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DETERMINATION OF THE THERMODYNAMIC PROPERTIES OF LIQUID *n*-HEXADECANE FROM THE MEASUREMENTS OF THE VELOCITY OF SOUND

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The density, the isobaric expansion coefficient, the specific heats at constant pressure and constant volume, and the isothermal compressibility coefficient of liquid n-hexadecane have been calculated in the range of temperatures 298–433 K and pressures 0.1–140 MPa from the data on the velocity of sound. The coefficients of the Tate equation in the above parametric range have been determined. The table of the thermodynamic properties of n-hexadecane has been presented.

Keywords: *n-hexadecane, velocity of sound, density, specific heat, isobaric expansion coefficient.*

Introduction. Normal hexadecane (C_{16}) has been the subject of numerous investigations of thermodynamic properties for years. A considerable volume of experimental data on the thermodynamic properties of *n*-hexadecane at atmospheric pressure is accumulated at present; also, the results of measurements of the density [1–10] and the velocity of sound [11–15] in the high-pressure range are available. However, there are no experimental data on the density at pressures and temperatures higher than 3.5 MPa and 393 K. It is only in [11] that the velocity of sound has been measured at pressures higher than 70 MPa. In the only experimental investigation [8], the specific heat at constant pressure has been determined at elevated pressures up to 10 MPa and temperatures of 318 to 373 K. The other thermodynamic properties at elevated pressure have virtually not been studied experimentally. At the same time, such properties as density, heat capacity, and compressibility can be determined by calculation from the experimental dependences of the density, specific heat at constant atmospheric pressure, and velocity of sound on temperature and pressure. The accuracy of calculation is comparable with the accuracy of direct measurements of the above quantities.

The present work seeks to calculate the thermodynamic properties of liquid *n*-hexadecane at temperatures of 298 to 433 K and pressures up to 140 MPa.

Initial Data. We have used, as the initial data on the velocity of sound, our measurements [4, 15] performed at temperatures of 298 to 433 K and pressures up to 100 MPa with an error of 0.1%, the results of experiments [11] at T = 293–473 K and p = 0.1–140.1 MPa having an accuracy of 0.1%, the data of [12] at T = 303–393 K and p = 0.1–70 MPa with an accuracy of 0.2%, and the results of [16–20] at atmospheric pressure, having an error of less than 0.1%. An analysis of the mentioned works has shown that their results are consistent with a deviation no higher than 0.1–0.2%.

The data array thus formed on the velocity of sound at T = 298–433 K and p = 0.1–140 MPa was approximated in the form of the dependence on temperature and pressure

$$\frac{10^6}{W^2} = E_0 + \frac{E_1}{E_2 + \frac{p}{100}} + \frac{E_3 T}{E_4 + \frac{p}{100}} . \quad (1)$$

Here $E_0 = 5.107 \cdot 10^{-2}$, $E_1 = 0.4696$, and $E_3 = 7.293 \cdot 10^{-4}$. The temperature dependences of the coefficients E_2 and E_4 have the form

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$$E_2 = 5.53\left(\frac{T}{100}\right)^{-0.9}, \quad E_4 = 7.59\cdot10^{-2}\left(\frac{T_c - T}{100}\right) + 4.457\cdot10^{-3}\left(\frac{T_c - T}{100}\right)^{3.2}.$$

The critical temperature is taken to be $T_c = 722$ K [21].

The standard deviation of the W values calculated from (1) from the initial values amounts to 0.06%.

At atmospheric pressure, the temperature dependences of the density ρ_0 and the specific heat c_{p0} were obtained by processing of the available values for ρ_0 at temperatures of 298 to 433 K ([21]), the data for c_{p0} at temperatures of 298 to 403 K ([22]), and the values of specific heat at constant pressure at temperatures of 403 to 433 K found by graphical-analytical interpolation of the dependence of c_{p0} on the number of carbon atoms in an *n*-alkane molecule. As a result of the processing, we have obtained the dependences

$$\rho_0 = 4.10701\cdot10^{2} + 1.13071\,(T_c - T) - 9.5509\cdot10^{-4}\,(T_c - T)^2 + 6.7842\cdot10^{-7}\,(T_c - T)^3, \tag{2}$$

$$c_{p0} = 8.7527 - 7.77203\cdot10^{-2}T + 3.28054\cdot10^{-4}T^2 - 5.92853\cdot10^{-7}T^3 + 4.0297\cdot10^{-10}T^4. \tag{3}$$

The estimated error of the initial data used for calculation of the thermodynamic properties is no higher than 0.1% for the density and the velocity of sound and 0.5% for the specific heat at constant pressure.

