

## Thermodynamic Properties of *n*-Hexane

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Specific volumes and isobaric heat capacity measurements are reported for *n*-hexane. The measurements were made in the liquid and vapor phases at temperatures from the triple point and also cover a wide region around the critical point. The thermal, caloric, and acoustic data from our own investigation as well as those of a number of other authors are fitted to a single equation of state with 32 constants. This equation yields to all thermodynamic properties of *n*-hexane in the temperature range 180 to 630 K and pressures up to 100 MPa. The data in the critical region have been analyzed in terms of a scaled equation of state.

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**KEY WORDS:** critical region; equation of state; *n*-hexane; sound velocity; specific heat; thermodynamic properties.

### 1. INTRODUCTION

The use of *n*-hexane in industry has made it one of the most important substances for which the development of tables of reference data is needed. To this end, experimental studies of thermal and caloric properties covering a wide range of states have been carried out in the thermophysical laboratory at our institute. An equation of state has been developed for calculating tables of thermodynamic properties based on our experimental data combined with the most reliable published density and sound velocity data. In the critical region, data have been processed by a scaled equation of state.

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## 2. EXPERIMENTAL PROPERTY DATA

*n*-Hexane is a substance which has been investigated in detail. The measurements of thermal, caloric, and acoustic properties cover the liquid and vapor phases and a wide region around the critical point (see Figs. 1 and 2) [1–21]. The present authors have also made a large number of experimental studies of the properties of *n*-hexane. The experimental procedures and the results obtained are briefly reviewed below.

### 2.1. Present Measurements

#### 2.1.1. Methods and Apparatus

The density,  $\rho$ , of *n*-hexane was measured in the temperature range from 223.15 to 623.15 K and at pressures up to 150 MPa with the aid of a spherical constant volume piezometer. The isobaric heat capacity,  $C_p$ , was measured in the temperature range 290 to 625 K and at pressures up to 60 MPa in an adiabatic flow calorimeter whose design feature is a closed circulation scheme and calorimetric measurement of the flow rate. A detailed discussion of the apparatus and the technique employed is available in Ref. 22.

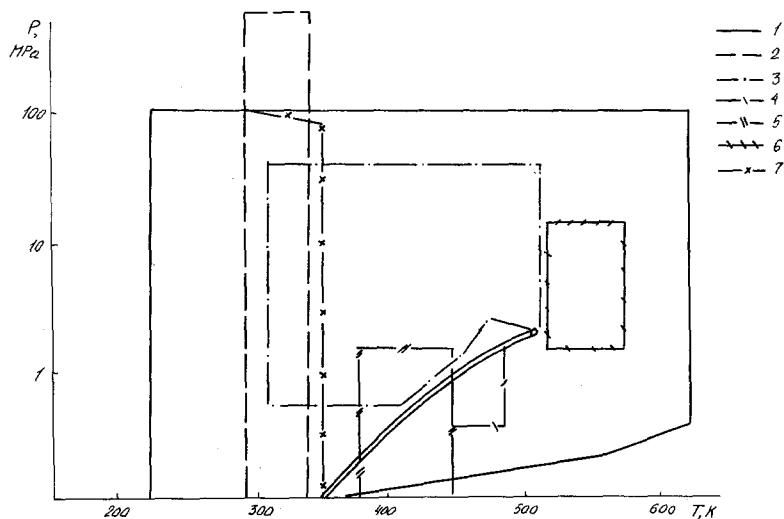


Fig. 1. Ranges of experimental data for the density of *n*-hexane: 1 [23–25]; 2 [4]; 3 [1]; 4 [12]; 5 [8]; 6 [5]; 7 [9].

