THERMODYNAMIC PARAMETERS OF BUTAN-1-OL AT ATMOSPHERIC PRESSURE

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Published data are examined on the density, speed of sound, and isobaric specific heat of liquid butan-1-ol. Values have been calculated for the isochoric specific heat and the adiabatic and isothermal compression coefficients for the range 183.85-390.81°K.

A promising method of drawing up tables of thermodynamic parameters for liquid is provided by using acoustic measurements as input data.

It has been shown [1-3] that one can use acoustic quantities (the speed of sound) along with data on the density and isobaric specific heat to calculate other thermodynamic quantities for liquids at atmospheric pressure with an accuracy greater than that of direct experimental measurement, particularly the isothermal and adiabatic compressibilities, the isochoric specific heat, and so on. As regards high pressures, calculations are also possible, where on the one hand one uses data on the speed of sound as a function of temperature and pressure and on the other hand employs data on the density and isobaric specific heat either at atmospheric pressure, or on the saturation curve, or on some other reference isobar in the liquid region where the pressure exceeds atmospheric, in accordance with the temperature interval involved. Clearly, data on the density and isobaric specific heat are basic in these calculations. Here it is desirable to represent the initial information as analytical relationships, particularly at atmospheric pressure, as this substantially simplifies the calculations.

Polynomials in powers of x = T/1000 have been used to approximate the temperature dependence of the specific volume (density), speed of sound, and isobaric specific heat for butan-1-ol at atmospheric pressure:

$$y = \sum_{i=0}^{n} a_i x^i , \tag{1}$$

where y is the function corresponding to the properties v, W, and C_p. The coefficients a_i in (1) for v, W, and C_p were determined by least-squares fitting with a Minsk-32 computer. We processed all the data selected from a careful analysis with statistical weights assigned in accordance with the estimated errors. The number of terms in (1) was restricted on the basis of the F test at the 0.95 confidence level.

Specific Volume and Density. There are numerous papers on the density of butan-1-ol at atmospheric pressure and on the saturation curve in the liquid state. Some of them covering the range 273-313°K have been systematized in works of reference [4, 5]. The fullest survey of the experimental studies is to be found in [6], which includes papers published up to 1970, and it gives not only the densities derived by various workers at 293.15 and 298.15°K but also recommendations on the average values for the density at these temperatures and on the saturation curve for the range 253-383°K. These density values were calculated from Francis's equation, which describes all the data used by those workers to derive the coefficients in this equation within limits of ±1 kg/m^3 and which is applicable in the range 240-420°K. Here it should be noted that the literature contains no equations describing the density of butan-1-ol from the melting point to the boiling point at atmospheric pressure. Also, firstly, not all the data on the density of butan-1-ol were incorporated in compiling the equation in [6], particularly for low temperatures, and secondly there are now new measurements at temperatures above 273°K. These features have led us to draw up a new interpolation equation. As a detailed list of the experimental studies is to be found in [6], we restrict ourselves to examining the basic ones.

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TABLE 1. Coefficients in (1)

Coefficient a_i	Property		
	v, см^3/g	W, m/sec	c_p, kJ/kg • K
a_0	1,00818772	149,538379	43,8452838
a_1	—0,387874078	45640,5669	—970,161826
a_2	9,81831033	—395928,986	9161,39538
a_3	—30,8128567	1477420,61	—45318,7506
a_4	36,9957188	—2619087,43	123696,167
a_5	—	1790311,97	—175909,444
a_6	—	—	101802,336
Temp. range, K	186—390,81	193—390,81	183,85—390,81

TABLE 2. Calculated Values of the Density and Specific Volume for Butan-1-ol

T, K	ρ, kg/m^3	$v \cdot 10^3$, m^3/kg	T, K	ρ, kg/m^3	$v \cdot 10^3$, m^3/kg
183,85	893,2*	1,1195	298,15	805,79	1,24102
193,15	885,7	1,1290	303,15	801,97	1,24693
203,15	877,8	1,1393	313,15	794,23	1,25909
213,15	869,9	1,1496	323,15	786,30	1,27178
223,15	862,2	1,1599	333,15	778,16	1,28509
233,15	854,5	1,1703	343,15	769,74	1,29914
243,15	846,9	1,1807	353,15	761,02	1,31403
253,15	839,5	1,1913	363,15	751,94	1,32990
263,15	832,0	1,2019	373,15	742,46	1,34687
273,15	824,56	1,21278	383,15	732,55	1,36509
283,15	817,10	1,22385	390,81	724,65	1,37998
293,15	809,58	1,23521			

*Extrapolation.

