Chapter 2
Equation of State Development and Use

Both historical and current functional forms for equations of state are described in the first edition of this book and other publications [1], and are not included here. Since the writing of the first edition, fundamental equations have become the standard source for accurate thermodynamic property information, and limited details are given in this section. The equations of state presented here all now use the same functional form, having replaced all historical forms.

2.1 Fundamental Equations of State

The term "fundamental equation" is generally reserved for those equations that contain calorimetric and datum state information so that absolute values of specified properties may be calculated directly by mathematical differentiation of a thermodynamic potential. The fundamental equations reported in this work are explicit in the Helmholtz energy a, which is the common form used for thermodynamic property formulations.

The use of fundamental equations of state in thermodynamic property correlations has several advantages over other forms:

1. The intrinsic advantage of a fundamental equation is that all properties may be calculated by differentiation. The typical pressure explicit equation of state must be integrated, for example, for the calculation of enthalpy and entropy.
2. The terms in the equation used for linear-fitting of property data may be changed by altering elements of an array defining the various coefficients and exponents. Section 2.2.2 gives details of the least-squares fitting process.
3. The temperature exponents can be fitted with the use of non-linear methods; further information on non-linear fitting is given in Sect. 2.2.3.
4. The form allows the correlator considerable flexibility in the design of an equation for a specific fluid.

5. Computer programs for fitting and for the calculation of thermodynamic prop-
   erty tables may be standardized so that few programming changes are required
   to calculate properties of several different fluids.

Schmidt and Wagner [2] developed a 32-term equation of state in 1985.
Although this form was developed for oxygen, it has been used by other investi-
gators for correlating properties of other fluids. The equation is explicit in
dimensionless Helmholtz energy,

$$a(\rho, T) = a^0(\rho, T) + a^r(\rho, T) \tag{2.1}$$

where $a^0(\rho, T)$ is the ideal gas contribution to the Helmholtz energy and the term
$a^r(\rho, T)$ represents the contribution due to the compressibility of the fluid. The
pressure derived from this expression is

$$P = \rho^2 \left(\frac{\partial a}{\partial \rho} \right)_T \tag{2.2}$$

The Helmholtz energy for the ideal gas is given by

$$a^0 = h^0 - RT - Ts^0 \tag{2.3}$$

Combining expressions for h^0 and s^0, the Helmholtz energy of the ideal gas is
given by

$$a^0 = h_0^0 + \int_{T_0}^{T} c_p^0 dT - RT - T \left[s_0^0 + \int_{T_0}^{T} \frac{c_p^0}{T} dT - R \ln\left(\frac{\rho T}{\rho_0 T_0} \right) \right] \tag{2.4}$$

An equation for the ideal-gas heat capacity as a function of temperature must be
used with this equation to develop the complete expression for a specific fluid:

$$\frac{c_p^0}{R} = a_0 + \sum_{i=3}^{m} a_i T^{k_i} + \sum_{i=100}^{n} a_i \frac{(k_i/T)^2 \exp(k_i/T)}{[\exp(k_i/T) - 1]^2} \tag{2.5}$$

Practical equations of state generally use reduced variables to simplify computer
calculations. Generally, properties are reduced by accepted critical state values or
other combinations of properties and constants to make the equations nondimen-
sional. The nondimensional variables used in this work are

$$\alpha = \frac{a}{RT}, \quad \delta = \frac{\rho}{\rho_c}, \text{ and } \tau = \frac{T_c}{T} \tag{2.6}$$

With these variables, Eq. (2.1) is written as

$$\alpha(\delta, \tau) = \alpha^0(\delta, \tau) + \alpha^r(\delta, \tau) \tag{2.7}$$

The ideal-gas contribution to the dimensionless Helmholtz energy can be derived from Eq. (2.4) as

$$\alpha^0 = \frac{h_0^0 \tau}{RT_c} - \frac{s_0^0}{R} - 1 + \ln\frac{\delta\tau_0}{\delta_0\tau} - \frac{\tau}{R}\int_{\tau_0}^{\tau}\frac{c_p^0}{\tau^2}\,d\tau + \frac{1}{R}\int_{\tau_0}^{\tau}\frac{c_p^0}{\tau}\,d\tau \tag{2.8}$$

