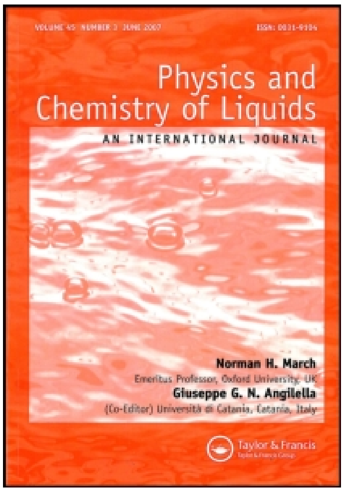

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Acoustic and Thermodynamic Properties of Benzene and Cyclohexane as a Function of Pressure and Temperature

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The phase comparison pulse echo method with two reflectors has been used to determine the velocity of sound in benzene and cyclohexane in the temperature range from 283–323 K and up to the freezing pressures of the liquids, the maximum pressures being 170 and 80 MPa respectively. The density, the isothermal compressibility, the isobaric thermal expansion and the specific heat at constant pressure of both liquids have been evaluated from the measured sound velocity, using a computational method similar to that of Davis and Gordon. The calculated densities for both liquids agree well with those found in the literature.

1 INTRODUCTION

In recent years, ultrasonic methods have been used increasingly for determining equations of state and thermodynamic properties of liquids and liquid mixtures at elevated pressures. Apart from the high precision attainable with ultrasonic measurements, these also have the advantage of simplicity and rapidity of the experimental procedure. Furthermore, the rapid development of computer systems during the last decade has simplified the problem of carrying out the tedious

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numerical calculations necessary for deriving other physical properties. It is thus possible to extract easily from the ultrasonic data such important thermodynamic properties as the adiabatic compressibility and specific heat of a liquid at high pressures, where direct measurements are extremely difficult.

These facts have led to the development at this laboratory of an ultrasonic apparatus for the measurement of sound velocity in liquids at pressures up to 3 kbar and at temperatures down to 77 K. The apparatus is based on the phase comparison pulse-echo method with two reflectors, the principle of which has been described in a previous paper.[1] The method has the advantage over the single reflector pulse-echo method that there are no unwanted reflections of the sound pulse from the transducer and, consequently, the uncertainty in the transit time determination is very small. Detailed measurements on n-heptane have shown that the method is capable of giving a very high precision for the sound velocity at high pressures. It has, therefore, been decided to use this method in the systematic investigation of acoustic and thermodynamic properties of a number of simple organic liquids as a function of pressure and temperature.

In the present work, we have chosen a pair of cyclic organic liquids, namely benzene and cyclohexane, which have similar physical properties but which differ in their molecular shapes. The sound velocity and pVT data of these liquids are of considerable interest in the analysis of both empirical and theoretical equations of state. To date, however, sound velocity measurements on benzene have not been carried out at pressures above 60 MPa, while for cyclohexane only scattered data are available in the literature. The purpose of the present work is, therefore, not only to increase the precision of the measurements but also to cover a wider range of pressure and temperature than that of previous studies.

2 EXPERIMENTAL

2.1 Materials

The materials for the present investigation, benzene and cyclohexane, were purchased from BDH Chemicals Ltd., Poole, U.K. According to the specifications of the supplier, the materials were of AnalaR grade and the purity of each compound was better than 99.5 % by volume. In the case of benzene, the main impurity was H_2O ($<0.05\,\%$) while for cyclohexane, the principal impurities were H_2O ($<0.03\,\%$), benzene ($<0.01\,\%$) and cyclohexene ($<0.01\,\%$). The materials were used without further purification.

2.2 Ultrasonic apparatus

As mentioned above, the present ultrasonic apparatus is based on a modified phase comparison pulse-echo method, which makes use of two reflectors placed at unequal distances from a transducer. With the exception of a few minor improvements, the design of the ultrasonic cell is the same as that described in Ref. 1. Essentially, it consists of a hollow copper cylinder in which an X-cut quartz transducer, serving both as the transmitter and the receiver, is clamped at its edges, while two copper reflectors are fixed at each end at distances of, respectively, 6 and 4 cm from the transducer. The electrodes on the transducer surface are formed by successive evaporation of a chromium and a gold layer which are not affected by the liquids under study. The apparatus is operated at a frequency of 2 MHz. In addition to several measures taken in the previous design to reduce unwanted reflections, the lower parts of the two reflectors now have a concave section, so that reflections from the corresponding surfaces are delayed in reaching the transducer and, consequently the pulse length can be increased.