Computational Procedure. For calculation, we have used the method of step computation of thermodynamic properties from the data on the velocity of sound under pressure with the use of successive approximations at each step [23, 24]. The difference of our computational method from that of [23, 24] is that we used new equations to describe the dependence of the velocity of sound on the temperature and the pressure and the density on the temperature at each step on isobars. This enabled us to improve the quality of the calculations carried out. The computational method is based on the well-known thermodynamic relations

$$\left(\frac{\partial\rho}{\partial p}\right)_T = \frac{1}{W^2} + \frac{T\alpha^2}{c_p}, \tag{4}$$

$$\left(\frac{\partial c_p}{\partial p}\right)_T = -\frac{T}{\rho}\left[\alpha^2 + \left(\frac{\partial\alpha}{\partial T}\right)_p\right], \tag{5}$$

in which $\alpha = -\frac{1}{\rho}\left(\frac{\partial\rho}{\partial T}\right)_p$.

The properties were computed from the isotherms by the iterative step method with a small pressure step (increment) $\Delta p = p_2 - p_1$. The density and the specific heat at constant pressure were found from the relations

$$\rho_2 = \rho_1 + \int_{p_1}^{p_2}\frac{1}{W^2}\,dp + \frac{T}{2}\left(\frac{\alpha_1^2}{c_{p1}} + \frac{\alpha_2^2}{c_{p2}}\right)\Delta p, \tag{6}$$

$$c_{p2} = c_{p1} - \frac{T}{2}\left\{\frac{1}{\rho_1}\left[\alpha_1^2 + \left(\frac{\partial\alpha}{\partial T}\right)_{p1}\right] + \frac{1}{\rho_2}\left[\alpha_2^2 + \left(\frac{\partial\alpha}{\partial T}\right)_{p2}\right]\right\}\Delta p. \tag{7}$$

The lower limit of integration was the initial pressure p_1 at which the density ρ_1 and the specific heat at constant pressure c_{p1} were known, whereas the isobaric expansion coefficient α_1 and the derivative $(\partial\alpha/\partial T)_{p1}$ could be determined by differentiating the temperature dependence of ρ. Atmospheric pressure, at which ρ and c_p were determined by expressions (2) and (3) and the values of α and $(\partial\alpha/\partial T)_p$ were obtained by differentiating (2), was initially taken as the pressure p_1. The values of ρ_2 and c_{p2} were calculated from (6) and (7) for the pressure $p_2 = p_1 + \Delta p$. In so doing, the integral $\int_{p_1}^{p_2} W^{-2} dp$ was computed analytically with Eq. (1), whereas further calculation was performed iterationally, since the unknown quantities c_{p2}, α_2, and $(\partial\alpha/\partial T)_{p2}$ were present on the right-hand sides of (6) and (7). In the first approximation, we took $c_{p2} = c_{p1}$, $\alpha_2 = \alpha_1$, and $(\partial\alpha/\partial T)_{p2} = (\partial\alpha/\partial T)_{p1}$.

The density values calculated from (6) at the pressure p_2 were approximated by a polynomial similar to that used for description of the density at atmospheric pressure

$$\rho_2 = \sum_{i=0}^{3} a_i (T_c - T)^i . \tag{8}$$

As the evaluations carried out have shown, Eq. (8) describes the computed density values in the range of high pressures as satisfactorily as the data at atmospheric pressure. Using this polynomial we computed α_2 and found the derivative $(\partial\alpha/\partial T)_{p2}$ at the pressure p_2. Thereafter we recalculated the density ρ_2 and the specific heat at constant pressure c_{p2} from Eqs. (6) and (7) with the new values of c_{p2}, α_2, and $(\partial\alpha/\partial T)_{p2}$ on the right-hand sides of the equations. The iterative process was continued until the density values calculated from Eq. (6) in two successive iterations agree to accuracy $1 \cdot 10^{-5}$%.

