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Speed of Sound and Some Thermodynamic Properties of Liquid Methylcyclopentane and Butylcyclohexane in a Wide Range of Pressure[1]

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Ultrasonic velocity measurements were performed on liquid methylcyclopentane and butylcyclohexane at pressures from atmospheric up to 150 MPa in the temperature range from 293 to 373 K using a pulse echo technique operating at 3 MHz. The data were used to evaluate various thermophysical properties such as density, and isentropic and isothermal compressibilities up to 150 MPa with the help of additional density measurements.

KEY WORDS: butylcyclohexane; compressibility; density; methylcyclopentane; pressure; speed of sound.

1. INTRODUCTION

Measurements of thermophysical properties such as density, compressibility, and heat capacities in pure liquids under pressure are of great interest not only for industrial applications (for example, in the petroleum industry), but also for fundamental aspects in view of designing equations of state able to represent dense fluids. As these measurements are difficult to perform under high pressure, it is often advantageous to measure the speed of sound to characterize the thermophysical properties of fluids. This property, which can be determined experimentally with a high degree of accuracy including at high pressures, presents the advantage of giving access to various physical properties [1, 2] if, at the same time, one has information about density and heat capacity at atmospheric pressure.

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Moreover, as both thermal and volumetric properties appear in its definition, the speed of sound can be used directly to develop and adjust equations of state [3, 4] or can, at least, be used to test the reliability with which they can represent derived properties [5].

It can readily be observed that, for the hydrocarbon components, most of the measurements of speed of sound under pressure has focused on the normal paraffins series. For the branched alkanes, the aromatics or naphthenes, other families which are of major interest for petroleum applications, studies are essentially restricted to compounds with few carbon atoms and at atmospheric pressure. It is therefore necessary to conduct specific experiments on components belonging to these families to set up a base which can then be used to define new models specially adapted to complex mixtures. With this aim in mind, an ultrasonic investigation of pure compounds belonging to the naphthene family (both alkylcyclopentane and alkylcyclohexane) has been initiated.

In this paper, which focuses on methylcyclopentane (C_6H_{12}) and butylcyclohexane ($C_{10}H_{20}$) in the liquid state, data of several thermophysical properties including density, speed of sound and isothermal and isentropic compressibilities determined from ultrasonic measurements are given at pressures ranging from atmospheric pressure to 150 MPa and at temperatures between 293.15 and 373.15 K.

2. EXPERIMENTAL

Speed-of-sound measurements were carried out using a pulse echo technique operating at 3 MHz. The experimental apparatus, which has been described in a previous publication [6], is made up of an autoclave cylindrical cell on which two piezoelectric (PZT) elements are fixed at each extremity. These transducers are connected to an ultrasonic emission/reception device (Panametrics 5055 PRM) which allows measurements by transmission and reflection. The speed of sound is deduced from a double measurement by transmission and by reflection of the transit time of the wave through the sample [7] using a digital oscilloscope with memory storage (Gould 4090). The path length of the wave through the sample was determined accurately at each pressure and temperature by calibration with water using the data of Del Grosso and Mader [8], Wilson [9], and of Petitet et al. [10]. To ensure thermal uniformity within the fluid, the cell is fully 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 determined using a platinum probe (Pt100) placed inside the experimental vessel. The pressure is generated by a pneumatic pump (Haskel) and measured by an HBM P3M gauge which is frequently checked against a dead weight tester (Bundenberg) to an uncertainty of better than 0.02 per cent. The propagation of all error sources leads to an uncertainty of the speed of sound better than 0.2 per cent over the entire pressure range (0.1 to 150 MPa), an uncertainty confirmed by several tests performed with hexane [11] and heptane [12].

Additional density measurements were carried out by means of an Anton Paar densimeter (DMA60 model) equipped with a high pressure cell (DMA 512 P) in the operating range of 0.1 to 60 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. [13] using reference data for water [9]. The overall uncertainty obtained by this apparatus is estimated to be better than $0.1\ \mathrm{kg \cdot m^{-3}}$.

Both compounds were supplied by Fluka with a stated purity higher than 99% and were used without further purification.