A computationally convenient parameterized form of Eq. (2.8) is

$$\alpha^0 = \ln\delta + (a_0 - 1)\ln\tau + a_1 + a_2\tau - \sum_{i=3}^{m} a_i[T_c/\tau]^{k_i}/[k_i(k_i + 1)]$$
$$+ \sum_{i=m+1}^{n} a_i \ln[1 - \exp(-k_i\tau/T_c)]. \tag{2.9}$$

The fluorine formulation is the one exception to this equation and replaces T_c/τ with $1/\tau$. The form of $\alpha^r(\delta, \tau)$ is based on theoretical and practical considerations required to represent experimental data for a fluid. The general form used in current practice is

$$\alpha^r(\tau, \delta) = \sum_{i=1}^{l} N_i\delta^{d_i}\tau^{t_i} + \sum_{i=l+1}^{m} N_i\delta^{d_i}\tau^{t_i}\exp(-\zeta_i\delta^{p_i})$$
$$+ \sum_{i=m+1}^{n} N_i\delta^{d_i}\tau^{t_i}\exp\left[-\phi_i(\delta - D_i)^2 - \beta_i(\tau - \gamma_i)^2\right] \tag{2.10}$$

The parameters in these terms are determined through fitting experimental data. The terms in the third summation are usually described as Gaussian bell-shaped terms. The term ζ_i is only used in the EOS of fluorine and has a value of one for all other fluids.

2.2 Development of Thermodynamic Property Formulations

2.2.1 Thermodynamic Property Data Correlation

As established in Chap. 1, equations of state rely extensively on experimental measurements, theoretical estimates, and molecular simulation. The data selection process is a matter of experience and requires several tools for comparison of data sets, including:

1. Graphical displays of data (data maps) to assist in identifying regions of over-lapping data from alternative sources.
2. Graphical plots that show deviations between experimental data and previously accepted correlations or to preliminary correlations.
3. Property algorithms for producing tables and charts that can be used to examine the behavior of properties that were not included in the correlation.

Contributions of individual data points are weighted before they are used in least-squares fitting. Weighting is based on estimated accuracy. Some form of the theorem of propagation of variance should be used to correct for different levels of random errors in data. Often additional weighting factors are applied to achieve a desired emphasis of one form or body of data over another.

2.2.2 Least-Squares Regression

Methods for performing simultaneous regression on multiple-property data are given by Hust and McCarty [3]. In the regression process, thermodynamic property relations are written to form fit variables to represent experimental data. The dimensionless fit variables listed in Table 2.1 are examples of such variables used in developing equations of state. In forming fit variables, it is necessary to "lin-earize" some data forms with the use of a preliminary equation of state. For example, velocity of sound data are often prepared for least-squares regression through the use of a density and a ratio of heat capacities calculated from a pre-liminary equation of state.

The objective of linear least-squares regression is to select the coefficients N_k that minimize the sum of squares of differences between each experimental fit variable

Table 2.1 Functions for fitting equations of state to various data forms

Data form	Experimental fit variable	Calculated fit variable
P-ρ-T	$F_p = \dfrac{P}{P_c} - \dfrac{\delta}{\tau Z_c}$	$F_P' = \dfrac{\delta^2}{\tau Z_c} \sum\limits_{q=1}^{M} \left(\dfrac{\partial \alpha_i^r}{\partial \delta} \right)_\tau$
C_v-ρ-T	$F_{C_v} = \dfrac{C_v}{R} + \tau^2 \dfrac{\partial^2 \alpha^0}{\partial \tau^2}$	$F_{C_v}' = -\tau^2 \sum\limits_{q=1}^{M} \left(\dfrac{\partial^2 \alpha^r}{\partial \tau^2} \right)_\delta$
W-ρ-T	$F_W = \left(\dfrac{w^2 \tau}{\gamma R T_c} \right) - 1$	$F_W' = \sum\limits_{q=1}^{M} \left[2\delta \left(\dfrac{\partial \alpha^r}{\partial \delta} \right)_\tau + \delta^2 \left(\dfrac{\partial^2 \alpha^r}{\partial \delta^2} \right)_\tau \right]$
Liquid-vapor phase boundary	$F_\sigma = (Z_{S_L} - Z_{S_V})$ $+ \ln(\delta_{S_L} / \delta_{S_V})$	$F_\sigma' = \alpha_{S_V}^r - \alpha_{S_L}^r$
B-T	$F_B = B\,\rho_c$	$F_B' = \sum\limits_{q=1}^{M} \left[\sum\limits_{k=1}^{m} \tau^{jk} \right]_{i_k=1}$

