Heat Capacity of Liquid Benzene and Hexafluorobenzene at Atmospheric Pressure

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Heat capacity, C_p, of benzene and hexafluorbenzene was measured in the liquid phase at atmospheric pressure in the temperature range from 280 K to the boiling point. The results are expressed by the following equations: for C_6H_6,

$$C_p = 1.5194 - 1.299 \times 10^{-3}T + 6.927 \times 10^{-6}T^2$$

for C_6F_6,

$$C_p = 1.1913 - 1.072 \times 10^{-3}T + 3.589 \times 10^{-6}T^2$$

where C_p is in $kJ \cdot kg^{-1} \cdot K^{-1}$ and T is in K. The limiting error of heat capacity calculated from these equations is 0.23% for benzene and 0.18% for hexafluorobenzene. From the data, equations for specific volume, thermal expansion coefficient, and the quantity $(\delta T/\delta p)_s$ were obtained for benzene and hexafluorobenzene.

KEY WORDS: Benzene; heat capacity; hexafluorobenzene; specific volume; thermal expansion coefficient.

1. INTRODUCTION

This paper describes the results of the measurements of the heat capacity of benzene and hexafluorobenzene in the liquid phase at atmospheric pressure in the temperature range from 280 K to the boiling point.

Benzene, technically an important organic compound, finds extensive application in various fields of science and technology. A thorough knowl-

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edge of its thermodynamic properties is essential for performing computations related to technological processes. In the recent years, some interest has also been directed towards the use of benzene and its fluorosubstituted compounds as working fluids, coolants, high-temperature lubricants and liquid dielectrics [1, 2].

In spite of the fact that considerable attention has been paid to the studies of the heat capacity of benzene, substantial disagreements exist in the data by various investigators. Practically no study has been conducted until now of fluorosubstituted hydrocarbons; and it appears rather interesting to investigate the effect of hydrogen substitution by fluorine in hydrocarbon compounds on the thermodynamic properties over a wide range of state parameters.

2. MEASURING TECHNIQUE

The heat capacity of benzene and hexafluorobenzene, was measured with a flow calorimeter. A schematic diagram of the apparatus is shown in Fig. 1. In this apparatus, the liquid under investigation is supplied from a tank (4) by a leak-free pump (1) to a calorimeter (2). The liquid is cooled down in a thermostated chamber (5) and passed through a device (3) for flow rate determination.

The heat capacity of the liquid is determined from the relation

$$C_p = \frac{Q - q}{G(\Delta T - \Delta T_0)} \tag{1}$$

where Q is the power supplied by the heater in the calorimeter, q is the heat loss in the calorimeter, ΔT is the increase of the temperature of the substance in the calorimeter, ΔT_0 is the correction term for throttling in the calorimeter and thermal emf of the lead wires of the differential thermocouple, and G is the flow rate of the substance. The construction of the adiabatic flow calorimeter was described in detail in ref. [3].

The adiabaticity of the calorimeter was monitored from the readings of a copper-constantan thermometer whose sensitivity had been estimated to be 4.0×10^{-4} V/μV as a result of special experiments.

