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A PREDICTIVE EXTENDED CORRESPONDING STATES MODEL FOR PURE AND MIXED REFRIGERANTS ¹

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ABSTRACT

We have developed a predictive corresponding states model for the thermophysical properties of pure refrigerants and refrigerant mixtures. The bulk phase properties such as the density, enthalpy and entropy are predicted using the principle of extended corresponding states. This theoretically based model uses shape factors to insure conformality among the various components and R134a is used as the reference fluid. The shape factors are found by mapping saturation boundaries of the fluids of interest onto the reference fluid. In the case where no saturation data are available, the shape factors are predicted. In addition to equilibrium properties, a one-fluid corresponding states model is used to predict the viscosity and thermal conductivity. Phase equilibria is predicted using a Peng-Robinson equation of state.

The corresponding states model is contained in an interactive microcomputer program, PROZPER, (PProperties of OZone Protecting Environmentally acceptable Refrigerants). The program performs various vapor-liquid equilibria calculations—dew point and bubble point pressures and temperatures, isothermal, isenthalpic and isentropic flashes—and gives the bulk phase properties (such as density, enthalpy, entropy, Joule-Thomson coefficient, C_p , C_p/C_v , sound speed, viscosity and thermal conductivity) of the coexisting phases. Calculations may be performed on pure fluids or on mixtures of up to 20 components. A built-in database contains many common refrigerants including fully-halogenated CFC's (for example R11, R114, R12 and R13) as well as environmentally acceptable alternatives (such as R123, R134a and others). In addition, the program can "learn" new components using only a minimum amount of information—the molecular weight, the normal boiling point and the critical parameters of the chemical species. Additional information such as vapor pressures, saturated liquid densities, saturated liquid thermal conductivities and viscosities may be input to improve the predictive capability of the model.

INTRODUCTION

Many authors have pointed out the need for accurate thermophysical property data and models. Models are especially needed for refrigerant mixtures because of the limited number of experimental studies which have been reported in the literature for these systems. The most accurate approach to modeling properties of

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single-phase fluid mixtures arises from combining a molecularly based corresponding states theory with a highly accurate empirical equation of state. This approach eliminates the need for detailed knowledge of the intermolecular potential while retaining the underlying theoretical rigor. In addition, the approach allows for the use of a more complex (and presumably more accurate) mathematical description of the reference fluid pVT surface, thereby removing restrictions associated with the inflexible forms of simpler engineering equation of state models.

In this manuscript we discuss the mathematical details, implementation, and preliminary application of the extended corresponding states model to the equilibrium properties of refrigerants. The extended corresponding states theory itself is not new. It was originally proposed by Leland and co-workers [1,2] in the early 1970's and was used by Rowlinson and co-workers [3,4] for simple mixtures, Parrish [5,6] for liquefied petroleum gas mixtures and by Mollerup [7,8] and McCarty [9] for cryogenic systems, particularly LNG. More recently, it has been applied to properties of carbon dioxide rich mixtures and air by Ely and coworkers [10,11,12]. As mentioned above, the accuracy of this approach lies in the accuracy of the empirical reference fluid equation of state and in the accuracy of the shape factors which are used in the definition of the equivalent substance state point. In this work we have used a 32-term Jacobsen-Stewart type modified Benedict-Webb-Rubin (MBWR-32) equation of state for R134a as the reference fluid and have calculated shape factors (discussed below) by mapping the saturation boundary of the fluids of interest onto the reference fluid. The results of these mappings were then used to develop a generalized expression for the shape factors of refrigerants relative to the R134a reference. We also present some preliminary comparisons of the predictions of the model with the PVT surface of R23.

MODEL DESCRIPTION

The corresponding states principle for simple spherically symmetric pure fluids has been known for many years and is well summarized in advanced texts. The basic result is that if two pure fluids are conformal (obey the same reduced intermolecular force law), straightforward scaling arguments lead one to the conclusion that

