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A Thermodynamic Property Formulation for Ethylene from the Freezing Line to 450 K at Pressures to 260 MPa[1]

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A new thermodynamic property formulation based upon a fundamental equation explicit in Helmholtz energy of the form $A = A(\rho, T)$ for ethylene from the freezing line to 450 K at pressures to 260 MPa is presented. A vapor pressure equation, equations for the saturated liquid and vapor densities as functions of temperature, and an equation for the ideal-gas heat capacity are also included. The fundamental equation was selected from a comprehensive function of 100 terms on the basis of a statistical analysis of the quality of the fit. The coefficients of the fundamental equation were determined by a weighted least-squares fit to selected P–ρ–T data, saturated liquid and saturated vapor density data to define the phase equilibrium criteria for coexistence, C_v data, velocity of sound data, and second virial coefficients. The fundamental equation and the derivative functions for calculating internal energy, enthalpy, entropy, isochoric heat capacity (C_v), isobaric heat capacity (C_p), and velocity of sound are included. The fundamental equation reported here may be used to calculate pressures and densities with an uncertainty of ±0.1%, heat capacities within ±3%, and velocity of sound values within ±1%, except in the region near the critical point. The fundamental equation is not intended for use near the critical point. This formulation is proposed as part of a new international standard for thermodynamic properties of ethylene.

KEY WORDS: equation of state; ethylene; saturation properties; thermodynamic properties.

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1. INTRODUCTION

The commercial importance of ethylene is well established. Recent efforts to improve both experimental measurements and correlation techniques have resulted in the development of an accurate thermodynamic property formulation for ethylene. This work has been a joint effort of the Center for Applied Thermodynamic Studies at the University of Idaho in Moscow and the Thermophysics Division of the National Bureau of Standards in Boulder, Colorado [1]. A companion formulation [2] on the thermodynamic properties of ethylene in the critical region has been completed as a joint effort of the Thermophysics Division of the National Bureau of Standards in Gaithersburg, Maryland, and the Institute for Physical Science and Technology at the University of Maryland.

Throughout this paper the word "data" is used to refer to experimental measurements. The term "formulation" refers to the equation or equations necessary to calculate fluid properties. The term "fundamental equation" is used to describe the equation, $A = A(\rho, T)$, used in this work. The ideal-gas heat capacity representation is an integral part of the fundamental equation. Ancillary equations for the vapor pressure, saturated liquid density, and saturated vapor density are used to calculate values to define liquid–vapor coexistence states and to estimate properties of saturated states in calculations applying the Maxwell criterion for phase equilibrium.

Discussions of prior correlations, experimental data sources, techniques of data analysis and correlation, and comparisons to experimental data are included in Ref. 1. This paper presents a wide-range fundamental equation explicit in Helmholtz energy and the associated ancillary equations in a form suitable for energy analysis of engineering systems. The reader who is interested in more detail about the correlation is referred to Ref. 1. The fixed points for ethylene used in this work are given in Table I. Sources and details of selection of these values are given in Ref. 1.

2. THE FUNDAMENTAL EQUATION

The fundamental equation used in this work is explicit in reduced Helmholtz energy, and other thermodynamic properties are derived from it by differentiation. The coefficients of the fundamental equation were determined using a least-squares regression procedure [3, 4] for selection of an optimum group of terms from an initial bank of 100 proposed terms. The range of validity of the fundamental equation for ethylene is from the freezing line to 450 K at pressures to 260 MPa. With a few exceptions, the equation presented here represents the selected experimental P–ρ–T data to within the estimated accuracies of these data.

Table I. Fixed Points for Ethylene

Symbol	Quantity	Value
T_c	Critical temperature	282.3452 ± 0.0017 K
P_c	Critical pressure	5.0401 ± 0.0002 MPa
ρ_c	Critical density	7.634 ± 0.0009 mol · dm^{-3}
T_{tp}	Triple-point temperature	103.986 ± 0.003 K
P_{tp}	Triple-point pressure	0.00012 ± 0.00001 MPa
ρ_{tpv}	Triple-point density (vapor)	1.425×10^{-4} mol · dm^{-3}
ρ_{tpl}	Triple-point density (liquid)	23.348 mol · dm^{-3}
T_{nbp}	Normal boiling-point temp.	169.35 K
ρ_{nbpv}	Normal boiling density (vapor)	0.07440 mol · dim^{-3}
ρ_{nbpl}	Normal boiling point (liquid)	20.247 mol · dm^{-3}
T_0	Reference temperature	298.15 K
P_0	Reference pressure	0.101325 MPa
H_0	Reference enthalpy at T_0	29610 J · mol^{-1}
S_0	Reference entropy at T_0 and P_0	219.225 J · mol^{-1} · K^{-1}

