatic compounds with more than 20 carbon atoms. On the basis of this observation, the behaviors of various thermophysical properties such as speed of sound, density (ρ), and isentropic (κ_s) and isothermal (κ_T) compressibilities were determined in aromatic components versus pressure and temperature from ultrasonic measurements up to 150 MPa and additional density measurements performed up to 60 MPa. In this paper the substances under study are two alkylbenzenes with 19 (tridecylbenzene) and 21 (pentadecylbenzene) carbon atoms.

Experimental Section

The speed of sound was measured up to 150 MPa using a pulse transmission−reflection apparatus working at 3 MHz. The apparatus, which has been described previously in detail,[1,2] is essentially made up of an autoclave cell closed at both ends by two identical piezoelectric transducers. One acts as pulse transmitter, and the second, as receiver. The ultrasonic speed is determined from the measurement, by direct chronometry,[3] 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 determined precisely by calibration with water using the data of Del Grosso et al.,[4] Wilson,[5] and Petitet et al.[6] The temperature is controlled by a thermostat with a stability of 0.02 K, and temperature measurements are carried out using a probe placed inside the experimental vessel. Several tests performed with hexane,[7] heptane,[2] and octane[3] have shown that an overall accuracy better than 0.2% is obtained over the entire pressure range of (0.1 to 150) MPa.

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.[8] using water reference data.[9] The overall accuracy obtained by this apparatus is estimated to be better than 0.1 kg/m^3.

The pressure is measured by an HBM P3M gauge which is frequently checked against a dead weight tester to an accuracy better than 0.02% whereas the temperature is measured with an accuracy of 0.1 K by means of a platinum probe (Pt100) placed inside the high-pressure cells and linked to an AOIP brand thermometer.

Both compounds were supplied by Fluka with a purity higher than 98% and used without further purification. The tridecylbenzene, also called 1-phenyltridecane, has the chemical formula $C_{19}H_{32}$ and a molar mass of 260.46 $g{\cdot}mol^{-1}$, whereas the pentadecylbenzene (1-phenylpentadecane) has the chemical formula $C_{21}H_{36}$ and a molar mass of 288.51 $g{\cdot}mol^{-1}$.

Results and Discussion

The tridecylbenzene was studied in the temperature range (293 to 373) K whereas pentadecylbenzene was investigated between (303 and 373) K, due to a higher melting temperature. The measurements of speed of sound

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10.1021/je025560n CCC: $22.00
Published on Web 10/08/2002

Table 1. Speed of Sound (*c*) of Tridecylbenzene as a Function of Pressure and Temperature

	c/m·s^{-1} at the following values of T/K								
P/MPa	293.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
0.1013	1437.8	1401.8	1366.3	1331.8	1297.4	1263.9	1230.8	1198.4	1166.8
10	1482.9	1448.3	1414.1	1381.1	1348.9	1316.6	1285.5	1255.1	1225.3
20	1525.3	1492.0	1459.2	1427.4	1396.6	1366.1	1336.3	1307.5	1279.1
30	1565.3	1532.7	1501.4	1471.1	1441.2	1412.0	1383.4	1355.9	1328.8
40	1603.1	1572.2	1541.2	1511.8	1482.7	1455.0	1427.5	1400.8	1374.7
50	1639.1	1608.8	1578.8	1550.4	1522.2	1495.3	1468.7	1443.0	1418.1
60	1673.6	1643.6	1615.0	1587.0	1559.9	1533.5	1507.4	1482.7	1458.5
70	1706.4	1677.3	1648.9	1622.0	1595.7	1569.6	1544.7	1520.6	1497.0
80	1737.4	1709.6	1681.6	1655.4	1629.4	1604.2	1580.0	1556.5	1533.6
90	1768.2	1740.5	1713.4	1687.8	1662.2	1637.5	1613.7	1590.8	1568.4
100	1797.5	1770.3	1744.0	1718.5	1693.6	1669.8	1646.4	1623.5	1602.0
110	1826.0	1799.0	1773.0	1748.5	1723.9	1700.3	1677.4	1655.6	1633.8
120	1853.2	1827.1	1801.4	1776.9	1753.1	1729.9	1707.4	1685.8	1664.9
130		1854.3	1828.6	1805.0	1781.3	1758.5	1736.7	1715.0	1694.8
140		1880.5	1855.3	1831.8	1808.6	1786.2	1764.8	1743.9	1723.7
150		1905.7	1881.1	1857.6	1835.4	1813.3	1791.7	1771.3	1751.6