$$a_j^r(\rho_j, T_j) = a_0^r(\rho_0, T_0). \quad (1)$$

In this equation, $a^r = [A(\rho, T) - A^*(\rho, T)]/RT$ where A is the Helmholtz free energy, the asterisk indicates an ideal gas value, ρ is the molar density, T is the absolute temperature, and R is the universal gas constant. The subscript "j" indicates the fluid of interest and "0" indicates a reference fluid whose thermodynamic properties are known, in principle, with great accuracy. The scaling arguments which lead to equation (1) provide a relation between the volume and temperature of the fluid j and the corresponding values for the reference fluid, namely $T_0 = T_j/f_j$ and $\rho_0 = \rho_j h_j$. The scale factors f_j and h_j are called the equivalent substance reducing ratios and are related on a microscopic scale to ratios of the intermolecular potential parameters, and on a macroscopic scale, to the critical parameters of the two fluids, namely, $f_j = (T_j^c/T_0^c)$ and $h_j = (\rho_0^c/\rho_j^c)$, where the superscript "c" denotes a critical point value. Other thermodynamic properties of

the fluid of interest follow from differentiation of equation (1). For example, since

$$da^r = d(A^r/RT) = -\frac{U - U^*}{RT^2}dT - \left(\frac{p}{RT} - \frac{1}{V}\right)dV + \sum_{i=1}^n \frac{\hat{\mu}_i - \hat{\mu}_i^*}{RT} dx_i$$

one finds for the residual compressibility factor $z^r = pV/RT - 1$

$$z_j^r(\rho_j, T_j) = z_0^r(\rho_j h_j, T_j / f_j).$$

The Extended Corresponding States Principle

The formalism discussed above was developed for spherically symmetric molecules whose intermolecular potentials are conformal. This is a somewhat limited class of materials and places severe restrictions on the applicability of the model, especially for refrigerants which are generally nonspherical and polar. In order to overcome this type of problem, Leland *et al.* [1] proposed the use of molecular shape factors. These shape factors are introduced in the equivalent substance reducing ratios for pure fluids and in principle are allowed to be functions of density and temperature. Thus, in effect, we assume that the intermolecular potential parameters are density and temperature dependent, but the potential is itself conformal to the potential of some reference fluid. Mathematically we have $f_j = (T_j^c/T_0^c)\theta_j(\rho, T)$ and $h_j = (\rho_0^c/\rho_j^c)\phi_j(\rho, T)$. The parameters θ and ϕ are called shape factors.

In the extended corresponding states model, the basic thermodynamic relationship obtained in the simple corresponding states model remains the same, *viz.*

$$a_j^r(\rho_j, T_j) = a_0^r(\rho_0, T_0) = a_0^r(\rho_j h_j, T_j / f_j). \quad (2)$$

One finds, however, that the thermodynamic relationships between various properties are not those observed in simple corresponding states. Reconsider, for example, the residual compressibility factor, z^r . Allowing for the fact that the shape factors might depend upon density and differentiating equation (2) with respect to ρ_j we find

$$z_j^r = z_0^r[1 + H_\rho] + u_0^r F_\rho,$$

where $u_0^r = (U_0 - U_0^*)/RT_0$, $F_\rho = (\partial f_j / \partial \rho_j)_{T_j} (\rho_j / f_j)$ and $H_\rho = (\partial h_j / \partial \rho_j)_{T_j} (\rho_j / h_j)$. Note that if the shape factors are independent of density that $F_\rho = H_\rho = 0$ and the result from simple corresponding states, $z_j^r = z_0^r$ is retained. Similar relations arise due to the temperature dependence of the shape factors. For example, the dimensionless residual internal energy is given by

$$u_j^r = u_0^r[1 - F_T] - z_0^r H_T,$$

where $F_T = (\partial f_j / \partial T_j)_{\rho_j} (T_j / f_j)$ and $H_T = (\partial h_j / \partial T_j)_{\rho_j} (T_j / h_j)$. These results are summarized in Table 1 for other thermodynamic properties of interest.

Property	Relation
Helmholtz Free Energy	$a_j^r = a_0^r$
Compressibility Factor	$z_j^r = z_0^r[1 + H_\rho] + u_0^r F_\rho$
Internal Energy	$u_j^r = u_0^r[1 - F_T] - z_0^r H_T$
Entropy	$s_j^r = s_0^r - u_0^r F_T - z_0^r H_T$
Enthalpy	$h_j^r = h_0^r + u_0^r[F_\rho - F_T] + z_0^r[H_\rho - H_T]$
Gibbs Free Energy	$g_j^r = g_0^r + z_0^r H_\rho + u_0^r F_\rho$
Fugacity Coefficient	$\ln(\hat{f}_k/x_k p) = \ln(f_0/p_0) + z_0^r H_{n_k} + u_0^r F_{n_k}$

