International Journal of Thermophysics, Vol. 16, No. 2, 1995

A Thermodynamic Property Formulation for Cyclohexane[1]

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A formulation for the thermodynamic properties of cyclohexane is presented. The equation is valid for single-phase and saturation states from the melting line to 700 K at pressures up to 80 MPa. It includes a fundamental equation explicit in reduced Helmholtz energy with independent variables of reduced density and temperature. The functional form and coefficients of the ancillary equations were determined by weighted linear regression analyses of evaluated experimental data. An adaptive regression algorithm was used to determine the final equation. To ensure correct thermodynamic behavior of the Helmholtz energy surface, the coefficients of the fundamental equation were determined with multiproperty fitting. Pressure–density–temperature (P–ρ–T) and isobaric heat capacity (C_p–P–T) data were used to develop the fundamental equation. Saturation P–ρ–T values, calculated from the estimating functions, were used to ensure thermodynamic consistency at the vapor–liquid phase boundary. Separate functions were used for the vapor pressure, saturated liquid density, saturated vapor density, ideal-gas heat capacity, and pressure on the melting curve. Comparisons between experimental data and values calculated using the fundamental equation are given to verify the accuracy of the formulation. The formulation given here may be used to calculate densities within ±0.1%, heat capacities to within ±2%, and speed of sound to within ±1%, except near the critical point.

KEY WORDS: cyclohexane; equation of state; thermodynamic properties.

1. INTRODUCTION

The design of engineering systems requires the knowledge of the thermodynamic properties of the working fluids in the system. In modern

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[1] Paper presented at the Twelfth Symposium on Thermophysical Properties, June 19–24, 1994, Boulder, Colorado, U.S.A.

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engineering practice it is desirable to have these properties computer-accessible for modeling or simulation work. This paper presents a preliminary thermodynamic property formulation for the fluid cyclohexane (C_6H_{12}) on the International Temperature Scale of 1990 [1].

Throughout this paper the word "formulation" refers to the equations required to calculate the thermodynamic properties of the fluid. All thermodynamic properties may be calculated by differentiating the fundamental equation with respect to the appropriate variables.

The formulation presented for cyclohexane is valid in the single-phase regions from the melting line to 700 K with pressures to 80 MPa. Ancillary functions for vapor pressure, saturated liquid density, and saturated vapor density are provided as estimating functions for iterative computer programs which are used to calculate properties of cyclohexane with various pairs of independent variables. Saturation properties can be calculated with the Maxwell criterion or by simultaneous solution of the vapor pressure equation with the equation of state. The method of simultaneous solution produces independent saturation properties which are not thermodynamically consistent; however, the calculated saturation properties are within $\pm 0.3\%$ of the Maxwell construction values.

2. EXPERIMENTAL DATA FOR CYCLOHEXANE

The available experimental data for cyclohexane are summarized in Tables I and II. Table I lists the single-phase data for cyclohexane. Vapor pressure, saturation densities, melting line, ideal-gas heat capacity, second virial coefficient, and saturated liquid heat capacity data are given in Table II. Not all of the data listed were used in the determination of the coefficients of the fundamental equation and ancillary functions; however, all data were compared to values calculated with the formulation. Those data used in the development of the fundamental equation are indicated with an asterisk in Table I.

The critical parameters are used as reducing parameters in the dimensionless fundamental equation and various ancillary functions. The selected critical parameters for this formulation correspond to the critical temperature reported by Hugill and McGlashan [32] and the critical pressure of Stryjek and Vera [68]. The selected value for the critical density was reported by Kobe and Lynn [69]. In the linear least-squares regression, the equation of state was constrained to pass through this point. The selected values, converted to a consistent set of units, are $P_c = 4.075$ MPa, $T_c = 553.60$ K, and $\rho_c = 3.244\ \mathrm{mol \cdot dm^{-3}}$. The triple-point temperature and pressure of cyclohexane are reported by Aston et al. [26] as $P_t = 5.388$ kPa

