

# Pressure and Density Series Equations of State for Steam as Derived from the Haar–Gallagher–Kell Formulation

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Two equations of state for the properties of steam, which are in the form of power series in pressure and density, are developed from the HGK84 formulation. These equations are of high accuracy in the equilibrium region where extensive measurements exist. They also accurately represent the extrapolated data in the metastable region between the vapor saturation and spinodal lines. The accuracy of the representations as a function of the number of terms of the series is presented. Their greatest utility is their use for high accuracy calculations that involve small to moderate departures from ideal-gas behavior. Conversion relationships for the second through the tenth coefficients of the pressure and density series, which apply to the corresponding virial coefficients, are presented. The pressure and density expansions are advantageous for efficient numerical calculations of water vapor properties in the equilibrium and metastable regions.

Key words: steam; virial equation; thermodynamic properties; metastable vapor; spinodal curve; HGK84; IAPS84.

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## 1. Introduction

The equilibrium thermodynamic properties for water have been recently summarized by an equation of state first

presented as an extended analytic equation,<sup>1</sup> subsequently in tabular form,<sup>2</sup> and finally in a dimensionless formulation.<sup>3</sup> All three of these representations are substantially identical as they are based on the same accumulation of recognized experimental data. Here, we will refer primarily to the second of these<sup>2</sup> which is frequently referred to as HGK84 and which, for scientific and general use, replaces earlier formulations of the properties of steam. For industrial use, the changes in property values may not be significant and the continued use of earlier formulations<sup>4</sup> has been recommended.<sup>5</sup> The new formulation has considerable advantage over its antecedents for certain applications.

An equation of state that is in the form of a power series has a number of advantages in several applications. The power series equation lends itself to efficient numerical calculations which can be executed with far greater speed than is possible with HGK84. Metastable steam vapor is observed in a number of applications of technical interest. Analyses of these processes require pressure-volume-temperature (PVT) information even though direct observations of these properties do not exist. In this case, the power series equations are of special relevance because they facilitate the extrapolation from the extensively measured superheated region into the metastable region.

The justification of the extrapolation into the metastable region of the equations which have been fitted in the equilibrium region is, at best, tentative. Noteworthy experiments have been conducted by Skripov and colleagues<sup>6-8</sup> which show that the density and speed of sound of liquid water and their derivatives are continuous when it passes from an equilibrium state into superheated metastable equilibrium. Comparable experimental information for the metastable water vapor is not available. However, experiments have been conducted with oxygen and nitrogen<sup>10</sup> and, in the instances of these gases, suggests that property values can be extrapolated from the equilibrium region.<sup>11</sup>

A number of experimental studies of the expansion of steam to the point at which supersaturation collapses have been conducted wherein careful measurement of pressure and nozzle area have been observed. In these measurements, the vapor clearly has been expanded to a metastable state well beyond the saturated vapor boundary prior to the onset of condensation. In these studies,<sup>12</sup> the measured pressure profile shows close agreement with theoretical predictions using the earlier IFC equation of state, thereby supporting the derivation of thermodynamic data by extrapolation from this equation of state into the metastable region.

## 2. Derivation of the Density Series Coefficients

### 2.1. Virial Equation

The virial equation of state was first used as an empirical equation for fitting PVT data. One of the earliest uses of a truncated form of the virial equation is due to Thiesen.<sup>13</sup> The formal development of the virial equation of state as an infinite series in density is attributed to Kamerlingh Onnes<sup>14</sup>:

$$\text{Pressure } P = RT \sum_{i=1}^{\infty} B_i \rho^i,$$

or

$$\text{Compressibility factor } Z = \frac{P}{\rho RT} = \sum_{i=1}^{\infty} B_i \rho^{i-1}, \quad (1)$$

where  $P$  is pressure,  $T$  is temperature,  $\rho$  is density, and  $R$  is the gas constant.  $B_1 = 1$  which represents the ideal-gas term in the expansion and functions  $B_i$  ( $i > 1$ ) are the *virial coefficients* which are function of temperature only. Equation (1) is also known as the Leiden series. A similar series expansion can be written in terms of pressure, instead of density:

$$\text{Compressibility factor } Z = \frac{P}{\rho RT} = \sum_{i=1}^{\infty} B'_i P^{i-1}. \quad (2)$$

The pressure series, also known as the Berlin series, is technically not a virial equation and the functions  $B'_i$  are not equal to but related to the virial coefficients  $B_i$  of (1) by a set of conversion (or inversion) formulas which will be developed later.