Table 1: Dimensionless residual thermodynamic properties in the extended corresponding states model with density and temperature dependent shape factors

Mixtures

In the case of mixtures, the application of the extended corresponding states model is complicated by the composition dependence of the equivalent substance reducing ratios. In our studies we have assumed that this composition dependence is given by the van der Waals one-fluid mixing rules. The van der Waals mixing rules are given by

$$h_{\mathbf{x}} = \sum_{i=1}^n \sum_{j=1}^n x_i x_j h_{ij} \quad (3)$$

and

$$f_{\mathbf{x}} h_{\mathbf{x}} = \sum_{i=1}^n \sum_{j=1}^n x_i x_j f_{ij} h_{ij}. \quad (4)$$

The cross terms are obtained from the combining rules

$$f_{ij} = \sqrt{f_i f_j} (1 - k_{ij}) \quad (5)$$

and

$$h_{ij} = (h_i^{1/3} + h_j^{1/3})^3 (1 - l_{ij}) / 8. \quad (6)$$

In these equations x_i is the concentration of component i in the mixture, $f_{\mathbf{x}}$ and $h_{\mathbf{x}}$ are the equivalent substance reducing ratios for the mixture and k_{ij} and l_{ij} are the binary interaction parameters and are nonzero when $i \neq j$.

Application of the formulas given in Table 1 to mixtures requires derivatives of $f_{\mathbf{x}}$ and $h_{\mathbf{x}}$ with respect to temperature, density and composition. An inspection of the mixing rules and the definitions of the equivalent substance reducing ratios shows that the arguments of the shape factors are the effective temperatures and densities of components in the mixture. These, in fact, do not correspond to the temperature and density of the mixture unless the shape factors are identically unity. Thus, in a mixture, the arguments of the shape factors are themselves functions of $f_{\mathbf{x}}$ and $h_{\mathbf{x}}$. The dependence of these is nominally given by $T_j = T_{\mathbf{x}} f_j / f_{\mathbf{x}}$ and $V_j = V_{\mathbf{x}} h_j / h_{\mathbf{x}}$. Differentiating these relations with respect to $T_{\mathbf{x}}$ one obtains two equations:

$$F_j(T_{\mathbf{x}}) = F_j(T_j)[1 + F_j(T_{\mathbf{x}}) - F_{\mathbf{x}}(T_{\mathbf{x}})] + F_j(V_j)[H_j(T_{\mathbf{x}}) - H_{\mathbf{x}}(T_{\mathbf{x}})] \quad (7)$$

and

$$H_j(T_x) = H_j(T_j)[1 + F_j(T_x) - F_x(T_x)] + H_j(V_j)[H_j(T_x) - H_x(T_x)] \quad (8)$$

where

$$W_\alpha(Y_\beta) = \frac{Y_\beta}{w_\alpha} \frac{\partial w_\alpha}{\partial Y_\beta} \quad \text{e.g.,} \quad H_j(T_x) = \frac{T_x}{h_j} \frac{\partial h_j}{\partial T_x}.$$

Differentiation of the mixing rules equations (3) and (4) with respect to temperature, T_x , yields

$$F_x(T_x) + H_x(T_x) = \frac{1}{f_x h_x} \sum_{i=1}^n \sum_{j=1}^n x_i x_j f_{ij} h_{ij} [F_i(T_x) + \frac{g_i}{g_{ij}} H_i(T_x)] \quad (9)$$

and

$$H_x(T_x) = \frac{1}{h_x} \sum_{i=1}^n \sum_{j=1}^n x_i x_j h_{ij} \frac{g_i}{g_{ij}} H_i(T_x) \quad (10)$$

where $g_i = h_i^{1/3}$ and $g_{ij} = (g_i + g_j)/2$.