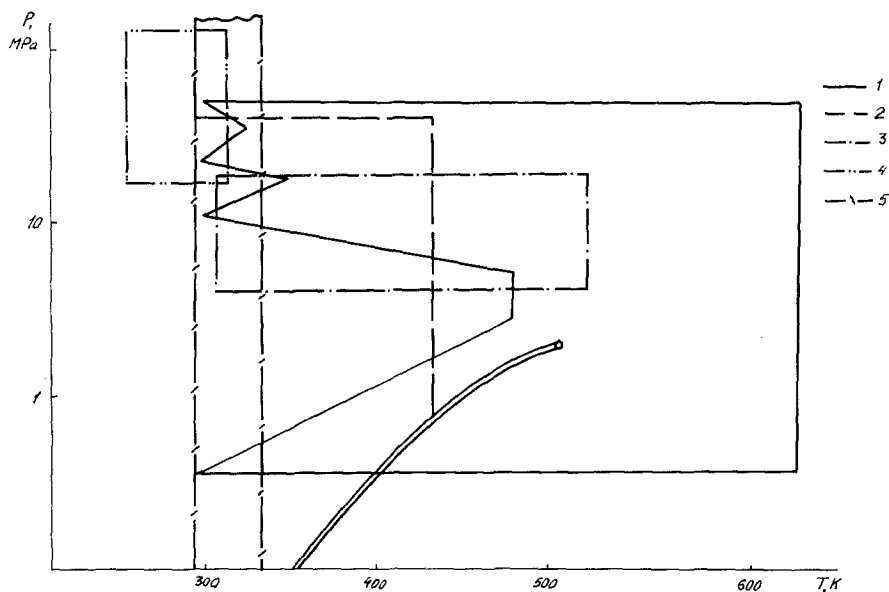


Fig. 2. Ranges of experimental data for the caloric and acoustic properties of *n*-hexane: 1 [26–28]; 2 [18]; 3 [14]; 4 [16]; 5 [2].

### 2.1.2. Materials

The sample of *n*-hexane used had a purity of at least 99.85–99.90 mol%; the impurities were the isomers of hexane. The simple properties were checked by absorption chromatography after every run.

### 2.1.3. Accuracy

The temperature was measured with an accuracy of 0.025 K. The pressure measurements were accurate to  $2.5 \times 10^{-4}$  and  $5.5 \times 10^{-4}$  MPa for  $\rho$  and  $C_p$ , respectively. The accumulated measurement error for the density was in the range of 0.05–0.07% at  $\rho > 2\rho_c$ , 0.07–0.1% at  $2\rho_c > \rho > 0.8\rho_c$ , and 0.1–0.2% at  $\rho < 0.8\rho_c$ , while the error for the isobaric heat capacity was 0.3–0.5% on a relative basis.

### 2.1.4. Results

More than 690 values of density and 540 values of isobaric heat capacity were obtained for the vapor and liquid phases, close to the saturation line and in a wide region around the critical point. These results are published in Refs. 23–28.

## 2.2. Analysis and Selection of Data

The experimental data analysis was performed according to the following scheme:

- (i) checking the reliability of the data for each property obtained by various authors on the basis of
  - (a) an analysis of the experimental procedure and the samples used and a check of the reliability of errors estimated by the authors,
  - (b) a graphic analysis of the data, and
  - (c) a check of the mutual consistency of the data of each pair of authors by using the homogeneity of dispersion criteria and the Kochran criteria of all selected authors; and
- (ii) evaluating the mutual correlation of heterogeneous data, which is done by
  - (a) comparing caloric and acoustic properties on the basis of local equations of state and
  - (b) comparing density values on the basis of well-known relations from caloric and acoustic properties with  $P$ - $V$ - $T$  data.

The present analysis made it possible to select for further handling the most reliable set of mutually correlated heterogeneous data of various authors. For  $n$ -hexane this set of data includes the following:

density [3, 4, 8, 9, 23–25],  
 heat capacity [14, 15, 26–28], and  
 ultrasonic velocity [2, 6, 17, 20, 21].