As a result we obtained the values of ρ_2, c_{p2}, α_2, and $(\partial\alpha/\partial T)_{p2}$ at a pressure p_2 Δp higher than the pressure p_1 at which these properties were known. Next we took the pressure p_2 at which the properties had already been determined as the pressure p_1, and the entire calculation was repeated again. Thus, passing successively from the lower pressure to the higher one, we calculated ρ, c_p, and α for the entire range of temperatures and pressures, in which the values of the velocity of sound were known.

Next, from the resulting values of ρ, c_p, α, and W, we computed the specific heat at constant volume and the isothermal compressibility coefficient with the relations

$$\beta_T = \frac{1}{\rho}\left(\frac{1}{W^2} + \frac{T\alpha^2}{c_p}\right), \tag{9}$$

$$c_v = \frac{c_p}{\left(1 + \frac{T\alpha^2 W^2}{c_p}\right)} . \tag{10}$$

The calculation results are presented in Table 1.

The evaluations carried out have shown that the error of the calculated values is determined by the accuracy of the initial data. It is no higher than 0.15% for ρ, 0.8% for c_p, 1.2% for c_v, 1.7% for α, and 0.8% for β_T.

We evaluated the error of calculation of the thermodynamic properties of C_{16} alkane due to the influence of the final pressure step Δp. The evaluations have shown that the influence of the step on the error of the computed quantities diminishes with it; this error becomes an order of magnitude lower than the error due to the inaccuracy of the initial data even for Δp = 10–20 MPa. We took a fairly small step Δp = 1 MPa, which ensured an acceptable degree of accuracy in calculating the properties.

Discussion of the Results. The thermodynamic properties of liquid C_{16} have experimentally been studied in rather limited ranges of state parameters. A comparison of the ρ values calculated with direct measurements at elevated pressure to the data of [1–10] and to the results of correlation [25] and generalization [26] is possible, whereas c_p can be compared only to the values in [8]. For ρ, the range of possible comparison at temperatures higher than 393 K is limited to a pressure of 3.5 MPa. A comparison of c_p values is possible at temperatures of 318 to 373 K and pressures of 0.1 to 10 MPa. Also, there are measurements of β_T [27] at atmospheric pressure and temperatures of 298 to 333 K.