Temperature difference of the liquid at the inlet and outlet of the flow calorimeter was measured with the aid of a four-junction copper-constantan differential thermocouple calibrated with the use of a standard platinum resistance thermometer in the temperature range from 290 to 400 K. The dependence of thermal emf (E) on the temperature of the differential thermocouple junctions, obtained during calibration, was approximated by the method of least squares (cubic equation). The maximum deviation
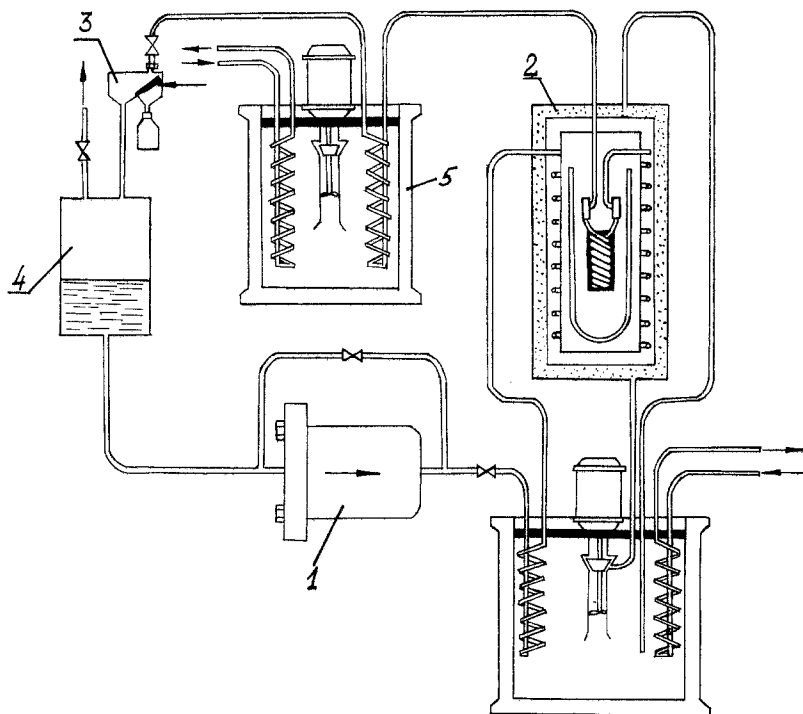
Fig. 1. Schematic diagram of the flow calorimeter.

of calibration values of temperature from those calculated from the equation is 0.023 K, and the root-mean-square deviation is 0.008 K.

The correction ΔT_0 for the thermal emf of the lead wires of the differential thermocouple and for the throttling effect was determined during the experiments with the calorimeter heat switched off, prior to each experiment aimed at determining the heat capacity of the liquid under investigation. This correction was insignificant in the experiments far from the saturation line (0.002 deg K) and reached 0.12 deg K for benzene at 351 K; in the case of hexafluorobenzene, the correction did not exceed 0.005 deg K over the entire temperature range under investigation.

In order to obtain a stable flow rate in the loop, a leak-free labyrinth pump was used. This pump was developed by the present authors for performing investigations of caloric properties of liquids at high pressures. The pump structure is shown in Fig. 2. The working portion of the pump consists of a multiple-thread screw (4) rotating in an inversely threaded sleeve (3). The magnitude of the head developed by the pump depends upon the length of the screw, depth and width of the groove profile, the
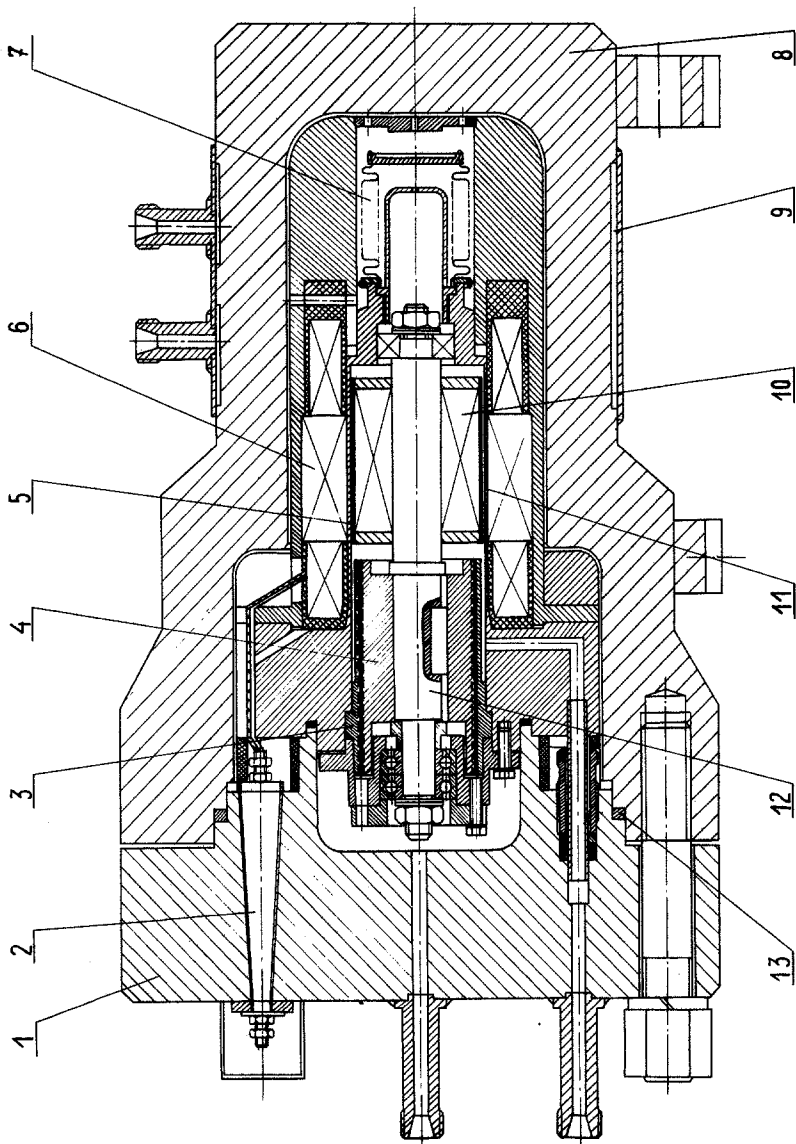
Fig. 2. Leak-free labyrinth pump.