## 3. CALCULATION OF THERMODYNAMIC PROPERTY TABLES IN THE VAPOR AND LIQUID PHASES

The tables of thermodynamic properties (density, enthalpy, entropy, isochoric and isobaric heat capacity, ultrasonic velocity) of  $n$ -hexane in the vapor and liquid phases were calculated from a single equation of state in a polynomial form:

$$z = 1 + \sum_{i=1}^r \sum_{j=0}^{s_i} b_{ij} \omega^i \theta^j \quad (1)$$

where  $z = P/\rho RT$  is the compressibility factor,  $\omega = \rho/\rho_c$  is the reduced density, and  $\theta = T_c/T$  is the reciprocal reduced temperature.

The coefficients in Eq. (1) were determined by a stepwise linear regression technique with the inclusion, exclusion, and exchange of variables. Some modifications of existing methods were made to increase the quality of the equation and to extend its boundary. These modifications are described later.

### 3.1. Observance of Maxwell's Rule

It has been shown in a special analysis that the application of techniques suggested by previous authors [29, 30] for satisfying Maxwell's rule reduces the stability of a stepwise regression algorithm, while the fit to initial *P-V-T* data also deteriorates. Techniques for satisfying Maxwell's rule with the use of stepwise linear regression have not been described in the literature so far. Therefore, a special technique was used in formulating a single equation of state for *n*-hexane [31].

The principal point of this technique is as follows. In the two-phase region the thermodynamic surface is presented in the form of a sum  $z_{\text{main}}(\omega, \theta) + z_{\text{add}}(\omega, \theta)$ , where  $z_{\text{main}}$  is the value calculated from the basic equation obtained without any restriction, while  $z_{\text{add}}$  is chosen so that the following conditions are satisfied:

$$\left( \frac{\partial^2 z_{\text{add}}}{\partial \theta^2} \right)_{\omega', \omega''} = \left( \frac{\partial z_{\text{add}}}{\partial \theta} \right)_{\omega', \omega''} = z_{\text{add}}(\omega', \theta) = z_{\text{add}}(\omega'', \theta) = 0 \quad (2)$$

This condition ensures smoothness of the summary surface along the boundary curves.

In Ref. 31 it has been shown that a proper choice of the functional form of  $z_{\text{add}}(\omega, \theta)$  yields a better description of the thermodynamic properties in the metastable region. If only Maxwell's rule is to be obeyed, the knowledge of the dependence of  $z_{\text{add}}$  on  $\omega$  is not necessary. In this case, the temperature correction function  $f(\theta)$  is applied, its coefficients being defined by minimizing the functional

$$S = \sum_{k=1}^N \left[ z''_{\text{exp}k} - z'_{\text{exp}k} - \int_{\omega_k}^{\omega'_k} z_{\text{main}} \frac{d\omega}{\omega} - f(\theta) \right]^2 \quad (3)$$

When calculations are performed in the liquid phase and on the saturated liquid curve, corrections

$$\int_0^\omega \frac{d\omega}{\omega} = f(\theta); \quad \int_0^\omega \frac{\partial z_{\text{add}}}{\partial \theta} \cdot \frac{d\omega}{\omega} = \frac{df(\theta)}{d\theta}; \quad (4)$$

$$\int_0^\omega \frac{\partial^2 z_{\text{add}}}{\partial \theta^2} \cdot \frac{d\omega}{\omega} = \frac{d^2f(\theta)}{d\theta^2}$$

are made in the property calculation formulas.

The present technique helps to avoid the difficult procedure of obeying the phase-equilibrium conditions at the intermediate stage of making the single equation of state. This, however, does not affect the description of the original data, as corrections are not applied in the density calculation. A proper choice of the  $f(\theta)$  functional form makes it possible to satisfy the phase-equilibrium conditions over the entire temperature range, from the triple point to the critical point.

Another advantage of this procedure, in comparison with the existing ones, is that one can satisfy Maxwell's rule for any equation (linear with relation to its coefficients and nonlinear one) without the need of solving nonlinear problems with restrictions.