2.3 Measurement of sound velocity

The sound velocity is determined from the transit time of an ultrasonic pulse through the liquid and the corresponding acoustic path length. In the present ultrasonic method with two reflectors, two electrical pulses are applied successively to the transducer with a time interval such that the echo of the first pulse reflected from the distant reflector and the echo of the second pulse reflected from the near reflector coincide at their arrival at the quartz transducer. The phase difference between the echoes is then equal to:

$$\Delta\phi = 2\pi f\tau \qquad (1)$$

where f is the frequency and τ is the difference in transit time of the echoes. The initial amplitudes of the pulses are such that their intensities are equal on their arrival at the transducer. The determination of the transit time involves the measurement of a set of null frequencies at which the two pulses completely cancel each other and the phase difference satisfies the relation:

$$\Delta\phi = (2n + 1)\pi \qquad (2)$$

where the integer n is the order of interference. An approximate value of n is easily found from the relation:

$$n = (f_n/\Delta f_n) - \tfrac{1}{2} \qquad (3)$$

derived from (1) and (2), $\Delta f_n = f_{n+1} - f_n$ being the difference in successive null frequencies. The exact value of n is then established by rounding off the approximate value. Subsequently, τ is found easily from (1) and (2). The corresponding acoustic path length l is determined by using a comparator with a precision better than 0.005 %. This is achieved by making a large number of measurements and taking an average. The value of the path length difference is 0.040042 m at 295.7 K and 0.1 MPa.

The measured transit time must be corrected in order to take account of the effect of diffraction. The appropriate corrections are determined at each experimental point using the method described by Papadakis.[4] In the present situation, diffraction corrections for the two liquids are estimated to be between 0.05 and 0.08 %, depending on the value of the sound velocity. Furthermore, for determining sound velocities at different pressures and temperatures, appropriate corrections are introduced to the measured path length using the following relation:

$$l(p, T) = l(0, 0)[1 + \alpha'(T)\,\Delta T][1 + \beta'(T)\,\Delta p],$$

$\alpha'(T)$ and $\beta'(T)$ being the coefficients of linear thermal expansion and compressibility of the construction material respectively.

2.4 Measurement of pressure and temperature

The high pressure system consists of a steel pressure vessel containing in it the ultrasonic cell, a mercury gas compressor coupled to a hydraulic oil press with which the liquids can be compressed to the required pressure and a pressure balance for controlling and measuring the pressure with an accuracy better than 1 in $10.^4$ Details of the gas compressor and the pressure balance can be found elsewhere.[2] The steel pressure vessel is surrounded by a demountable vacuum jacket and, together, they form a vacuum cryostat. A detailed description of this type of cryostat has been given elsewhere.[3] In this experiment, adequate measures were taken to keep the temperature gradient over the whole length of the ultrasonic cell below 10 mK. The temperature was measured by means of a calibrated Pt resistance thermometer with an estimated accuracy of 1 mK. All temperatures were recorded on the IPTS '68 Scale.

3 EXPERIMENTAL RESULTS

The velocity of sound in benzene and cyclohexane has been measured isothermally as a function of pressure in the temperature range between

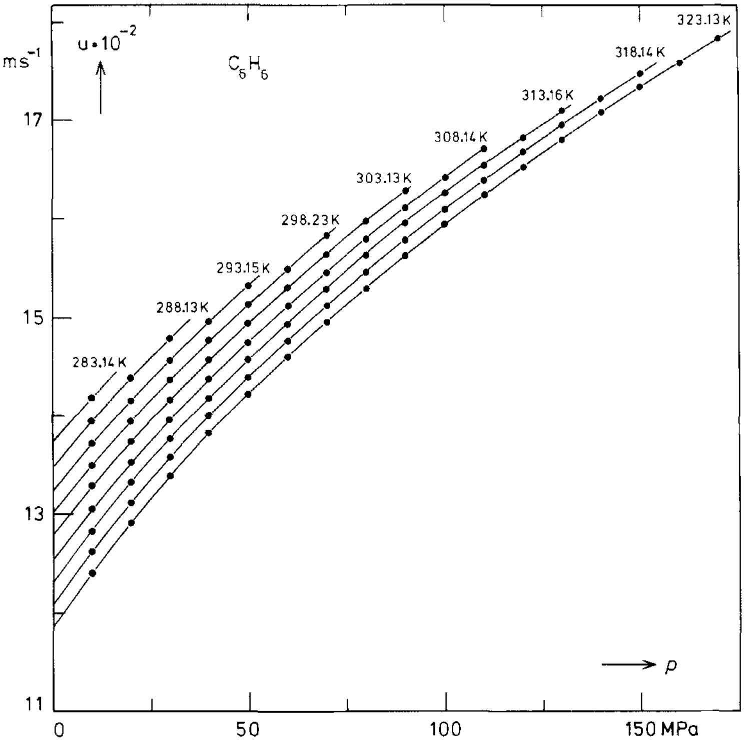

FIGURE 1 Sound velocity in benzene as a function of pressure at several temperatures.

283 and 323 K with intervals of about 5 K. For both liquids, the experimental data are taken from 0.1 MPa upwards with a pressure interval of 5 MPa, except for benzene at lower temperatures where the intervals are smaller, namely 0.5 to 1.0 MPa. In both cases, an isotherm is terminated at a pressure which is close to the freezing pressure of the liquids. The results are shown in Figures 1 and 2, in which the measured sound velocities (in ms^{-1}) are plotted as a function of pressure (in MPa) at several temperatures.