3. RESULTS AND DISCUSSION

The speed of sound u was measured at 10 K intervals from 293.15 to 373.15 K and from atmospheric pressure to 150 MPa. The results are reported in Table I and plotted as a function of temperature and pressure in Figs. 1 and 2. The data were fitted to a two-dimensional rational function which correlates $1/u^2$ instead of u:

$$\frac{1}{u^2} = \frac{\mathrm{A} + \mathrm{B}P + \mathrm{C}P^2 + \mathrm{D}P^3}{\mathrm{E} + \mathrm{F}P} \tag{1}$$

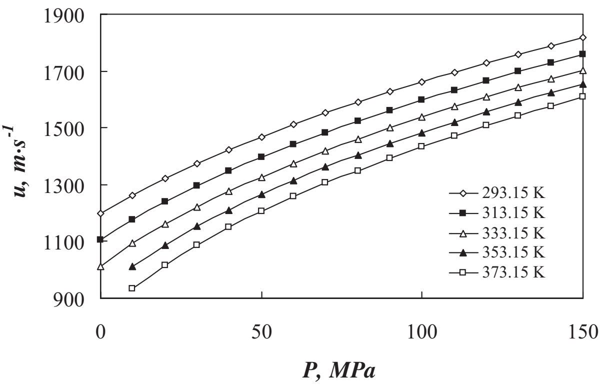

Fig. 1. Speed of sound u in liquid methylcyclopentane as a function of pressure along various isotherms.

Table I. Speed of Sound u ($m \cdot s^{-1}$) of Methylcyclopentane and Butylcyclohexane as a Function of Pressure and Temperature

	T (K)								
P (MPa)	293.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
methylcyclopentane									
0.1013	1200.3	1152.0	1104.9	1057.9	1012.5				
10	1263.6	1219.3	1175.5	1132.5	1092.2	1051.9	1011.6	974.5	934.0
20	1321.3	1279.2	1238.5	1198.6	1160.2	1123.4	1086.2	1051.7	1014.2
30	1373.9	1333.2	1295.2	1257.3	1221.2	1186.2	1151.9	1118.9	1086.8
40	1422.2	1384.0	1347.0	1311.1	1276.1	1242.9	1210.7	1179.3	1149.1
50	1467.9	1431.4	1395.5	1360.3	1327.2	1295.5	1264.5	1234.0	1205.5
60	1511.0	1474.3	1440.7	1406.7	1374.5	1345.3	1314.1	1284.8	1257.4
70	1551.7	1515.9	1482.8	1449.6	1418.7	1389.2	1361.0	1331.8	1305.2
80	1589.9	1555.1	1522.6	1491.4	1460.7	1431.5	1403.4	1376.0	1348.4
90	1626.9	1592.5	1561.0	1529.9	1500.5	1472.0	1444.1	1417.4	1392.2
100	1661.2	1627.9	1597.1	1567.5	1538.1	1510.1	1483.6	1456.9	1432.8
110	1695.1	1661.9	1632.0	1602.5	1574.1	1546.3	1520.6	1494.8	1470.2
120	1727.3	1694.8	1665.4	1636.6	1608.2	1581.5	1556.2	1530.6	1506.8
130	1759.0	1726.6	1697.4	1669.2	1641.4	1615.0	1589.9	1565.0	1541.7
140	1787.6	1757.5	1728.1	1700.5	1673.2	1647.5	1622.5	1598.1	1575.4
150	1817.1	1786.6	1757.4	1731.0	1704.0	1678.6	1654.6	1629.8	1607.5
butylcyclohexane									
0.1013	1328.7	1289.0	1247.7	1210.4	1169.8	1133.6	1097.1	1063.6	1028.7
10	1383.5	1346.7	1307.2	1270.4	1234.7	1199.7	1165.2	1131.7	1098.7
20	1432.8	1396.4	1360.6	1325.3	1291.7	1258.4	1225.7	1194.5	1164.4
30	1479.1	1443.6	1410.2	1375.8	1343.5	1312.1	1281.6	1251.7	1221.3
40	1522.5	1488.2	1455.5	1423.3	1391.7	1361.5	1332.3	1304.0	1274.9
50	1564.1	1530.3	1498.6	1467.6	1438.4	1408.2	1380.3	1351.8	1323.6
60	1603.4	1570.5	1539.4	1509.3	1479.6	1451.0	1421.9	1395.0	1368.8
70	1639.8	1607.5	1578.0	1547.9	1519.3	1492.6	1464.6	1438.1	1414.7
80	1675.4	1643.7	1614.8	1585.4	1558.3	1531.5	1504.9	1481.4	1454.5
90	1709.2	1678.0	1649.8	1621.0	1594.4	1568.1	1542.6	1518.6	1494.5
100	1741.3	1711.2	1683.5	1655.8	1629.7	1604.1	1579.0	1555.0	1531.3
110	1772.6	1743.4	1715.9	1688.7	1663.0	1637.7	1613.2	1589.3	1567.0
120	1802.7	1774.4	1747.0	1720.5	1694.9	1669.8	1646.5	1623.0	1600.2
130	1832.6	1803.3	1777.3	1751.4	1726.1	1701.9	1677.8	1655.3	1632.7
140	1860.9	1832.9	1805.5	1780.1	1755.1	1732.4	1708.5	1686.6	1664.0
150	1888.2	1860.7	1834.6	1808.4	1785.1	1761.4	1738.7	1715.7	1694.3