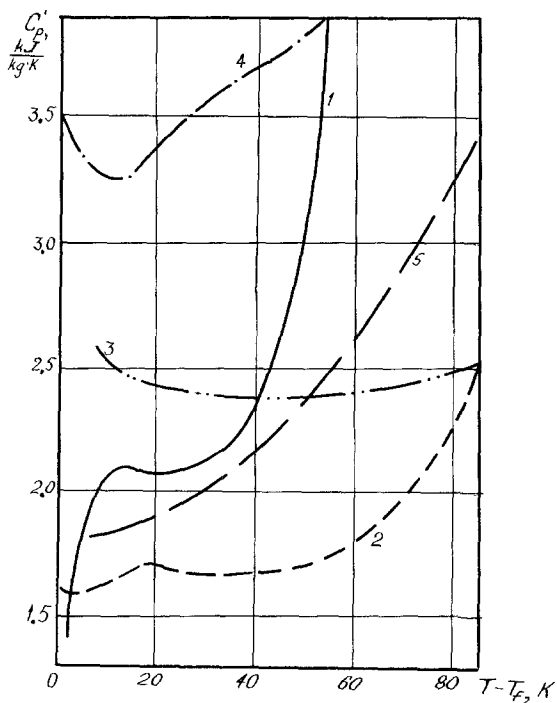
### 3.2. $P$ - $V$ - $T$ Surface Near the Boundaries of Validity of the Equation of State

Many equations of state in the literature are semiempirical or empirical approximations of the actual thermodynamic surface of the substance. Extrapolation with these equations is known to be restricted to the region of state parameters of the initial experimental data, and the shapes of isolines of the thermodynamic functions when extrapolated may be opposite to those predicted theoretically [32]. As a consequence, errors in thermophysical properties calculated near the boundaries within which the equation holds may increase significantly.

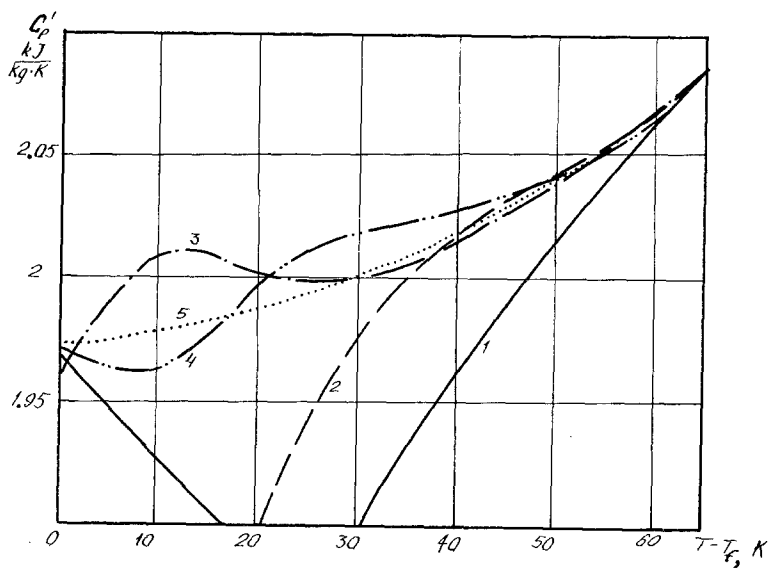
Often abnormalities in thermodynamic calculations are observed near the triple point,  $T_r$ . Our investigation reveals that in this region the calculated  $C_p'$  isobars have arbitrary shapes for all the substances studied [29, 30, 33–35] (Fig. 3). The shapes of the isolines for other thermodynamic properties are similar to the  $C_p'$  isobars but less vivid.

A calculated thermodynamic surface is often deformed near the upper temperature boundary, especially for hydrocarbons whose thermal transformation temperature is close to the critical point. For instance, the form of  $C_p'$  isotherms for  $n$ -hexane at  $T > 600$  K is different from those obtained by experiment [26–28].