The accuracy of the measured sound velocities is determined by the accuracy with which the difference in acoustic path lengths l and the difference in transit times τ are determined. The accuracy of the measurement of τ depends mainly on the accuracy of the diffraction corrections, since the null frequencies can be measured with a precision better than 1 in $10,^5$ while the value of the integer n is known exactly. The analysis for the diffraction corrections, as carried out by the method of Papadakis,[4] shows that the maximum error in τ is less than 0.01 %. Considering that the error in the determination of the acoustic

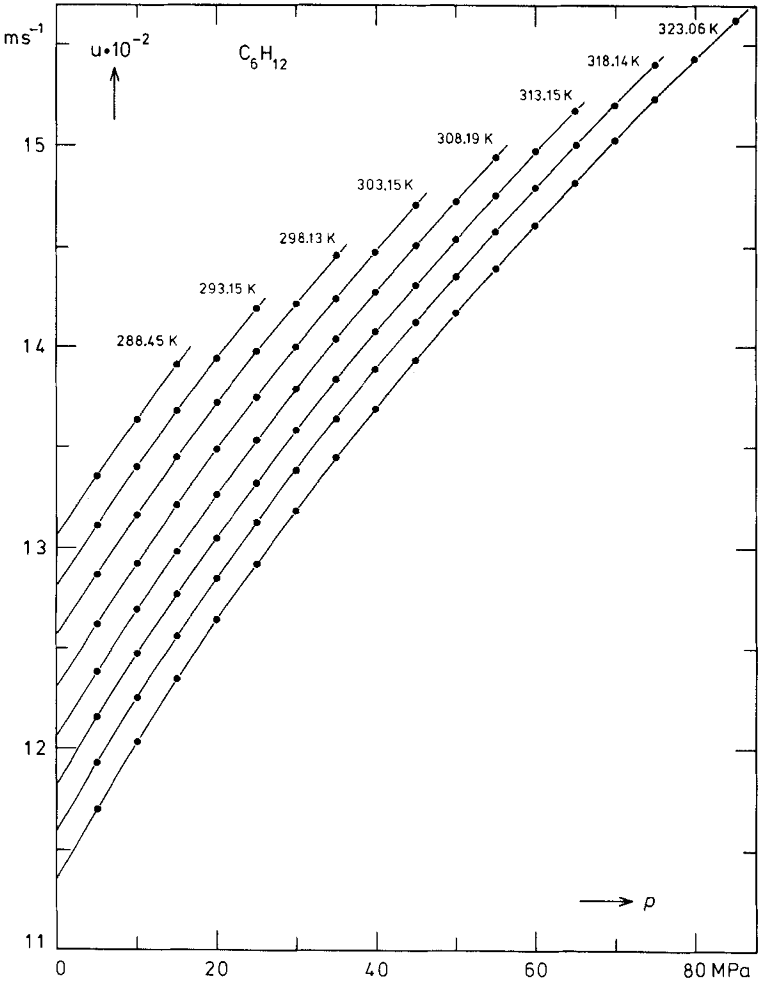

FIGURE 2 Sound velocity in cyclohexane as a function of pressure at several temperatures.

path length l is less than 0.01% and that the inaccuracies in pressure and temperature corrections of l are very small, one may conclude that the overall accuracy of the sound velocity measurements is better than 0.02%.

The sound velocity data for each isotherm are smoothed by fitting into an equation of the form:

$$p = \sum_{i=0}^{3} a_i u^i \tag{4}$$

by a least squares method. Here, p is the pressure in MPa, u is the sound velocity in ms^{-1} and a_i are the coefficients which are functions of the temperature only. It turns out from the least squares analysis that the standard deviations for the isotherms are between 0.05 and 0.1 ms^{-1} for benzene and 0.05 to 0.2 ms^{-1} for cyclohexane, while the maximum deviations of the experimental results from the smoothed curves are mostly in the range 0.1 to 0.2 ms^{-1}. It has, therefore, been used later in deriving from the sound velocity data the density and other physical properties of the two liquids.

The complete sound velocity data for each liquid can be fitted to a double polynomial equation of the form

$$p - p_0 = \sum_{i=1}^{m} \sum_{j=0}^{n} a_{ij}(u - u_0)^i T^j \tag{5}$$

by a least squares method. Here $p_0 = 0.1$ MPa, u_0 is the sound velocity at 0.1 MPa, T is the temperature in K and a_{ij} are the coefficients. The analysis shows that for both liquids, the best fit can be achieved with $m = 3$ and $n = 2$. The resulting coefficients a_{ij} are presented in Tables I and II. The average deviation for the best fit is found to be 0.1 ms^{-1}, which is less than the experimental accuracy of the present measurement.