angle of inclination and the clearance between the screw and the sleeve. The screw is set to rotation by an asynchronous motor (6), which has a sealed stator, pressure relieved and placed in an oil chamber. In order to relieve the pressure on a thin-wall sleeve (11) accommodating the stator, a bellows (7) is used, which separates the "working" space of the pump from the oil chamber. The pump is cooled by the circulation of water on the outer surface of the housing. The pump provided a flow rate of 6×10^{-3} kg · s^{-1} for benzene at a pressure of 0.2 MPa.

A pump of this type is characterized by the possibility of developing high heads at relatively low flow rates of the liquid, perfect tightness in the absence of any seals or gaskets, high stability of the flow rate, absence of a reducer, and compact arrangement. The pump is capable of operating at a high total pressure of the loop (30 MPa), its housing and parts are manufactured from stainless steel, and the rotor is protected from environmental effects by a stainless steel jacket.

The flow rate of substances under investigation in the apparatus loop was determined by the weighing technique. For this purpose, a sealed chamber (3) in Fig. 1 was used to sample the substance for weighing. The direction of the flow of material (to measuring flask or to collector tank) was controlled with a cutoff device, which operated with an electromagnetic coil located outside of the chamber. The time of filling of the measuring flask was registered automatically with an electronic stopwatch upon switching of the position of the cutoff device.

3. MEASUREMENT RESULTS AND EXPERIMENTAL ERROR

The working capacity of the apparatus was checked in the course of the experiments aimed at measuring the heat capacity of water in the temperature range from 303 to 326 K. In this temperature range, 11 experimental values have been obtained for water. The maximum deviation from the most accurate data on the heat capacity of water [4] is 0.14%, and the mean deviation is 0.05%.

The results of heat capacity measurements of benzene and hexafluorobenzene are presented in Tables I and II, respectively. The purity of benzene was 99.9%, while that of hexafluorobenzene was 99.74%. The molecular weight of benzene was taken as 78.11, and that of hexafluorobenzene was taken as 186.056. The experiments yielded 12 values of heat capacity of benzene and 18 values of heat capacity of hexafluorobenzene in the temperature range from the melting point to the boiling point of the substances under investigation.

The experimental values of the heat capacity, C_p, of benzene and hexafluorobenzene were fitted by polynomial functions by the method of

Table I. Experimental Results on the Heat Capacity of Benzene

T (K)	Q (W)	q (W)	$10^3 G$ (kg·s^{-1})	$(\Delta T - \Delta T_0)$ (K)	C_p (kJ·kg^{-1}·K^{-1})	$\dfrac{C_p(\exp) - C_p}{C_p} \times 100$ (%)
283.78	8.023	0.101	1.845	2.509	1.7094	+0.05
283.78	8.024	0.103	1.848	2.505	1.7087	0.00
293.58	18.593	0.090	2.550	4.182	1.7331	−0.11
298.28	18.580	0.097	2.574	4.120	1.7500	+0.10
298.50	10.474	0.017	2.080	2.872	1.7489	0.00
303.26	18.588	0.0004	2.608	4.037	1.7625	0.00
312.94	10.465	0.097	2.144	2.701	1.7883	−0.17
319.19	7.270	0.063	1.931	2.060	1.8045	−0.06
327.86	10.459	0.120	2.187	2.558	1.8453	+0.39
341.27	10.451	0.075	2.223	2.474	1.8773	−0.29
347.62	10.386	0.064	2.242	2.419	1.8992	−0.3
348.47	8.474	0.056	3.426	1.225	1.9154	+0.39

