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EXPERIMENTAL STUDY OF THE VELOCITY OF SOUND IN LIQUID *n*-TETRADECANE AT TEMPERATURES FROM 303.15 TO 433.15 K AND PRESSURES TO 100 MPa

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The velocity of sound in liquid n-tetradecane has been studied experimentally in the interval of temperatures from 303 to 433 K and pressures to 100 MPa. The maximum measurement error amounts to 0.1%. Experimental data on the velocity of sound in liquid n-tetradecane in the region of the state variables p = 50–100 MPa and T > 373 K have been obtained for the first time. It has been shown that the new data demonstrate the reliability of the structure–property quantitative correlation proposed earlier for the acoustic quantity in the series of n-alkanes.

The velocity of sound in liquid *n*-tetradecane (C_{14}) has been the focus of a number of works. We performed such measurements earlier [1] at temperatures of 303 to 433 K and pressures to 50 MPa with an error of 0.1%. In [2], the velocity of sound has been determined at temperatures from 293 to 373 K and pressures from 0.1 to 150 MPa with an error of 0.1%. In [3], the measurements have been performed with an error of 0.1% on the 298.15 K isotherm at pressures of 0.1 and 50 MPa. Such measurements have been carried out in [4, 5] at atmospheric pressure in the temperature intervals 293–373 K and 283–393 K respectively with an error of 0.05 and 0.2%. Furthermore, with the use of the structure–property quantitative relation, in the homogeneous series of *n*-alkanes at T = 293–433 K and p = 0.1–140 MPa and for the number of the carbon atoms from 5 to 16 in the homolog we have calculated ([1, 6]) the values of the velocity of sound in C_{14} alkane in the above range of variation of the state variables.

The analysis of the available experimental data and calculated values has shown that they are consistent with a deviation no higher than 0.1–0.2%, except for the results of [5], which are 0.5–2.9% lower than most of the interconsistent data [1–4] at temperatures above 344 K. We should note that the set of given experimental data covers a limited region of both temperatures and pressures. In particular, the region above 373 K and 50 MPa is not covered by the experiments.

The present work is devoted to determination of the velocity of sound in the liquid phase in the interval of temperatures from 303 to 433 K and pressures to 100 MPa and to evaluation of the procedure proposed for calculation and prediction of the velocity of sound in the series of *n*-alkanes [1, 6].

Experimental Setup and Measurement Procedure. To investigate the velocity of sound in liquid *n*-tetradecane we have selected the method of application of echo pulses. The basic element of the setup shown in Fig. 1 was the acoustic cell representing two TLZ-19 piezoceramic plates of diameter 0.02 m and resonant frequency 3 MHz, which were separated by a stainless-steel tube with an inside diameter of 0.018 m and a wall thickness of 0.001 m. The acoustic cell was depressurized and placed in the high-pressure autoclave 3. The seals of the electric lead-in wires 1 to the piezoelectric elements were performed in the cold zone. The autoclave with an acoustic cell was thermostatted in the liquid thermostat 4. We used PES-5 as the thermostatting liquid. The autoclave was located at the center of the thermostat's large cylinder. In the smaller cylinder, there was a propeller mixer. In the vicinity of the exterior surface of the autoclave, there were two diametrically opposite sleeves (of 1Kh18N10T stainless steel) into which the resistance thermometers 2 were inserted. The thermostat had five heaters of total power 10 kW. The prescribed temperature of the thermostat was maintained automatically accurate to 0.01 K. The temperature was measured by two PRT-10 platinum resistance thermometers of the first category. The balanced method with the use of R363 double-row semi-