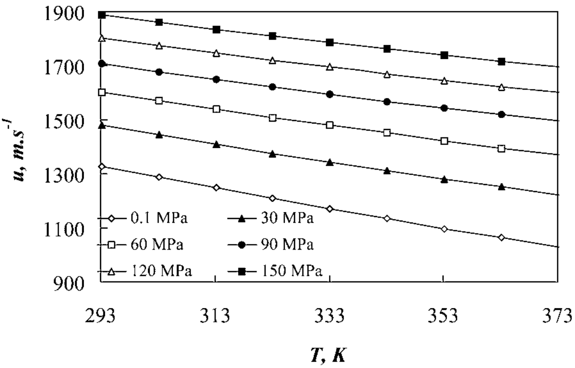

Fig. 2. Speed of sound u in liquid butylcyclohexane as a function of temperature along various isobars.

in which

$$A = A_0 + A_1 T + A_2 T^2 + A_3 T^3 \tag{2}$$

and

$$E = 1 + E_1 T \tag{3}$$

The coefficients of this equation, obtained by a least squares fit for each set of data, are given in Table II along with the fit characteristics. As can be seen, the maximum deviation observed between the experimental speed-of-sound data and the smoothing function is of the same magnitude as the experimental error. Moreover, an analysis of the average deviation shows that the smoothing function does not introduce any systematic error. Therefore, the function is appropriate to interpolate the speed-of-sound measurements and to calculate the integration of $1/u^2$ with respect to pressure which accounts for the main contribution to the variation of density ρ with pressure:

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

where C_P is the heat capacity at constant pressure and α_P is the isobaric expansion coefficient. Thus, by evaluating numerically the last integral of Eq. (5), it is possible to evaluate the density as a function of pressure from speed-of-sound data since the density and heat capacity are known at atmospheric pressure [1]. This was done by using a predictor-corrector procedure [14] in which the initialization procedure proposed by Denielou et al. [15] was used. The atmospheric density data required to initiate the

Table II. Parameters of Eqs. (1) to (3) with T in K, P in MPa, and u in $m \cdot s^{-1}$

Parameters			Deviations of u
methylcyclopentane			
$A_0 = -2.58380 \times 10^{-7}$	$A_3 = 3.45293 \times 10^{-15}$	$D = 1.51233 \times 10^{-14}$	$AD(\%) = -2.1 \times 10^{-3}$
$A_1 = 3.03499 \times 10^{-9}$	$B = 1.90545 \times 10^{-9}$	$E_1 = -2.02272 \times 10^{-3}$	$AAD(\%) = 4.5 \times 10^{-2}$
$A_2 = -5.06490 \times 10^{-12}$	$C = -6.84460 \times 10^{-12}$	$F = 7.51656 \times 10^{-3}$	$MD(\%) = 1.4 \times 10^{-1}$
butylcyclohexane			
$A_0 = 8.50788 \times 10^{-8}$	$A_3 = -3.40960 \times 10^{-15}$	$D = 9.56359 \times 10^{-15}$	$AD(\%) = 1.3 \times 10^{-3}$
$A_1 = 2.66863 \times 10^{-10}$	$B = 1.43645 \times 10^{-9}$	$E_1 = -1.74891 \times 10^{-3}$	$AAD(\%) = 3.5 \times 10^{-2}$
$A_2 = 2.31173 \times 10^{-12}$	$C = -4.53230 \times 10^{-12}$	$F = 6.77444 \times 10^{-3}$	$MD(\%) = 1.5 \times 10^{-1}$

iterative procedure were measured with the Anton Paar densimeter, whereas the atmospheric values of C_P come from the compilation of Zabransky et al. [16].