Simultaneous solution of (7) and (8) for $F_i(T_x)$ and $H_i(T_x)$ and substitution into (9) and (10) and subsequent solution for $F_x(T_x)$ and $H_x(T_x)$ yields $F_x(T_x) = 1 - S_7/R$ and $H_x(T_x) = S_6/R$. Similar procedures yield for the volume derivatives $F_x(V_x) = (S_2 + S_4 - S_7)/R$ and $H_x(V_x) = (S_6 + S_1 - S_3 + R)/R$ and for the composition derivatives $F_x(n_k) = [S_5^{(k)} S_7 - S_8^{(k)} (S_2 + S_4)]/R$ and $H_x(n_k) = [S_8^{(k)} (S_1 + S_3) - S_5^{(k)} S_6]/R$ where $R = (S_1 + S_3) S_7 - (S_2 + S_4) S_6$. The definitions of the sums S_m which appear in these results are given below:

$$\begin{aligned} S_1 &= \frac{1}{f_x h_x} \sum_{i=1}^n \sum_{j=1}^n x_i x_j f_{ij} h_{ij} [1 - H_i(V_i)] D_i \\ S_2 &= \frac{1}{f_x h_x} \sum_{i=1}^n \sum_{j=1}^n x_i x_j f_{ij} h_{ij} F_i(V_i) D_i \\ S_3 &= \frac{1}{f_x h_x} \sum_{i=1}^n \sum_{j=1}^n x_i x_j f_{ij} h_{ij} \frac{g_i}{g_{ij}} H_i(T_i) D_i \\ S_4 &= \frac{1}{f_x h_x} \sum_{i=1}^n \sum_{j=1}^n x_i x_j f_{ij} h_{ij} \frac{g_i}{g_{ij}} [1 - F_i(T_i)] D_i \\ S_5^{(k)} &= \frac{2x_k}{f_x h_x} \sum_{i=1}^n x_i f_{ik} h_{ik} \\ S_6 &= \frac{1}{h_x} \sum_{i=1}^n \sum_{j=1}^n x_i x_j h_{ij} \frac{g_i}{g_{ij}} H_i(T_i) D_i \\ S_7 &= \frac{1}{h_x} \sum_{i=1}^n \sum_{j=1}^n x_i x_j h_{ij} \frac{g_i}{g_{ij}} [1 - F_i(T_i)] D_i \\ S_8^{(k)} &= \frac{2x_k}{h_x} \sum_{i=1}^n x_i h_{ik} \end{aligned}$$

$$\text{where} \quad D_i = ([1 - H_i(V_i)][1 - F_i(T_i)] - F_i(V_i) H_i(T_i))^{-1}.$$

RESULTS

Application of the extended corresponding states model in a predictive mode requires a knowledge of a component's shape factors given a relatively minor amount of information, e.g., the critical point parameters and perhaps the acentric factor. To develop such a correlation, one would ideally use high accuracy equations of state and broadly based experimental data for homologous series in conjunction with the results given in Table 1 to calculate "exact" shape factors. These could then be correlated in terms of appropriately reduced variables. Unfortunately, the "super" equations of state which are necessary to perform this mapping are not readily available for refrigerants, especially the HFC's.

Currently, most of the data which are available for refrigerants are for the saturation boundary, *viz.* vapor pressures and saturated liquid densities. According to the results given in Table 1, these properties are related to the reference fluid properties through the equations

$$p_j^s(T) = p_0^s(T/f_j) f_j / h_j \quad (11)$$

and

$$\rho_j^s(T) = \rho_0^s(T/f_j) h_j \quad (12)$$

Simultaneous solution of these two equations provides the equivalent substance reducing ratios along the saturation boundary. Since the saturation boundary only depends upon temperature, all of the density derivatives given in Table 1 are identically zero. As mentioned in the introduction, the reference fluid used in this work is R134a. The equation of state used to represent the reference fluid is the Jacobsen-Stewart [13] modified 32 term BWR equation (MBWR-32). This equation has been used extensively to represent the properties of hydrocarbons and common inorganics. Its functional form is given by

$$p = \sum_{n=0}^9 a_n(T) \rho^n + e^{-\gamma \rho^2} \sum_{n=10}^{15} a_n(T) \rho^{2n-17}$$

and temperature dependence of the $a_n(T)$ is summarized in Table 2. The coefficients for R134a used in this work are given in reference [14].