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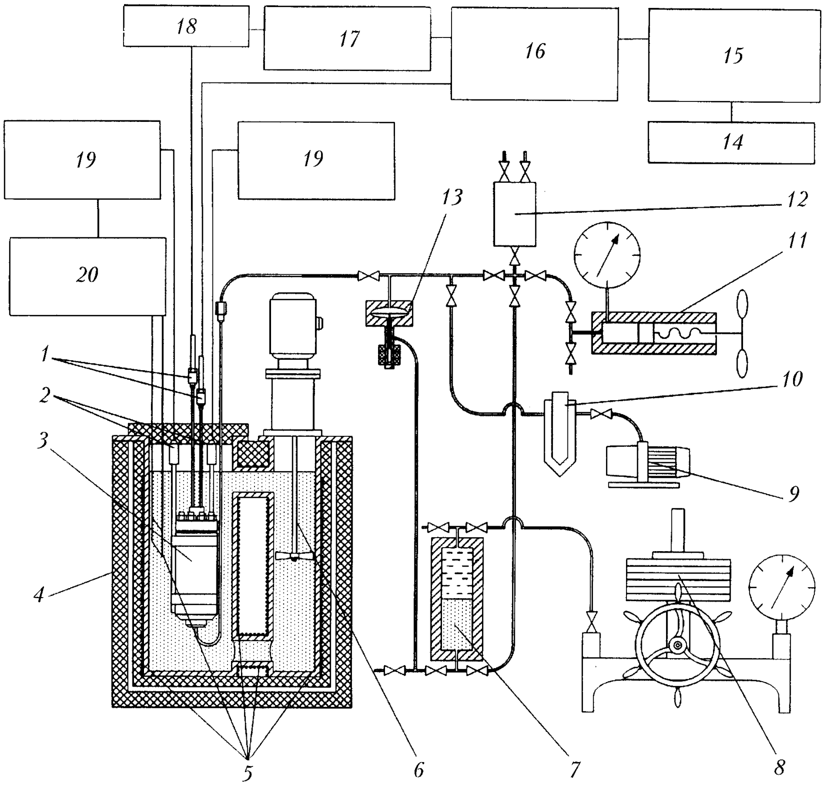

Fig. 1. Diagram of the experimental setup: 1) seals of the electric lead-in wires to the piezoelectric elements; 2) platinum resistance thermometers; 3) autoclave; 4) liquid thermostat; 5) electric heaters; 6) propeller mixer; 7) separating vessel; 8) dead-weight pressure gauge; 9) vacuum pump; 10) nitrogen trap; 11) high-pressure press; 12) deaerator; 13) differential pressure gauge; 14) Ch3-34 frequency meter; 15) G3-34 sine-wave oscillator; 16) G5-27 pulse generator; 17) S1-70 oscilloscope; 18) U3-29 amplifier; 19) potentiometer; 20) temperature-control system.

automatic potentiometers of the 0.002 class was employed to measure the resistance of the thermometers. Reference resistance coils and the standard cell were thermostatted at 293.15 K. In the experiment, the readings of the two resistance thermometers coincided accurate to 0.01 K in all the regimes. The measurement error for the temperature was no higher than 0.02 K. The pressure in the setup was measured by an MP-2500 dead-weight pressure gauge of the 0.05 class with allowance for barometric pressure and corrections for the hydrostatic pressures of the oil and water columns and the liquid under study. The highly sensitive diaphragm differential pressure gauge 13 operating in the zero-meter regime was used to separate the volume occupied by the liquid under study from the pressure-gauge system. The castor oil filling the pressure gauge and the water filling the lower cavity of the differential pressure gauge were separated in a separating vessel. The pressure of the liquid under study was produced by press 11.

The electron-acoustic system operates as follows. The pulse generator 16 producing rectangular pulses which excite the radiating piezoelectric element is triggered by the sine-wave oscillator 15. The acoustic pulse converted to the electric pulse by the detecting piezoelectric element is amplified by the wide-band amplifier 18 and is observed on the screen of oscilloscope 17. Varying the pulse repetition frequency, one can bring into coincidence a pulse transmitted once by the substance under study and a pulse doubly reflected from the detecting and radiating piezoelectric elements. The frequency obtained in such a manner was measured with a high degree of accuracy by frequency meter 14 in the regime of measurement of the ratio of the 10-MHz frequency of the reference generator to the frequency measured.

The velocity of sound was calculated from the formula

TABLE 1. Velocity of Sound in *n*-Tetradecane

p, MPa	T, K						
	303.15	323.15	353.15	373.15	393.15	413.15	433.15
0.1	1293.4	1219.1	1113.5	1045.3	979.5	915.8	854.3
10.1			1182.2	1119.4	1059.6	1002.5	947.9
20.1	1402.2	1336.2	1243.4	1184.6	1128.8	1076.0	1026.1
30.1			1299.0				
40.1	1495.4	1434.7	1350.0	1296.8	1246.7	1199.5	1155.2
50.1			1397.6				
60.1	1577.7	1520.8	1441.9	1392.7	1346.2	1302.7	1261.9
70.1			1484.0				
80.1		1598.1	1523.6	1477.1	1433.4	1392.5	1354.3
100.1		1668.4	1597.4	1553.5	1511.6	1472.8	1436.4

$$W = 2 \cdot \frac{L_{p,T} \cdot 10^7}{n} - \Delta W_{\mathrm{diffr}} + \Delta W_{\mathrm{wave}} \,. \tag{1}$$