In addition to the fundamental equation, ancillary functions including a vapor pressure equation, equations for the density of the saturated liquid and saturated vapor, and an equation for the ideal-gas heat capacity are given.

The fundamental equation may be used for the calculation of accurate tables of thermodynamic properties of ethylene within its range of applicability. This equation may also be readily used for system analysis where iterative solutions are required to solve the equation for known variable pairs other than density and temperature. The fundamental equation was developed to conform to the Maxwell criterion for liquid–vapor phase equilibrium. The fundamental equation given here is accurate in the critical region to within 0.1% in pressure. However, for accurate property determination in the critical region, the revised and extended scaling formulation of Levelt Sengers et al. [2] should be used.

The Helmholtz energy is given by the fundamental equation

$$A(\rho, T) = A^0(\rho, T) + \bar{A}(\rho, T) \tag{1}$$

where $A^0(\rho, T)$ is the ideal-gas contribution to the Helmholtz energy. The term $\bar{A}(\rho, T)$ is the contribution represented by the compressibility of the real gas.

The pressure derived from this expression is

$$P = \rho^2 \left(\frac{\partial A}{\partial \rho}\right)_T \tag{2}$$

The Helmholtz energy for the ideal gas is given by

$$A^0 = H_0^0 + \int_{T_0}^{T} C_\mathrm{p}^0 \, dT - RT - T\left[S_0^0 + \int_{T_0}^{T} \frac{C_\mathrm{p}^0}{T} \, dT - R \ln \frac{\rho T}{\rho_0 T_0} \right] \tag{3}$$

where H_0^0 is the ideal-gas enthalpy at $T_0 = 298.15$ K, C_p^0 is the ideal-gas heat capacity, ρ_0 is the ideal-gas density at T_0 and $P_0 = 0.101325$ MPa, and S_0^0 is the ideal-gas entropy at T_0 and P_0.

The functional form used for the fundamental equation for ethylene is a nondimensional function,

$$\alpha(\delta, \tau) = \frac{A(\rho, T)}{RT} = \alpha^0(\delta, \tau) + \bar{\alpha}(\delta, \tau) \tag{4}$$

$$\alpha^0 = \frac{H_0^0 \tau}{RT_\mathrm{c}} - \frac{S_0^0}{R} - 1 + \ln \frac{\delta \tau_0}{\delta_0 \tau} - \frac{\tau}{R} \int_{\tau_0}^{\tau} \frac{C_\mathrm{p}^0}{\tau^2} \, d\tau + \frac{1}{R} \int_{\tau_0}^{\tau} \frac{C_\mathrm{p}^0}{\tau} \, d\tau \tag{5}$$

where $\tau = T_\mathrm{c}/T$; $\delta = \rho/\rho_\mathrm{c}$; $\tau = T_\mathrm{c}/T_0$; $\delta_0 = \rho_0/\rho_\mathrm{c}$; ρ_c is the critical density; T_c is the critical temperature; T_0, P_0, ρ_0, H_0^0, and S_0^0 are as defined previously; and R is the gas constant, 0.00831434 $\mathrm{MPa \cdot dm^3 \cdot mol^{-1} \cdot K^{-1}}$. The real-fluid contribution to dimensionless Helmholtz energy is given by

$$\bar{\alpha}(\delta, \tau) = \sum_{k=1}^{100} N_k \, \delta^i \tau^j \exp(-\gamma \delta^l) \tag{6}$$

where the N_k are the coefficients of the fundamental equation, and γ has a value of 0 or 1. The values of i and j are generally greater than zero, and l is an integer greater than or equal to zero. The terms in Eq. (6) are similar to those used by Schmidt and Wagner for oxygen [5].