The following calculation was applied to improve the quality of the thermodynamic surface near the boundaries of the equation of state. Arrays of additional data were calculated by the Lennard-Jones and Devonshire local equation of state in the liquid phase and by the virial equation in the vapor phase. We believe that pressure and temperature extrapolation may be made with more confidence due to theoretical substantiation of these equations. These "quasiexperimental" data extending the vapor and liquid phase of  $n$ -hexane to 660 and 140 K, respectively, used for determining equation-of-state coefficients make it possible to realize the following:



**Fig. 3.** Calculated  $C_p'$  values as a function of temperature for various substances: (1) nitrogen; (2) oxygen; (3) ethylene; (4) methane; (5) carbon dioxide.



**Fig. 4.** Calculated  $C_p'$  values as a function of temperature for *n*-hexane: (1, 2) without additional data; (3, 4) with additional data; (5) smoothed experimental data.

at temperature near  $T_f$  deviations of calculated property values from experimental values did not exceed the errors of the latter (for  $C_p$ , for example, see Fig. 4); and

calculated  $C'_p$  isotherms at  $TT_c$  are in complete agreement with experimental isotherms.

### 3.3. Equation-of-State Parameters and Its Validity

Several variants of the equation were obtained, which differed in the values of summation indexes, in the inclusion and exclusion criteria levels, and in the procedure of the weight calculation of experimental points.

The final variant of the single equation of state selected for  $n$ -hexane has the following parameters.

$$r = 8, \quad s = \{8, 8, 8, 2, 2, 1, 8, 8\}$$

$$\begin{aligned} b_{10} &= 1.418961 & b_{30} &= 8.874639 \times 10^{-1} & b_{60} &= 1.015613 \\ b_{11} &= -4.460154 & b_{31} &= -1.058535 \times 10^1 & b_{61} &= -6.151667 \times 10^{-1} \\ b_{12} &= 2.838127 & b_{32} &= -8.860190 \times 10^1 & & \\ b_{14} &= -1.155566 & b_{38} &= 1.969848 \times 10^{-2} & b_{70} &= 2.018526 \times 10^{-1} \\ b_{18} &= 2.981982 \times 10^{-2} & & & b_{71} &= 8.836143 \times 10^{-2} \\ & & b_{40} &= 1.355310 & b_{74} &= -4.836143 \times 10^{-3} \\ b_{20} &= -2.971036 \times 10^{-1} & b_{41} &= 5.780836 & b_{78} &= -9.908889 \times 10^{-5} \\ b_{21} &= 5.298878 & b_{42} &= -6.441410 & & \\ b_{22} &= -5.467548 & & & b_{80} &= 1.552810 \times 10^{-2} \\ b_{24} &= 1.200501 & b_{50} &= -2.132277 & b_{81} &= -6.039342 \times 10^{-3} \\ b_{28} &= -5.833447 \times 10^{-2} & b_{51} &= -2.660180 \times 10^{-1} & b_{84} &= 1.331250 \times 10^{-3} \\ & & b_{52} &= 1.482821 & b_{85} &= -8.367965 \times 10^{-5} \\ & & & & b_{88} &= 1.851536 \times 10^{-5} \end{aligned}$$

$$T_c = 507.85 \text{ K}, \quad \rho = 233.6 \text{ kg} \cdot \text{m}^{-3}, \quad R = 0.9649 \text{ kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$$

The weights of experimental points for these equations were calculated by

$$w = \left[ \frac{z_{\text{exp}}^2}{P^2} \sigma_P^2 + \left( \frac{z_{\text{exp}}}{T} + \frac{\partial z_{\text{calc}}}{\partial T} \right)^2 \sigma_T^2 + \left( \frac{z_{\text{exp}}}{\rho} + \frac{\partial z_{\text{calc}}}{\partial \rho} \right)^2 \sigma_\rho^2 \right]^{-1} \quad (5)$$

which takes into account the errors in  $T$ ,  $P$ , and  $\rho$  by the methods of error propagation. The derivatives  $\partial z_{\text{calc}}/\partial T$  and  $\partial z_{\text{calc}}/\partial \rho$  appearing in Eq. (5)



