

CRITICAL PARAMETERS: ALKANES AND CERTAIN COMPOUNDS CONTAINING  
CO GROUPS

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UDC 542.3

The input data in thermophysical calculations often includes critical parameters.

In [1] one finds the fullest survey of critical-parameter researchers up to the 1950s, but considerable progress has been made since that time because purer substances have been produced and new methods have been developed for measuring critical-state properties.

Critical properties for many organic compounds published up to 1968 have been examined in [2]; a review [3] contains data on inorganic substances appearing between 1951 and 1970. Studies published before 1951 were considered in [3] only if they did not appear in [1, 2]. In [4], critical parameters were surveyed for some hydrocarbons published in the thirteen years after the publication of [2]. Here we consider only studies that have made it necessary to reconsider the recommendations of [2-4] and some publications appearing in the 1970s not mentioned in [4].

There are two basic methods for measuring critical temperatures. The first is visual identification of the temperature at which the meniscus vanishes, the second is to examine the P-V-T curves in the critical region. One identifies zero-curvature isotherm points, and the least of these isotherms is taken as the critical one. Even precise P-V-T measurements leave it difficult to establish which isotherm corresponds to  $T_{CR}$ , and therefore the first method is preferable.

Preference is given to quantities found from the Calliet-Mathias diameter method for the critical density; this method has the exceptional advantage [4] that it is not necessary to make measurements directly around the critical point, where it is necessary to correct for gravitational forces and minor impurities as well as other minor effects having a substantial influence on the results.

If  $T_{CR}$  is determined by temperature identification, with  $\rho_{CR}$  calculated from the diameter method,  $P_{CR}$  can be derived by extrapolating the saturation vapor pressure to  $T_{CR}$  or from the critical isotherm in P- $\rho$  coordinates.

There is no generally accepted set of principles for selecting the best data; critical parameters are evaluated and chosen mainly from the accuracy and reliability.

For example, the quoted purity may be confirmed by the method used as well as by the number and form of the purification procedures. Purity is often evaluated indirectly from some property readily measured accurately such as the normal boiling point, refractive index, or density, which is compared with published values for high-purity specimens.

Substances with complicated structures such as members of homologous series can be evaluated also via certain empirical or semiempirical correlations.

Here we present a survey of critical parameters widely used in engineering calculations and theories for three inorganic substances containing carbon (carbon monoxide, carbon dioxide, and carbonyl sulfide), as well as for alkanes.

Carbon Monoxide. For more than 20 years there have been no papers on the critical parameters of this. Therefore, the measurements of [5] are traditionally given in reference works [1-3] and other sources appearing during that time.

Compressibility measurements at 100-300°K up to 35 MPa have been used [6] in thermodynamic parameter tables based on the literature; it was concluded that the published data require refinement. The  $T_{CR}$  of [5] given in the handbooks was reduced somewhat to give the best agreement with the P-V-T data, while  $P_{CR}$  was calculated with this  $T_{CR}$  from the measured and published saturation vapor pressures. The critical density was determined by matching the dens-

ity data for the saturated liquid to an analytic equation. The values proposed in [6] are the most reliable.

Carbon Dioxide. The published data have been examined comprehensively [7]; after that work of reference appeared, some other studies were published.

In [8],  $T_{CR}$  was determined by the meniscus method with an error of  $\pm 1$  mK. The reported error in measuring  $T_{CR}$  was related primarily to the high purity. The discrepancy between this  $T_{CR}$  [8] and other values did not exceed  $\pm 20$  mK when all the error sources were considered.

In [9], a specimen containing 95.6%  $^{13}\text{CO}_2$  was used in a closed vessel with critical filling ( $\rho_{CR} = 468 \text{ kg/m}^3$ ). The meniscus vanishing and the critical opalescence were observed with an error of  $\pm 0.05^\circ\text{K}$ . The temperature was measured to  $\pm 0.03^\circ\text{K}$  and was maintained to  $\pm 0.05^\circ\text{K}$  over a long period. The system was equilibrated for up to 10 h near the critical point.

The critical temperature was determined by the first method in [10]; the gas was carefully purified from water, air, and hydrocarbons, temperature-measurement error  $\pm 0.005^\circ\text{K}$ .