The bank of terms used for ethylene is given in Table II. The coefficients of Eq. (6) given in Table III were determined by a least-squares fit to 2088 selected data points including P–ρ–T data, velocity of sound data, and second virial coefficients. In addition, calculated smoothed values from the revised and extended scaling equation of Ref. 2, from the low-density vapor equation of Ref. 6, and from ancillary equations for the phase equilibrium properties were used. Details of the data selection are given in Ref. 1.

The functions used for calculating thermodynamic properties including the internal energy, enthalpy, entropy, isochoric heat capacity, isobaric heat capacity, Gibbs energy, and velocity of sound from Eq. (6) are given as Eqs. (7) through (15).

Table II. Parameters Considered in the Determination of the Equation of State for Ethylene[a]

k	i	j	l	k	i	j	l
1	1	0.25	0	51	1	1.00	3
2	1	0.50	0	52	1	1.50	3
3	1	0.75	0	53	1	2.00	3
4	1	1.00	0	54	1	3.00	3
5	1	1.25	0	55	1	4.00	3
6	1	1.50	0	56	1	5.00	3
7	1	1.75	0	57	2	0.50	2
8	1	2.00	0	58	2	1.00	2
9	1	2.25	0	59	2	1.50	2
10	1	2.50	0	60	2	2.00	2
11	1	2.75	0	61	2	3.00	2
12	1	3.00	0	62	2	4.00	2
13	1	3.25	0	63	2	5.00	2
14	1	3.50	0	64	2	1.00	4
15	1	3.75	0	65	2	2.00	4
16	1	4.00	0	66	2	3.00	4
17	2	0.25	0	67	2	4.00	4
18	2	0.50	0	68	2	5.00	4
19	2	0.75	0	69	2	6.00	4
20	2	1.00	0	70	2	1.00	6
21	2	1.50	0	71	2	2.00	6
22	2	2.00	0	72	2	3.00	6
23	2	2.50	0	73	2	4.00	6
24	2	3.00	0	74	3	0.50	3
25	2	3.50	0	75	3	1.00	3
26	2	4.00	0	76	3	1.50	3
27	2	5.00	0	77	3	2.00	3
28	2	6.00	0	78	3	3.00	3
29	3	0.25	0	79	3	4.00	3
30	3	0.50	0	80	3	5.00	3
31	3	0.75	0	81	4	0.50	2
32	3	1.00	0	82	4	1.00	2
33	3	1.50	0	83	4	1.50	2
34	3	2.00	0	84	4	2.00	2
35	3	3.00	0	85	4	3.00	2
36	3	4.00	0	86	4	4.00	2
37	3	5.00	0	87	4	5.00	2
38	4	0.25	0	88	4	1.00	4
39	4	0.50	0	89	4	2.00	4
40	4	1.00	0	90	4	3.00	4
41	4	1.50	0	91	4	4.00	4
42	4	2.00	0	92	4	5.00	4
43	4	3.00	0	93	4	6.00	4
44	6	0.50	0	94	8	0.50	2
45	6	1.00	0	95	8	1.00	2
46	6	1.50	0	96	8	1.50	2
47	6	2.00	0	97	8	2.00	2
48	6	2.50	0	98	8	3.00	2
49	6	3.00	0	99	8	4.00	2
50	1	0.50	3	100	8	5.00	2

[a] $\gamma = 0$ for terms with $l = 0$; $\gamma = 1$ for terms with l greater than 0.

Table III. Coefficients for the Fundamental Equation [Eq. (6)] for Ethylene[a] (Coefficients Not Listed Are Zero)