Table I. Experimental Single-Phase Data for Cyclohexane[a]

Ref.	Year	No. of points	Pressure range (MPa)	Temperature range (K)
P–ρ–T data				
* Bich *et al.* [2]	1984	40	0.06–0.19	372–622
* Grigor'ev *et al.* [3]	1975	119	2–79	498–698
Kerimov and Apaev [4]	1974	543	0.1–69	293–748
Kerimov and Apaev [5]	1973	717	0.1–69	283–748
Kerns [6]	1972	92	0.01–2.8	423–523
Kuss and Taslimi [7]	1970	10	0.1–118	298–353
* Rastorguev *et al.* [8]	1975	136	0.1–85	298–473
Reamer and Sage [9]	1957	78	0.4–69	311–511
Sun *et al.* [10]	1987	88	0.1–85	288–323
Sun *et al.* [11]	1988	12	0.1	282–336
Wisotzki and Würflinger [12]	1982	33	0.1–110	287–338
Overall		1868	0.01–118	282–748
C_v–ρ–T data				
Asenbaum and Wilhelm [13]	1982	7	8.7–9.4	283–343
Moelwyn-Hughes and Thorpe [14]	1963	36	8.8–9.4	280–333
Overall		43	8.7–9.4	280–343
C_p–P–T data				
McCullough *et al.* [15]	1951	4	0.04–0.08	371–469
Montgomery and De Vries [16]	1942	3	0.1	370–410
Murdaev [17]	1980	44	5–50	323–533
Rastorguev *et al.* [18]	1976	253	4.5–50	547–669
Safir [19]	1978	7	0.10	299–312
Safir and Grigor'ev [20]	1979	10	4.5–10	561–641
* Safir *et al.* [21]	1975	120	0.5–50	295–541
* Safir and Grigor'ev [22]	1976	91	0.5–4	418–675
Spitzer and Pitzer [23]	1946	8	0.03–0.1	384–544
Sun *et al.* [10]	1987	88	0.1–85	288–323
Overall		628	0.03–85	280–675
w–P–T (speed of sound) data				
Kiyohara *et al.* [24]	1978	1	0.1	298–298
Sun *et al.* [10]	1987	88	0.1–85	288–323
Takagi [25]	1976	39	0.1–107	283–333
Overall		128	0.1–107	283–333

[a] Asterisk indicates data used in development of the fundamental equation.

Table II. Vapor–Liquid Equilibrium, Melting Line, Isobaric Ideal-Gas Heat Capacity, and Second Virial Coefficient Data for Cyclohexane

Ref.	Date	No. of points	Temperature range (K)
Vapor pressure data			
Aston *et al.* [26]	1943	5	279–294
Brown and Ewald [27]	1950	6	303–354
Cruickshank and Cutler [28]	1967	30	298–354
Dejardin [29]	1919	15	297–354
Ewing [30]	1974	5	288–328
Glaser and Ruland [31]	1957	19	354–552
Hugill and McGlashan [32]	1978	12	451–554
Kay and Albert [33]	1956	4	435–550
Kerns [6]	1974	9	323–523
Palczewska-Tulinska *et al.* [34]	1983	9	300–353
Reamer and Sage [9]	1957	7	311–511
Rotinjanz and Nagornow [35]	1934	7	280–289
Scatchard *et al.* [36]	1939	6	303–353
Scatchard *et al.* [37]	1939	6	303–353
Schmitt [38]	1934	8	302–354
Shibata and Sandler [39]	1989	2	366–411
Weclawski and Bylicki [40]	1983	15	298–349
Willingham *et al.* [41]	1945	17	293–355
Young *et al.* [42]	1977	4	298–313
Overall		186	279–554
Saturated liquid density data			
Asenbaum and Wilhelm [43]	1982	7	283–343
French [44]	1983	5	288–318
Karvo [45]	1980	4	303–333
Kurumov *et al.* [46]	1977	9	280–349
Massart [47]	1936	6	288–372
Reamer and Sage [9]	1957	7	311–511
Rotinjanz and Nagornow [35]	1934	21	280–553
Shibata and Sandler [39]	1989	2	366–411
Sun *et al.* [11]	1987	12	282–336
Wilhelm *et al.* [48]	1968	3	293–313
Overall		76	280–554
Saturated vapor density data			
Shibata and Sandler [39]	1989	2	366–411
Young and Fortey [49]	1899	21	363–547
Overall		23	363–547