The present sound velocity data for benzene at 0.1 MPa are found to be in very good agreement with those of Bobik[5] and Langguth *et al.*[6] and in fair agreement with those reported by Takagi *et al.*[7] and Klyohara *et al.*[8] At higher pressures, however, the agreement with previous measurements is less satisfactory, as can be seen in Figure 3 where deviations of the present data at 298 K from those reported previously are plotted as a function of the pressure. Except for a few data points of Takagi, which fall on the velocity *vs.* pressure isotherm, the present data are 0.1 to 0.2% higher than the previous ones. A comparison of the complete data for benzene with the previous data over the full experimental range shows that the agreement is fair, the

TABLE I

Coefficients a_{ij} of Eq. (5) for benzene in MPa $\mathrm{m}^{-i}\mathrm{s}^{i}\mathrm{K}^{-j}$

j \ i	1	2	3
0	0.5435209	2.627405×10^{-3}	-7.283321×10^{-6}
1	-1.359705×10^{-3}	-1.525326×10^{-5}	4.671585×10^{-8}
2	6.298076×10^{-7}	2.368336×10^{-8}	$-7.449643 \times 10^{-11}$

TABLE II

Coefficients a_{ij} of Eq. (5) for cyclohexane in MPa $m^{-i}s^{i}K^{-j}$

$j \backslash i$	1	2	3
0	0.2525400	2.791449×10^{-3}	-5.681965×10^{-6}
1	1.016006×10^{-4}	-1.703754×10^{-5}	3.685258×10^{-8}
2	-1.415322×10^{-6}	2.732418×10^{-8}	$-5.951442 \times 10^{-11}$

deviation being less than 0.3%. In the case of cyclohexane, the present data at 0.1 MPa agree well with those of Tamura *et al.*[9] and Wilhelm *et al.*[10] However, the velocity data reported by Takagi[11] are lower than the present data by 0.1 to 0.3% at higher temperatures and about 0.4% at lower temperatures.

4 DENSITY AND THERMODYNAMIC PROPERTIES

The pVT and thermodynamic properties of benzene and cyclohexane have been computed from the present sound velocity data following a

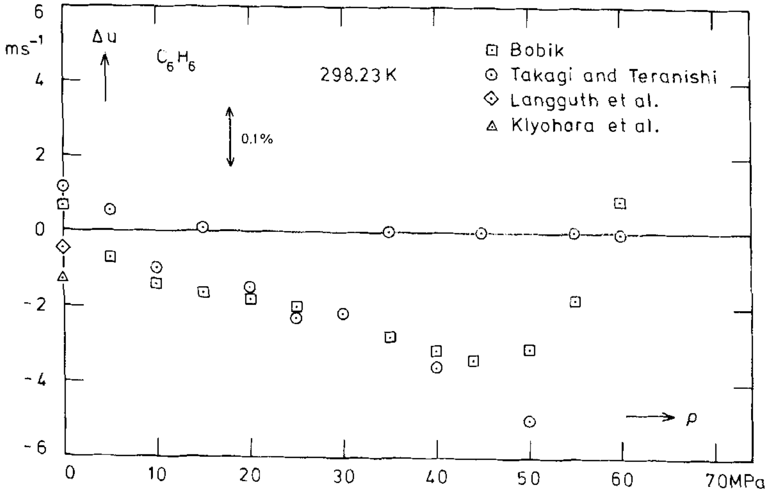

FIGURE 3 Comparison of previous and present sound velocity data for benzene.

method similar to that of Davis and Gordon.[12] This is based on the sequential operation of the following thermodynamic relations:

$$\beta_T = \frac{1}{\rho}\left(\frac{1}{u^2} + \frac{T\alpha^2}{C_p}\right) \tag{6}$$

$$\left(\frac{\partial \alpha}{\partial p}\right)_T = -\left(\frac{\partial \beta_T}{\partial T}\right)_p \tag{7}$$

$$\left(\frac{\partial \rho}{\partial p}\right)_T = \frac{1}{u^2} + \frac{T\alpha^2}{C_p} \tag{8}$$

$$\left(\frac{\partial C_p}{\partial p}\right)_T = \frac{T}{\rho}\left[\left(\frac{\partial \alpha}{\partial T}\right)_p + \alpha^2\right] \tag{9}$$

Here, β_T is the isothermal compressibility, α is the volume thermal expansion coefficient, ρ the density, u the sound velocity and C_p is the specific heat at constant pressure.

Equations (7)–(9) form a set of first order differential equations with initial values. The quantities $\rho_0(T)$ and $C_{p_0}(T)$ at 0.1 MPa are taken from literature,[20,21] while $\alpha_0(T)$ is derived from $\rho_0(T)$. By a simultaneous, step by step integration of the Eqs (7)–(9) with respect to pressure, and applying Eq. (6) at each step, the values of ρ, α, C_p and β_T are computed at elevated pressures. In contrast to the procedure of Davis and Gordon where a simple Euler method was used for the solution of the differential equation, the present computational procedure consists of using a more accurate method, namely Adams extrapolation and interpolation method[13] with pressure steps of 1.0 MPa for benzene and 0.5 MPa for cyclohexane. For the first three steps, however, a modified Euler method has to be employed since these cannot be calculated with the Adams' method. For these steps, smaller pressure intervals, namely 0.5 MPa for benzene and 0.25 MPa for cyclohexane, are taken in order to match the accuracies. In both cases, the appropriate values for the pressure steps are selected by trial and error. The derivatives $(\partial\beta_T/\partial T)_p$ and $(\partial\alpha/\partial T)_p$ in Eqs (7) and (9) respectively, are obtained by fitting the values of $\beta_T(T)$ and $\alpha(T)$ into appropriate polynomials in T using a least squares method.

