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SPEED OF SOUND IN *n*-HEXANE, *n*-OCTANE, *n*-DECANE, AND *n*-HEXADECANE IN THE LIQUID STATE

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This paper describes an improved experimental facility for measuring the speed of sound in liquids with an accuracy of up to 0.1%. Measurements of the speed of sound in liquid n-hexane, n-octane, n-decane, and n-hexadecane at temperatures of 298–433 K and pressures of 0.1–100 MPa have been made. It has been shown that in the possible comparison range the obtained values of the speed of sound are in good agreement with the literature data.

Introduction. In this work, we continued our studies [1–4] on further development and improvement of the acoustic method of investigating matter properties (experimental technique, computational procedure). The current methods of investigating matter properties presuppose wide use in thermophysical practice of automatic data acquisition and processing systems and systems for automatization and computerization of investigations. This called for the realization of new approaches of methodological and experimental character in investigating the speed of sound in liquids [3]. Significant changes were made in the electron-acoustic system and in the temperature maintenance and measurement system.

The present work describes an improved experimental facility and measurements of the speed of sound in liquid *n*-hexane (C_6), *n*-octane (C_8), *n*-decane (C_{10}), and *n*-hexadecane (C_{16}) performed on it in the temperature range of 298–433 K and at pressures up to 100 MPa. We also present the analytical temperature and pressure dependences of the speed of sound obtained by us.

Liquid *n*-alkanes were chosen for the following reason. First, we are planning to investigate mixtures of the above *n*-alkanes. In this case, it is desirable that pure substances and mixtures based on them are investigated on the same experimental facility. Second, new measurements for alkanes C_6, C_8, and C_{10} will widen the temperature range.

Description of the Experimental Facility and Measurement Procedure. In the new facility, the method of direct measurement of the time of passage of an acoustic pulse through the investigated medium has been realized. The time intervals are determined by means of the electron-acoustic system schematically represented in Fig. 1. The system consists of an acoustic cell placed in the liquid being investigated and a UNIPRO multifunctional measuring complex incorporating a V-131 generator of signals of arbitrary form and a V-121 digital oscilloscope. The latter are connected to a personal computer, with whose aid control of their operation and display and storage of information coming from these devices are carried out.

The acoustic cell 1 represents two parallel piezoceramic plates made of TsTS-19 of diameter 0.02 m and with a resonance frequency of 3 MHz separated by a stainless steel tube. One of the plates is transmitting and the other is receiving.

By means of the control program the generator 2 forms sinusoidal pulses of width five periods, their frequency corresponding to the resonance frequency of the piezoelectric plates. The electric pulses are applied to the transmitting piezoelectric element placed in the acoustic cell. This element transforms the electric signal into an acoustic pulse which passes through the investigated liquid and strikes the receiving piezoelectric plate. Here the acoustic pulse is transformed to an electric pulse which goes to the input of the digital oscilloscope 3 and is displayed on the screen of the personal computer 4. Locking of the oscilloscope is realized by a delayed sync pulse of the generator.

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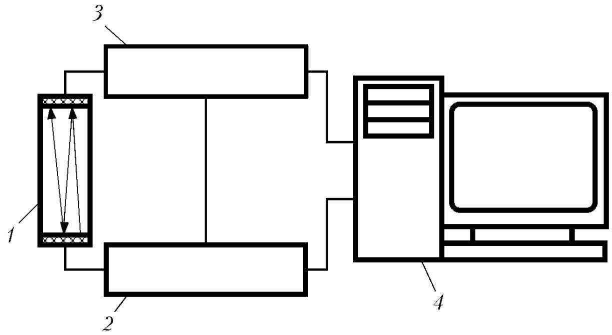

Fig. 1. Block diagram of the electron-acoustic system: 1) acoustic cell; 2) V-131 generator of pulses of arbitrary form; 3) V-121 digital oscilloscope; 4) personal computer.

Since the acoustic pulse is multiply reflected from the receiving and transmitting piezoelectric elements, the computer screen displays both the pulse that has passed once through the investigated liquid and the multitude of echo pulses.

The digital oscilloscope is used to measure the time intervals between the pulse that has passed once through the investigated liquid and the twice-reflected pulse. The temporal positions of the pulse peaks were determined by the oscilloscope automatically, which excluded the possible errors associated with the visual matching of pulses with the marks on the screen of the analog oscilloscope. The measurement error of the time intervals does not exceed 0.01%. The speed of sound is defined by the formula

$$W = \frac{2L_{p,T}}{\tau} + \Delta W_{\text{w}} - \Delta W_{\text{d}} .$$

The length of the sound base at atmospheric pressure and at a temperature of 298.15 K was determined by calibration with the aid of precision data on the speed of sound in water [5]. In calculating the experimental values, corrections for the change in the sound base length depending on the temperature and pressure, for the diffraction, and for the wave effect [6, 7] were introduced. As experiments [8, 9] have shown, the influence of dispersion on the speed of sound in organic liquids at frequencies of up to 5 MHz is negligibly small.