The isochoric specific heat was examined over a wide range near the critical point in [11] with an adiabatic calorimeter and discrete heat input by means of an apparatus described in detail in [12]. The temperature was measured with a platinum resistance thermometer calibrated on the IPTS-68 scale.

In [13], one finds critical parameters derived from gauge theory. Precision measurements [8] agree well with those of [10] and are the most accurate and are recommended in [14].

Carbonyl Sulfide. The method used in [15] was that pure carbonyl sulfide was placed in a constant-volume container fitted with a window, when the pressure was measured corresponding to preset temperatures while there was a visible meniscus between the vapor and liquid phases. The critical volume of COS had not previously been measured. The specimen was carefully purified and was analyzed with a gas chromatograph. The liquid contained 99.952% COS and small amounts of  $\text{CO}_2$ ,  $\text{N}_2$ , and  $\text{O}_2$ . The temperature was determined to  $\pm 0.04\%$  with a copper-constantan thermocouple, which was attached directly to the cell and was calibrated directly before the experiment by reference to the platinum resistance thermometer calibrated at the Canadian National Research Center. The pressure was measured with a piston gauge and load checked at the National Bureau of Standards (NBS) in the USA. The pressure-measurement error was  $\pm 0.5\%$ .

This study [15] has been the only one on the critical parameters of carbonyl sulfide in the last 30 years, when the results are quite accurate.

Methane. After the publication of a handbook [16] and a review [4], several new papers appeared on the critical point of methane.

In [17], the critical temperature was derived from the slope of the isochoric specific heat against the logarithmic temperature; the  $\rho$ - $T$  dependence was derived along the coexistence curve and from the isochoric specific heat over a wide density range, including the critical value. The makers' purity specification for the methane was 99.89%, and it contained 0.03% ethane and 0.08% nitrogen. The temperature was measured from quasistatic thermograms with an error or not more than  $0.0005^\circ\text{K}$ . The density-measurement error was in the range 0.05-0.10%. The critical density was determined at the critical temperature from the original  $\rho$ - $T$  data.

Measurements have been made [18] on the saturation-line density and isochoric specific heat with the calorimetric system described in [12]; the critical parameters were selected in accordance with the current theory on the basis of the original measurements on the isochoric specific heat in the critical region; the  $T_{CR}$  and  $\rho_{CR}$  from [17] and [18] were in good agreement and represent the most accurate values.

We recommend the critical pressure derived from the skeletal saturation vapor-pressure data [16] at the critical temperature.

Ethane. In [19], the behavior was examined on the coexistence curve in the critical region; the  $T_{CR}$  and  $\rho_{CR}$  derived in [20] were used in calculating the critical indices.

A method has been given [20] for determining  $\rho_{CR}$  based on the motion of the meniscus for  $(T - T_{CR})/T_{CR} \rightarrow 0$  at several densities; the critical density corresponds to the filling density for which the temperature line is a symmetry one. This requires measurement of the

meniscus-position temperature dependence for a series of densities, from which the symmetry line is derived. The slow-neutron transmission apparatus of [21] was used to measure the meniscus displacement from the center at various temperatures and average densities. A cascade thermostat stabilized the temperature to  $\pm 0.002^\circ\text{K}$ . The density was altered by allowing batches of gas to escape, whose volumes were determined by normal pressure. It was possible to measure  $\rho_{\text{CR}}$  to  $\pm 0.4 \text{ kg/m}^3$ . Chromatography showed the purity to be 99.96%. The critical temperature was determined by meniscus loss from neutron transmission as the beam was displaced from one phase to the other. The error in  $\rho_{\text{CR}}$  was estimated [19] at 0.1%.

In [10], the critical temperature was identified visually from meniscus loss at the middle of a glass tube; the specimen was carefully purified from water, air, and higher homologues, temperature-measurement error  $\pm 0.005^\circ\text{K}$ .

In [22, 23], it was concluded from measurements on the saturation-line density that the best results were obtained with the  $T_{\text{CR}}$  and  $\rho_{\text{CR}}$  from the skeletal tables [24] and from the recommendations of [25].

The critical pressure has been derived [24] from the equation for the saturation-vapor pressure at the given  $T_{\text{CR}}$  of [26]. The density was derived from the critical isotherms and the rectilinear-diameter method directly by the critical point. The data of [24, 25] are currently the most accurate.