It should be pointed out that the major contribution to the density increment due to pressure actually comes from the term $1/u^2$ in Eq. (8) since the difference between the adiabatic and isothermal compressibility of liquids is small. Accordingly, it is important to evaluate the integral

$$I = \int \frac{\mathrm{d}p}{u^2} \tag{10}$$

TABLE III

The experimental values of sound velocity and derived physical properties of benzene as a function of pressure and temperature

T K	p MPa	u m s^{-1}	ρ kg m^{-3}	$\beta_T \times 10^4$ MPa^{-1}	$\alpha \times 10^4$ K^{-1}	C_p J kg^{-1} K^{-1}
323.125	0.1	1185.7	846.92	11.874	12.878	1820.0
	10.0	1240.9	856.37	10.590	11.989	1803.7
	20.0	1291.5	865.03	9.574	11.255	1790.6
	30.0	1337.8	872.98	8.756	10.645	1779.8
	40.0	1380.9	880.35	8.082	10.126	1770.9
	50.0	1421.1	887.24	7.514	9.676	1763.3
	60.0	1459.1	893.71	7.034	9.294	1756.5
	70.0	1495.0	899.82	6.612	8.939	1750.3
	80.0	1529.1	905.63	6.249	8.638	1743.8
	90.0	1561.7	911.15	5.919	8.339	1737.6
	100.0	1593.0	916.42	5.613	8.036	1734.0
	110.0	1623.0	921.45	5.354	7.805	1731.3
	120.0	1651.8	926.29	5.120	7.595	1728.7
	130.0	1679.7	930.94	4.907	7.397	1726.3
	140.0	1706.7	935.43	4.712	7.214	1724.2
	150.0	1732.8	939.77	4.534	7.042	1722.2
	160.0	1758.1	943.96	4.369	6.881	1720.3
	170.0	1782.7	948.02	4.217	6.730	1718.4
318.135	0.1	1208.4	852.35	11.382	12.720	1804.1
	10.0	1262.1	861.48	10.195	11.868	1788.4
	20.0	1311.5	869.88	9.249	11.161	1775.7
	30.0	1357.0	877.61	8.482	10.570	1765.4
	40.0	1399.3	884.80	7.846	10.066	1756.7
	50.0	1438.9	891.52	7.308	9.627	1749.4
	60.0	1476.3	897.85	6.848	9.246	1742.8
	70.0	1511.8	903.84	6.445	8.898	1736.7
	80.0	1545.5	909.52	6.093	8.586	1730.4
	90.0	1577.8	914.93	5.776	8.290	1724.4
	100.0	1608.7	920.10	5.494	8.027	1720.9
	110.0	1638.4	925.05	5.245	7.800	1718.2
	120.0	1667.0	929.81	5.018	7.589	1715.7
	130.0	1694.6	934.39	4.812	7.393	1713.4
	140.0	1721.3	938.80	4.624	7.210	1711.3
	150.0	1747.2	943.07	4.451	7.039	1709.3
313.156	0.1	1231.0	857.73	10.917	12.566	1788.5
	10.0	1283.5	866.56	9.818	11.749	1773.4
	20.0	1331.8	874.71	8.936	11.068	1761.2
	30.0	1376.4	882.23	8.216	10.497	1751.2
	40.0	1418.0	889.24	7.616	10.007	1742.8
	50.0	1457.0	895.80	7.106	9.579	1735.7
	60.0	1493.9	901.99	6.667	9.199	1729.3
	70.0	1528.8	907.84	6.282	8.856	1723.4
	80.0	1562.2	913.40	5.940	8.534	1717.3
	90.0	1594.0	918.70	5.636	8.241	1711.5
	100.0	1624.6	923.78	5.377	8.018	1708.0
	110.0	1654.0	928.65	5.137	7.795	1705.4
	120.0	1682.4	933.32	4.918	7.582	1702.9
	130.0	1709.8	937.83	4.719	7.388	1700.7

TABLE III (*continued*)