Kamerlingh Onnes was careful to refer to the functions  $B_i(T)$  as virial coefficients only for the infinite expansion. He noted that if a truncated form of the virial equation is used to fit experimental data, only the lower order resultant functions  $B_i$  approximate the "true" virial coefficients. He suggested the name "virial remainder function" for the higher order term to clearly denote that they are not virial coefficients.

Since series expansions which are developed herein are truncated expansions from data of limited accuracy, we cannot correctly call the derived  $B_i$  functions virial coefficients. However, the method of determining the  $B_i$  functions and the inversion formulas does apply to virial coefficients. Only the lowest order term  $B_2$  contains any valid virial coefficient data.

### 2.2. HGK84 Equation

HGK84<sup>2</sup> is a complex form of the Helmholtz free-energy equation of state  $A(\rho, T)$  which accurately describes thermodynamic properties of fluid water from the dilute vapor to compressed liquid states. Unlike previous equations of state for steam, HGK84 is a single analytic formulation covering the entire fluid state, rather than a collection of equations describing various regions.<sup>4</sup> This provides a higher degree of thermodynamic consistency. This single equation consists of four terms: ideal-gas equation ( $i$ ), base function ( $b$ ), residual function ( $r$ ), and critical region correction function ( $c$ ).

$$A(\rho, T) = A_i(T) + A_b(\rho, T) + A_r(\rho, T) + A_c(\rho, T). \quad (3)$$

In the following derivation, the critical region correction will be ignored. As will be shown later, this omission precludes accurate agreement between HGK84 and the series expansion near the critical point. The ideal-gas function of Ref. 2 is used directly. The virial coefficients (or  $B_i$  functions of a finite polynomial) can be found by first writing the  $i$ th density derivative of pressure from Eq. (1):

$$\left( \frac{\partial^i P}{\partial \rho^i} \right)_T = (i!) RT B_i + [(i+1)!] RT B_{i+1} \rho + \dots \quad (4)$$

The individual virial coefficients are found by taking the limit of the equation above:

$$B_i(T) = [(i!)RT]^{-1} \lim_{\rho \rightarrow 0} \left( \frac{\partial^{(i)} P}{\partial \rho^{(i)}} \right)_T \quad (5)$$

The pressure derivatives can be related to the Helmholtz function by  $P = \rho^2 \partial A / \partial \rho$ .

We see from Eq. (3) that the virial coefficient depends on both the base and residual functions. These components will be denoted by the subscripts  $b$  and  $r$ :

$$B_i = B_{i,b} + B_{i,r}. \quad (6)$$

The derivation of each component is described in the following sections.

### 2.3. Base Function

The base function of HGK84 is

$$A_b(\rho, T) = RT \{ -\ln(1-y) - (\beta-1)/(1-y) + (\alpha + \beta + 1)/[2(1-y)^2] + 4y(\bar{B}/b - \gamma) - (\alpha - \beta + 3)/2 + \ln(\rho RT/P_0) \}, \quad (7)$$

where  $\alpha$ ,  $\beta$ ,  $\gamma$ , and  $P_0$  are constants,  $y = b\rho/4$ . The two molecular parameters  $b$  and  $\bar{B}$  are functions of temperature:

$$b = b_1 \ln(T_r) + \sum_{j=0,1,3,5} b_j T_r^{-j}, \quad (8)$$

$$A_r(\rho, T) = \sum_{i=1}^{36} g_i T_r^{-l(i)} (1 - e^{-\rho})^{k(i)}, \quad (12)$$

where  $g_i$ ,  $l(i)$ , and  $k(i)$  are constants. Defining  $G_n$  as

$$G_n = \sum_{i=1}^{36} g_i T_r^{-l(i)} |k(i) = n, \quad (13)$$

we are able to express the residual function contribution to the virial coefficients by