Table 2. Speed of Sound (*c*) of Pentadecylbenzene as a Function of Pressure and Temperature

	c/m·s^{-1} at the following values of T/K							
P/MPa	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
0.1013	1414.0	1378.9	1344.0	1310.4	1277.5	1244.8	1213.8	1182.9
10	1460.0	1426.7	1393.3	1361.7	1330.1	1298.8	1269.3	1240.3
20	1503.0	1470.7	1439.3	1408.6	1378.6	1348.9	1321.3	1293.2
30	1544.2	1512.8	1482.4	1452.3	1424.0	1396.1	1369.0	1342.4
40	1582.5	1552.1	1523.1	1494.4	1466.7	1440.2	1413.3	1387.9
50	1619.5	1590.3	1561.6	1533.9	1507.3	1480.9	1455.1	1431.0
60	1654.6	1626.0	1598.2	1571.3	1545.6	1519.7	1495.0	1471.5
70	1688.2	1660.4	1633.1	1606.7	1581.5	1556.4	1531.9	1509.6
80	1720.0	1693.0	1666.6	1640.6	1616.0	1591.6	1568.3	1545.8
90	1750.7	1724.4	1698.8	1673.6	1649.2	1625.3	1602.6	1580.7
100	1780.6	1754.7	1729.5	1704.7	1680.8	1657.5	1635.4	1613.7
110	1809.7	1784.3	1759.6	1734.3	1711.6	1688.7	1667.0	1645.7
120	1838.0	1812.1	1787.8	1763.9	1740.9	1718.6	1697.2	1676.5
130	1864.6	1839.6	1815.9	1792.4	1770.2	1747.5	1726.6	1706.3
140	1890.9	1866.2	1842.8	1819.1	1797.6	1775.5	1754.8	1735.0
150	1916.6	1892.5	1869.0	1846.3	1824.4	1802.3	1782.5	1762.8

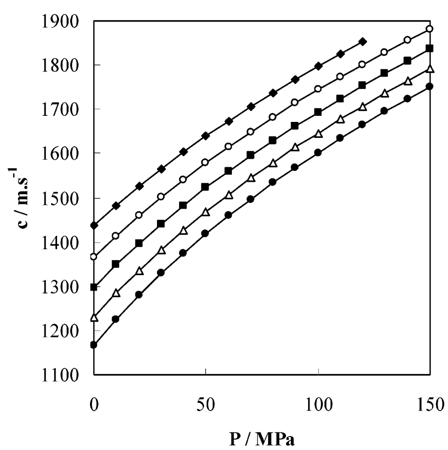

Figure 1. Speed of sound (c) in liquid tridecylbenzene as a function of pressure along various isotherms: ◆, 293.15 K; ○, 313.15 K; ■, 333.15 K; △, 353.15 K; ●, 373.15 K.

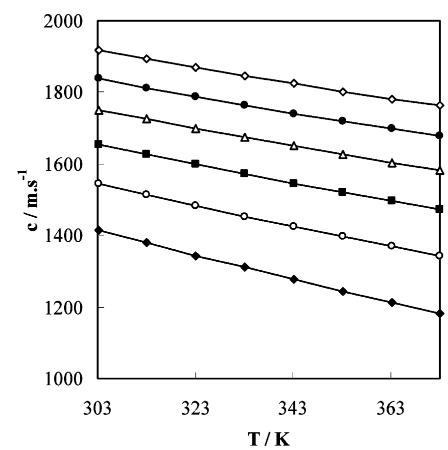

Figure 2. Speed of sound (c) in liquid pentadecylbenzene as a function of temperature along various isobars: ◆, 0.1 MPa; ○, 30 MPa; ■, 60 MPa; △, 90 MPa; ●, 120 Mpa; ◇, 150 MPa.