T K	p MPa	u m s^{-1}	ρ kg m^{-3}	$\beta_T \times 10^4$ MPa^{-1}	$\alpha \times 10^4$ K^{-1}	C_p J kg^{-1} K^{-1}
308.143	0.1	1254.0	863.12	10.470	12.413	1773.1
	10.0	1305.1	871.65	9.454	11.630	1758.6
	20.0	1352.4	879.56	8.632	10.976	1746.8
	30.0	1396.2	886.88	7.958	10.424	1737.1
	40.0	1437.0	893.70	7.392	9.948	1729.0
	50.0	1475.4	900.11	6.909	9.531	1722.2
	60.0	1511.7	906.16	6.488	9.152	1715.9
	70.0	1546.2	911.89	6.122	8.815	1710.3
	80.0	1579.1	917.33	5.789	8.482	1704.3
	90.0	1610.6	922.52	5.498	8.193	1698.7
	100.0	1640.8	927.50	5.261	8.009	1695.3
	110.0	1669.9	932.28	5.031	7.790	1692.8
303.129	0.1	1278.1	868.47	10.034	12.262	1758.0
	10.0	1328.1	876.72	9.095	11.513	1744.1
	20.0	1374.3	884.38	8.331	10.886	1732.7
	30.0	1417.1	891.49	7.700	10.352	1723.4
	40.0	1457.2	898.13	7.167	9.890	1715.5
	50.0	1495.0	904.38	6.711	9.484	1708.9
	60.0	1530.7	910.29	6.310	9.106	1702.8
	70.0	1564.8	915.89	5.960	8.774	1697.3
	80.0	1597.3	921.21	5.637	8.431	1691.6
	90.0	1628.5	926.28	5.358	8.145	1686.1
298.228	0.1	1300.4	873.67	9.642	12.117	1743.5
	10.0	1349.2	881.66	8.772	11.401	1730.1
	20.0	1394.6	889.10	8.057	10.798	1719.2
	30.0	1436.9	896.02	7.463	10.282	1710.1
	40.0	1476.5	902.50	6.959	9.833	1702.6
	50.0	1513.8	908.60	6.526	9.438	1696.1
	60.0	1549.0	914.37	6.142	9.061	1690.3
	70.0	1582.6	919.85	5.808	8.734	1684.9
293.149	0.1	1324.5	879.04	9.248	11.969	1728.8
	10.0	1372.0	886.76	8.444	11.286	1715.9
	20.0	1416.2	893.97	7.781	10.708	1705.4
	30.0	1457.5	900.70	7.226	10.210	1696.7
	40.0	1496.3	907.01	6.752	9.775	1689.4
	50.0	1533.0	912.96	6.343	9.390	1683.1
288.129	0.1	1348.4	884.30	8.876	11.825	1714.5
	10.0	1394.5	891.77	8.136	11.173	1702.2
	20.0	1438.0	898.77	7.518	10.620	1692.1
	30.0	1478.6	905.31	6.996	10.141	1683.7
283.143	0.1	1372.4	889.50	8.524	11.683	1700.7
	10.0	1417.5	896.72	7.838	11.063	1688.8

TABLE IV

The experimental values of sound velocity and derived physical properties of cyclohexane as a function of pressure and temperature

T K	p MPa	u m s^{-1}	ρ kg m^{-3}	$\beta_T \times 10^4$ MPa^{-1}	$\alpha \times 10^4$ K^{-1}	C_p J kg^{-1} K^{-1}
323.061	0.1	1135.8	750.20	14.024	13.011	1974.9
	5.0	1170.2	755.18	13.034	12.428	1963.7
	10.0	1203.1	759.95	12.175	11.906	1953.7
	15.0	1234.2	764.45	11.434	11.444	1944.7
	20.0	1263.7	768.70	10.789	11.037	1936.7
	25.0	1291.8	772.75	10.218	10.663	1929.3
	30.0	1318.7	776.61	9.712	10.327	1922.6
	35.0	1344.6	780.30	9.256	10.016	1916.4
	40.0	1369.4	783.83	8.832	9.700	1911.1
	45.0	1393.5	787.23	8.460	9.441	1906.7
	50.0	1416.7	790.49	8.117	9.191	1902.8
	55.0	1439.2	793.65	7.807	8.970	1899.5
	60.0	1461.1	796.69	7.515	8.747	1897.1
	65.0	1482.3	799.64	7.254	8.564	1895.5
	70.0	1503.0	802.50	7.013	8.394	1894.3
	75.0	1523.2	805.27	6.789	8.235	1893.2
	80.0	1542.9	807.97	6.581	8.086	1892.4
	85.0	1562.1	810.59	6.386	7.947	1891.8
318.139	0.1	1159.1	754.98	13.404	12.817	1952.7
	5.0	1192.6	759.78	12.494	12.259	1941.9
	10.0	1224.8	764.39	11.699	11.758	1932.2
	15.0	1255.3	768.74	11.009	11.313	1923.6
	20.0	1284.2	772.86	10.406	10.917	1915.9
	25.0	1311.9	776.79	9.870	10.555	1908.8
	30.0	1338.4	780.53	9.393	10.227	1902.3
	35.0	1363.9	784.12	8.962	9.924	1896.3
	40.0	1388.5	787.56	8.569	9.635	1891.2
	45.0	1412.2	790.88	8.217	9.383	1886.9
	50.0	1435.2	794.07	7.895	9.149	1883.1
	55.0	1457.4	797.15	7.601	8.935	1879.9
	60.0	1479.0	800.13	7.332	8.743	1877.6
	65.0	1500.0	803.02	7.084	8.566	1876.0
	70.0	1520.5	805.82	6.854	8.401	1874.8
	75.0	1540.4	808.54	6.640	8.246	1873.8
313.149	0.1	1182.7	759.79	12.812	12.623	1930.1
	5.0	1215.4	764.41	11.975	12.090	1919.8
	10.0	1246.8	768.86	11.240	11.610	1910.5
	15.0	1276.7	773.06	10.599	11.182	1902.2
	20.0	1305.1	777.06	10.035	10.797	1894.8
	25.0	1332.3	780.87	9.533	10.447	1887.9
	30.0	1358.4	784.51	9.083	10.127	1881.7
	35.0	1383.5	788.00	8.677	9.833	1875.9
	40.0	1407.8	791.35	8.313	9.570	1870.9
	45.0	1431.2	794.58	7.980	9.325	1866.8
	50.0	1453.9	797.70	7.678	9.106	1863.1
	55.0	1475.9	800.71	7.399	8.899	1860.0
	60.0	1497.2	803.63	7.152	8.739	1857.7
	65.0	1518.0	806.46	6.916	8.568	1856.2