**Table I.** Experimental  $P$ ,  $\rho$ ,  $T$  Data Used in Developing a Single Equation of State for *n*-Hexane

| Parameter interval |               | Number of points |      | Accepted maximum errors (%) |            |               |                    | Reference No.          |
|--------------------|---------------|------------------|------|-----------------------------|------------|---------------|--------------------|------------------------|
| $P$ (MPa)          | $T$ (K)       | Total            | Used | $\delta P$                  | $\delta T$ | $\delta \rho$ | $\delta \rho_{ms}$ |                        |
| 0.1–500            | 273.15–333.15 | 44               | 44   | 0.05                        | 0.05       | 0.05          | 0.1                | 3                      |
| 0.1–900            | 293.15–338.15 | 64               | 63   | 0.05                        | 0.05       | 0.08          | 0.08               | 4                      |
| 0.5–7.0            | 507.85–516.15 | 194              | 168  | 0.08                        | 0.10       | 1.00          | 1.23               | 24 <sup>a</sup>        |
| 0.1–150            | 223.15–623.15 | 527              | 524  | 0.05                        | 0.05       | 0.15          | 0.18               | 25                     |
| 0.1–2.0            | 389.35–475.25 | 222              | 222  | 0.06                        | 0.05       | 0.25          | 0.17               | 8 <sup>a</sup>         |
| 2.5–20             | 513.15–573.15 | 58               | 58   | 0.06                        | 0.05       | 0.25          | 0.26               | 10 <sup>a</sup>        |
| 0.1–150            | 298.15–353.15 | 57               | 57   | 0.05                        | 0.05       | 0.15          | 0.11               | 9                      |
| 0.1–60             | 140–170       | 64               | 64   | 0.05                        | 0.05       | 0.05          | 0.25               | This work <sup>b</sup> |
| 10–80              | 630–650       | 36               | 36   | 0.05                        | 0.05       | 0.05          | 0.24               | This work <sup>b</sup> |

<sup>a</sup> Data for the critical region.

<sup>b</sup> "Pseudoexperimental" data.

<sup>c</sup> Data for the metastable region.

were determined by a special equation of state deduced without allowance for the weights of experimental points. An estimate of dispersion was calculated as

$$\sigma_x^2 = \left( \frac{x \delta x}{2} \right)^2 \quad (6)$$

where  $\delta x$  is the maximum relative error in the variable  $x$ . The values of  $\delta P$ ,  $\delta T$ , and  $\delta \rho$  obtained for the data of various authors are given in Table I.

With the equation obtained it is possible to calculate thermal, caloric, and acoustic properties of *n*-hexane in the temperature range from  $T_i$  to the beginning of thermal transformation and at pressures up to 100 MPa.

The standard deviations of calculated values from experimental values (outside the critical region) are as follows: for density, 0.18%; for isochoric heat capacity, 4.2%; for isobaric heat capacity, 1.3%; and for sound velocity, 1.3%.

#### 4. CALCULATION OF THERMODYNAMIC PROPERTIES IN THE CRITICAL REGION

As an analytic equation of state is invalid in the critical region, the thermodynamic properties of *n*-hexane in the range  $|(T - T_c)/T_c| < 0.1$  and

$|(\rho - \rho_c)/\rho_c| < 0.25$  were calculated by a scaled equation of state in the "linear model" variables with corrections to the asymptotical laws [36],

$$\Delta\bar{p} = \frac{P - P_c}{P_c} = ar^{\beta\delta}\theta(1 - \theta^2) + cr^{\beta\delta + \Delta}\theta + aKr^{2-\alpha}[\theta^2(1 - \theta^2) - f_0(\theta)] \\ + cKr^{2-\alpha+\Delta}[\theta^2 - f_1(\theta)] - \tilde{\phi}(t) \quad (7)$$