were carried out in 10 MPa steps from atmospheric pressure up to 150 MPa. The results are given in Tables 1 and 2 and are plotted as a function of temperature and pressure in Figures 1 and 2. These data were fitted to a rational function which correlates c^2 with nine 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 two sets of parameters determined by a least-squares method are listed in Table 3 along with the average deviation (AD%), the average absolute deviation (AAD%), and the maximum deviation (MD%). These deviations, which are less than the experimental error, show that the function leads to a good interpolation of the speed of sound data. Moreover, this expression leads to a simple analytical

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

parameters			deviations of *c*
Tridecylbenzene			
$A_0 = 1.333\ 99 \times 10^{-7}$	$A_3 = -3.186\ 50 \times 10^{-15}$	$D = 4.314\ 02 \times 10^{-15}$	AD% = -3.0×10^{-4}
$A_1 = 3.540\ 08 \times 10^{-11}$	$B = 9.801\ 30 \times 10^{-10}$	$E_1 = -1.637\ 16 \times 10^{-3}$	AAD% = 1.2×10^{-2}
$A_2 = 2.190\ 21 \times 10^{-12}$	$C = -2.377\ 70 \times 10^{-12}$	$F = 5.466\ 20 \times 10^{-3}$	MD% = 5.4×10^{-2}
Pentadecylbenzene			
$A_0 = 2.891\ 11 \times 10^{-7}$	$A_3 = -7.435\ 80 \times 10^{-15}$	$D = 5.384\ 85 \times 10^{-15}$	AD% = 2.7×10^{-4}
$A_1 = -1.384\ 80 \times 10^{-9}$	$B = 1.044\ 52 \times 10^{-9}$	$E_1 = -1.588\ 72 \times 10^{-3}$	AAD% = 1.7×10^{-2}
$A_2 = 6.501\ 45 \times 10^{-12}$	$C = -2.741\ 90 \times 10^{-12}$	$F = 5.664\ 12 \times 10^{-3}$	MD% = 5.2×10^{-2}

Table 4. Density (*ρ*) of Tridecylbenzene as a Function of Pressure and Temperature

	ρ/kg·m^{-3} at the following values of *T*/K								
P/MPa	293.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
0.1013[a]	855.19	847.78	840.87	834.11	827.32	820.46	813.62	806.78	800.41
5[a]	857.41	850.70	843.81	837.23	830.57	823.99	817.34	813.72	804.43
10[a]	860.13	853.52	846.93	840.40	833.78	827.44	820.97	814.38	808.48
15[a]	862.84	856.39	849.85	843.36	837.08	830.75	824.42	817.92	812.26
20[a]	865.41	859.01	852.66	846.31	840.19	833.93	827.73	821.49	815.83
25[a]	867.91	861.71	855.38	849.19	843.11	836.97	830.91	824.75	819.27
30[a]	870.38	864.24	858.03	851.93	845.90	839.89	833.95	828.00	822.67
35[a]	872.72	866.68	860.55	854.60	848.63	842.73	837.11	831.05	825.68
40[a]	875.09	869.06	863.11	857.19	851.43	845.54	839.76	833.97	828.75
45[a]	877.28	871.36	865.43	859.71	853.95	848.29	842.53	836.77	831.77
50[a]	879.57	873.68	867.78	862.21	856.40	850.85	845.32	839.53	834.62
55[a]	881.72	875.79	870.11	864.47	858.82	853.38	847.93	842.22	837.37
60[a]	883.73	878.13	871.68	866.83	861.27	855.90	850.52	844.89	840.12
70[b]	887.98	882.16	876.50	870.99	865.62	860.38	855.27	850.27	845.37
80[b]	891.88	886.18	880.64	875.25	870.01	864.90	859.92	855.06	850.30
90[b]	895.64	890.04	884.61	879.34	874.21	869.22	864.36	859.62	854.99
100[b]	899.27	893.77	888.44	883.27	878.25	873.37	868.61	863.98	859.47
110[b]	902.78	897.37	892.13	887.06	882.13	877.35	872.70	868.17	863.76
120[b]	906.17	900.85	895.70	890.72	885.88	881.19	876.63	872.19	867.88
130[b]		904.23	899.16	894.25	889.50	884.89	880.42	876.07	871.84
140[b]		907.50	902.51	897.68	893.00	888.47	884.08	879.81	875.66
150[b]		910.69	905.76	901.00	896.40	891.94	887.62	883.43	879.35