Table II. *(Continued).*

Ref.	Date	No. of points	Temperature range (K)
Melting line data			
Deffet [50]	1935	11	280–328
Sun *et al.* [51]	1987	22	280–323
Takagai [25]	1976	7	280–333
Wisotzki and Würflinger [12]	1982	2	279–330
Overall		42	279–333
Ideal-gas C_p values			
Beckett *et al.* [52]	1947	14	298–1501
Dorofeva *et al.* [53]	1986	16	100–1500
Montgomery and De Vries [16]	1942	3	370–410
Spitzer and Pitzer [23]	1947	6	384–544
Overall		39	100–1501
Second virial coefficient data			
Bich *et al.* [2]	1984	11	373–623
Bottomley and Coopes [54]	1962	3	308–343
Bottomley and Remmington [55]	1953	4	295–308
Brostow [56]	1978	4	323–400
Cox and Stubley [57]	1969	1	373
Grigor'ev *et al.* [58]	1983	10	473–698
Hajjar *et al.* [59]	1969	11	317–473
Karvo [45]	1980	4	303–333
Lambert *et al.* [60]	1949	12	324–404
Rowlinson [61]	1969	5	327–405
Waelbroeck [62]	1955	9	315–348
Overall		74	295–698
Saturated liquid C_p and C_v data			
Aston *et al.* [26] (C_p)	1943	4	282–292
Dejardin [29] (C_p)	1919	4	296–323
Moelwyn-Hughes and Thorpe [14] (C_p)	1963	4	298–328
Parks *et al.* [63] (C_p)	1930	5	283–291
Ruehrwein and Huffman [64] (C_p)	1943	7	282–301
Vesley *et al.* [65] (C_p)	1979	3	298–318
Wilhelm *et al.* [48] (C_p)	1968	3	293–313
Wilhelm *et al.* [48] (C_v)	1968	3	293–313
Overall		33	282–328
Saturated liquid speed-of-sound data			
Nath and Narain [66]	1982	2	293–303
Nath and Dixit [67]	1984	2	298–308
Overall		4	293–308

and $T_t = 279.47$ K. The molecular weight of cyclohexane is $84.1608\ \mathrm{g \cdot mol^{-1}}$ and the universal gas constant used in this work is $8.31434\ \mathrm{J \cdot mol^{-1} \cdot K^{-1}}$.

3. ANCILLARY FUNCTIONS FOR CYCLOHEXANE

Ancillary functions for the vapor pressure, saturated liquid density, and saturated vapor density have been developed. The vapor pressure and orthobaric density equations were determined by linear least-square fits to selected data. The functional form of the vapor pressure equation used in this work is given by

$$\ln\left(\frac{P}{P_c}\right) = \left(\frac{T_c}{T}\right)(N_1\theta + N_2\theta^{1.5} + N_3\theta^{3.5} + N_4\theta^{7} + N_5\theta^{10.5}) \tag{1}$$

where $\theta = 1 - T/T_c$. The experimental data selected for the development of this equation span the temperature range from 293 to 553 K. The coefficients of Eq. (1) determined from these data are listed in Table III.

The saturated liquid density, ρ', of cyclohexane can be represented by

$$\frac{\rho'}{\rho_c} - 1 = N_1\theta^{1/3} + N_2\theta^{2/3} + N_3\theta^{7/3} + N_4\theta^{20/3} \tag{2}$$

where $\theta = 1 - T/T_c$. The data used in the determination of the coefficients and functional form of this equation cover the temperature range from 293 to 511 K. The coefficients of Eq. (2) are listed in Table III.