where: σ = Saturation state; S_V = Saturated vapor; S_L = Saturated liquid; γ = Ratio of heat capacities (C_p/C_v); M = Number of data points; and i_k, j_k, and m come from Eq. (2.10)

F and the equivalent value calculated from the equation of state F'. The coefficients N_k are computed to minimize the sum of squares of the deviations S,

$$S = \sum Y_P \left(F_P - F'_P\right)^2 + \sum Y_{C_v}\left(F_{C_v} - F'_{C_v}\right)^2 + \sum Y_w \left(F_W - F'_W\right)^2$$
$$+ \sum Y_\sigma \left(F_\sigma - F'_\sigma\right)^2 + \sum Y_B \left(F_B - F'_B\right)^2 \tag{2.11}$$

where Y_p, Y_{C_v}, Y_W, Y_σ, and Y_B are weights applied to each data point. The subscripts P, C_v, W, σ, and B refer to weights for P-ρ-T data, C_v-ρ-T data, W-P-T data, Maxwell criterion data, and virial data, respectively. Often equations of state are fitted to the data with a fixed functional form with specified values for density and temperature exponents. However, more accurate equations of state are possible if these exponents are determined in a selection process. Schmidt and Wagner [2], de Reuck and Armstrong [4], and Bjornn [5] have developed stepwise regression algorithms for this purpose.

2.2.3   Non-linear Least-Squares Regression

Nonlinear fitting is currently the most effective method used to develop equations of state [6]. The techniques used continue to expand as new information is learned about fluid properties at extremely low temperatures and/or high pressures, and as the limits for the temperature exponents and Gaussian bell-shaped terms are refined. The method requires an initial starting point, which is often taken from recently developed equations for another molecularly similar fluid that demonstrates favorable behavior.

Nonlinear fitting does not require the need to linearize experimental data as described in the previous section. Experimental data can be used directly, instead of, for example, requiring a preliminary equation of state to transform pressure and temperature measured experimentally to density and temperature as required by the equation of state. The most important advantage in nonlinear fitting is the ability to use "greater than" or "less than" operators in constraints that control the extrapolation behavior of properties to regions outside that of the range of validity (usually determined as the range of experimental data). Linear fitting can only use equality conditions, and areas without data but with incorrect behavior are often extrapolated graphically to obtain the proper shape. Nonlinear fitting controls the behavior by coercing calculated values to be greater or less than those for lower or higher temperatures (or densities).

Nonlinear algorithms reduce the overall sum of squares of the deviations by adjusting the parameters of the equation of state with a residual sum of squares expression given as

$$S = \sum W_\rho F_\rho^2 + \sum W_p F_p^2 + \sum W_{c_v} F_{c_v}^2 + \cdots \qquad (2.12)$$

where W is the weight assigned to each data point and F is the function used to minimize the deviations. The deviations from all data types are summed and weighted such that each property contributes according to its uncertainty.

Because nonlinear methods can fit the temperature exponents and Gaussian-bell-shaped parameters simultaneously with the coefficients of the equations of state, the number of terms that are required to both fit the experimental data and to obtain correct extrapolation is generally half that required in linear fitting. This results in faster calculations and less inter-correlation between the terms.