Propane.  $T_{\text{CR}}$ ,  $P_{\text{CR}}$ , and  $\rho_{\text{CR}}$  have been measured [27] with the apparatus described in [28]; purity 99.99%, temperature error  $\pm 0.1^\circ\text{K}$ , pressure  $\pm 0.005 \text{ MPa}$ , density  $\pm 0.1\%$ .

The apparatus of [28] was used in [29] to determine the critical parameters; the makers' purity was 99.9%, and the specimen was further purified. A small amount of the material was placed in a thick-walled glass tube above mercury in a steam thermostat. Thermostatic accuracy  $\pm 0.01^\circ\text{K}$ . The pressure was measured with a piston gauge having a sensitivity of 0.3 kPa.

P-V-T measurements have been made [30] in the range 258.15-623.15 $^\circ\text{K}$  at 0-40 MPa by the pycnometer method. The makers' purity was 99.993%, and chromatographic analysis after further purification gave 99.998%.

The critical temperature was taken as the point on the P-V-T surface satisfying the condition that the first and second derivatives are zero:  $(\partial P/\partial V)_T = (\partial^2 P/\partial V^2)_T = 0$ .

The critical density was determined from the diameter rule with this  $T_{\text{CR}}$ ; the critical pressure was derived from the P-V-T measurements at the chosen  $T_{\text{CR}}$  and  $\rho_{\text{CR}}$ .

Critically evaluated measurements have been used [31, 32] in novel equations, which were employed in thermodynamic and critical parameter tables.

Measurements have been made on the isochoric specific heat [12] in the two-phase and single-phase ranges, including the critical region; the makers' purity was 99.98%. The temperatures were measured with a platinum resistance thermometer calibrated on IPTS-68 with an error of  $\pm 0.01^\circ\text{K}$ . The critical temperature corresponded to the maximum specific heat in the two-phase range.

The values chosen for  $T_{\text{CR}}$  in [33, 34] were identical with that obtained in [30];  $P_{\text{CR}}$  was calculated from the equations for the vapor pressure and matched to the P-V-T data. The critical density in [34] was taken directly from [30], while that in [33] was derived from the orthobaric densities of [30]. The data of [30] are the most accurate.

Butane. The  $T_{\text{CR}}$  of [36] has been used [35] to derive  $P_{\text{CR}}$  by extrapolating the original saturation vapor-pressure measurements.

The  $T_{\text{CR}}$  selected from [38] has been used in [37] in deriving  $P_{\text{CR}}$  and  $\rho_{\text{CR}}$  giving the best match to the orthobaric densities, vapor pressure, and P-V-T data.

Critically evaluated measurements have given [39] and [40] equations from which tables have been compiled for the thermodynamic parameters in a state of saturation and in the single-phase region, including the critical point. Critical parameters were recommended whose source was not given. The data of [37] are the most accurate.

Isobutane. Critical-point parameters are quoted in [41] as derived in [42] from gauge-transformation theory via the data of [43].

The results of [42] were revised somewhat in [44], although it was observed there that the theory does not give a very accurate critical temperature, so  $T_{cr}$  was measured by the meniscus method, while the critical density was determined by weighing. The makers' purity was 99.9%. The amount was determined by weighing the cell before and after filling with a balance having an accuracy of 0.5 mg. The temperature was measured with a platinum resistance thermometer calibrated at NBS in the USA to  $\pm 0.01^\circ\text{K}$ .

The result from  $T_{cr}$  agreed well with gauge theory; the critical pressure was considered as an adjustable parameter in fitting to the P-V-T data. Table 1 gives the results; these measurements are the most accurate.

Pentane. Phase equilibria in the system formed by n-pentane with pent-1-ene have been considered [45], where the critical parameters for pure n-pentane were determined by the meniscus method.

The makers' purity was 99.9%, and further purification was provided by passing the gas through a molecular sieve to remove traces of water and air. The temperature was measured with an iron-constantan thermocouple calibrated from a tested resistance thermometer to  $\pm 0.01^\circ\text{K}$ , while the pressure was measured with a precision spring gauge to  $\pm 3.4$  kPa. The critical density was determined from the diameter law. These data are the most reliable.