	i	j	l
N_2 = 3.248937034	1	0.50	0
N_4 = −10.17278862	1	1.00	0
N_5 = 7.386604053	1	1.25	0
N_7 = −1.568916359	1	1.75	0
N_{16} = −0.08884514287	1	4.00	0
N_{22} = 0.06021068143	2	2.00	0
N_{26} = 0.1078324588	2	4.00	0
N_{27} = −0.02004025211	2	5.00	0
N_{28} = 0.001950491412	2	6.00	0
N_{29} = 0.06718006403	3	0.25	0
N_{35} = −0.04200451469	3	3.00	0
N_{38} = −0.001620507626	4	0.25	0
N_{44} = 0.0005555156795	6	0.50	0
N_{48} = 0.0007583671146	6	2.50	0
N_{49} = −0.0002878544021	6	3.00	0
N_{50} = 0.06258987063	1	0.50	3
N_{51} = −0.06418431160	1	1.00	3
N_{57} = −0.1368693752	2	0.50	2
N_{60} = 0.5179207660	2	2.00	2
N_{62} = −0.3026331319	2	4.00	2
N_{66} = 0.7757213872	2	3.00	4
N_{67} = −2.639890864	2	4.00	4
N_{68} = 2.927563554	2	5.00	4
N_{69} = −1.066267599	2	6.00	4
N_{71} = −0.05380471540	2	2.00	6
N_{72} = 0.1277921080	2	3.00	6
N_{73} = −0.07450152310	2	4.00	6
N_{76} = −0.01624304356	3	1.50	3
N_{81} = 0.1476032429	4	0.50	2
N_{83} = −0.2003910489	4	1.50	2
N_{86} = 0.2926905618	4	4.00	2
N_{87} = −0.1389040901	4	5.00	2
N_{88} = 5.913513541	4	1.00	4
N_{89} = −38.00370130	4	2.00	4
N_{90} = 96.91940570	4	3.00	4
N_{91} = −122.6256839	4	4.00	4
N_{92} = 77.02379476	4	5.00	4
N_{93} = −19.22634672	4	6.00	4
N_{94} = −0.003800045701	8	0.50	2
N_{95} = 0.01118003813	8	1.00	2
N_{100} = 0.002945841426	8	5.00	2

[a] $\gamma = 0$ for terms 2 through 49, and $\gamma = 1$ for terms 50 through 100.

The compressibility factor is given by[4]

$$Z = \frac{P}{\rho RT} = 1 + \delta \frac{\partial \bar{\alpha}}{\partial \delta} \tag{7}$$

and

$$\frac{P}{P_c} = \frac{\delta}{\tau Z_c}\left[1 + \frac{\partial \bar{\alpha}}{\partial \delta}\right] \tag{8}$$

Similarly

$$\frac{U}{RT} = \tau\left[\frac{\partial \alpha^0}{\partial \tau} - \frac{\partial \bar{\alpha}}{\partial \tau}\right] \tag{9}$$

$$\frac{S}{R} = \tau\left[\frac{\partial \alpha^0}{\partial \tau} + \frac{\partial \bar{\alpha}}{\partial \tau}\right] - \alpha^0 - \bar{\alpha} \tag{10}$$

$$\frac{H}{RT} = \tau\left[\frac{\partial \alpha^0}{\partial \tau} + \frac{\partial \bar{\alpha}}{\partial \tau}\right] + \delta \frac{\partial \bar{\alpha}}{\partial \delta} + 1 \tag{11}$$

$$\frac{G}{RT} = 1 + \alpha^0 + \bar{\alpha} + \delta \frac{\partial \bar{\alpha}}{\partial \delta} \tag{12}$$

$$\frac{C_v}{R} = -\tau^2\left[\frac{\partial^2 \alpha^0}{\partial \tau^2} + \frac{\partial^2 \bar{\alpha}}{\partial \tau^2}\right] \tag{13}$$

$$\frac{C_p}{R} = \frac{C_v}{R} + \frac{\left[1 + \delta \frac{\partial \bar{\alpha}}{\partial \delta} - \delta\tau \frac{\partial^2 \bar{\alpha}}{\partial \delta\, \partial \tau}\right]^2}{\left[1 + 2\delta \frac{\partial \bar{\alpha}}{\partial \delta} + \delta^2 \frac{\partial^2 \bar{\alpha}}{\partial \delta^2}\right]} \tag{14}$$

and

$$\frac{W^2}{RT} = 1 + 2\delta \frac{\partial \bar{\alpha}}{\partial \delta} + \delta^2 \frac{\partial^2 \bar{\alpha}}{\partial \delta^2} - \frac{\left[1 + \delta \frac{\partial \bar{\alpha}}{\partial \delta} - \delta\tau \frac{\partial^2 \bar{\alpha}}{\partial \delta\, \partial \tau}\right]^2}{\left[\tau^2 \frac{\partial^2 \alpha^0}{\partial \tau^2} + \tau^2 \frac{\partial^2 \bar{\alpha}}{\partial \tau^2}\right]} \tag{15}$$