Measurements of the speed of sound were made in an acoustic cell placed in a high-pressure vessel filled with the liquid under investigation. On the outer surface of the vessel there are two diametrically opposite shells with resistance thermometers. The temperature was determined with the help of PTS-10M platinum resistance thermometers of the first category and an MIT-8.15 multichannel precision thermometer. The measurement error did not exceed ±0.02 K. Thermostating of the autoclave with an acoustic cell was carried out in a liquid thermostat of volume 15 liters. Temperatures above the ambient temperature were created by means of electrical heaters arranged along the thermostat perimeter. On the outside, the thermostat is insulated by two layers of a heat-insulating material with an air interlayer between them. Temperature control was carried out by means of a quick-response heater and an RTP-8.1 precision temperature regulator that permits holding the temperature within the limits of ±0.01 K. The RTP-8.1 and MIT-8.15 instruments are equipped with unified RS-232C outputs. This makes it possible to control them and display and store information on the temperature in the computer.

The manometer pressure is measured by an MP-2500 dead-weight pressure gauge tester of accuracy class 0.05 through a membrane separator, and atmospheric pressure is measured by an IR mercury bellows-cup barometer. The absolute pressure is determined as their sum with account for the corrections for the hydrostatic pressure of the oil, water, and investigated liquid columns.

The estimates made have shown that the maximum error in determining the speed of sound with account for the reference errors does not exceed 0.1%.

To confirm the stated error in determining the speed of sound, the experimental facility was calibrated against the reference substance. For the latter, we chose water, the speed of sound in which has been well studied. Measurements were taken at temperatures of 298–433 K and pressures of 0.1–100 MPa. The discrepancy between the values

TABLE 1. Experimental Values of the Speed of Sound in *n*-Alkanes

p, MPa	*T*, K							
	298.15	313.15	333.15	353.15	373.15	393.15	413.15	433.15
				n-hexane				
0.1	1078.0	1010.7	921.8	—	—	—	—	—
10.1	—	—	—	940.5	866.8	795.8	727.0	660.2
20.1	1223.8	1167.9	1095.9	1026.7	961.0	898.7	839.5	783.1
40.1	—	1290.2	1226.8	1166.5	—	1056.2	1006.1	958.4
50.1	1391.6	—	—	—	1171.2	—	—	—
60.1	1438.4	1392.6	1334.1	1279.1	1227.5	1178.9	1133.6	1091.2
80.1	1525.6	1482.0	1427.2	1375.8	1327.8	1282.5	1240.4	1200.8
100.1	1602.5	1561.9	1510.1	1461.4	1416.0	1373.1	1333.2	1295.7
				n-octane				
0.1	1172.6	1110.7	1029.9	950.6	872.8	797.2	—	—
10.1	—	—	—	—	—	904.5	841.5	780.5
20.1	1300.2	1246.7	1178.3	1112.6	1049.8	991.4	934.8	881.1
40.1	1405.8	1357.5	1295.9	1237.1	1181.7	1130.7	1081.6	1035.5
60.1	1496.4	1451.5	1394.7	1341.0	1290.1	1243.5	1198.9	1156.9
80.1	1577.1	1535.1	1481.6	1431.2	1383.7	1340.2	1298.4	1259.3
100.1	1650.4	1610.3	1559.6	1511.9	1466.7	1425.5	1386.1	1350.6
				n-decane				
0.1	1234.7	1175.4	1098.8	1024.6	952.2	881.5	812.8	743.7
20.1	1352.2	1300.2	1233.7	1170.6	1110.3	1052.7	998.4	945.9
40.1	1451.8	1403.7	1343.4	1287.0	1233.1	1182.1	1134.7	1088.6
60.1	1537.8	1493.5	1437.4	1384.9	1335.2	1288.4	1244.9	1202.8
80.1	1615.4	1573.5	1520.7	1471.3	1424.8	1380.7	1340.3	1300.6
100.1	1686.1	1646.1	1595.9	1548.8	1504.6	1462.9	1424.8	1387.0
				n-hexadecane				
0.1	1338.8	1282.8	1211.2	1142.3	1076.1	1012.1	950.1	890.0
20.1	1442.8	1392.0	1328.2	1267.4	1209.8	1154.9	1102.8	1053.2
40.1	—	1485.7	1426.8	1371.1	1318.8	1269.2	1222.3	1178.2
60.1	—	1568.3	1513.0	1461.2	1412.4	1366.3	1323.0	1282.3
80.1	—	1643.0	1590.5	1541.4	1495.4	1451.8	1411.1	1372.8
100.1	—	—	1660.9	1614.0	1570.2	1528.8	1490.0	1453.3

of the speed of sound obtained by us and the data of [5] and the results of [10, 11] in the investigated range of parameters is within the limits of 0.05 and 0.04%, respectively, which does not exceed the error of our experiments.