Hexane. The meniscus method has been used [46] to determine the critical parameters; the makers' purity was 99.96%. The temperature was measured with a copper-constantan thermocouple to  $\pm 0.1^\circ\text{K}$ , and the pressure with a piston gauge to  $\pm 0.002$  MPa. The critical density was derived from the diameter law.

The apparatus described in [28] was used in [27]; purity 99.96%, temperature-measurement error  $\pm 0.1^\circ\text{K}$ , pressure  $\pm 0.005$  MPa, density  $\pm 0.1\%$ .

Comprehensive measurements have been made [47-49] on the properties of n-hexane over wide temperature and pressure ranges, including the critical region. These measurements and published data were used in skeletal tables. The critical temperature was taken as the value from [50], which is recommended in several handbooks. The critical pressure was derived by extrapolating the measurements on vapor pressure to that temperature. The critical density was determined from the P-V-T measurements in two ways: by the Planck-Gibbs method and from the rectilinear diameter, the results agreeing within 0.2%.

The recommended  $T_{cr}$  and  $P_{cr}$  in [51] coincide with the values from [2].

The data of [42-44] are the most reliable and have been certified by the State Standard Commission for the category of standard reference data.

2-Methylpentane. Measured critical parameters given in [27] are close to ones recommended in the literature; the method and apparatus used in [27] have been described in [28]. Purity 99.83%. Temperature-measurement error  $\pm 0.1^\circ\text{K}$ , pressure  $\pm 0.005$  MPa, density  $\pm 0.1\%$ . These data are fairly reliable.

Heptane. The critical temperature adopted in [52] was used in [4]. This value represents an underestimate in relation to virtually all measured critical temperatures for n-heptane. In [53], which dealt with the properties of heptane-propyl alcohol mixtures, measurements were reported on the critical parameters of pure heptane on the apparatus described in [54]. The heptane of initial purity 99.99% was additionally purified.

Critical-temperature measurements for other homologues made by reliable methods indicate that the most accurate value was obtained in [55], which virtually coincides with the results of [56] and [57] and which is recommended in many reference publications such as [2, 51].

The critical pressures found in [56, 58, 59] by different methods are virtually the same; they are recommended in works of reference.

The critical density has been measured directly [52] and is the most accurate.

Heavy Alkanes. Critical temperatures and pressures have been given [60] for n-alkanes containing from 5 to 50 C atoms; in particular, these values for the  $C_5$ - $C_{10}$  n-alkanes agree with the measurements of [55].

A study has been made [61] on how the solubility of mercury used as a separator affects the critical parameters for high-boiling n-alkanes; a modified apparatus was used [62], purity 99.7%. Further purification consisted in removing traces of water by filtration through