least squares. The resultant equations are, for C_6H_6,

$$C_p = 1.5194 - 1.299 \times 10^{-3}T + 6.927 \times 10^{-6}T^2 \qquad (2)$$

for C_6F_6,

$$C_p = 1.1913 - 1.072 \times 10^{-3}T + 3.589 \times 10^{-6}T^2 \qquad (3)$$

where C_p is in kJ·kg^{-1}·K^{-1}, and T is in K. The root-mean-square deviation of the experimental heat capacity values from those calculated from the equations is 0.2% for benzene and 0.12 for hexafluorobenzene. The random error in the heat capacity evaluated for an individual experiment from the random errors of values being measured amounted to 0.2%.

The systematic error in measuring the heat capacity with the aid of the aforementioned method is dependent on the accuracy of measuring the temperature difference of the substance under investigation in the calorimeter which, in turn, depends upon the error of calibrating the differential thermocouple of the calorimeter with the standard platinum resistance thermometer. The systematic error of determining the temperature difference was evaluated from the error in the parameters of the calibration equation obtained by the least squares method; it did not exceed 0.1% over the entire temperature range investigated. The limiting systematic error in measuring the heat capacity C_p, due to systematic errors in measuring flow rate of the substance G in the apparatus loop, power Q of the electric heater in the calorimeter, and calorimeter heat loss q, is estimated to be 0.05%.

Table II. Experimental Results on the Heat Capacity of Hexafluorobenzene

T (K)	Q (W)	q (W)	$10^3 G$ (kg·s^{-1})	$(\Delta T - \Delta T_0)$ (K)	C_p (kJ·kg^{-1}·K^{-1})	$\dfrac{C_p(\exp) - C_p}{C_p} \times 100$ (%)
284.00	7.155	0.020	5.296	1.146	1.1750	− 0.12
290.99	7.160	0.020	5.418	1.116	1.1808	− 0.22
292.28	7.169	0.099	3.225	1.846	1.1876	+ 0.24
297.91	7.166	0.069	3.290	1.811	1.1926	+ 0.17
297.90	7.178	0.029	5.248	1.142	1.1908	+ 0.01
298.76	7.158	0.029	5.514	1.087	1.1892	− 0.20
303.14	7.161	0.017	3.314	1.796	1.2001	+ 0.32
309.98	7.151	0.017	5.606	1.059	1.2016	− 0.20
314.69	7.156	0.022	2.782	2.115	1.2125	+ 0.24
318.61	7.223	− 0.012	5.565	1.072	1.2143	0.00
320.72	7.149	0.018	5.595	1.047	1.2173	+ 0.04
328.60	7.227	0.016	5.581	1.057	1.2230	− 0.31
330.65	7.156	0.0158	5.687	1.020	1.2303	+ 0.07
334.29	7.220	0.018	5.522	1.059	1.2313	− 0.23
337.31	7.222	0.024	5.469	1.065	1.2353	− 0.24
342.82	7.140	0.016	5.783	0.988	1.2473	+ 0.12
343.42	7.218	0.0156	5.665	1.016	1.2509	+ 0.34
349.67	7.213	0.010	5.715	1.004	1.2549	− 0.05

Considering the systematic random errors at a confidence level of 0.95, the limiting total error of heat capacity obtained from Eqs. (2) and (3) is 0.25% for benzene and 0.18% for hexafluorobenzene.

4. ANALYSIS OF DATA

4.1. Benzene

Several papers [5–20] were devoted to the study of heat capacity of benzene in the liquid phase. However, Burlew [17] showed that, in spite of the adequate accuracy (better than 1%) cited by the authors of these papers, substantial disagreements (reaching 3%) exist in the reported data. Burlew notes, with good reason, that the disagreements in the data could not be attributed to the presence of impurities in the investigated benzene. He performed extensive measurements of the heat capacity of benzene by the piezothermometric technique [17] which, in his opinion, showed an S-like temperature dependence of heat capacity. Our measurements, as well as data by other authors [14, 15] did not reveal the temperature dependence of heat capacity suggested by Burlew.