TABLE 1. Thermodynamic Properties of *n*-Hexadecane

T, K	W, m/sec	ρ, kg/m^3	c_p, kJ/(kg·K)	c_v, kJ/(kg·K)	$\alpha \cdot 10^3$, K^{-1}	$\beta_T \cdot 10^3$, MPa^{-1}
			p = 0.1 MPa			
298.15	1338.4	770.0	2.214	1.858	0.891	0.864
313.15	1282.9	759.7	2.254	1.896	0.908	0.951
333.15	1211.4	745.9	2.313	1.954	0.933	1.082
353.15	1142.5	731.9	2.376	2.015	0.961	1.234
373.15	1076.1	717.7	2.439	2.077	0.992	1.413
393.15	1011.8	703.4	2.504	2.141	1.027	1.624
413.15	949.7	688.8	2.571	2.207	1.067	1.875
433.15	889.7	674.0	2.643	2.277	1.113	2.176
			p = 5 MPa			
298.15	1365.5	773.2	2.210	1.860	0.865	0.824
313.15	1311.5	763.2	2.250	1.898	0.880	0.903
333.15	1242.2	749.7	2.309	1.956	0.901	1.020
353.15	1175.8	736.2	2.370	2.017	0.923	1.155
373.15	1111.9	722.5	2.433	2.079	0.948	1.310
393.15	1050.5	708.8	2.497	2.143	0.975	1.490
413.15	991.6	694.9	2.562	2.208	1.006	1.699
433.15	934.9	680.8	2.632	2.278	1.041	1.942
			p = 10 MPa			
298.15	1391.9	776.3	2.207	1.862	0.841	0.788
313.15	1339.3	766.5	2.247	1.901	0.854	0.860
333.15	1272.1	753.4	2.305	1.958	0.871	0.966
353.15	1207.8	740.3	2.366	2.018	0.889	1.085
373.15	1146.2	727.1	2.428	2.081	0.909	1.221
393.15	1087.3	713.8	2.491	2.144	0.931	1.376
413.15	1030.9	700.5	2.555	2.209	0.955	1.553
433.15	977.1	687.1	2.624	2.278	0.981	1.756
			p = 15 MPa			
298.15	1417.3	779.3	2.204	1.865	0.819	0.755
313.15	1366.0	769.8	2.244	1.903	0.830	0.821
333.15	1300.5	757.0	2.301	1.960	0.844	0.917
353.15	1238.1	744.2	2.362	2.020	0.859	1.025
373.15	1178.5	731.4	2.424	2.083	0.875	1.146
393.15	1121.7	718.6	2.486	2.146	0.892	1.281
413.15	1067.6	705.8	2.549	2.211	0.911	1.434
433.15	1016.0	692.9	2.617	2.279	0.932	1.605
			p = 20 MPa			
298.15	1441.7	782.2	2.202	1.867	0.799	0.725
313.15	1391.6	772.9	2.241	1.905	0.808	0.786
333.15	1327.7	760.4	2.298	1.962	0.820	0.874
353.15	1266.9	747.9	2.359	2.022	0.832	0.972
373.15	1209.1	735.5	2.420	2.084	0.845	1.080
393.15	1154.1	723.1	2.481	2.147	0.858	1.200
413.15	1101.9	710.7	2.545	2.212	0.873	1.333
433.15	1052.3	698.2	2.612	2.280	0.889	1.481
			p = 25 MPa			
313.15	1416.2	775.8	2.239	1.907	0.788	0.755
333.15	1353.7	763.6	2.296	1.963	0.798	0.835
353.15	1294.5	751.5	2.356	2.024	0.808	0.924
373.15	1238.2	739.4	2.417	2.086	0.818	1.022
393.15	1184.8	727.3	2.478	2.149	0.829	1.129
413.15	1134.2	715.3	2.540	2.213	0.840	1.247
433.15	1086.2	703.2	2.607	2.281	0.853	1.377
			p = 30 MPa			
313.15	1439.9	778.7	2.236	1.909	0.769	0.726
333.15	1378.8	766.8	2.293	1.965	0.777	0.801
353.15	1320.9	754.9	2.353	2.026	0.785	0.882
373.15	1266.0	743.0	2.414	2.088	0.794	0.971
393.15	1214.0	731.3	2.475	2.151	0.802	1.068
413.15	1164.8	719.6	2.537	2.215	0.811	1.173
433.15	1118.3	707.9	2.603	2.283	0.821	1.288
			p = 40 MPa			
313.15	1485.1	784.2	2.233	1.913	0.736	0.675
333.15	1426.2	772.7	2.290	1.969	0.741	0.740
353.15	1370.7	761.3	2.349	2.030	0.746	0.809
373.15	1318.2	750.0	2.409	2.092	0.751	0.884