TABLE 1. Critical Parameters

Substance	Ref.	$T_{cr}$ , K	$P_{cr}$ , MPa	$\rho_{cr}$ , kg/m <sup>3</sup>
CO	[3]	132,92	3,499	301
	[6]	132,75	3,477	306
CO <sub>2</sub>	[3]	304,2	7,38	468
	[8]	304,127	7,3753	467,8
	[7]	304,19	7,383	468
	[9]	304,13	—	—
	[10]	304,16	—	—
	[13]	304,13	7,375	467
	[11]	304,126	—	467,4
COS	[14]	304,13	7,3753	467,8
	[3]	375	5,9	440
CH <sub>4</sub>	[15]	378,8	6,349	445
	[16]	190,77	4,627	163,5
	[4]	190,53	4,595	162,8
C <sub>2</sub> H <sub>6</sub>	[17]	190,66	—	161
	[18]	190,663	—	161
	[4]	305,36	4,873	204,5
	[10]	305,345	—	—
	[19]	305,347	—	205,8
C <sub>3</sub> H <sub>8</sub>	[25]	305,33	4,8714	204,5
	[22, 23]	305,33	—	204,5
	[4]	369,78	4,246	220,1
	[27]	369,72	4,26	—
	[29]	370,02	4,260	223,2
	[30]	369,85	4,24709	218,5
	[11, 31, 32]	369,9	4,242	220,0
	[33]	369,85	4,24746	220,5
	[34]	369,85	4,2460	218,5
	[4]	425,09	3,790	228,7
C <sub>4</sub> H <sub>10</sub>	[35]	425,14	3,78385	—
	[37]	425,16	3,7960	227,85
	[39]	425,16	3,798	225
	[40]	425,16	3,798	225
	[4]	407,95	3,640	226,4
<i>i</i> -C <sub>4</sub> H <sub>10</sub>	[41]	407,851	3,6306	227,0
	[44]	407,865	3,6306	226,85
	[44]	407,84	3,629	225,5
C <sub>5</sub> H <sub>12</sub>	[4]	469,62	3,363	232
	[45]	469,8	3,38	231
C <sub>6</sub> H <sub>14</sub>	[46]	507,86	—	235
	[4]	507,54	3,028	233
	[27]	507,95	3,017	233
	[49]	507,85	3,058	233,6
	[48]	507,85	3,0576	233,6
	[51]	507,3	3,015	—
<i>i</i> -C <sub>6</sub> H <sub>14</sub>	[2]	497,45	3,010	235
	[27]	497,85	3,031	235
	[55]	540,28	—	—
C <sub>7</sub> H <sub>16</sub>	[2]	540,2	2,736	232
	[4]	539,86	2,721	234
	[53]	540,68	2,775	235,6
	[51]	540,2	2,730	—
<i>n</i> -C <sub>8</sub> H <sub>18</sub>	[60]	568,56	2,497	—
	[4]	568,76	2,486	232
	[63]	568,80	—	—
<i>n</i> -C <sub>9</sub> H <sub>20</sub>	[60]	594,56	2,28	—
	[61]	595,85	2,2911	234
	[63]	593,61	—	—
<i>n</i> -C <sub>10</sub> H <sub>22</sub>	[60]	617,55	2,10	—
	[63]	617,54	—	—

silica gel. Strong light scattering was observed, as is characteristic of the critical state. The temperature and pressure were read when the critical opalescence occurred. The temperature was measured with a Chromel-constantan thermocouple calibrated from a platinum resistance thermometer checked at NBS, while the pressure was measured with a precision Bourdon gauge having an error of  $\pm 1.72$  kPa.

In [63], the critical temperatures for *n*-octane and higher homologues were measured by the first method on sealed tubes filled with the material and heated until opalescence occurred. Purity of *n*-octane specimen 99.85%, *n*-nonane 99.85%, *n*-decane 99.49%.

There are differences of 2°K in the critical temperatures for *n*-nonane measured by different workers, which is more than the experimental error, and this applies also to some recent studies [61] and [63].

TABLE 2. Recommended Critical Parameters

Substance	$T_{cr}$ , K	$\Delta T_{cr}$ , K	$P_{cr}$ , MPa	$\Delta P_{cr}$ , MPa	$\rho_{cr}$ , kg/m <sup>3</sup>	$\Delta \rho_{cr}$ , kg/m <sup>3</sup>
Carbon monoxide	132,75	0,05	3,477	0,005	306	3
Carbon dioxide	304,13	0,005	7,375	0,005	467,8	1,5
Carbonyl sulfide	378,8	0,1	6,349	0,005	445	2
Methane	190,66	0,01	4,61	0,005	161	1
Ethane	305,33	0,02	4,871	0,005	204,5	0,5
Propane	369,85	0,05	4,247	0,001	220,5	0,7
n-Butane	425,16	0,05	3,796	0,002	227,9	0,5
i-Butane	407,84	0,05	3,629	0,002	225,5	0,5
n-Pentane	469,8	0,05	3,38	0,01	231	1
n-Hexane	507,85	0,05	3,06	0,02	233,6	1,5
i-Hexane	497,85	0,05	3,03	0,02	235	3
n-Heptane	540,28	0,05	2,736	0,005	234	2
n-Octane	568,8	0,1	2,50	0,01	235	3
n-Nonane	593,6	0,1	2,29	0,01	235,5	2
n-Decane	617,5	0,1	2,10	0,01	236	2

The critical parameters of n-octane, n-nonane, and n-decane show that reliable critical temperatures were obtained in [63], and the same applies to the critical pressures in [4, 61, 60] correspondingly. As there are no reliable critical-density measurements for these alkanes, we selected values on the basis of the data for entire homologous series.

Table 2 gives recommended critical parameters and error limits; the errors are usually based on the accuracy in the original papers and the discrepancies between the different sources.

Those recommended values can be used in theoretical and engineering treatments.

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