TABLE IV (*continued*)

T K	p MPa	u m s^{-1}	ρ kg m^{-3}	$\beta_T \times 10^4$ MPa^{-1}	$\alpha \times 10^4$ K^{-1}	C_p J kg^{-1} K^{-1}
308.187	0.1	1206.5	764.52	12.253	12.434	1907.7
	5.0	1238.3	768.98	11.483	11.925	1897.8
	10.0	1269.1	773.27	10.803	11.465	1888.9
	15.0	1298.4	777.34	10.206	11.053	1880.9
	20.0	1326.3	781.22	9.678	10.680	1873.7
	25.0	1353.1	784.91	9.207	10.341	1867.1
	30.0	1378.8	788.45	8.784	10.029	1861.1
	35.0	1403.5	791.84	8.401	9.742	1855.5
	40.0	1427.4	795.11	8.065	9.506	1850.7
	45.0	1450.5	798.26	7.750	9.268	1846.7
	50.0	1472.8	801.30	7.468	9.064	1843.1
	55.0	1494.4	804.24	7.203	8.864	1840.1
303.154	0.1	1230.8	769.29	11.715	12.245	1884.9
	5.0	1262.0	773.58	11.006	11.760	1875.4
	10.0	1292.1	777.72	10.377	11.319	1866.9
	15.0	1320.8	781.66	9.823	10.924	1859.2
	20.0	1348.1	785.41	9.330	10.563	1852.3
	25.0	1374.4	788.99	8.890	10.235	1846.0
	30.0	1399.7	792.43	8.491	9.930	1840.2
	35.0	1424.1	795.73	8.131	9.652	1834.8
	40.0	1447.7	798.90	7.821	9.441	1830.2
	45.0	1470.5	801.97	7.522	9.211	1826.2
298.132	0.1	1256.7	774.00	11.189	12.059	1862.2
	5.0	1287.0	778.13	10.539	11.598	1853.1
	10.0	1316.5	782.12	9.958	11.177	1844.9
	15.0	1344.5	785.92	9.445	10.798	1837.6
	20.0	1371.4	789.55	8.985	10.447	1830.9
	25.0	1397.2	793.02	8.573	10.130	1824.8
	30.0	1422.1	796.35	8.199	9.834	1819.2
	35.0	1446.1	799.56	7.860	9.563	1814.0
293.145	0.1	1281.6	778.63	10.706	11.877	1839.7
	5.0	1311.2	782.61	10.109	11.439	1831.0
	10.0	1340.0	786.47	9.572	11.037	1823.1
	15.0	1367.5	790.14	9.095	10.674	1816.0
	20.0	1393.9	793.66	8.664	10.334	1809.6
	25.0	1419.2	797.03	8.279	10.028	1803.8
288.150	0.1	1306.4	783.23	10.251	11.698	1817.1
	5.0	1335.5	787.06	9.700	11.282	1808.8
	10.0	1363.6	790.79	9.203	10.899	1801.2
	15.0	1390.6	794.35	8.760	10.551	1794.4

as accurately as possible. By using Eq. (4) and changing the integration variable, the integral (10) becomes

$$I = \int_{u_0}^{u} \frac{a_1 + 2a_2u + 3a_3u^2}{u^2}\,du. \tag{11}$$

The calculated values of the density ρ (in kg m^{-3}), isothermal compressibility β_T (in MPa^{-1}), cubic thermal expansion coefficient α (in K^{-1}) and specific heat at constant pressure C_p (in J kg^{-1} K^{-1}) together with the smoothed values of the sound velocity u (in ms^{-1}) for the two organic liquids are presented in Tables III and IV respectively. The