The density data derived from these speed-of-sound measurements are listed in Table III. The uncertainty of these data which has been estimated to be 0.1% on the basis of tests performed with hexane [11], was checked up to 60 MPa (Table III) by comparison with direct measurements performed with the high pressure DMA device. On reading this table one can see that the maximum deviation observed between the two sets of data is actually less than 0.1%.

The data for the speed of sound and density were used to evaluate the compressibilities of both components. The isentropic compressibility κ_S was determined (Table IV) with an uncertainty of 0.3% by using the following relation:

$$\kappa_S = \frac{1}{\rho u^2} \tag{5}$$

whereas the isothermal compressibility κ_T was correlated by means of a Tait-like equation:

$$\kappa_T = -\rho \frac{\mathrm{a}}{P + \mathrm{b}} \tag{6}$$

with

$$\mathrm{a} = \mathrm{a}_0 + \mathrm{a}_1 T + \mathrm{a}_2 T^2 \tag{7}$$

$$\mathrm{b} = \mathrm{b}_0 + \mathrm{b}_1 T + \mathrm{b}_2 T^2 \tag{8}$$

in which the coefficients a_0, a_1, a_2 and b_0, b_1, b_2 were adjusted by fitting the data of the volume change with respect to pressure $(v - v_{atm})$ to the integral

Table III. Density ρ ($kg \cdot m^{-3}$) Derived from Speed of Sound Measurements for Methylcyclopentane and Butylcyclohexane as a Function of Pressure and Temperature

	T (K)								
P (MPa)	293.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
methylcyclopentane									
0.1013	748.54	739.48	730.31	720.57	710.76				
10	756.83	748.54	739.95	731.11	722.06	712.86	703.55	694.18	684.79
20	764.65	756.91	748.91	740.69	732.31	723.83	715.29	706.76	698.30
30	771.77	764.48	756.95	749.22	741.37	733.44	725.49	717.59	709.80
40	778.33	771.41	764.27	756.95	749.52	742.03	734.56	727.16	719.89
50	784.43	777.83	771.01	764.03	756.96	749.84	742.75	735.75	728.91
60	790.13	783.80	777.27	770.59	763.82	757.02	750.25	743.59	737.10
70	795.49	789.41	783.13	776.70	770.19	763.66	757.18	750.80	744.61
80	800.56	794.70	788.64	782.44	776.16	769.87	763.62	757.50	751.56
90	805.37	799.70	793.84	787.84	781.77	775.69	769.66	763.76	758.05
100	809.94	804.46	798.78	792.96	787.08	781.18	775.35	769.64	764.13
110	814.31	808.99	803.47	797.83	792.11	786.39	780.73	775.19	769.86
120	818.50	813.33	807.96	802.47	796.91	791.34	785.83	780.46	775.29
130	822.51	817.48	812.26	806.90	801.48	796.06	790.70	785.47	780.45
140	826.38	821.48	816.38	811.16	805.87	800.58	795.35	790.26	785.37
150	830.11	825.32	820.35	815.25	810.08	804.92	799.81	794.84	790.08
Deviations from DMA results (up to 60 MPa)			AD(%): 4.8×10^{-5}		AAD(%): 1.4×10^{-2}		MD(%): 5.2×10^{-2}		
butylcyclohexane									
0.1013	799.20	791.88	784.28	776.80	769.14	761.31	753.59	745.87	737.77
10	805.82	798.81	791.73	784.60	777.43	770.20	762.94	755.64	748.31
20	811.96	805.28	798.55	791.79	784.99	778.17	771.32	764.46	757.60
30	817.66	811.27	804.84	798.38	791.90	785.41	778.91	772.41	765.91
40	823.00	816.85	810.68	804.49	798.28	792.07	785.86	779.65	773.46
50	828.03	822.10	816.15	810.19	804.22	798.25	792.28	786.33	780.40
60	832.78	827.05	821.31	815.55	809.79	804.03	798.28	792.54	786.83
70	837.30	831.75	826.18	820.61	815.03	809.46	803.90	798.36	792.84
80	841.60	836.21	830.81	825.41	820.00	814.59	809.20	803.83	798.49
90	845.72	840.48	835.23	829.97	824.71	819.46	814.22	809.00	803.81
100	849.66	844.56	839.44	834.33	829.21	824.10	819.00	813.92	808.87
110	853.45	848.47	843.49	838.50	833.51	828.52	823.55	818.60	813.67
120	857.10	852.24	847.37	842.50	837.63	832.76	827.91	823.07	818.26
130	860.61	855.87	851.11	846.35	841.59	836.83	832.08	827.36	822.66
140	864.01	859.37	854.72	850.06	845.40	840.75	836.10	831.48	826.87
150	867.31	862.76	858.21	853.64	849.08	844.52	839.97	835.44	830.93
Deviations from DMA results (up to 60 MPa)			AD(%): 3.1×10^{-2}		AAD(%): 3.2×10^{-2}		MD(%): 8.2×10^{-2}		