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

Table 5. Density (*ρ*) of Pentadecylbenzene as a Function of Pressure and Temperature

	ρ/kg·m^{-3} at the following values of *T*/K							
P/MPa	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
0.1013[a]	848.87	842.12	835.41	828.78	821.97	815.34	808.61	801.81
5[a]	851.73	845.00	838.54	831.97	825.50	818.90	812.21	805.72
10[a]	854.50	848.07	841.59	835.18	828.84	822.38	815.94	809.72
15[a]	857.31	850.88	844.50	838.32	832.04	825.77	819.54	813.34
20[a]	859.94	853.64	847.40	841.27	835.22	829.03	822.89	816.84
25[a]	862.53	856.25	850.16	844.20	838.21	832.10	826.15	820.24
30[a]	865.00	858.90	852.85	846.93	841.08	835.14	829.23	823.47
35[a]	867.39	861.42	855.52	849.66	843.86	838.25	832.23	826.60
40[a]	869.82	863.87	858.01	852.30	846.57	840.84	835.05	829.61
45[a]	872.06	866.19	860.47	854.81	849.15	843.55	837.95	832.52
50[a]	874.34	868.54	862.92	857.26	851.82	846.18	840.66	835.37
55[a]	876.44	870.77	865.34	859.63	854.30	848.79	843.30	838.12
60[a]	878.68	872.34	867.54	862.03	856.71	851.11	845.86	840.82
70[b]	882.59	877.11	871.73	866.44	861.22	856.06	850.94	845.85
80[b]	886.56	881.20	875.94	870.77	865.67	860.64	855.66	850.71
90[b]	890.39	885.12	879.97	874.91	869.93	865.02	860.16	855.34
100[b]	894.08	888.91	883.85	878.89	874.02	869.22	864.47	859.76
110[b]	897.64	892.56	887.59	882.73	877.95	873.25	868.60	864.00
120[b]	901.09	896.09	891.20	886.43	881.74	877.13	872.57	868.07
130[b]	904.44	899.51	894.70	890.00	885.40	880.87	876.41	871.99
140[b]	907.69	902.82	898.09	893.47	888.94	884.49	880.11	875.77
150[b]	910.85	906.05	901.38	896.82	892.37	887.99	883.69	879.43

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

form of the integral of $1/c^2$:

$$\int 1/c^2\, dp = p(B/F - CE/F^2 + DE^2/F^3) + p^2(C/F - DE/F^2)/2 + p^3(D/F)/3 + (A/F - BE/F^2 + CE^2/F^3 - DE^3/F^4)\ln(E + Fp) \quad (4)$$

which is the most significant contribution of the change of density with pressure from a reference pressure P_{ref} (0.1013 MPa):

$$\rho(P,T) - \rho(P_{\mathrm{ref}},T) = \int_{P_{\mathrm{ref}}}^{P} 1/c^2\, dP + T\int_{P_{\mathrm{ref}}}^{P} (\alpha_P{}^2/C_P)\, dP \quad (5)$$

where α_P represents the isobaric expansion coefficient and C_P the heat capacity at constant pressure. By calculating numerically the last integral of eq 5, it is possible to evaluate the density as a function of pressure from speed of sound data from the moment that the density and heat capacity are known at the reference pressure.[10] Unfortunately, no measurement of heat capacity has been made for these components. To overcome this lack of data, the density was measured up to 60 MPa (Tables 4 and 5). These moderate pressure density data were used to initiate the numerical calculation of the integral by an inverse technique.[11] The densities were then determined from integration throughout the speed of sound measurement pressure