Table III gives a summary of the measurements reported in the literature on the heat capacity of benzene, which were used in the determination of the recommended equation for the heat capacity of liquid

Table III. Summary of Measurements on the Heat Capacity of Benzene
Reported in the Literature

Ref.	Year	Experimental technique	Range of measurements (K)	Error (%)	Statistical weight
[9]	1919	Adiabatic calorimeter	293–323	0.4	9
[10]	1921	Adiabatic calorimeter	293–323	0.3	16
[14]	1931	Adiabatic calorimeter	323–353	0.5	6
[17]	1940	Piezothermometric method	281–353	0.4–1.2	10–1
[18]	1948	Adiabatic calorimeter	287–337	0.2	35
[19]	1975	Flow calorimeter	293–353	0.5	6
present paper		Flow calorimeter	283–348	0.25	20

benzene at atmospheric pressure. The heat capacity values [5–7], character-ized by low accuracy, were not included in the above process.

In spite of a high accuracy claimed by some of the authors [8, 12, 16], the heat capacity values appear to have significantly great errors. In these papers, heat capacity measurements were performed in a relatively narrow temperature range, and the resulting values of the temperature dependence of heat capacity differed considerably from these obtained in more recent studies. Extrapolation of heat capacity data reported in refs. [8, 12, 16] leads to heat capacity values different from those obtained by other investigators by 3% and even more.

Williams and Daniels [11, 20] reported the existence of a minimum in the heat capacity of benzene at a temperature of about 293 K. Neither our measurements nor the data by other investigators [13, 17, 18, 23] have confirmed the presence of the minimum heat capacity of liquid benzene. Apparently, the accuracy (0.1%) of the data by Williams and Daniels was considerably overestimated.

The heat capacity values obtained by Richards and Wallace [15] using the piezothermometric technique are in good agreement with the data of the present paper, as well as with the data of some investigators [17, 18]. Nevertheless, these data have not been included in the process of obtaining the recommended equation because the investigators had not taken into account a series of corrections in determining $(\partial T/\partial P)_s$; also, inaccurate values for $(\partial V/\partial T)_p$ were used in calculating the heat capacity of benzene. The data of ref. [15] are markedly inferior to those by Burlew [17] with respect to the accuracy of measuring $(\partial T/\partial P)_s$. In our opinion, the error in the data of ref. [15] was underestimated.

The early data by Huffman et al. [13] on the study of heat capacity in the temperature range from 281 to 300 K have not been taken into

consideration because of more detailed and accurate measurements of heat capacity performed at a later date with his participation [18]. The accuracy of the heat capacity values of the latter investigation exceeds that of the earlier work by 0.3–0.9%.

Based on the results of heat capacity measurements in the papers listed in Table III, we have used the least squares method to obtain the following recommended equation for heat capacity of liquid benzene:

$$C_p = 1.3943 - 5.857 \times 10^{-4}T + 5.89 \times 10^{-6}T^2 \qquad (4)$$

where C_p is in kJ · kg^{-1} · K^{-1} and T is in K.

The statistical weight of the experimental values of heat capacity used in obtaining the above equation was assumed to be inversely proportional to the square of the measurement error estimated by the investigators of the original data.

Deviations of the experimental values of the heat capacity of benzene from those calculated from Eq. (4) are presented in Fig. 3. The dashed line indicates the error in heat capacity computed from the uncertainty in the coefficients that result from the least squares method. At the ends of the investigated temperature range, the error in heat capacity amounts to 0.005–0.006 kJ · kg^{-1} · K^{-1} (0.3%). As may be seen from Fig. 3, the agreement between the experimental values by the investigators of refs. [10,
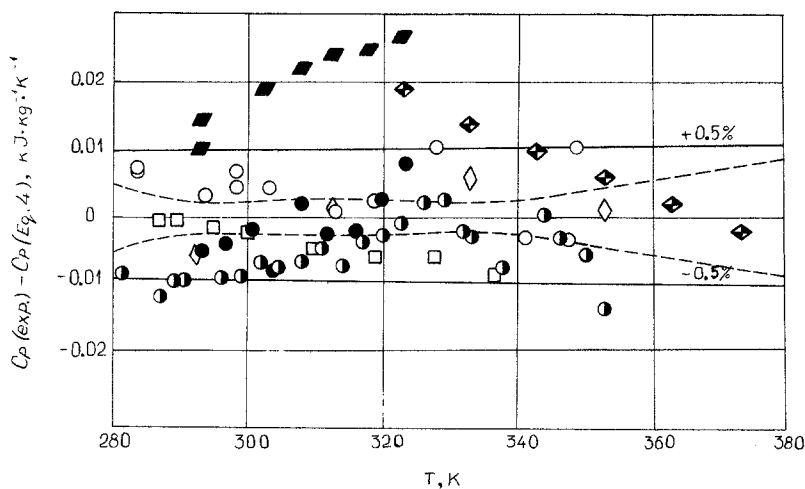