Continued

T, K	W, m/sec	ρ, kg/m^3	c_p, kJ/(kg·K)	c_v, kJ/(kg·K)	$\alpha \cdot 10^3$, K^{-1}	$\beta_T \cdot 10^3$, MPa^{-1}
393.15	1268.6	738.7	2.470	2.154	0.756	0.964
413.15	1221.7	727.6	2.531	2.218	0.761	1.051
433.15	1177.6	716.6	2.596	2.285	0.768	1.144
			p = 50 MPa			
313.15	1527.5	789.3	2.230	1.916	0.706	0.632
333.15	1470.7	778.2	2.286	1.973	0.710	0.689
353.15	1417.1	767.2	2.346	2.033	0.713	0.749
373.15	1366.5	756.3	2.406	2.095	0.715	0.813
393.15	1318.9	745.6	2.466	2.158	0.718	0.881
413.15	1273.9	734.9	2.527	2.221	0.720	0.954
433.15	1231.7	724.4	2.591	2.287	0.724	1.031
			p = 60 MPa			
313.15	1567.6	794.2	2.227	1.920	0.680	0.594
333.15	1512.5	783.4	2.284	1.976	0.683	0.645
353.15	1460.6	772.8	2.343	2.037	0.684	0.698
373.15	1411.7	762.3	2.403	2.099	0.685	0.754
393.15	1365.7	751.9	2.463	2.161	0.685	0.813
413.15	1322.4	741.7	2.524	2.224	0.686	0.875
433.15	1281.6	731.6	2.587	2.290	0.688	0.940
			p = 70 MPa			
313.15	1605.7	798.8	2.225	1.924	0.657	0.562
333.15	1552.1	788.3	2.282	1.980	0.658	0.607
353.15	1501.7	778.0	2.341	2.040	0.658	0.654
373.15	1454.3	767.9	2.401	2.102	0.658	0.703
393.15	1409.6	757.8	2.461	2.164	0.657	0.755
413.15	1367.6	747.9	2.521	2.227	0.656	0.809
433.15	1328.1	738.2	2.584	2.292	0.656	0.866
			p = 80 MPa			
313.15	1642.1	803.2	2.223	1.927	0.636	0.533
333.15	1589.8	793.0	2.280	1.983	0.636	0.574
353.15	1540.7	783.0	2.339	2.044	0.635	0.616
373.15	1494.5	773.1	2.399	2.105	0.634	0.660
393.15	1451.1	763.4	2.459	2.167	0.632	0.706
413.15	1410.2	753.8	2.519	2.230	0.630	0.754
433.15	1371.8	744.4	2.582	2.295	0.629	0.803
			p = 90 MPa			
313.15	1676.8	807.3	2.221	1.930	0.617	0.507
333.15	1625.8	797.4	2.278	1.987	0.616	0.544
353.15	1577.8	787.7	2.338	2.047	0.615	0.582
373.15	1532.7	778.1	2.398	2.109	0.613	0.622
393.15	1490.4	768.6	2.457	2.171	0.610	0.663
413.15	1450.5	759.3	2.517	2.233	0.607	0.706
433.15	1413.0	750.2	2.580	2.298	0.606	0.750
			p = 100 MPa			
333.15	1660.2	801.7	2.277	1.990	0.598	0.518
353.15	1613.3	792.2	2.337	2.050	0.596	0.553
373.15	1569.2	782.8	2.397	2.112	0.593	0.589
393.15	1527.8	773.6	2.456	2.174	0.590	0.626
413.15	1488.8	764.5	2.516	2.236	0.587	0.664
433.15	1452.2	755.6	2.578	2.300	0.584	0.703
			p = 120 MPa			
333.15	1725.0	809.7	2.275	1.996	0.567	0.473
353.15	1680.0	800.6	2.335	2.056	0.564	0.503
373.15	1637.6	791.6	2.395	2.118	0.560	0.533
393.15	1597.8	782.8	2.455	2.180	0.556	0.564
413.15	1560.3	774.2	2.514	2.242	0.552	0.595
433.15	1525.1	765.7	2.576	2.305	0.548	0.627
			p = 140 MPa			
333.15	1785.3	817.0	2.273	2.001	0.539	0.436
353.15	1741.8	808.3	2.334	2.062	0.536	0.462
373.15	1700.9	799.7	2.395	2.124	0.532	0.487
393.15	1662.4	791.3	2.454	2.186	0.527	0.514
413.15	1626.2	783.0	2.514	2.247	0.522	0.540
433.15	1592.1	774.9	2.575	2.310	0.518	0.567