$$\begin{aligned} (0!)RTB_{2,r} &= G_1, \\ (1!)RTB_{3,r} &= -G_1 + G_2, \\ (2!)RTB_{4,r} &= G_1 - 3G_2 + 2G_3, \\ (3!)RTB_{5,r} &= -G_1 + 7G_2 - 12G_3 + 6G_4, \\ (4!)RTB_{6,r} &= G_1 - 15G_2 + 50G_3 - 60G_4 + 24G_5, \\ (5!)RTB_{7,r} &= -G_1 + 31G_2 - 180G_3 + 390G_4 - 360G_5 + 120G_6, \\ (6!)RTB_{8,r} &= G_1 - 63G_2 + 602G_3 - 2100G_4 + 3360G_5 - 2520G_6 + 720G_7, \\ (7!)RTB_{9,r} &= -G_1 + 127G_2 - 1932G_3 + 10206G_4 - 25300G_5 + 31920G_6 - 20160G_7 + 5040G_8, \\ (8!)RTB_{10,r} &= G_1 - 255G_2 + 6050G_3 - 46620G_4 + 166824G_5 - 317520G_6 + 332640G_7 - 181440G_8 + 440320G_9. \end{aligned} \quad (14)$$

### 3. Conversion to Pressure Series

Epstein,<sup>15</sup> in 1952, published an algebraic method for the calculation of pressure-series virial coefficients as functions of the density-series virial coefficients. Epstein's paper also gives the pressure-series relations for the second to fifth virial coefficients ( $B'_2$  to  $B'_5$ ). A year later, Putnam and Kilpatrick<sup>16</sup> presented a different solution to the same problem and noted an error in Epstein's formula for  $B'_5$ . Putnam

$$\bar{B} = \sum_{j=0,1,2,4} C_j T_r^{-j}, \quad (9)$$

where  $T_r$  is the reduced temperature  $T/T_0$  ( $T_0 = 647.03$  K).  $C_j$  and  $b_j$  are constants.

The base function contribution to the virial coefficients are

$$\begin{aligned} B_{2,b} &= \bar{B}, \\ B_{3,b} &= (b/4)^2 [1 - 2(\beta - 1) + 3(\alpha + \beta + 1)], \\ B_{4,b} &= (b/4)^3 [1 - 3(\beta - 1) + 6(\alpha + \beta + 1)], \\ B_{5,b} &= (b/4)^4 [1 - 4(\beta - 1) + 10(\alpha + \beta + 1)], \\ &\dots \end{aligned} \quad (10)$$

where  $\bar{B}$  is the second virial coefficient which has been incorporated into the base function of HGK84. The higher order terms can be generalized for  $B_{i,b}$  ( $i > 2$ ) as

$$B_{i,b} = (b/4)^{i-1} [1 - (i-1)(\beta - 1) + \left( \sum_{j=1}^{i-1} j \right) (\alpha + \beta + 1)]. \quad (11)$$

### 2.4. Residual Function

The residual function consists of a 36-term expansion determined by a global, least-squares fit to experimental data.

and Kilpatrick's solution has now been implemented by a computer algorithm and used to develop the conversion formula for the first nine pressure-series virial coefficients. These formulas are given in Table 1. Values for both the density and pressure series coefficients are given in Table 2. The derivation of the density expansion and some of the pressure series conversion formulas have been verified by two investigators.