Fig. 3. Deviation of the experimental values of the heat capacity of benzene from the values computed from Eq. (4). ○—present work, ■—[9], ●—[10], □—[18], ◑—[17], ◇—[19], ⊠—[14].

Table IV. Comparison of Heat Capacity of Benzene Computed from Eqs. (2) and (4)
with Values Recommended by Chao [21]

T (K)	C_p[21] (kJ · kg^{-1} · K^{-1})	$\dfrac{C_p - C_p[21]}{C_p[21]} \times 100$ from Eq. (4) (%)	$\dfrac{C_p - C_p[21]}{C_p[21]} \times 100$ from Eq. (2) (%)
280	1.694	− 0.13	0.27
290	1.719	0.06	0.33
300	1.747	0.12	0.37
310	1.775	0.22	0.42
320	1.804	0.33	0.47
330	1.835	0.44	0.53
340	1.867	0.48	0.59
350	1.901	0.52	0.65
360	1.935	0.62	0.72

17–19] and those by the present authors is 0.5%. However, disagreement between the data of refs. [9, 14] and those of the present work reaches 1.5%.

A comparison of heat capacity values computed from Eqs. (2) and (4) with the values recommended by Chao [21] is given in Table IV. It may be seen that the agreement of the results of ref. [21] with the heat capacity values calculated from Eq. (4), as recommended by us, is within 0.13–0.6%, and the disagreement with the smoothed experimental heat capacity data obtained in the present paper is 0.3–0.7%.

Based on the well-known thermodynamic relation,

$$C_p = T \frac{(\partial V / \partial T)_p}{(\partial T / \partial P)_s} \tag{5}$$

we computed thermodynamic properties for liquid benzene at atmospheric pressure. In the calculations, use was made of the experimental data on specific volumes obtained in refs. [22–26], experimental measurements of the quantity $(\Delta T / \Delta P)_s$ performed by the authors of ref. [27], and of the heat capacity values recommended in the present paper.

Temperature dependence of the thermal expansion coefficient of benzene was found by the method of least squares from the values of $(\delta V / \delta T)_p$ obtained from the data on specific volume $V - V_{T=298.15}$ reported in refs. [22–26] and from the data on $(\delta V / \delta T)_p$ calculated from Eq. (5) using the experimental values of $(\Delta T / \Delta P)_s$ [27] and the recommended heat capacity of the present paper. The statistical weight was assumed to be inversely proportional to the square of the error, 1–8 for $(\delta V / \delta T)_p$ obtained on the

basis of specific volume measurements, 1 for $(\delta V/\delta T)_p$ found from the data on $(\Delta T/\Delta P)_s$ and C_p.

As a result of data processing, the following equations have been obtained for specific volume (in $m^3 \cdot kg^{-1}$), thermal expansion coefficient (in $m^3 \cdot kg^{-1} \cdot K^{-1}$), and the quantity $(\partial T/\partial P)_s$ (in $K \cdot MPa^{-1}$) for benzene:

$$(V - V_0) \times 10^3 = -0.3618 + 1.5184 \times 10^{-3}T - 2.658 \times 10^{-6}T^2$$

$$+ 5.48 \times 10^{-9}T^3 \tag{6}$$

$$(\delta V/\delta T)_p \times 10^3 = 1.5184 \times 10^{-3} - 5.316 \times 10^{-6}T$$

$$+ 1.644 \times 10^{-8}T^2 \tag{7}$$

$$(\delta T/\delta P)_s = 1.256 \times 10^{-1} - 4.099 \times 10^{-4}T + 2.645 \times 10^{-6}T^2 \tag{8}$$

where T is in K.