Table 6. Isentropic Compressibility (κ_S) of Tridecylbenzene as a Function of Pressure and Temperature

	κ_S/GPa^{-1} at the following values of T/K								
P/MPa	293.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
0.1013	0.5657	0.6002	0.6370	0.6759	0.7181	0.7630	0.8113	0.8630	0.9178
10	0.5287	0.5586	0.5905	0.6239	0.6591	0.6972	0.7371	0.7795	0.8238
20	0.4967	0.5230	0.5508	0.5799	0.6102	0.6426	0.6766	0.7120	0.7491
30	0.4689	0.4925	0.5170	0.5424	0.5692	0.5972	0.6265	0.6569	0.6884
40	0.4446	0.4655	0.4877	0.5104	0.5342	0.5586	0.5844	0.6111	0.6385
50	0.4232	0.4422	0.4623	0.4825	0.5039	0.5257	0.5485	0.5721	0.5958
60	0.4040	0.4216	0.4399	0.4581	0.4771	0.4968	0.5174	0.5384	0.5596
70	0.3868	0.4029	0.4196	0.4364	0.4537	0.4717	0.4900	0.5087	0.5278
80	0.3715	0.3861	0.4015	0.4169	0.4329	0.4493	0.4658	0.4828	0.5000
90	0.3571	0.3709	0.3851	0.3992	0.4140	0.4291	0.4443	0.4597	0.4755
100	0.3442	0.3570	0.3701	0.3833	0.3970	0.4107	0.4247	0.4391	0.4534
110	0.3322	0.3443	0.3566	0.3688	0.3814	0.3943	0.4072	0.4202	0.4337
120	0.3213	0.3325	0.3440	0.3556	0.3673	0.3792	0.3913	0.4034	0.4157
130		0.3216	0.3326	0.3432	0.3543	0.3655	0.3766	0.3881	0.3993
140		0.3116	0.3219	0.3320	0.3424	0.3528	0.3632	0.3737	0.3844
150		0.3024	0.3120	0.3216	0.3311	0.3410	0.3509	0.3608	0.3707

Table 7. Isentropic Compressibility (κ_S) of Pentadecylbenzene as a Function of Pressure and Temperature

	κ_S/GPa^{-1} at the following values of T/K							
P/MPa	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
0.1013	0.5892	0.6245	0.6627	0.7027	0.7454	0.7915	0.8394	0.8914
10	0.5490	0.5793	0.6121	0.6457	0.6820	0.7208	0.7607	0.8029
20	0.5148	0.5416	0.5697	0.5991	0.6300	0.6630	0.6961	0.7321
30	0.4848	0.5087	0.5335	0.5598	0.5863	0.6144	0.6435	0.6739
40	0.4591	0.4805	0.5024	0.5254	0.5491	0.5734	0.5995	0.6258
50	0.4361	0.4553	0.4752	0.4958	0.5167	0.5389	0.5618	0.5846
60	0.4157	0.4336	0.4513	0.4699	0.4886	0.5088	0.5289	0.5493
70	0.3975	0.4135	0.4301	0.4471	0.4643	0.4823	0.5008	0.5188
80	0.3813	0.3959	0.4110	0.4267	0.4423	0.4587	0.4752	0.4920
90	0.3665	0.3800	0.3938	0.4080	0.4226	0.4376	0.4527	0.4679
100	0.3528	0.3654	0.3782	0.3915	0.4050	0.4188	0.4325	0.4466
110	0.3402	0.3519	0.3639	0.3766	0.3888	0.4016	0.4143	0.4274
120	0.3285	0.3398	0.3511	0.3626	0.3742	0.3860	0.3979	0.4099
130	0.3180	0.3285	0.3389	0.3497	0.3604	0.3718	0.3828	0.3939
140	0.3081	0.3181	0.3279	0.3382	0.3481	0.3587	0.3690	0.3793
150	0.2989	0.3082	0.3176	0.3271	0.3367	0.3467	0.3561	0.3659