TABLE 1. Pressure series virial coefficients

$(RT)^1 B' = B$
$(RT)^2 C' = C - B^2$
$(RT)^3 D' = D - 3BC + 2B^3$
$(RT)^4 E' = E - 2C^2 - 4BD + 10B^2C - 5B^4$
$(RT)^5 F' = F - 5CD - 5BE + 15BC^2 + 15B^2D - 35B^3C + 14B^5$
$(RT)^6 G' = G - 3D^2 - 6CE + 7C^3 - 6BF + 42BCD + 21B^2E - 84B^2C^2 - 56B^3D + 126B^4C - 42B^6$
$(RT)^7 H' = H - 7DE - 7CF + 28C^2D - 7BG + 28BD^2 + 56BCE - 84BC^3 + 28B^2F - 252B^2CD - 84B^3E + 420B^3C^2 + 210B^4D - 462B^5C + 132B^7$
$(RT)^8 I' = I - 4E^2 - 8DF - 8CG + 36CD^2 + 36C^2E - 30C^4 - 8BH + 72BDE + 72BCF - 360BC^2D + 36B^2G - 180B^2D^2 - 360B^2CE + 660B^2C^3 - 120B^3F + 1320B^3CD + 330B^4E - 1980B^4C^2 - 792B^5D + 1716B^6C - 429B^8$
$(RT)^9 J' = J - 9EF - 9DG + 15D^3 - 9CH + 90CDE + 45C^2F - 165C^3D - 9BI + 45BE^2 + 90BDF + 90BCG - 495BCD^2 - 495BC^2E + 495BC^4 + 45B^2H - 495B^2DE - 495B^2CF + 2970B^2C^2D - 165B^3G + 990B^3D^2 + 1980B^3CE - 4290B^3C^3 + 495B^4F - 6435B^4CD - 1287B^5E + 9009B^5C^2 + 3003B^6D - 6435B^7C + 1430B^9$

$$B = B_2, C = B_3, D = B_4, E = B_5, \dots, J = B_{10}.$$

#### 4. Thermodynamic Relations

Thermodynamic properties such as enthalpy, internal energy, and entropy are readily determined from the classical thermodynamic relationships. Each property can be represented as the sum of an ideal gas ( $i$ ) and real gas or excess ( $*$ ) quantity.

$$\text{Pressure} \quad P = \rho^2 \frac{\partial A}{\partial \rho} = Z\rho RT = RT \sum_{i=1} B_i \rho^i, \quad (15)$$

$$\begin{aligned} \text{Internal energy} \quad u &= A + Ts, \\ u &= u_i + u_*, \\ u_i &= \int c_{vi} dT, \\ u_* &= \int \left( P - T \frac{\partial P}{\partial \rho} \right) \rho^{-2} d\rho \\ &= -RT^2 \sum_{i=2} \left( \frac{dB_i}{dT} \right) \frac{\rho^{i-1}}{i-1}, \end{aligned} \quad (16)$$

$$\text{Enthalpy} \quad h = u + P/\rho = u + ZRT, \quad (17)$$

$$\begin{aligned} \text{Isochoric heat capacity} \quad c_v &= -T \frac{\partial^2 A}{\partial T^2} \\ c_v &= c_{vi} + c_{v*}, \\ c_{vi} &= c_{vi}(T), \\ c_{v*} &= - \int T \rho^{-2} \frac{\partial^2 P}{\partial \rho^2} d\rho \\ &= -RT \sum_{i=2} \left( 2 \frac{dB}{dT} + T \frac{d^2 B}{dT^2} \right) \rho^{i-1}, \end{aligned} \quad (18)$$

$$\text{Isobaric heat capacity} \quad c_p = c_v + \frac{T}{\rho^2} \left( \frac{\partial P}{\partial T} \right)^2 / \left( \frac{\partial P}{\partial \rho} \right), \quad (19)$$

$$\begin{aligned} \text{Entropy} \quad s &= - \left( \frac{\partial A}{\partial T} \right)_\rho \\ s &= s_i + s_*, \\ s_i &= \int \frac{c_{pi}}{T} dT - R \ln \left( \frac{P}{P_{\text{ref}}} \right), \\ s_* &= \int \left[ R\rho - \frac{\partial P}{\partial T} \right] \frac{d\rho}{\rho^2} \\ &= -R \sum_{i=2} \left( B + T \frac{dB}{dT} \right) \frac{\rho^{i-1}}{(i-1)}, \end{aligned} \quad (20)$$

$$\text{Speed of sound} \quad c^2 = \left( \frac{c_p}{c_v} \right) \left( \frac{\partial P}{\partial \rho} \right)_T. \quad (21)$$

#### 5. Comparison of Density Expansion to HGK84

The region within the coexistence boundary in the PV plane is divided into three regions: metastable liquid, unstable, and metastable vapor. The unstable region is characterized by the positive slope of the (theoretical) isotherm,  $(\partial P / \partial V)_T$  as shown in Fig. 1. Two regions of metastable fluid exist between the unstable region and the equilibrium coexistence boundary. The focus of the present investigation is directed toward the estimation of thermodynamic properties in the stable and metastable vapor regions. As noted above, no accurate experimental PVT data for metastable vapor are available. Therefore, our basis for comparison must be the HGK84 equation of state from which the density and pressure series expansion were derived.