The thermodynamic properties of liquid benzene at atmospheric pressure recommended by us are listed in Table V. In computing the specific volume, the value $V_0 = 1.14456 \times 10^{-3} m^3 \cdot kg^{-1}$ for 298.15 K was taken from ref. [21], which agrees within $\pm 0.01\%$ with the values of well-known measurements by a number of investigators. The error in $(\delta V/\delta T)_p$ over the entire investigated temperature range is 0.07–0.25%, and that in $(\delta T/\delta P)_s$ is 0.25–0.5%.

A comparison of the values of specific volume recommended by us with the data of refs. [21–26] is presented in Fig. 4. It may be seen that the values of specific volume agree within 0.04%. The disagreement of the

Table V. Thermodynamic Properties of Benzene

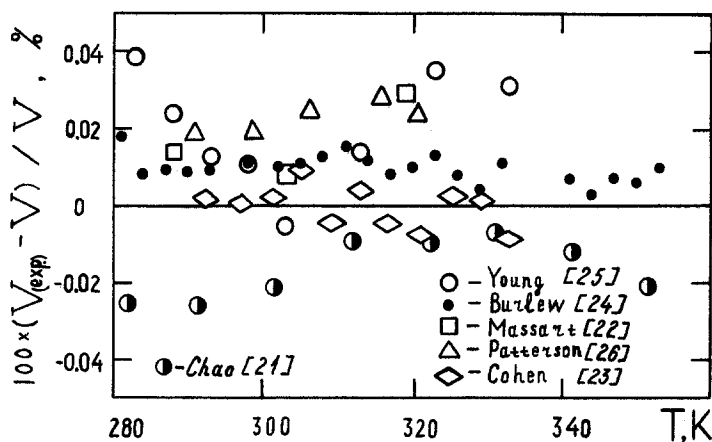| T (K)	C_p (kJ·kg^{-1}·K^{-1})	10^3V (m^3·kg^{-1})	$10^6(\delta V/\delta T)_p$ (m^3·kg^{-1}·K^{-1})	$(\delta T/\delta P)_s$ (K·MPa^{-1})
278.68	1.6885	1.118084	1.314	0.2168
280.00	1.6921	1.119822	1.319	0.2182
290.00	1.7198	1.133210	1.359	0.2292
300.00	1.7487	1.147020	1.403	0.2407
310.00	1.7788	1.161285	1.450	0.2527
320.00	1.8100	1.176037	1.501	0.2653
330.00	1.8424	1.191311	1.554	0.2784
340.00	1.8761	1.207137	1.611	0.2920
350.00	1.9108	1.223250	1.672	0.3062
353.25	1.9224	1.229016	1.692	0.3109

Fig. 4. Deviation of the experimental values of the specific volume of benzene from the values computed from Eq. (6).

values of $(\delta T/\delta P)_s$ with the experimental data of ref. [27] is in the range 0.3–0.6%.

4.2. Hexafluorobenzene

Only one paper [28] is known to us which is devoted to the measurement of the heat capacity of liquid hexafluorobenzene. This paper describes the measurements of heat capacity in the solid and liquid phases in the temperature range from 11 to 310 K. The error, as estimated by the investigators, is 0.2%.

The processing of the combined data of Counsell and Green [28] and those of the present authors by the least squares method, giving equal statistical weight, yielded the following equation for the heat capacity of hexafluorobenzene:

$$C_p = 1.1975 - 1.105 \times 10^{-3}T + 3.636 \times 10^{-6}T^2 \qquad (9)$$

where C_p is in $kJ \cdot kg^{-1} \cdot K^{-1}$ and T is in K.

The deviation of the experimental data by Counsell and Green and by the present authors from the smoothed results of Eq. (9) is presented in Fig. 5. As may be seen, the agreement is within the experimental error; the maximum deviation from the smoothed curve does not exceed 0.35%.