$a_1 = RT$	$a_9 = b_{19}/T^2$
$a_2 = b_1 T + b_2 T^{1/2} + b_3 + b_4/T + b_5/T^2$	$a_{10} = b_{20}/T^2 + b_{21}/T^3$
$a_3 = b_6 T + b_7 + b_8/T + b_9/T^2$	$a_{11} = b_{22}/T^2 + b_{23}/T^4$
$a_4 = b_{10} T + b_{11} + b_{12}/T$	$a_{12} = b_{24}/T^2 + b_{25}/T^3$
$a_5 = b_{13}$	$a_{13} = b_{26}/T^2 + b_{27}/T^4$
$a_6 = b_{14}/T + b_{15}/T^2$	$a_{14} = b_{28}/T^2 + b_{29}/T^3$
$a_7 = b_{16}/T$	$a_{15} = b_{30}/T^2 + b_{31}/T^3 + b_{32}/T^4$
$a_8 = b_{17}/T + b_{18}/T^2$	

Table 2: Temperature dependence of the MBWR-32 coefficients

Solution of equations (11) and (12) requires that experimental saturation data for the fluids of interest be fit to predetermined functional forms. The vapor

pressures were fit to the Wagner [15] equation

$$\ln \left(\frac{p^s}{p^c} \right) = \frac{a\tau + b\tau^{1.5} + c\tau^3 + d\tau^6}{T_r}$$

and the liquid densities were fit to a function proposed by Ely *et al.* [10]

$$\frac{\rho^s}{\rho^c} - 1 = \frac{k_1\tau^\beta + k_2\tau^2 + k_3\tau^3}{1 + k_4\tau^{1-\beta}}$$

where $T_r = 1 - T/T_c$ and $\tau = 1 - T_r$. In the case where fewer than five experimental vapor pressure or liquid density points were available, the Frost-Kalkwarf vapor pressure equation [16] and/or COSTALD [17] density equation were used. Table 3 summarizes the fluids which have been studied and the experimental data used in this work are summarized in reference [18].

R11	R12	R13	R13B1	R14	R22	R23
R32	R113	R114	R115	R123	R124	R125
R134	R134a	R141b	R142b	R143a	R152a	R218

Table 3: Refrigerants contained in PROZPER

The shape factors which were determined by mapping the saturation boundaries were used to develop generalized correlations which only require a knowledge of the Pitzer acentric factor and critical point constants. The functional forms of these correlations are

$$\theta_i = 1 + (\omega_i - \omega_0)(\alpha_1 + \alpha_2 \ln T_{r_i}) \quad (13)$$

and

$$\phi_i = \frac{Z_0^c}{Z_i^c} [1 + (\omega_i - \omega_0)(\beta_1 + \beta_2 \ln T_{r_i})]. \quad (14)$$

These functional forms were chosen so that the correlated shape factors would extrapolate smoothly into the one-phase region. Figures 1 and 2 illustrate the deviations of the shape factors which were determined along the saturation boundary to the correlations for twelve different refrigerants. The results of the generalization are good for the θ shape factor and less satisfactory for the ϕ shape factor. The only problem apparent with the θ correlation is the "hook" at low reduced temperatures. A redetermination of the reference fluid equation of state incorporating new low temperature data should remove this problem. The ϕ shape factor is highly sensitive to the values of the critical point parameters and its generalization can be improved with the availability of more accurate data. In particular, the systematic offsets for some of the fluids can be directly attributed to the critical compressibility factor ratio which appears in equation (14). Errors of a few dm^3/mol in the critical volume can cause this ratio to change substantially. Finally, the correlation of the ϕ shape factor can also be improved by refitting the saturation boundaries using the correlated (rather than calculated) θ shape factors. The values of ϕ obtained in that manner would then be the optimal values for use in the generalization.

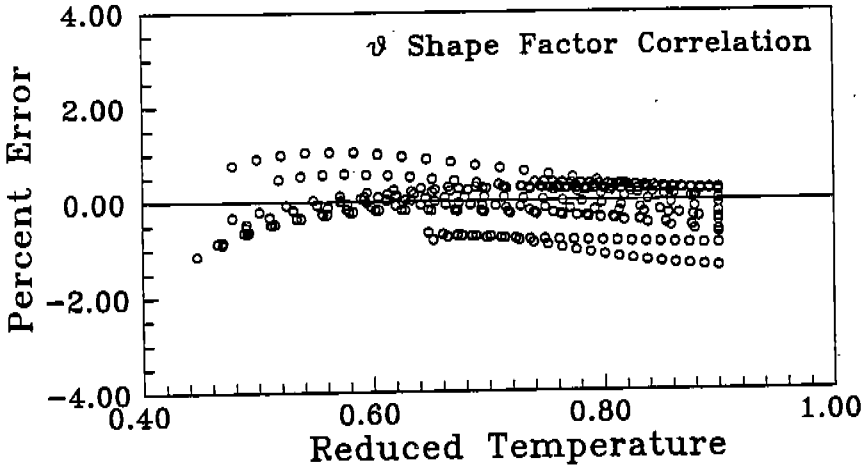


Figure 1: Comparison of generalized and fit v shape factors for twelve refrigerants.