3. ANCILLARY EQUATIONS

A new vapor pressure equation and equations for the density of the saturated liquid and the saturated vapor as functions of temperature have

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[4] In the expressions for derived properties [Eqs. (7) to (15)] the subscripts for the properties held constant during differentiation are omitted.

been developed to include the critical-region data. New equations for coexistence properties consistent with the revised and extended scaling formulation of Levelt Sengers et al. [2] in the critical region are given in this section. Coefficients for the liquid–vapor coexistence property equations in this section are given in Table IV.

3.1. The Vapor Pressure Equation

The vapor pressure equation is

$$\ln(P/P_c) = (T_c/T) \sum_{i=1}^{17} N_i(1 - T/T_c)^{(i+1)/2} \tag{16}$$

where P_c and T_c are the critical pressure and the critical temperature, respectively.

Table IV. Coefficients for Liquid–Vapor Coexistence Property Equations for Ethylene[a]

Vapor pressure equation [Eq. (16)]

$N_1 = -6.373572165$
$N_2 = 1.363317303$
$N_3 = -0.3197269070$
$N_5 = -1.157656259$
$N_{10} = -1.899024189$

Saturated vapor density equation [Eq. (17)]

$N_1 = -1.722832379$
$N_3 = 19.03195837$
$N_4 = 30.38649257$
$N_5 = -41.14655323$
$N_7 = 12.75383046$
$N_8 = -3.767668608$
$N_{25} = 31.80542637$

Saturated liquid density equation [Eq. (18)]

$N_{10} = -2.973241426 \times 10^{-6}$
$N_{11} = -1.759597892$
$N_{12} = 1.297770957$
$N_{14} = -1.735448063$
$N_{15} = 0.9422348274$
$N_{18} = -0.04226244872$
$N_{25} = 3.131180969$

[a] Coefficients not listed are zero.

3.2. The Equation for the Saturated Vapor Density

The equation for the saturated vapor density of ethylene is

$$\ln(\rho_{\mathrm{sv}}/\rho_{\mathrm{c}}) = \sum_{i=1}^{24} N_i \tau^{i/3} + N_{25} \ln \theta \tag{17}$$

where $\tau = (T_{\mathrm{c}} - T)/T$, $\theta = T/T_{\mathrm{c}}$, $\rho_{\mathrm{c}} = 7.634\ \mathrm{mol \cdot dm^{-3}}$, the critical density, and ρ_{sv} is the density of the saturated vapor.

3.3. The Equation for the Saturated Liquid Density

The functional form of the equation for the saturated liquid density is

$$[(\rho_{\mathrm{sl}}/\rho_{\mathrm{c}}) - 1] = \sum_{i=-10}^{-1} N_{i+11} \tau^{i/3} + \sum_{i=1}^{13} N_{i+10} \tau^{i/3} + N_{24} \ln \theta + N_{25} \tau^{0.325} \tag{18}$$

where $\tau = (T_{\mathrm{c}} - T)/T$, $\theta = T/T_{\mathrm{c}}$, and ρ_{sl} is the density of the saturated liquid.

3.4. The Melting Pressure Equation

The melting pressure equation for ethylene used in this work is taken from Straty [7]. The melting pressure is given by

$$P = P_{\mathrm{tp}} + a[(T/T_{\mathrm{tp}})^{\varepsilon} - 1] \tag{19}$$

where $P_{\mathrm{tp}} = 0.00012\ \mathrm{MPa}$, $a = 357.924\ \mathrm{MPa}$, $\varepsilon = 2.0645$, and $T_{\mathrm{tp}} = 103.986\ \mathrm{K}$.