PRESSURE AND DENSITY EQUATIONS OF STATE FOR STEAM

TABLE 2. Values of density and pressure series coefficients

Temp. K	300	400	500	600
<u>Density Series Expansion Coefficients</u>				
B <sub>2</sub>	-6.2144E-002	-1.9610E-002	-9.5719E-003	-5.6032E-003
B <sub>3</sub>	-5.7842E-003	-4.7928E-004	-5.0752E-005	-5.7015E-006
B <sub>4</sub>	5.80276E-005	5.04810E-006	7.15806E-007	1.90502E-007
B <sub>5</sub>	-1.7577E-007	-1.4188E-008	-2.4975E-009	-1.0493E-009
B <sub>6</sub>	7.70390E-012	-7.9138E-012	2.53185E-012	3.60712E-012
B <sub>7</sub>	1.43229E-012	1.55473E-013	7.39633E-015	-9.4649E-015
B <sub>8</sub>	-5.0440E-015	-5.0946E-016	-3.4114E-017	1.97863E-017
B <sub>9</sub>	1.02578E-017	1.00963E-018	7.27149E-020	-3.2775E-020
B <sub>10</sub>	-1.4540E-020	-1.4142E-021	-1.0465E-022	4.21568E-023
dB <sub>2</sub> /dT	9.72370E-004	1.69740E-004	5.79027E-005	2.68441E-005
dB <sub>3</sub> /dT	1.57469E-004	1.11295E-005	1.11688E-006	1.23314E-007
dB <sub>4</sub> /dT	-1.5773E-006	-1.1110E-007	-1.1951E-008	-1.8897E-009
dB <sub>5</sub> /dT	4.96842E-009	3.11055E-010	3.08480E-011	6.62999E-012
dB <sub>6</sub> /dT	-1.6763E-012	1.80799E-013	3.99724E-014	-6.7732E-015
dB <sub>7</sub> /dT	-3.3444E-014	-3.4787E-015	-4.4674E-016	-1.8673E-017
dB <sub>8</sub> /dT	1.23904E-016	1.14431E-017	1.39325E-018	8.64019E-020
dB <sub>9</sub> /dT	-2.5592E-019	-2.2760E-020	-2.7121E-021	-1.8329E-022
dB <sub>10</sub> /dT	3.65184E-022	3.19454E-023	3.76751E-024	2.62900E-025
d <sup>2</sup> B <sub>2</sub> /dT <sup>2</sup>	-2.2315E-005	-2.2407E-006	-5.1389E-007	-1.7994E-007
d <sup>2</sup> B <sub>3</sub> /dT <sup>2</sup>	-4.6432E-006	-2.6950E-007	-2.4776E-008	-2.7191E-009
d <sup>2</sup> B <sub>4</sub> /dT <sup>2</sup>	4.67317E-008	2.67305E-009	2.46471E-010	2.95784E-011
d <sup>2</sup> B <sub>5</sub> /dT <sup>2</sup>	-1.5214E-010	-7.8845E-012	-6.2591E-013	-6.5704E-014
d <sup>2</sup> B <sub>6</sub> /dT <sup>2</sup>	8.74972E-014	-1.2618E-015	-8.9250E-016	-1.8063E-016
d <sup>2</sup> B <sub>7</sub> /dT <sup>2</sup>	8.47723E-016	7.22110E-017	9.56379E-018	1.40914E-018
d <sup>2</sup> B <sub>8</sub> /dT <sup>2</sup>	-3.3221E-018	-2.4787E-019	-2.9856E-020	-4.1739E-021
d <sup>2</sup> B <sub>9</sub> /dT <sup>2</sup>	6.97034E-021	5.00238E-022	5.82913E-023	7.96609E-024
d <sup>2</sup> B <sub>10</sub> /dT <sup>2</sup>	-1.0012E-023	-7.0673E-025	-8.1131E-026	-1.0964E-026
<u>Pressure Expansion Coefficients</u>				
B <sub>2</sub> '	-4.4883E-007	-1.0623E-007	-4.1480E-008	-2.0234E-008
B <sub>3</sub> '	-5.0318E-013	-2.5347E-014	-2.6737E-015	-4.8378E-016
B <sub>4</sub> '	-5.6525E-019	-6.0765E-021	-2.0309E-022	-1.2111E-023
B <sub>5</sub> '	-9.5406E-025	-2.2903E-027	-2.4233E-029	-6.0597E-031
B <sub>6</sub> '	-1.7248E-030	-9.0877E-034	-3.0629E-036	-2.8131E-038
B <sub>7</sub> '	-3.3884E-036	-3.9456E-040	-4.2327E-043	-1.5245E-045
B <sub>8</sub> '	-6.9528E-042	-1.7853E-046	-6.1023E-050	-8.3616E-053
B <sub>9</sub> '	-1.4772E-047	-8.3721E-053	-9.1177E-057	-4.8212E-060
B <sub>10</sub> '	-3.2189E-053	-4.0255E-059	-1.3972E-063	-2.8334E-067
<u>Saturated Vapor</u>				
P, kPa	3.53570	245.533	2,637.29	12,339.4
d, kg/m <sup>3</sup>	0.0255775	1.36792	13.1896	72.8105
Z(HGE84)	0.998407	0.972291	0.866489	0.612009
Z(dens.)*	0.998407	0.972291	0.866489	0.612009
Z(pres.)*	0.998407	0.972291	0.866495	0.618314