where

$$f_0(\theta) = f_{00} + f_{02}\theta^2 + f_{04}\theta^4; \quad f_1(\theta) = f_{10} + f_{12}\theta^2; \\ f_{00} = -\frac{\beta(\delta - 3) - b^2\alpha\gamma}{2b^4(1 - \alpha)\alpha(2 - \alpha)}; \quad f_{02} = \frac{\beta(\delta - 3) - b^2\alpha(1 - 2\beta)}{2b^2(1 - \alpha)\alpha}; \\ f_{04} = -\frac{1 - 2\beta}{2\alpha}; \quad f_{10} = \frac{\gamma + \Delta}{2b^2(2 - \alpha + \Delta)(1 - \alpha + \Delta)}; \\ f_{12} = -\frac{1 - 2\beta}{2(1 - \alpha + \Delta)}; \quad \tilde{\phi}(t) = \sum_{i=1}^3 a_i t^i$$

$T_c$ ,  $P_c$ , and  $\rho_c$  are the critical temperature, pressure, and density, respectively;  $\alpha$ ,  $\beta$ ,  $\gamma$ ,  $\delta$ , and  $\Delta$  are the conventional critical exponents [37];

**Table II.** Coefficients in Eqs. (7)–(10)

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|                                       |
|---------------------------------------|
| $T_c = 507.37$ K                      |
| $P_c = 3.036$ MPa                     |
| $\rho_c = 233.6$ kg · m <sup>-3</sup> |
| $\alpha = 0.108$                      |
| $\beta = 0.339$                       |
| $\gamma = 1.214$                      |
| $\delta = 4.561$                      |
| $\Delta = 0.5$                        |
| $B_0 = 1.9$                           |
| $B_1 = 0.44$                          |
| $K = 1.358$                           |
| $a = 26.8842$                         |
| $c = -11.7907$                        |
| $a_1 = -7.61632$                      |
| $a_2 = 31.3323$                       |
| $a_3 = -93.2$                         |

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$b^2 = (\gamma - 2\beta)/\gamma(1 - 2\beta)$ ; and  $a$ ,  $K$ , and  $c$  are individual constants for each substance. The dimensionless variables  $r$  and  $\theta$  in Eq. (7) were determined from

$$\begin{aligned} \Delta \bar{u} &= ar^{\beta\delta}\theta(1 - \theta^2) + cr^{\beta\delta} + \Delta\theta \\ t &= r(1 - b^2\theta^2) \end{aligned} \tag{8}$$

$$\Delta \tilde{p} = Kr^{\beta}\theta$$

where

$$\Delta \tilde{p} = (\rho - \rho_c)/\rho_c; \quad t = (T - T_c)/T_c; \quad \Delta \bar{u} = \frac{\rho_c}{P_c} [\mu(\rho, T) - \mu(\rho_c, T)]$$