Table IV. Isentropic Compressibility κ_S (GPa^{-1}) of Methylcyclopentane and Butylcyclohexane as a Function of Pressure and Temperature

	T (K)								
P (MPa)	293.15	303.15	313.15	323.15	333.15	343.15	353.15	363.15	373.15
methylcyclopentane									
0.1013	0.9272	1.0190	1.1216	1.2400	1.3724				
10	0.8276	0.8986	0.9781	1.0664	1.1609	1.2678	1.3889	1.5168	1.6740
20	0.7491	0.8074	0.8705	0.9397	1.0146	1.0947	1.1850	1.2793	1.3923
30	0.6864	0.7359	0.7875	0.8443	0.9044	0.9691	1.0388	1.1132	1.1929
40	0.6352	0.6768	0.7212	0.7685	0.8193	0.8724	0.9288	0.9889	1.0520
50	0.5916	0.6275	0.6661	0.7073	0.7500	0.7946	0.8421	0.8925	0.9440
60	0.5543	0.5870	0.6198	0.6558	0.6930	0.7299	0.7719	0.8147	0.8581
70	0.5221	0.5512	0.5808	0.6127	0.6451	0.6785	0.7130	0.7510	0.7883
80	0.4942	0.5203	0.5470	0.5746	0.6039	0.6338	0.6649	0.6972	0.7318
90	0.4691	0.4931	0.5170	0.5423	0.5681	0.5950	0.6230	0.6517	0.6806
100	0.4474	0.4691	0.4908	0.5132	0.5370	0.5614	0.5859	0.6121	0.6374
110	0.4274	0.4476	0.4673	0.4881	0.5095	0.5318	0.5540	0.5774	0.6009
120	0.4095	0.4281	0.4462	0.4653	0.4852	0.5052	0.5255	0.5469	0.5681
130	0.3929	0.4103	0.4273	0.4448	0.4631	0.4816	0.5003	0.5198	0.5391
140	0.3787	0.3941	0.4102	0.4263	0.4432	0.4602	0.4776	0.4955	0.5130
150	0.3648	0.3796	0.3947	0.4094	0.4251	0.4409	0.4567	0.4737	0.4898
butylcyclohexane									
0.1013	0.7087	0.7600	0.8190	0.8787	0.9501	1.0222	1.1025	1.1851	1.2808
10	0.6484	0.6903	0.7391	0.7898	0.8438	0.9021	0.9655	1.0332	1.1070
20	0.5999	0.6368	0.6765	0.7191	0.7635	0.8115	0.8629	0.9169	0.9736
30	0.5590	0.5915	0.6248	0.6617	0.6996	0.7396	0.7817	0.8263	0.8754
40	0.5242	0.5528	0.5823	0.6136	0.6468	0.6811	0.7168	0.7543	0.7955
50	0.4936	0.5194	0.5456	0.5731	0.6010	0.6318	0.6625	0.6959	0.7314
60	0.4671	0.4902	0.5138	0.5383	0.5641	0.5907	0.6196	0.6484	0.6784
70	0.4442	0.4653	0.4861	0.5086	0.5316	0.5545	0.5799	0.6056	0.6302
80	0.4233	0.4426	0.4616	0.4820	0.5022	0.5234	0.5457	0.5669	0.5920
90	0.4047	0.4226	0.4399	0.4585	0.4770	0.4963	0.5161	0.5360	0.5570
100	0.3881	0.4043	0.4203	0.4372	0.4541	0.4716	0.4897	0.5081	0.5272
110	0.3729	0.3878	0.4027	0.4182	0.4338	0.4500	0.4666	0.4837	0.5005
120	0.3590	0.3727	0.3867	0.4010	0.4156	0.4307	0.4455	0.4613	0.4773
130	0.3460	0.3593	0.3720	0.3852	0.3988	0.4126	0.4269	0.4411	0.4560
140	0.3342	0.3464	0.3589	0.3712	0.3840	0.3963	0.4097	0.4228	0.4368
150	0.3234	0.3348	0.3462	0.3582	0.3696	0.3817	0.3938	0.4066	0.4192