Table V. Vibrational Wave Numbers for Ethylene

Mode	ω_i
1	3026
2	1623
3	1342
4	1023
5	3103
6	1236
7	949
8	943
9	3106
10	826
11	2989
12	1444

Table VI. Thermodynamic Properties of Ethylene

Temperature (K)	Density $(\mathrm{mol \cdot dm^{-3}})$	Internal energy $(\mathrm{J \cdot mol^{-1}})$	Enthalphy $(\mathrm{J \cdot mol^{-1}})$	Entropy $(\mathrm{J \cdot mol^{-1} \cdot K^{-1}})$	C_v $(\mathrm{J \cdot mol^{-1} \cdot K^{-1}})$	C_p $(\mathrm{J \cdot mol^{-1} \cdot K^{-1}})$	Velocity of sound $(\mathrm{m \cdot s^{-1}})$
			0.101325-MPa isobar				
110	23.077	7011.3	7016.0	88.04	46.01	69.81	1732
150	21.208	9777.5	9782.2	109.53	39.29	66.67	1457
200	0.06214	24081.	25712.	203.53	27.43	36.43	275
250	0.04923	25532.	27591.	211.91	30.45	39.09	305
275	0.04464	26322.	28592.	215.73	32.50	41.06	319
300	0.04085	27166.	29646.	219.39	34.77	43.29	331
350	0.03494	29030.	31930.	226.42	39.68	48.13	353
400	0.03053	31142.	34461.	233.18	44.69	53.10	274
450	0.02712	33501.	37238.	239.71	49.54	57.93	394
			6.00-MPa isobar				
110	23.181	6957.6	7216.5	87.54	46.11	69.52	1761
150	21.370	9686.1	9966.8	108.92	39.50	66.06	1496
200	18.894	12991.	13308.	128.12	37.24	68.98	1135
250	15.701	16644.	17026.	144.66	38.17	83.16	729
275	13.089	18950.	19408.	153.72	40.27	116.8	475
300	4.5957	24149.	25454.	174.65	43.62	148.3	249
350	2.6412	27399.	296.70.	187.77	42.16	66.02	313
400	2.0659	29930.	32834.	196.22	45.97	62.24	351
450	1.7367	32521.	35975.	203.62	50.33	63.78	381

Thermodynamic properties of ethylene for the coexistence states

Temperature (K)	Pressure (MPa)	Density $(\mathrm{mol \cdot dm^{-3}})$	Enthalpy $(\mathrm{J \cdot mol^{-1}})$	Entropy $(\mathrm{J \cdot mol^{-1} \cdot K^{-1}})$	C_v $(\mathrm{J \cdot mol^{-1} \cdot K^{-1}})$	C_p $(\mathrm{J \cdot mol^{-1} \cdot K^{-1}})$	Velocity of sound $(\mathrm{m \cdot s^{-1}})$
110	0.00033	23.075	7012.6	88.05	46.01	69.81	1732
		0.00037	22751.	231.12	25.00	33.33	208
150	0.02748	21.206	9779.9	109.54	39.29	66.68	1457
		0.02231	24039.	204.60	25.76	34.56	241
200	0.45585	18.589	13190.	129.01	37.13	71.01	1070
		0.30289	25325.	189.68	29.46	42.12	262
250	2.3298	15.048	17143.	146.08	38.57	94.60	627
		1.6031	25673.	180.20	37.47	74.61	249

3.5. The Ideal-Gas Heat Capacity Equation

The equation used for the ideal-gas heat capacity of ethylene is

$$C_p^0/R = 4 + \sum_{i=1}^{n} \frac{u_i^2 e^{u_i}}{(e^{u_i} - 1)^2} \tag{20}$$

where $u_i = \theta_i/T$, $\theta_i = hc\omega_i/k$, n is the number of vibrational modes (for ethylene, $n = 12$), h is Planck's constant, c is the speed of light in vacuum, and k is Boltzmann's constant. The value of hc/k used in this work is 1.4388325 cm · K. The values of C_p^0 were calculated using Eq. (20) with the vibrational wave numbers, ω_i, from Shimanouchi [8] given in Table V.

4. COMPARISONS OF CALCULATED PROPERTIES TO EXPERIMENTAL DATA

Comparisons of calculated properties to experimental data are used as the basis for the validity and estimated accuracy of the correlation. Detailed comparisons of calculated thermodynamic properties to experimental data are given in Ref. 1. Space limitations do not permit the inclusion of graphical comparisons in this paper. Table VI is an abbreviated table of thermodynamic properties of ethylene. The property table may be used to verify programming of the equations presented here for system analysis.

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