\* 10 term expansion

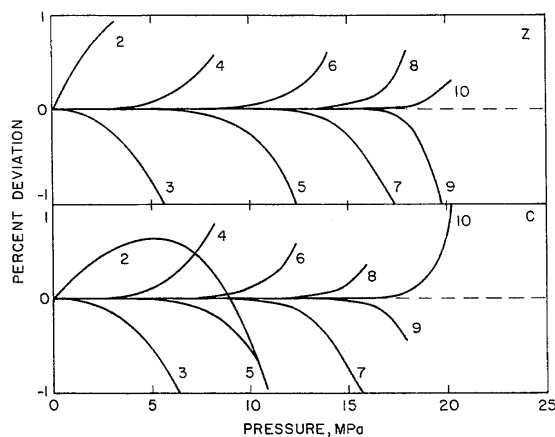


FIG. 6. Deviation in predicting the compressibility factor  $Z$  and sound speed  $c$  along the saturated vapor boundary. Numbers indicate the number of terms in the density series.

viation from ideal-gas behavior. For the determination of properties for low-pressure steam, a series expansion containing only a few terms can achieve sufficient precision for most applications. For example, in the metering of low-pressure steam by means of an orifice or flow nozzle, the uncertainty in the discharge coefficient of the primary flow element is typically 0.6%. If a sufficient number of terms are included in the series expansion to reduce the deviation between the truncated expansion and the full HGK84 equation to within, say, 0.1% along the saturated vapor boundary, then the systemic bias introduced in the superheated vapor region will be insignificant compared to other sources of errors and uncertainties. Figure 6 indicates that only four terms in the density expansion are required to keep the deviation within 0.1% for pressure 5 MPa, and eight terms for pressures to 15 MPa.

Since the sound speed depends on derivatives of the

thermodynamic properties, the magnitude of its derivation is often much larger than that for  $Z$ . In some instances, even the general nature of the deviation curves are quite different. One unexpected result of the present investigation is that deviations in sound speed are comparable in magnitude and the deviation curves resemble those for  $Z$ . This, perhaps fortuitous, result indicates that polytropic flow processes can be accurately calculated using the present pressure or density series.

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