The most extensive density measurements, which cover also the low-temperature range, are to be found in the following: [7] from 192 to 273°K, [8] from 213 to 333°K, [9] from 242 to 265°K, [10] from 213 to 453°K, and [11] from 186 to 298°K. The errors of these measurements were estimated by these workers as not exceeding 0.1%. The density above 273°K has been examined in [2, 12-17]. Finally, Hales and Ellender [18] in 1976 made new measurements of the density on the saturation curve over the range 293-490°K with an error of ±0.15 kg/m^3. The data of [18] are the most accurate ones on the density above 293°K.

The density data given by different workers in the regions where they can be compared often show large discrepancies, which frequently exceed the overall errors. For example, the new density data [18] may be compared with the densities recommended in [6], and it is seen that the latter are larger, with a deviation of 0.02% at 293.15°K, but with the difference increasing with temperature and attaining 0.08% at the normal boiling point. As regards the data of [2, 9, 10, 14-17], it has been shown in [18] and is further demonstrated here that there are large discrepancies from the values of [18]. The discrepancies between the values of [7, 8, 11] at low temperatures are less than the overall errors and in the main do not exceed 0.05%. The data of [10] are lower than these by 0.10-0.25%. The densities of [6] are also lower, and the discrepancies increase as the temperature is reduced ($T < 273$°K). Most of the data sets agree within 0.01-0.03% in the range 273-303°K.

The data of [4, 5, 7, 8, 11, 18-20] were selected for final joint processing. The densities on the saturation curve of [18] were referred to atmospheric pressure. The corrections were not more than 0.01%.

Processing for (1) gave the following a_i for the specific volume, whose values are given in Table 1 for the case where v is in cm^3/g. The recommended values for the specific volume and density, which is defined as $\rho = 1/v$, are given in Table 2 between the melting point (183.85°K [6]) and the boiling point (390.81°K [6]) at atmospheric pressure. Figure 1 shows the deviations of the calculated densities from the measured values, and it is evident that (1) with the coefficients for the specific volume describes the initial experimental data with an error not exceeding the errors in the measurements from the individual sources. For comparison, we show the deviations in the data of [6, 12, 13, 17, 21, 22].

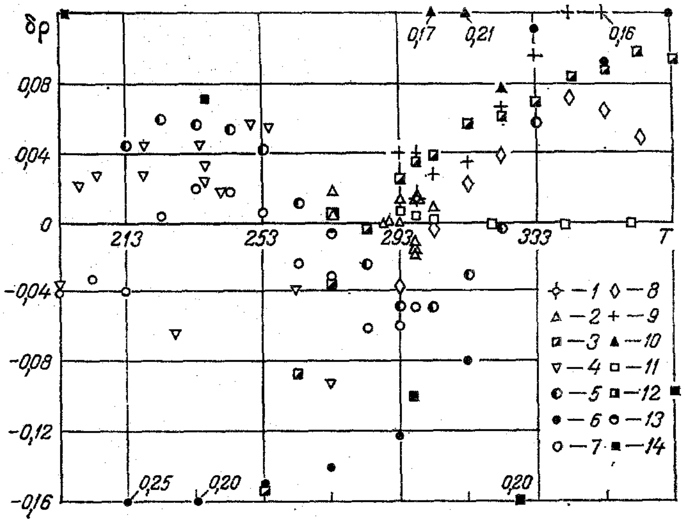

Fig. 1. Comparison of calculated densities with experimental values given by other workers: (δρ[i] = (ρ[i] — ρ[MTI])·100/ρ[MTI], %): 1) [4]; 2) [5]; 3 [6]; 4) [7]; 5) [8]; 6) [10]; 7) [11]; 8) [12]; 9) [13]; 10) [17]; 11) [18]; 12) [19]; 13) [20]; 14) [21, 22]. T, K.

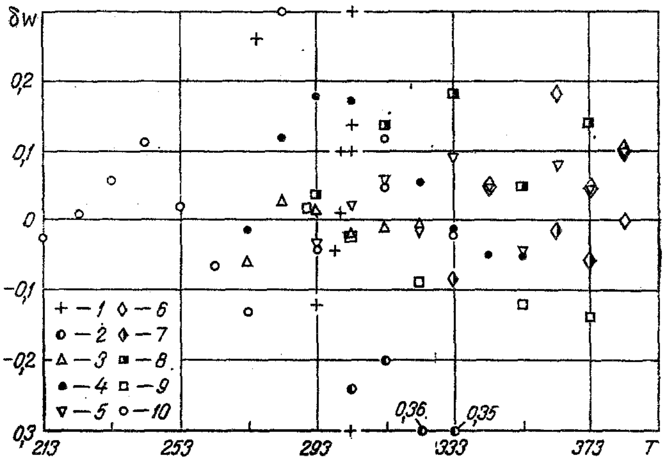

Fig. 2. Comparison of calculated speeds of sound with data given by other workers: (δW[i] = (W[i] — W[MTI])·100/W[MTI], %): 1) [23-32]; 2) [12]; 3) [37]; 4) [38]; 5) [33]; 6) [34]; 7) [2]; 8) [35]; 9) [3]; 10) [36].