Table V. Parameters of the Tait Equation (Eqs. (6)–(8)) with T in K, P in MPa, and ρ in $kg \cdot m^{-3}$

Parameters			Deviations		
methylcyclopentane			$\boldsymbol{V - V_{atm}}$	$\boldsymbol{\rho}$	$\boldsymbol{\kappa_T}$ **(Eq. 9)**
$\rho_0 = 7.03784 \times 10^2$	$a_0 = -5.94830 \times 10^{-5}$	$b_0 = 3.53339 \times 10^2$	AD(%) = 3.3×10^{-2}	AD(%) = 4.5×10^{-6}	AD(%) = 0.2
$\rho_1 = 1.62195$	$a_1 = -1.93120 \times 10^{-7}$	$b_1 = -1.31764$	AAD(%) = 8.4×10^{-2}	AAD(%) = 3.6×10^{-3}	AAD(%) = 0.3
$\rho_2 = -6.53415 \times 10^{-3}$	$a_2 = -1.74920 \times 10^{-11}$	$b_2 = 1.22935 \times 10^{-3}$	MD(%) = 1.0	MD(%) = 1.9×10^{-2}	MD(%) = 1.6
$\rho_3 = 5.18684 \times 10^{-6}$					
butylcyclohexane			$\boldsymbol{V - V_{atm}}$	$\boldsymbol{\rho}$	$\boldsymbol{\kappa_T}$ **(Eq. 9)**
$\rho_0 = 9.73126 \times 10^2$	$a_0 = -2.67250 \times 10^{-5}$	$b_0 = 3.54501 \times 10^2$	AD(%) = 8.5×10^{-2}	AD(%) = 8.4×10^{-4}	AD(%) = 0.2
$\rho_1 = -4.24368 \times 10^{-1}$	$a_1 = -3.57690 \times 10^{-7}$	$b_1 = -1.16908$	AAD(%) = 1.5×10^{-1}	AAD(%) = 4.9×10^{-3}	AAD(%) = 0.5
$\rho_2 = -6.61410 \times 10^{-4}$	$a_2 = 3.05042 \times 10^{-10}$	$b_2 = 9.96142 \times 10^{-4}$	MD(%) = 9.1×10^{-1}	MD(%) = 1.6×10^{-2}	*MD*(%) = 1.7
$\rho_3 = 2.91536 \times 10^{-7}$					

of $a/(p+b)$. The parameters determined by a least-squares method are listed in Table V along with the deviations observed between the data and the values of $v-v_{atm}$ calculated from the integration of Eq. (6) with respect to pressure. These deviations, which are less than the experimental error, show that the function leads to a good representation of volume change and thus to the isothermal compressibility. To check the accuracy, the isothermal compressibilities determined from Eq. (6) were compared (Table V) to those calculated from speed of sound using the following relation:

$$\kappa_T = \frac{1}{\rho u^2} + \frac{T\alpha_P^2}{\rho C_P} \tag{9}$$

in which the isobaric expansion α_P was evaluated from derivation of density data, whereas the values of heat capacity come from:

$$C_P = C_{P_{atm}} - T\int_{P_{atm}}^{P} \frac{\partial^2 1/\rho}{\partial T^2}\, dp \tag{10}$$

The maximum deviation observed between the two procedures is 1.7%. This value shows that Eq. (6) adjusted in this way yields very good predictions of isothermal compressibility.

Comparison of the present density data with those reported by the API Research Project 42 at atmospheric pressure shows an absolute average deviation of 0.06% and a maximum deviation of only 0.1% for methylcyclopentane. For butylcyclohexane, a deviation of 0.02% is observed at low temperature (293 to 333 K) whereas a deviation of 0.5% is obtained at higher temperature. Excellent agreement was found with the data of Tatevskiy [18] for methylcyclopentane with an average deviation of 0.01% and a maximum deviation of 0.02%.

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