Constraints form a very essential part of nonlinear fitting equations of state. These have the ability to smooth or change the slope and curvature of any constant property line on the surface of state, as long as such changes do not conflict with behavior caused by fitting accurate experimental data or by other valid constraints. When experimental data are available for derived thermodynamic properties, such as speed of sound and isobaric or isochoric heat capacities, there may be conflicts that must be avoided in the characteristics of parts of the surface, particularly the low temperature liquid or low density states approaching atmospheric pressure. Each of these properties is a function of derivatives of the Helmholtz energy with respect to temperature and/or density, and thus each property requires that the others have the correct characteristic behavior.

On the other hand, fitting the slope and curvature of the speed of sound and other properties below the triple point is possible, and generally required, and the person fitting the equation must be aware of how such properties should behave. Unrealistic shapes far from the region of validity still impact calculated values within the valid region because second order derivatives are part of heat capacity or speed of sound equations. To eliminate small bumps in the surface, non-linear fitting also allows the ability to control the third and fourth derivatives of the equation so that very smooth behavior can be obtained at all conditions outside and inside the range of validity.

2.2.4 Criteria for Equations of State

The quality of a thermodynamic property formulation is determined by its ability to model the physical behavior of the fluid as represented by the available data and its conformance to theory. Published correlations should include estimates of the accuracy of calculated properties as well as a careful definition of the range of validity. A modern thermodynamic property formulation is generally capable of representing all data values within the estimated experimental uncertainty of the measurements (see Table 1.2). The criteria listed below are generally used by correlators in developing equations of state for pure fluids:

1. *Pv* approaches *RT* in the limit as ρ approaches zero (i.e., the ideal-gas limit is approached).
2. At low densities the equation of state degenerates to a truncated form of the virial equation.
3. At the critical point

    (a) $(\partial P/\partial v)_T = 0$
    (b) $(\partial^2 P/\partial v^2)_T = 0$
    (c) $(\partial P/\partial T)_{\rho=\rho_c} = dP/dT$ of the vapor pressure curve

4. At the coexistence boundary, the Maxwell criteria (equal Gibbs energies in the liquid and vapor phases at the saturation temperature and pressure) is applied to the equation of state, i.e., coexistence property values predicted by the equation of state should be consistent with those from the ancillary equations for the vapor pressure and the liquid and vapor saturated densities.
5. Numerical requirements (constraints) may be imposed in fitting the equation of state for state points known to high accuracy.

2.2.5   Critical Region Behavior

Correct behavior of the equation of state in the critical region is of concern to some users of property formulations. Because of the nonanalytic behavior of certain fluid properties such as C_v in the critical region, special correlations (e.g., the scaled equations of state presented by Tang et al. [7], Sengers [8]) that use models that account for the fluid behavior at the critical point have been developed for critical region properties. Some provisions can be added to wide range equations of state to approximate critical-region behavior. Wide range models may be improved by including calculated properties near the critical point in the data set used for fitting. Special empirical terms may also be included in equations of state to improve the representation of calculated fluid properties near the critical point. The following statements summarize the considerations used in correlating thermodynamic data in the critical region:

1. The critical point is a singular point (e.g., $C_v \rightarrow \infty$, $C_p \rightarrow \infty$, $W \rightarrow 0$). However, an analytic equation of state cannot fully represent the critical point behavior. Non-analytic terms are required in an equation of state that correctly models the critical region behavior.
2. Most equations of state are analytic and properties may be calculated with differential or integral equations. A non-analytic equation requires numerical solutions for derived property calculations.

3. Critical point phenomena are reflected in the derived properties. The critical region anomalous behavior observed in C_p, C_v, and W generally distort the property surfaces as defined by an analytic equation of state.

2.3   Use of Fundamental Equations of State for Cryogenic Fluids

The equation of state described in this chapter is a general form used for the calculation of properties of a large number of fluids. Although the original references reporting the equations of state for cryogenic fluids use a wide variety of formats, the equations have been transformed to the fundamental form given in Sect. 2.1. Details of the implementation of this equation as a generalized form for cryogenic fluids follow.

Table 2.2 gives the thermodynamic property relations for the fundamental equation summarized in the previous section. The functions in this table are readily used in computer programs for the calculation of thermodynamic properties as given by Lemmon et al. [9, 10]. Equations of state for each of the cryogenic fluids discussed in subsequent chapters have been transformed, if necessary, to this format for consistency.