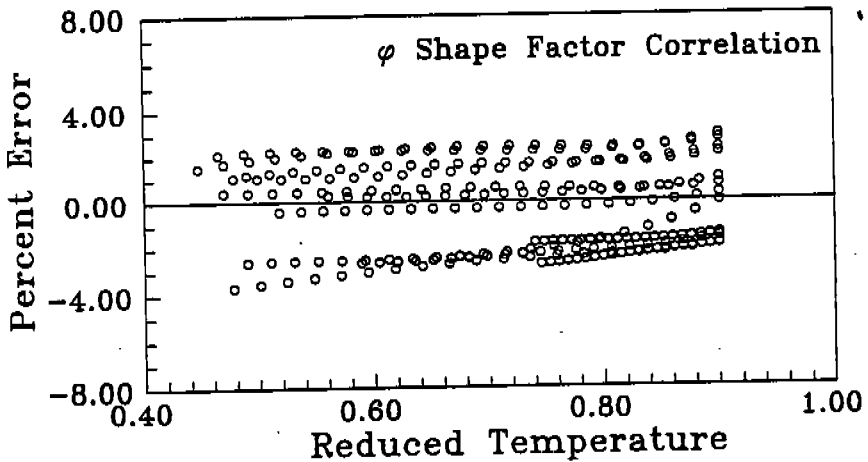


Figure 2: Comparison of generalized and fit ϕ shape factors for twelve refrigerants. The large systematic scatter is associated with uncertainty in the critical points of the fluids.

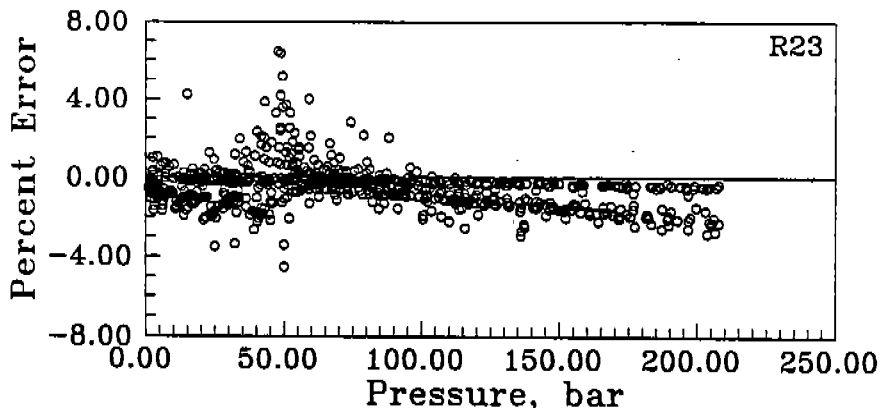


Figure 3: Comparison of experimental and predicted densities of R23. Experimental results are included from four authors with data covering the entire pVT surface.

As an illustration of the application of the model, Figure 3 compares the predicted and experimental densities for R23. This figure contains data from four different experimental sources and covers the temperature range of 130 to 400 K. Thus, results for both the low temperature compressed liquid and dense gas phases are shown. The increased scatter in the vicinity of 50 bar is associated with the critical point. As can be seen, the results are good with relatively little bias and an average absolute error of prediction of less than two percent. The deviations at high pressure are primarily for the low temperature, compressed liquid. Indications are that by improving the reference fluid in this region, the predictions of the model will be improved. Comparisons for other pure fluids and mixtures are in process.

SUMMARY

We have developed a predictive corresponding states program for refrigerants. Using generalized shape factors, the minimum input is the critical point and other material constants such as the molecular weight, normal boiling point, etc. Preliminary comparisons with pure refrigerants have shown the approach to be accurate to within a few percent for the pVT relationship. Work is in progress to further improve and expand the model by refitting the reference fluid and incorporating new data into the model.

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