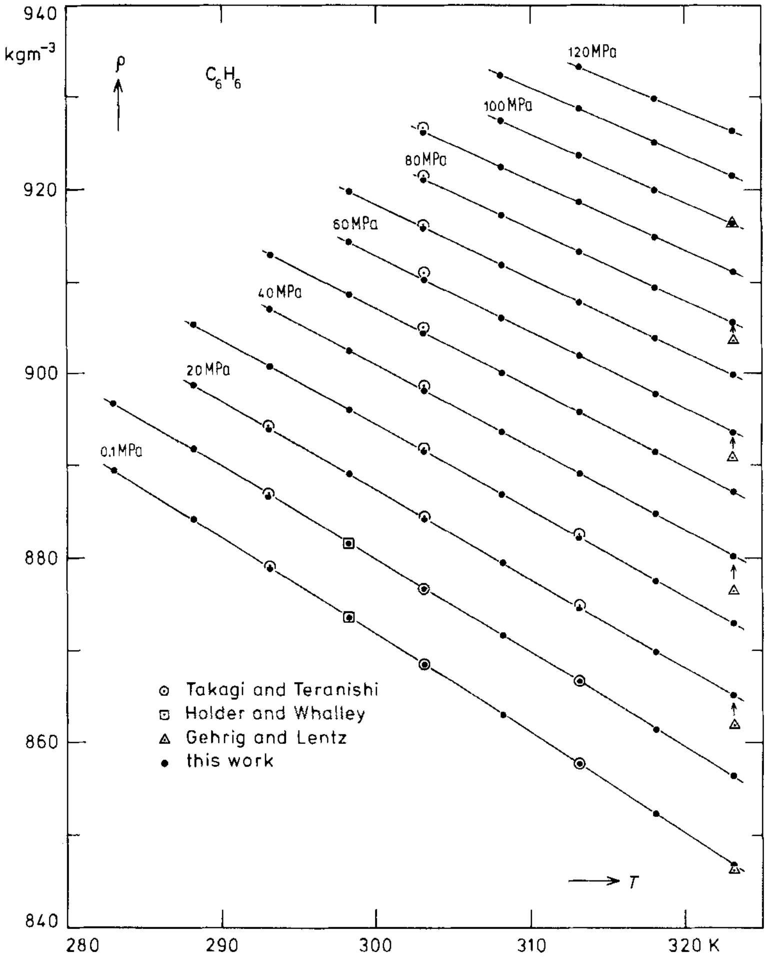

FIGURE 4 Density of benzene as a function of temperature for several isobars.

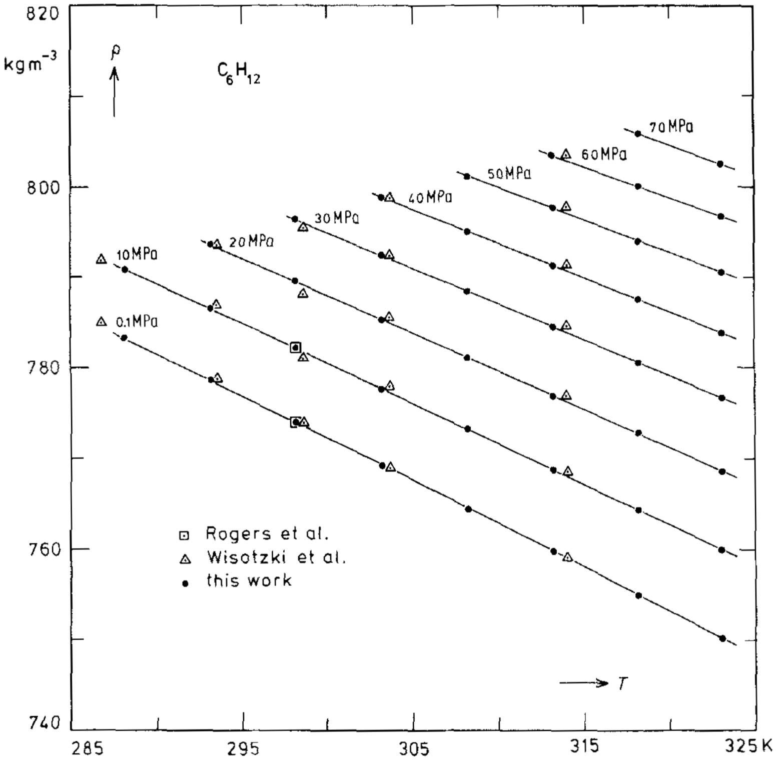

FIGURE 5 Density of cyclohexane as a function of temperature for several isobars.

variation of the density with temperature for the two liquids at several constant pressures is shown in Figures 4 and 5 respectively. In both cases, this variation is found to be nearly linear. This behaviour is similar to that on toluene and heptane reported earlier.

A comparison of the present density data for benzene and cyclohexane with the previous measurement is also shown in Figures 4 and 5 respectively. As shown, the present density data for benzene agree well with those reported by Holder and Whalley[14] and Takagi and Teranishi.[7] A detailed comparison of the present data with those of the latter authors shows that the difference between the two sets of data is less than 0.1 %. The present data for benzene also agree well with those of Bridgman.[15] However, the density data of benzene reported by Figuière *et al.*[16] and Gehrig *et al.*[17] differ from the present ones by about 0.3 to 0.4 %. In the case of cyclohexane, the agreement with those of Rogers *et al.*[18] and Wisotzki *et al.*[19] is very good, namely within 0.1 %.

5 CONCLUSIONS

From an analysis of the experimental results of this investigation, one may draw the conclusion that the present ultrasonic method is very suitable for the precise determination of liquid densities at elevated pressures. Furthermore, the method is capable of providing useful information on the pressure and temperature dependence of a number of important thermodynamic properties, such as the thermal expansivity, the isothermal compressibility and the specific heat at constant pressure, for which a direct measurement is extremely difficult to perform.

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