2.4   Ancillary Functions

In the development of a thermodynamic property formulation for a pure fluid or fluid mixture, it is desirable to determine separate ancillary functions for the liquid-vapor coexistence properties and for the ideal-gas heat capacity. There are many functional forms and methods for determining these ancillary functions. Equations for the vapor pressure, saturated liquid density, and saturated vapor density are used to define the saturation (coexistence) states during the development of the equation of state and as initial estimates in the iterative calculation of saturation properties used in computer programs for the completed property formulation. Equations for the freezing liquid line (also known as the melting line) and sublimation line are not provided here. If calculations exceed the ranges of tabulated properties, the user is encouraged to consult REFPROP or the original publication.

The ancillary functions for vapor pressure, saturated liquid density, and saturated vapor density are given as estimating functions for those properties. These functions are also useful in computer programs for calculating properties iteratively with an equation of state with input properties other than the independent variables of the equation. The default form for most equations is presented here, with the exceptions to these default formats described below and in the fluid specific chapters.

Table 2.2 Functions for calculation of thermodynamic properties

Property	Relation
Compressibility factor	$Z = \frac{P}{\rho RT} = 1 + \delta \left(\frac{\partial \alpha^r}{\partial \delta} \right)$
Pressure	$\frac{P}{P_c} = \frac{\delta}{\tau Z_c} \left[1 + \delta \left(\frac{\partial \alpha^r}{\partial \delta} \right) \right]$
Fugacity coefficient	$\phi = \exp[Z - 1 - \ln(Z) + \alpha^r]$
Second virial coefficient	$B(T) = \frac{1}{\rho_c} \left(\frac{\partial \alpha^r}{\partial \delta} \right)_{\delta=0}$
Third virial coefficient	$C(T) = \frac{1}{\rho_c^2} \left(\frac{\partial^2 \alpha^r}{\partial \delta^2} \right)_{\delta=0}$
Internal energy	$\frac{U}{RT} = \tau \left[\left(\frac{\partial \alpha^o}{\partial \tau} \right) + \left(\frac{\partial \alpha^r}{\partial \tau} \right) \right]$
Enthalpy	$\frac{H}{RT} = \tau \left[\left(\frac{\partial \alpha^o}{\partial \tau} \right) + \left(\frac{\partial \alpha^r}{\partial \tau} \right) \right] + \delta \left(\frac{\partial \alpha^r}{\partial \delta} \right) + 1$
Gibbs energy	$\frac{G}{RT} = 1 + \alpha^o + \alpha^r + \delta \left(\frac{\partial \alpha^r}{\partial \delta} \right)$
Helmholtz energy	$\frac{a}{RT} = \alpha^0 + \alpha^r$
Entropy	$\frac{S}{R} = \tau \left[\left(\frac{\partial \alpha^o}{\partial \tau} \right) + \left(\frac{\partial \alpha^r}{\partial \tau} \right) \right] - \alpha^o - \alpha^r$
Speed of sound	$\frac{W^2}{RT} = \frac{C_p}{C_v} \left[1 + 2\delta \left(\frac{\partial \alpha^r}{\partial \delta} \right) + \delta^2 \left(\frac{\partial^2 \alpha^r}{\partial \delta^2} \right) \right]$
Isochoric heat capacity	$\frac{C_v}{R} = -\tau^2 \left[\left(\frac{\partial^2 \alpha^o}{\partial \tau^2} \right) + \left(\frac{\partial^2 \alpha^r}{\partial \tau^2} \right) \right]$
Isobaric heat capacity	$\frac{C_p}{R} = \frac{C_v}{R} + \frac{\left[1 + \delta \left(\frac{\partial \alpha^r}{\partial \delta} \right) - \delta \tau \left(\frac{\partial^2 \alpha^r}{\partial \delta \partial \tau} \right) \right]^2}{\left[1 + 2\delta \left(\frac{\partial \alpha^r}{\partial \delta} \right) + \delta^2 \left(\frac{\partial^2 \alpha^r}{\partial \delta^2} \right) \right]}$
First pressure derivative with respect to density	$\left(\frac{\partial P}{\partial \rho} \right)_T = RT \left[1 + 2\delta \left(\frac{\partial \alpha^r}{\partial \delta} \right) + \delta^2 \left(\frac{\partial^2 \alpha^r}{\partial \delta^2} \right) \right]$
Second pressure derivative with respect to density	$\left(\frac{\partial^2 P}{\partial \rho^2} \right)_T = \frac{RT}{\rho} \left[2\delta \left(\frac{\partial \alpha^r}{\partial \delta} \right) + 4\delta^2 \left(\frac{\partial^2 \alpha^r}{\partial \delta^2} \right) + \delta^3 \left(\frac{\partial^3 \alpha^r}{\partial \delta^3} \right) \right]$
First pressure derivative with respect to temperature	$\left(\frac{\partial P}{\partial T} \right)_\rho = R \left[1 + \delta \left(\frac{\partial \alpha^r}{\partial \delta} \right) - \delta \tau \left(\frac{\partial^2 \alpha^r}{\partial \delta \partial \tau} \right) \right]$
Joule-Thomson coefficient	$\mu_j = \left(\frac{\partial T}{\partial P} \right)_h = \frac{T\beta - 1}{\rho C_p}$
Isentropic expansion coefficient	$k = -\frac{v}{P} \left(\frac{\partial P}{\partial v} \right)_s = \frac{W^2 \rho}{P}$
Isothermal expansion coefficient	$k_T = -\frac{v}{P} \left(\frac{\partial P}{\partial v} \right)_T = \frac{\rho}{P} \left(\frac{\partial P}{\partial \rho} \right)_T$
Volume expansivity	$\beta = \frac{1}{v} \left(\frac{\partial v}{\partial T} \right)_P = \frac{1}{\rho} \left(\frac{\partial P}{\partial T} \right)_\rho \left(\frac{\partial P}{\partial \rho} \right)_T$
Adiabatic compressibility	$\beta_s = \frac{1}{kP} = -\frac{1}{v} \left(\frac{\partial v}{\partial P} \right)_s$
Adiabatic bulk modulus	$B_s = kP = -v \left(\frac{\partial P}{\partial v} \right)_s$
Isothermal compressibility	$\kappa = \frac{1}{k_T P} = -\frac{1}{v} \left(\frac{\partial v}{\partial P} \right)_T$
Isothermal bulk modulus	$K_T = k_T P = -v \left(\frac{\partial P}{\partial v} \right)_T$