The deviations in the calculated densities from the most reliable experimental values are not more than ±0.02% above 273°K or ±0.05% below 273°K, so one can assume that the recommended values for the density and specific volume have the same accuracy.

Speed of Sound. There are about 30 papers on the speed of sound in liquid butan-1-ol at atmospheric pressure. We consider the main ones below. We do not consider papers in which the information on the speed of sound is given only in graphical form or those where the measurements were made with low accuracy. In [23-32], the measurement was made at one point at room temperature. The errors of measurement were estimated as 0.1-0.2%. Otpusuchennikov et al. [2, 3, 32-36] have made numerous measurements of the speed of sound at atmospheric pressure and on the saturation curve at 193-553°K. The errors in their measurements did not exceed 0.2%. In [12], the speed of sound was determined at 293-333°K, and in [37] between 273 and 323°K with an accuracy of ±0.56 m/sec, or in [38] at 273-353°K. The last paper gives not the experimental values but the coefficients in the interpolation equation. At low temperatures, measurements have been made [8] between 213 and 333°K and in [36] between

TABLE 3. Calculated Values of the Thermodynamic Parameters for Butan-1-ol

T, K	c_p, $\frac{\text{kJ}}{\text{kg}\cdot\text{K}}$	c_V, $\frac{\text{kJ}}{\text{kg}\cdot\text{K}}$	$\beta_S\cdot10^5$, 1/MPa	$\beta_T\cdot10^5$, 1/MPa	W, m/sec
183,85	1,8245*	1,4632	37,72	47,04	1722,7*
193,15	1,8405	1,4820	40,18	49,91	1676,2
203,15	1,8654	1,5114	43,05	53,14	1626,7
213,15	1,8953	1,5467	46,14	56,54	1578,5
223,15	1,9290	1,5859	49,43	60,12	1571,9
233,15	1,9666	1,6287	52,91	63,89	1487,2
243,15	2,0093	1,6755	56,59	67,87	1444,4
253,15	2,0585	1,7272	60,46	72,05	1403,7
263,15	2,1156	1,7848	64,51	76,47	1364.9
273.15	2,1819	1,8493	68,79	81,16	1327,8
283,15	2,2581	1,9210	73,30	86,17	1292,1
293,15	2,3443	1,9999	78,11	91,56	1257,5
298,15	2,3911	2,0418	80,64	94,43	1240,5
303,15	2,4401	2,0852	83,27	97,44	1223,7
313,15	2,5441	2,1755	88,85	103,91	1190,4
323,15	2,6545	2,2689	94,97	111,10	1157,2
333,15	2,7690	2,3631	101,72	119,20	1124.0
343,15	2,8851	2,4554	109,26	128,38	1090,4
353,15	3,000	2,544	117,73	138,86	1056,5
363,15	3,112	2,626	127,29	150,89	1022,2
373,15	3,220	2,700	138,10	164,67	987,6
383,15	3,323	2,768	150,30	180,45	953,0
390,81	3,401	2,817	160,65	193,99	926,8

*Extrapolation

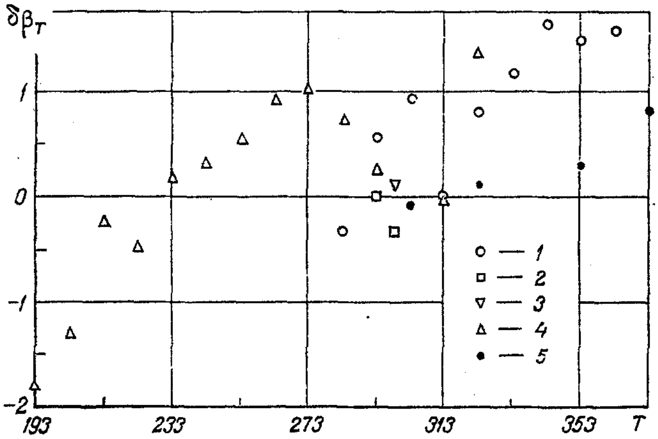

Fig. 3. Comparison of calculated values for the isothermal compressibility with data given by other workers: ($\delta\beta_T[i] = (\beta_T[i] - \beta_T[\text{MTI}])\cdot 100/\beta_T[\text{MTI}]$, %): 1) [45]; 2) [46]; 3) [47]; 4) [36]; 5) [3].