There are very few data for the saturated vapor phase of cyclohexane. Therefore, pseudodata were calculated from the triple point to the critical point. In the range from the triple point to 450 K, the saturated vapor density was determined by simultaneously solving the vapor pressure equation with a previously developed virial equation of state for cyclohexane. In the range from 450 K to the critical point, the critical liquid volume fraction method of Van Poolen et al. [70] was used. The estimated saturated vapor density ρ'' can be represented by,

$$\ln\left(\frac{\rho''}{\rho_c}\right) = N_1\theta^{1/3} + N_2\theta^{2/3} + N_3\theta^{5/3} + N_4\theta^{8/3} + N_5\theta^{22/3} \tag{3}$$

where $\theta = 1 - T/T_c$ and the coefficients are given in Table III.

Table III. Coefficients of the Ancillary Functions for Cyclohexane

Vapor pressure equation, Eq. (1)
$N_1 = -7.013076840$
$N_2 = 1.605995414$
$N_3 = -2.145174141$
$N_4 = -16.21218440$
$N_5 = 69.77103310$

Saturated liquid density equation, Eq. (2)
$N_1 = 1.744987527$
$N_2 = 0.6809805909$
$N_3 = 0.4994644939$
$N_4 = -0.8916325138$

Saturated vapor density equation, Eq. (3)
$N_1 = 0.4657794656$
$N_2 = -8.407267114$
$N_3 = 6.351041449$
$N_4 = -25.81512773$
$N_5 = -65.13864581$

The solid–liquid equilibrium curve was represented with an equation determined by Wisotzki and Würflinger [12]. This equation is reported as

$$\frac{P}{a} = \left(\frac{T}{T_0}\right)^c - 1 \tag{4}$$

where $a = 383.4$ MPa^{-1}, $c = 1.41$, and $T_0 = 279.7$ K.

4. THE FUNDAMENTAL EQUATION FOR CYCLOHEXANE

The functional form of the fundamental equation used for this formulation was developed by Jacobsen et al. [71]. This equation is a potential function explicit in dimensionless Helmholtz energy with reduced density and reciprocal reduced temperature as the independent variables. The form of this equation is

$$\alpha(\delta, \tau) = \alpha^0(\delta, \tau) + \bar{\alpha}(\delta, \tau) \tag{5}$$

where $\delta = \rho/\rho_c$, $\tau = T_c/T$, and $\alpha = A/RT$. The superscript zero signifies the ideal-gas contribution and the overbar indicates the real-fluid contribution.

The ideal-gas Helmholtz function is given by

$$\alpha^0(\delta, \tau) = \frac{H_0^0 \tau}{RT_c} - \frac{S_0^0}{R} - 1 + \ln\frac{\delta \tau_0}{\delta_0 \tau} - \frac{\tau}{R}\int_{\tau_0}^{\tau} \frac{C_p^0}{\tau^2}\,d\tau + \frac{1}{R}\int_{\tau_0}^{\tau} \frac{C_p^0}{\tau}\,d\tau \tag{6}$$

Table IV. Coefficients of the Ideal-Gas Isobaric Heat Capacity Equation for Cyclohexane, Eq. (7)

$N_1 = -56214088$

$N_2 = 9.368327211$

$N_3 = 0.01526155409$

$N_4 = -0.000003635246755$

$N_5 = 23.76658940$

$N_6 = 2000$

where $\delta_0 = \rho_0/\rho_c$ and $\tau_0 = T_c/T_0$. The value of ρ_0 is calculated as the ideal-gas density at $T_0 = 279.47$ K, the triple-point temperature, and $P_0 = 0.101325$ MPa. The values of H_0^0 and S_0^0 were arbitrarily determined such that the values of enthalpy and entropy were zero for saturated liquid at the triple-point temperature. The resulting values for this datum state are $H_0^0 = 33884.8\ \mathrm{J \cdot mol^{-1}}$ and $S_0^0 = 96.612\ \mathrm{J \cdot mol^{-1} \cdot K^{-1}}$.