The generic form of the vapor pressure ancillary function is

$$\ln \left(\frac{p_\sigma}{p_c} \right) = \tau \sum_{i=1}^{q} N_i \theta^{k_i} \tag{2.13}$$

where $\theta = (1 - T/T_c)$, p_σ is the saturated vapor pressure, and the values for the coefficients N_i and the exponents k_i are given in each fluid chapter.

The generic form of the saturated liquid density ancillary function is

$$\frac{\rho'}{\rho_c} = 1 + \sum_{i=1}^{q} N_i \theta^{k_i} \qquad (2.14)$$

where ρ' is the saturated liquid density. The ancillary equations for the saturated liquid density for argon, methane, and nitrogen use a different form, as given by

$$\ln\left(\frac{\rho'}{\rho_c}\right) = \sum_{i=1}^{q} N_i \theta^{k_i} \qquad (2.15)$$

The generic form of the saturated vapor density ancillary function is

$$\ln\left(\frac{\rho''}{\rho_c}\right) = \sum_{i=1}^{q} N_i \theta^{k_i} \qquad (2.16)$$

where ρ'' is the saturated vapor density. The ancillary equations for the saturated vapor density for argon and nitrogen use a different form, as given by

$$\ln\left(\frac{\rho''}{\rho_c}\right) = \tau \sum_{i=1}^{q} N_i \theta^{k_i} \qquad (2.17)$$

The default form of the ideal-gas reduced Helmholtz ancillary function was given previously in Eq. (2.9).

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