For samples of the limiting hydrocarbons to be investigated, we used Fluka and Aldrich *n*-alkanes of mass purity of the basic product over 99%.

Measurements were made by isotherms with increasing and decreasing pressure. In forward and backward motion, the difference between the values of the speed of sound did not exceed 0.01%. Measurements were begun 60–90 min after the experimental temperature was attained and 30–60 min after the given pressure was created. To confirm the absence of pollutants and a change in the composition of the investigated liquids, we performed their chromatographic analysis before and after measurements. The analysis has shown that their composition remained unchanged. Moreover, on completion of a set of experiments on one sample we took a repeated measurement on the initial isotherm. The discrepancy between the values of the speed of sound did not exceed 0.03%.

Experimental Data. Measurements were made on eight isotherms: 298.15, 313.15, 333.15, 353.15, 373.15, 393.15, 413.15, and 433.15 K in the pressure range from 0.1 to 100.1 MPa (Table 1). The experimental data obtained were compared to the most reliable data of [2, 12–19]. The discrepancy is within 0.1–0.3%, which does not exceed the total error of experiments.

To obtain the generalized dependence of the speed of sound on the temperature and pressure, we used our measurement data and the results of [2, 12–19]. The thus-formed data array for the speed of sound at temperatures of 293–433 K and pressures of up to 150 MPa was approximated by the least-squares technique by the equation

TABLE 2. Parameters of Eqs. (2)–(6)

i	a_i	b_i	c_i	d_i	e_i
n-hexane, $n = -0.2$, $k = 2.6$, $T_c = 507.9$ K					
0	0.00414	0.6041	–12.0825	0.0858	–0.025
1	0.01608	–0.0477	—	0.0461	0.1154
2	—	—	16.929	—	0.0324
n-octane, $n = -0.35$, $k = 2.8$, $T_c = 568.91$ K					
0	0.021829	0.5968	–5.9266	0.07396	–0.02573
1	0.00894	–0.03805	—	0.05305	0.10892
2	—	—	11.393	—	0.01761
n-decane, $n = -0.4$, $k = 3$, $T_c = 617.61$ K					
0	0.008347	0.651	–7.1516	0.18853	–0.02626
1	0.01066	–0.0464	—	0.02932	0.10003
2	—	—	14.9215	—	0.01159
n-hexadecane, $n = -0.9$, $k = 3.2$, $T_c = 722$ K					
0	0.05107	0.4696	—	—	—
1	—	—	—	0.07293	0.0759
2	—	—	5.53	—	0.004457

$$\left(\frac{1000}{W}\right)^2 = A + \frac{B}{C + \frac{p}{100}} + \frac{D}{E + \frac{p}{100}}. \tag{1}$$

The temperature dependences A, B, C, D, and E are of the form

$$A = a_0 + a_1 \frac{T}{100}, \tag{2}$$

$$B = b_0 + b_1 \frac{T}{100}, \tag{3}$$

$$C = c_0 + c_2 \left(\frac{T}{100}\right)^n, \tag{4}$$

$$D = d_0 + d_1 \frac{T}{100}, \tag{5}$$

$$E = e_0 + e_1 \left(\frac{T_c - T}{100}\right) + e_2 \left(\frac{T_c - T}{100}\right)^k. \tag{6}$$

As a result of the statistical analysis with account for the weighting functions, the parameters of Eqs. (2)–(6), whose values are given in Table 2, were calculated. Table 2 also presents the values of the critical temperatures [20]. Equation (1) approximates the initial data within the limits of their estimated error. It is applicable in the domain of existence of the liquid phase in the temperature range 293.15–433.15 K at pressures for *n*-hexane, *n*-octane, and *n*-decane up to 150 MPa and for *n*-hexadecane up to 140 MPa.

Conclusions. Thus, our experiments point to the reliability of the operation of the improved experimental facility and the possibility of taking measurements of the speed of sound on it in organic liquids in a wide range of variation of the state parameters.

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NOTATION

A, B, C, D, E, a_0, a_1, b_0, b_1, c_0, c_1, d_0, d_1, e_0, e_1, e_2, coefficients of Eqs. (1)–(6); $L_{p,T}$, sound base length at experimental temperature and pressure, m; p, pressure, MPa; T and T_c, temperature and critical temperature, K; ΔW_w, waveguide correction, m/sec; W, speed of sound, m/sec; ΔW_d, diffraction correction, m/sec; τ, time of pulse passage through the investigated liquid, sec. Subscripts: w, waveguide; d, diffraction; c, critical; n and k, exponents of Eqs. (4) and (6).

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