In order to evaluate Eq. (6), an equation for the ideal-gas isobaric heat capacity C_p^0 is required. For our work, the function

$$\frac{C_p^0}{R} = \frac{N_1}{T^3} + N_2 + N_3 T + N_4 T^2 + N_5 \left[\frac{u^2 e^u}{(e^u - 1)^2} \right] \tag{7}$$

was used. In Eq. (7), $u = N_6/T$. We arbitrarily assigned N_6 a value of 2000. The data used to determine the remaining coefficients cover a range from 298 to 1500 K. The coefficients of Eq. (7), determined by a least-squares fit to the data, are given in Table IV.

The real-fluid compressibility contribution to the fundamental equation is given by

$$\bar{\alpha}(\delta, \tau) = \sum_{k=1}^{m} N_k \delta^{i_k} \tau^{j_k} \exp(-\gamma \delta^{l_k}) \tag{8}$$

where the N_k values are the coefficients of the fundamental equation and γ is a constant which has a value of zero when l_k is zero, or one when l_k is greater than zero. The values of i_k, j_k, and l_k are arbitrary; however, it is generally expected that i_k is an integer, j_k is a real number, and l_k is an integer greater than or equal to zero. The values of N_k, i_k, j_k, and l_k determined for cyclohexane are given in Table V.

The least-squares algorithm used to determine the N_k values of Eq. (8) is a linear stepwise regression incorporating a search procedure based on statistical parameters monitored during the fitting procedure. This algorithm was developed by Wagner [72] and further modified for equations of state by de Reuck and Armstrong [73]. It is capable of simultaneously

Table V. Coefficients and Exponents of the Fundamental Equation for Cyclohexane, Eq. (8)

k	i	j	l	N
1	1	0.0	0	0.8425412659
2	1	1.5	0	−3.138388327
3	1	2.5	0	1.679072631
4	2	1.5	0	−0.1538192490
5	3	1.0	0	0.1984911143
6	3	2.5	0	−0.1445325940
7	7	2.0	0	$0.3746346428 \times 10^{-3}$
8	1	5.0	2	−0.6427428062
9	1	6.0	2	0.2280757615
10	2	5.5	2	−1.868116802
11	3	3.0	2	−1.028243711
12	3	7.0	2	0.5821457418
13	5	6.0	2	−0.2558911520
14	8	6.5	2	$0.1276844113 \times 10^{-1}$
15	10	5.5	2	$-0.5158613166 \times 10^{-2}$
16	3	11.0	4	$0.6334794755 \times 10^{-1}$
17	4	11.0	4	$-0.6014686589 \times 10^{-1}$
18	6	0.5	0	$0.1861479616 \times 10^{-3}$
19	6	3.0	0	$0.1745721652 \times 10^{-3}$
20	1	0.5	3	0.4439056828
21	1	1.0	3	−0.6264920642
22	2	4.0	2	2.132589969
23	2	4.0	6	$-0.3620300991 \times 10^{-2}$
24	4	1.5	2	0.2534453992
25	4	2.0	4	$0.1669144715 \times 10^{-1}$
26	8	0.5	2	$0.3985052291 \times 10^{-2}$

fitting several data forms. For cyclohexane, the final fundamental equation was determined by simultaneously fitting selected P–ρ–T data, calculated isochoric heat capacity data, and saturation pseudodata for the Maxwell criterion. The C_p–P–T data and a preliminary equation of state were used to estimate values of C_v–ρ–T for use in the linear least-squares regression procedure used to develop the fundamental equation. The transformation of C_p–P–T data into C_v–ρ–T data was done prior to each regression using the most recently correlated equation of state.