The coefficient  $K$  in Eqs. (7) and (8) was calculated by

$$K = b_0(b^2 - 1)^P \tag{9}$$

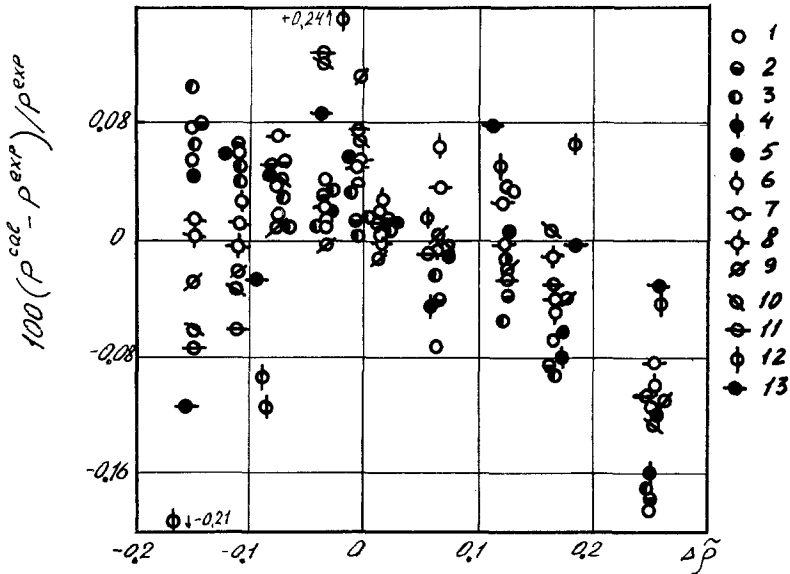


Fig. 5. Deviations of the pressures calculated with Eq. (7) from the experimental pressures at various temperatures: (1) 507.85 K; (2) 507.95 K; (3) 508.15 K; (4) 509.15 K; (5) 510.15 K; (6) 511.15 K; (7) 512.15 K; (8) 513.15 K; (9) 514.15 K; (10) 515.15 K; (11) 516.15 K; (12) 523.15 K; (13) 548.15 K.

where  $B_0$  is determined by the saturated vapor and liquid density data in the range  $|\Delta\tilde{\rho}| \leq 0.5$  via the equation

$$(\rho' - \rho'')/2\rho_c = B_0 t^\beta + B_1 t^{\beta+\Delta} \quad (10)$$

The coefficients in Eq. (7) were obtained from the selected experimental  $P$ - $V$ - $T$  data taken from Refs. 24 and 25 ( $0 < t < 0.08$ ,  $|\Delta\tilde{\rho}| \leq 0.25$ ). The values of the critical exponents were taken from Ref. 38. The parameters of the equation are listed in Table II.

The standard deviation between experimental pressure values and those calculated by Eq. (7) is 0.07%. The deviations are shown in Fig. 5.

The equations for the caloric and acoustic properties were obtained from Eq. (7) by differentiation. The calculated  $C_p'$  values were compared with those obtained by the authors [27, 28] (see Fig. 6). The standard deviation on the 3.3-, 3.4-, 3.5-, 4.0-, 4.5-, and 5.0-MPa isobars equaled 3.5%. The deviations on the 3.203-MPa isobar are 10–11%. The 3.103-MPa isobar close to the critical point still showed large deviations. This deviation can be explained by the errors of determining critical parameters in  $P$ - $V$ - $T$  and  $C_p$ - $P$ - $T$  experiments and may be due in part to the presence of impurities.

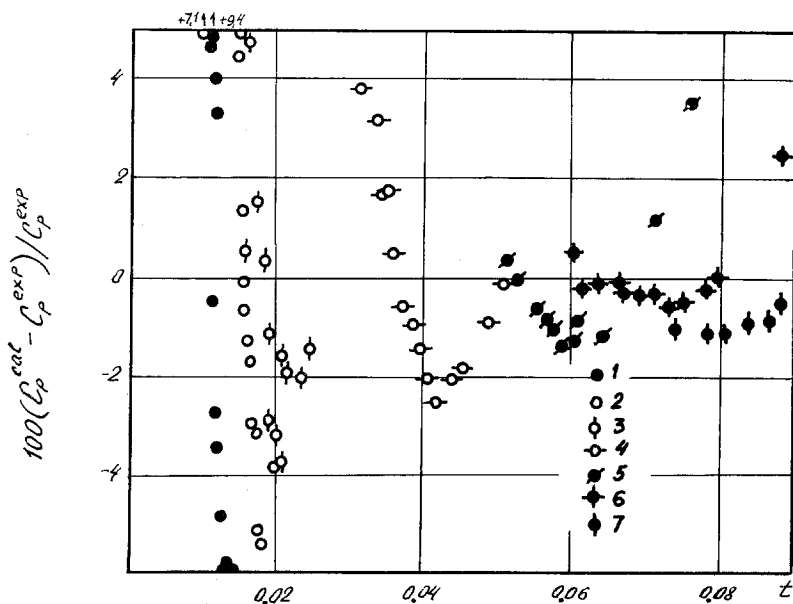


Fig. 6. Deviations of calculated  $C_p'$  values from experimental values at various pressures: (1) 3.302 MPa; (2) 3.402 MPa; (3) 3.503 MPa; (4) 4.003 MPa; (5) 4.503 MPa; (6) 5.002 MPa; (7) 5.500 MPa.

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