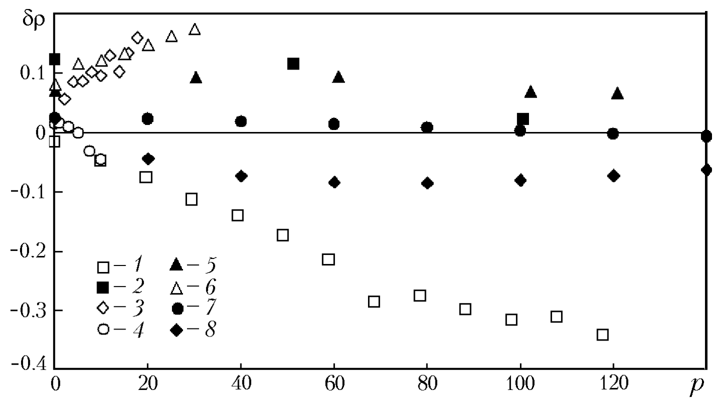

Fig. 1. Deviation $\delta\rho = (\rho - \rho_{calc})/\rho_{calc} \cdot 100\%$ of the literature data on the density ρ from the calculated ρ_{calc}: 1) [1], T = 363.15 K; 2) [4], T = 373.15 K; 3) [5], T = 360.15 K; 4) [8], T = 373.15 K; 5) [9], T = 348.15 K; 6) [10], T = 373.15 K; 7) [25], T = 373.15 K; 8) [26], T = 373.15 K. $\delta\rho$, %; p, MPa.

The computed values of ρ are consistent with direct density measurements [4, 8, 9] and correlation results [25] within the estimated error of our calculation (see Fig. 1). The remaining experimental data and generalization results [26] have a deviation within 0.4%, which is no higher than the total experimental and computational error. The agreement with the data on c_p [8] is within 0.4%, which is lower than the computational error. The obtained values of β_T are consistent with the data of [27] with a deviation no higher than 1.5%, which is within the total error of the compared calculated and experimental values.

Generalizing Dependences. The computed density values were generalized by an equation of state of the Tate-equation type

$$\rho = \frac{\rho_0}{1 - A \ln\left(\dfrac{B + p}{B + p_0}\right)} . \tag{11}$$

Here A = 0.08744. The temperature dependence of B has the form

$$B = -96.3 + 82.37\left(\frac{T_c}{T}\right) - 0.07\left(\frac{T_c}{T}\right)^2 . \tag{12}$$

Equation (11) describes the density values (see Table 1) in the range of parameters T = 298–433 K and p = 0.1–140 MPa with a deviation no higher than 0.03% and possesses good extrapolation potentialities as far as pressure is concerned. This is demonstrated by the reproduction, by it, of the results of [4] at temperatures of 323 to 373 K and pressures up to 300 MPa accurate to 0.3% and of the density values [25, 26] at temperatures of 298 to 393 K and pressures up to 300 MPa. Therefore, it is likely that the density values computed from (1), too, for temperatures of 373 to 433 K and pressures of 140 to 300 MPa (parametric range not investigated before) will have an error of the order of 0.3–0.4%. Equation (11) can be recommended for practical calculations of the density for pressures of 0.1 to 300 MPa and temperatures of 298 to 433 K.

Conclusions. Thus, the performed calculations and evaluations demonstrate the reliability of the computational procedure proposed and of the results obtained on their basis. The proposed tables of thermodynamic properties of *n*-hexadecane at temperatures of 298 to 433 K and pressures up to 140 MPa are unique.

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NOTATION

a_i, coefficients of Eq. (8); c_p, specific heat at constant pressure, kJ/(kg·K); c_{p0}, c_{p1}, and c_{p2}, specific heats at constant atmospheric pressure and at constant initial and final pressures of each pressure step respectively, kJ/(kg·K); c_v, specific heat at constant volume, kJ/(kg·K); p, pressure, MPa; Δp, pressure step (increment), MPa; p_0, atmospheric pressure; p_1 and p_2, initial and final pressure of each pressure step respectively, MPa; T, temperature, K; T_c, critical temperature, K; W, velocity of sound, m/sec; α, isobaric expansion coefficient, K^{-1}; α_1 and α_2, isobaric expansion coefficients at the initial and final pressures of each pressure step respectively, K^{-1}; $(\partial\alpha/\partial T)_{p1}$ and $(\partial\alpha/\partial T)_{p2}$, derivatives of the isobaric expansion coefficient at the initial and final pressures of each pressure step respectively, K^{-2}; β_T, isothermal compressibility coefficient, MPa^{-1}; ρ, density, kg/m^3; ρ_0, ρ_1, and ρ_2, densities at atmospheric pressure and at the initial and final pressures of each pressure step respectively, kg/m^3; $\delta\rho$, deviation of the literature data on the density ρ from those calculated. Subscripts: c, critical state; calc, calculated value; p, v, and T values, at constant pressure, volume, and temperature respectively; i, coefficient No.

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