Table 8. Parameters of the Tait Equation (Eqs 7−10) with T in K, P in MPa, and ρ in kg·m^{-3}

parameters			deviations
Tridecylbenzene			
$\rho_0 = 1.175\ 62 \times 10^{3}$	$a_0 = -1.271\ 80 \times 10^{-4}$	$b_0 = 5.722\ 85 \times 10^{2}$	AD% = 3.9×10^{-4}
$\rho_1 = -1.671\ 24$	$a_1 = 1.889\ 39 \times 10^{-7}$	$b_1 = -2.162\ 15$	AAD% = 1.2×10^{-2}
$\rho_2 = 2.659\ 13 \times 10^{-3}$	$a_2 = -3.565\ 00 \times 10^{-10}$	$b_2 = 2.259\ 87 \times 10^{-3}$	MD% = 5.8×10^{-2}
$\rho_3 = -2.347\ 30 \times 10^{-6}$			
Pentadecylbenzene			
$\rho_0 = 1.149\ 34 \times 10^{3}$	$a_0 = -7.551\ 20 \times 10^{-5}$	$b_0 = 3.867\ 28 \times 10^{2}$	AD% = 3.1×10^{-4}
$\rho_1 = -1.546\ 51$	$a_1 = -5.668\ 90 \times 10^{-8}$	$b_1 = -1.141\ 46$	AAD% = 7.9×10^{-3}
$\rho_2 = 2.625\ 81 \times 10^{-3}$	$a_2 = -7.834\ 10 \times 10^{-11}$	$b_2 = 8.859\ 79 \times 10^{-4}$	MD% = 7.2×10^{-2}
$\rho_3 = -2.618\ 80 \times 10^{-6}$			

range using a predictor−corrector method.[1] The values of density deduced from speed of sound are also listed in Tables 4 and 5. The accuracy of these data has been estimated to be 0.1% on the basis of comparisons with literature data for hexane.[7] The knowledge of speed of sound and density at the same conditions makes it possible to evaluate the isentropic compressibility κ_S according to the following relation:

$$\kappa_S = \frac{1}{\rho c^2} \qquad (6)$$

The resulting data, evaluated with an error less than 0.3%, are given in Tables 6 and 7. The full set of $P-\rho-T$ data was used to adjust the parameters of the Tait-like equation:

$$\frac{1}{\rho} = \frac{1}{\rho_{\text{ref}}} + a \ln\left(\left[\frac{P + b}{P_{\text{ref}} + b}\right]\right) \qquad (7)$$

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

$$\rho_{\text{ref}} = \rho_0 + \rho_1 T + \rho_2 T^2 + \rho_3 T^3 \qquad (8)$$

$$a = a_0 + a_1 T + a_2 T^2 \qquad (9)$$

$$b = b_0 + b_1 T + b_2 T^2 \qquad (10)$$

This equation, with the best-fit parameters (Table 8), matches the density data within the experimental uncertainty. It can be used to calculate the isentropic compressibility by derivation with respect to pressure:

$$\kappa_T = -\rho \frac{a}{P + b} \qquad (11)$$

with an accuracy of 2%.

Conclusion

In this work, the speed of sound in liquid tridecylbenzene and pentadecylbenzene was measured up to 150 MPa in the temperature range (293.15 to 373.15) K. In addition, measurements of density were performed on the same components up to 60 MPa. From both sets of measurements, the density was evaluate up to 150 MPa and the isentropic compressibility was determined in the same $P-T$ domain. Moreover, the density data were fit to a Tait-like equation within the experimental error in order to allow the determination of the isothermal compressibility of these aromatics. These data constitute a part of a program of systematic investigation of hydrocarbons with high molecular weight. Through the diversity of the properties studied and the extent of the pressure domain covered, these data will be of use for the thermodynamic characterization of these compounds, which have an importance in the petroleum industry due to their presence in significant amount in heavy oils.

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Received for review May 31, 2002. Accepted September 10, 2002.

JE025560N