The length of the acoustic base at atmospheric pressure and a temperature of 303.15 K was determined indirectly with the use of precision data on the velocity of sound in water [7]. Its value was 0.049593 m. In calculating the experimental data, we introduced corrections for variation in the acoustic-base length with temperature and pressure and for diffraction and the waveguide effect [8, 9]. According to the estimates made, the influence of the dispersion of sound is negligible and can be disregarded in calculation of the velocity of sound.

Before filling the autoclave, the press, the deaerator, and the pipelines connecting them with the liquid under study, we evacuated them through a nitrogen trap by a vacuum pump. After filling the deaerator and deaerating the liquid under study by heating of the latter to boiling, we filled the autoclave. The measurements were carried out by the isotherms in both the regime of increase of the pressure from atmospheric pressure to the maximum one and its decrease from the maximum pressure to atmospheric one. The difference in the velocities of sound on all the isotherms under study was no larger than 0.01%.

The specimen under study had a purity of more than 98% for the mass of the basic product.

The maximum total relative error of determination of the velocity of sound, as the evaluation of the accuracy of measurements has shown, was no higher than 0.1%.

Before the experiments, we carried out check measurements with ordinary water. The velocity of sound was determined in the interval of temperatures from 303 to 433 K and pressures to 100 MPa. A comparison of the resulting values of the velocity of sound in water and the precision data of [7] and the results of [10, 11] has shown that they are consistent within 0.03 and 0.08% respectively. The deviations lie within the error of our experiment.

Discussion of the Results. The measurements have been performed on seven isotherms (303.15, 323.15, 353.15, 373.15, 393.15, 413.15, and 433.15 K) in the pressure interval 0.1–100 MPa. We have obtained 48 experimental values of the velocity of sound (see Table 1). A comparison has shown that the disagreement between our new data and the data obtained earlier [1] and also the data of other authors [2–4] is within 0.1–0.15%, which is no higher than the total experimental error. As far as the results of correlation [1, 6] are concerned, they are consistent with the data obtained in the region which has not been studied earlier (p = 50–100 MPa and $T > 373$ K) with deviations of 0.1 to 0.2%.

The new experimental results on the velocity of sound in combination with the data of [1–4] were approximated by the equation

$$\frac{1}{W^2} = A + \frac{B}{C/T + p} + \frac{DT}{E + p} \,. \tag{2}$$

The temperature dependence of E has the form

$$E = E_0 + E_1 (T_{cr} - T)^{2.5} . \qquad (3)$$

Here T_{cr} is equal to 693 K [12].

As a result of processing, we have computed the coefficients of Eqs. (2) and (3): $A = 5.3069 \cdot 10^{-8}$, $B = 4.60745 \cdot 10^{-5}$, $C = 5.6838 \cdot 10^4$, $D = 7.05439 \cdot 10^{-8}$, $E_0 = 8.897$, and $E_1 = 2.082071 \cdot 10^{-5}$.

Equation (2) well describes the initial data on the velocity of sound in the intervals of temperatures 293–433 K and pressures 0.1–150 MPa studied. The standard and maximum deviations of W values calculated from (2) from the initial ones are no higher than 0.028 and 0.08% respectively.

Thus, our experiments demonstrate the reliability of the procedure proposed earlier ([1, 6]) for calculation and prediction of the velocity of sound in the series of *n*-alkanes.

NOTATION

W, velocity of sound, m/sec; $L_{p,T}$, acoustic-base length at experimental temperature and pressure, m; $\bar{n}$, arithmetic mean of the series of successive measurements (10–15 measurements) of the ratio of the reference frequency to the measured frequency; ΔW_{diffr}, correction for diffraction, m/sec; ΔW_{wave}, waveguide correction, m/sec; p, pressure, MPa; A, B, C, D, E, E_0, and E_1, coefficients of Eqs. (2) and (3); T and T_{cr}, temperature and critical temperature, K; TLZ, titanium lead zirconate; PES, polyethyl siloxane; PRT, Platinum resistance thermometer. Subscripts: diffr, diffraction; wave, waveguide; cr, critical.

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