An initial bank of 75 combinations of the exponents i_k, j_k, and l_k for Eq. (8) were selected as initial input information to the regression algorithm. The statistical search procedure selected only those terms deemed statistically significant in the representation of the thermodynamic surface with the given input data sets. The final fundamental equation for cyclohexane contained 26 terms.

The accuracy of the fundamental equation was determined by statistical comparisons of property values calculated with the equation of state to experimental data. The statistics used in these comparisons are the absolute average deviation (AAD), the bias (BIAS), the standard deviation (SDV), and the root-mean-square deviation (RMS). These statistics are based on percent deviation in any property X, defined as

$$\%\Delta X = 100\left(\frac{X_{\text{data}} - X_{\text{calc}}}{X_{\text{data}}}\right) \tag{9}$$

Using this definition, the statistics above are defined as

$$\text{AAD} = \frac{1}{n}\sum_{i=1}^{n} |\%\Delta X_i| \tag{10}$$

$$\text{BIAS} = \frac{1}{n}\sum_{i=1}^{n} (\%\Delta X_i) \tag{11}$$

$$\text{SDV} = \left[\frac{1}{n-1}\sum_{i=1}^{n} (\%\Delta X_i - \text{BIAS})^2\right]^{1/2} \tag{12}$$

$$\text{RMS} = \left[\frac{1}{n}\sum_{i=1}^{n} (\%\Delta X_i)^2\right]^{1/2} \tag{13}$$

The results of the statistical comparisons are shown in Table VI. The overall statistics for all reported experimental data are shown. In the development of the fundamental equation, a subset of the complete P–ρ–T data set was used. The statistical comparisons for only these fitted data are also shown in Table VI. No C_v–ρ–T, w–P–T, or second virial data were used in the determination of the fundamental equation. The C_p–P–T data

Table VI. Statistical Comparisons of Property Values Calculated with the Equation of State for Cyclohexane to Experimental Data[a]

Data type	Deviation in	Number of points	AAD	BIAS	SDV	RMS	Max AAD	ADD > 10%
Fitted P–ρ–T	ρ	295	0.066	−0.008	0.299	0.298	−4.276	0
All P–ρ–T	ρ	1868	0.226	−0.007	0.547	0.547	−7.325	0
C_v–ρ–T	C_v	47	2.310	1.460	3.105	3.396	9.904	7
C_p–P–T	C_p	662	2.398	1.675	2.566	3.062	−9.812	44
w–P–T	w	136	1.105	−0.165	1.392	1.396	−4.819	0
Second virial[b]	B	74	0.088	−0.079	0.097	0.125	−0.354	0

[a] See text for abbreviations.

[b] Second virial values are based on differences ($\text{dm}^3 \cdot \text{mol}^{-1}$) rather than percent deviation.

(transformed into C_v–ρ–T data) were used, except in the critical region. The number of data points that exhibit an AAD greater than 10% are indicated in the last column of Table VI.

5. CONCLUSION

We estimate that the fundamental equation has an uncertainty of ±0.1% in density, ±2.5% in heat capacity, and ±1% in speed of sound and that calculated enthalpy and entropy values have uncertainties of ±1% from the melting line to 700 K at pressures up to 80 MPa.

The fundamental equation is not valid in the critical region. This region is bounded by isotherms ±5% of T_c and isochores ±25% of ρ_c. For cyclohexane, the critical region is bounded by $256 < T_c < 581$ K and $2.4 < \rho_c < 4.1$ mol · dm^{-3}.

The saturation values can be calculated from the fundamental equation by invoking the Maxwell equal-area principle. The simultaneous solution of the vapor pressure equation with the equation of state produces results that are within ±0.3% of the Maxwell construction over the entire saturation line. Therefore, either method is viable for the determination of saturation properties based upon the probable uncertainties of the measured and calculated values used to define these states in the least-squares fit. As mentioned previously, the Maxwell construction is the preferred method for determination of saturation properties which are thermodynamically consistent.

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