In the computation of the quantities $(\delta V/\delta T)_p$ and $(\delta T/\delta P)_s$, we have used the values of specific volume of hexafluorobenzene provided in refs. [29] and [28] for the temperature range from 293 to 353 K. In computing the specific volume for hexafluorobenzene, which was obtained by us in the
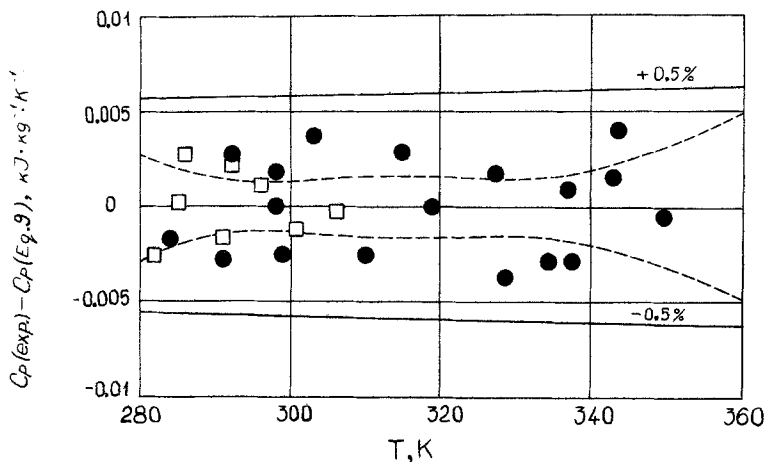
Fig. 5. Deviation of the experimental values of the heat capacity of hexafluorobenzene from the values computed from Eq. (9). ●, present work; □, [28].

course of studies on the enthalpy of liquid hexafluorobenzene over a wide range of state parameters. The computation was performed using the well-known thermodynamic relation

$$V/T = V_0/T_0 - \int_{T_0}^{T} \frac{(\delta i/\delta P)_T}{T^2} \, dT \tag{10}$$

The quantity $(\delta T/\delta P)_s$ was obtained from Eq. (5) using the heat capacity data and the computed values of $(\delta V/\delta T)_p$. The values of specific volume (in $m^3 \cdot kg^{-1}$), thermodynamic quantities $(\delta V/\delta T)_p$ (in $m^3 \cdot kg^{-1} \cdot K^{-1}$), and $(\delta T/\delta P)_s$ (in $K \cdot MPa^{-1}$) were approximated by the following equations:

$$V \times 10^3 = 0.4628 + 6.05 \times 10^{-4}T - 1.22 \times 10^{-6}T^2$$

$$+ 3.30 \times 10^{-9}T^3 \tag{11}$$

$$(\delta V/\delta T)_p \times 10^3 = 6.05 \times 10^{-4} - 2.44 \times 10^{-6}T$$

$$+ 9.90 \times 10^{-9}T^2 \tag{12}$$

$$(\delta T/\delta P)_s = 1.14467 \times 10^{-1} - 8.307 \times 10^{-4}T$$

$$+ 3.632 \times 10^{-6}T^2 \tag{13}$$

Table VI. Thermodynamic Properties of Hexafluorobenzene

T (K)	C_p (kJ · kg^{-1} · K^{-1})	$10^3 V$ (m^3 · kg^{-1})	$10^6 (\delta V / \delta T)_p$ (m^3 · kg^{-1} · K^{-1})	$(\delta T / \delta P)_s$ (K · MPa^{-1})
278.35	1.1716	0.6079	0.693	0.1646
280.00	1.1732	0.6090	0.698	0.1666
290.00	1.1828	0.6161	0.730	0.1790
300.00	1.1932	0.6236	0.764	0.1921
310.00	1.2044	0.6314	0.800	0.2060
320.00	1.2162	0.6396	0.838	0.2206
330.00	1.2288	0.6482	0.878	0.2359
340.00	1.2421	0.6572	0.920	0.2519
350.00	1.2562	0.6666	0.964	0.2686
353.41	1.2611	0.6699	0.979	0.2745

The thermodynamic properties of liquid hexafluorobenzene at atmospheric pressure, calculated from Eqs. (9) and (11)–(13) are listed in Table VI.

It may be seen from Tables V and VI that substantial differences exist in the thermodynamic properties of benzene and hexafluorobenzene; namely, the specific volume of benzene is 1.5 times and the heat capacity is 1.3 times greater than those of hexafluorobenzene, and the quantities $(\delta V / \delta T)_p$ and $(\delta T / \delta P)_s$ differ by 42 and 18%, respectively.

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