193 and 323°K with errors of 0.5 and 0.2% correspondingly. The values of [8, 36] are in poor agreement with one another, particularly below 273°K, where the discrepancies substantially exceed the overall errors. Also, the discrepancies increase as the temperature is reduced and attain 3% at T = 213.15°K. Preference was given to the values of [36], since in contrast to those of [8] they agree well with the data from other sources above 273°K.

The coefficients a_i of Table 1 were obtained by processing the selected data for (1).

Figure 2 shows the deviations in the calculated speeds from the initial experimental values. The deviations in the main lie within the experimental error limits. As the convergence between the calculated results and the most reliable data lies within the range ±0.10%, one can say that the recommended speeds of sound (Table 3) have the same accuracy.

Isobaric Specific Heat. We know of only five papers where C_p for the liquid phase has been measured. In [39] there are measurements over the range 194–294°K with an error of

0.5%. In [40], the results are presented as the coefficients in an interpolation equation covering the range T = 303-348°K. The most careful and extensive studies of C_p on the saturation curve at 188-322°K are to be found in [41], where the error level is 0.1-0.2%. In that paper, C_p at the triple point (184.51°K) was determined by graphical extrapolation. The measurements on C_p of [42] apply to the liquid state, but close to the saturation curve, for the range 303-462°K and pressure of 0.098-0.92 MPa, error level 0.9%. Values for C_p have been derived [43] for the range 293-533°K at 0.1-60 MPa with errors of 1.5%. The value of C_p recommended in [6] for the normal boiling point has an estimated error of 1.65%.

All these data were used in the processing.

Table 1 gives the coefficients of (1) for the isobaric specific heat; (1) with the coefficients for C_p describes the initial data from the melting point to the boiling point at atmospheric pressure within the workers' accuracy estimates. An exception is represented by the data of [43], which lie below the calculated values of C_p at 313-373°K by 1.6-2.2%.

We used (1) with the coefficients for v, W, and C_p to calculate the isochoric specific heat C_V, together with the adiabatic compressibility β_S and the isothermal compressibility β_T for the range 183.85-390.81°K at atmospheric pressure from the equations given in [44]. Table 3 gives the results. Figure 3 shows the deviations of the calculated β_T from the experimental values [45-47] and the values of β_T [35, 36] calculated from velocity measurements. The discrepancies do not exceed 1-1.5%. As regards the β_T calculated in [2], these lie systematically below our calculated values by on average 4%.

So far as we are aware, no experimental values have been published for the isochoric specific heat, so a comparison was made with the calculated values [3, 36]. The values of C_V of [36] lie below ours on average by 0.3-0.4%. The discrepancies from the C_V of [3] at 303, 323, 353, and 373°K are correspondingly 0.29, −1.45, −4.63, and −10.4%.

NOTATION

T, temperature; v, specific volume; ρ, density; W, speed of sound; C_V, C_p, isochoric and isobaric heat capacities; β_S, β_T, adiabatic and isothermal compression coefficients.

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COMPRESSIBILITY AND VIRIAL COEFFICIENTS OF METHANE—ARGON MIXTURES

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Using the Barnett method we have measured the compressibility of methane—argon mixtures at temperatures of 373.15 and 423.15°K and pressures up to 30 MPa. The methane concentrations in the mixtures were varied from 8 to 82 mole %. We calculated the second, third, and second mixed virial coefficients.

The present article is a continuation of systematic studies of the bulk behavior of gases and gas mixtures by the Barnett method [1-4]. We measured the compressibility of methane—argon mixtures, and calculated the second and third virial coefficients of the mixtures. The values obtained for the second virial coefficients were used to calculate the second mixed virial coefficient B_{12}, which furnishes important information on the nature of the intermolecular interaction of dissimilar molecules.

The apparatus and measurement procedure were described earlier [1, 2]. The pressure was measured with MP-600 and MP-60 piston manometers of grade 0.05 accuracy, and the temperature was measured to within 0.01°C by a calibrated Beckmann thermometer. The temperature was maintained constant to within 0.005-0.01°C.

We used helium, methane, and argon with a minimum purity of 99.97% in the experiments. The mixtures were prepared in a tilting mixer provided with an electric heater. Mixing was

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