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A Next Generation Refrigerant Properties Database

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A NEXT GENERATION REFRIGERANT PROPERITES DATABASE[1]

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ABSTRACT

A completely revised version of the REFPROP refrigerant properties database is described. This program is based on the most accurate pure fluid and mixture models currently available. It implements three models for the thermodynamic properties of pure fluids: the modified Benedict-Webb-Rubin (MBWR) and Helmholtz equations of state, and an extended corresponding states (ECS) model. Mixture calculations employ a new model which applies mixing rules to the pure-fluid Helmholtz energies. Viscosity and thermal conductivity are modeled with either fluid-specific correlations or a new variation on the ECS method. These models are implemented in a suite of subroutines written in standard FORTRAN. A separate graphical user interface provides a convenient means of accessing the models. It will generate tables and/or plots for any user-specified mixture. Numerous options to customize the output are available as well as copy and paste capabilities to and from other applications.

INTRODUCTION

Knowledge of the thermophysical properties is essential for the evaluation of alternative refrigerants and the design of equipment using them. The REFPROP computer database from the National Institute of Standards and Technology (NIST) (Huber et al. 1995) has been one of the more widely used tools designed to provide these data. In the initial versions of REFPROP (Gallagher et al. 1993), the intent was to provide data on a wide variety of fluids to allow screening studies of possible replacements for the CFC or HCFC refrigerants. For many of these fluids, only sparse data were available, and, consequently, the database relied primarily on a simple model with few adjustable parameters—the Carnahan-Starling-DeSantis (CSD) equation of state. As the alternative refrigerants move from the laboratory to use in commercial equipment, highly accurate properties are required for a more limited set of fluids. We describe a completely revised REFPROP program (designated as Version 6) based on the most accurate pure fluid and mixture models currently available.

THERMODYNAMIC MODELS

REFPROP 6 calculates the thermodynamic properties using comprehensive equations of state. This approach ensures thermodynamic consistency and allows calculations at all conditions. Other approaches, such as the combination of a vapor-phase model with vapor pressure and liquid density equations may not be applicable in the compressed liquid and supercritical regions and do not always give reliable results for derived properties such as heat capacity and speed of sound.

Pure Fluid Models

Three models are used for the thermodynamic properties of pure components, depending on the availability of data. The first is the modified Benedict-Webb-Rubin (MBWR) equation of state. This model was first proposed by Jacobsen and Stewart (1973) and has been applied to a wide variety of fluids, including hydrocarbons, cryogenic fluids, and refrigerants. It is capable of accurately representing the properties of a fluid over wide ranges of temperature, pressure, and density. The MBWR equation is the basis for the current international standard for the properties of R123 (Younglove and McLinden, 1994).

The MBWR equation expresses pressure as an explicit function of temperature and molar density and is of the form,

$$P = \sum_{n=1}^{9} \alpha_n \rho^n + \exp\left[\left(\rho/\rho^{crit}\right)^2\right] \sum_{n=10}^{15} \alpha_n \rho^{2n-17} \quad , \tag{1}$$

where the α_i are simple functions of temperature resulting in a total of 32 adjustable parameters. For a complete description of the energy quantities (enthalpy, entropy, etc.), the MBWR equation is combined with an expression for the molar heat capacity of the ideal-gas state, that is, vapor in the limit of zero pressure. A form combining polynomial and theoretical terms is used:

$$C_p^{id} = \sum_i c_i T^{t_i} + \sum_k \frac{u_k^2 \exp(u_k)}{\left[\exp(u_k)-1\right]^2} \quad , \text{ with } \qquad u_k = \frac{c_k}{T} \quad . \tag{2, 3}$$

All of the thermodynamic properties can be computed from Eqs. (1–3), as detailed by Younglove and McLinden (1994).

The second high-accuracy pure-fluid equation of state is in terms of reduced molar Helmholtz free energy:

$$a = \frac{A}{RT} = a^{id} + a^r = \ln\delta + \sum_i \alpha_i \tau^{t_i} + \sum_k \alpha_k \tau^{t_k}\delta^{d_k} \exp\left(-\gamma\delta^{l_k}\right) \quad ,$$ (4)

where the first two terms on the right side of Eq. (4) constitute the ideal-gas contribution a^{id}; they are analogous to the combination of the α_1 term in the MBWR equation of state ($\alpha_1 = RT$) and the ideal-gas heat capacity (Eqs. 2 and 3). The second summation is the residual, or real-fluid, contribution a^r. The temperature and density are expressed in the dimensionless variables $\tau = T^*/T$ and $\delta = \rho/\rho^*$, where T^* and ρ^* are reducing parameters which are often, but not always, equal to the critical parameters. The α_i and α_k are numerical coefficients fitted to experimental data and the exponents t_i, t_k, d_i, and d_k are typically determined by a selection algorithm starting with a large bank of terms. The parameter γ is equal to 0 for terms with $l_k = 0$; it is equal to 1 for terms with $l_k \neq 0$. This "Helmholtz model" is the basis for the international standard formulation for R134a (Tillner-Roth and Baehr, 1994).

This model is sometimes termed the "fundamental equation" because it gives a complete description of the thermodynamic properties, as discussed by Tillner-Roth and Baehr (1994), but the MBWR equation of state combined with a C_p^{id} equation is entirely equivalent. The sources for the high-accuracy formulations implemented in the database are summarized in Table 1. The accuracy of these formulations vary, but those for R123 are typical: experimental data are reproduced with average absolute deviations of 0.04 % for densities, 0.05 % for vapor pressures, and 0.75 % for heat capacities.

The third pure-fluid model is the extended corresponding states (ECS) model of Huber and Ely (1994). It is used for fluids with limited experimental data. Simple corresponding states is based on the assumption that different fluids obey, in reduced coordinates, the same intermolecular force law. This assumption leads to the conclusion that, with the appropriate scaling of temperature and density, the reduced residual Helmholtz energies and compressibilities ($Z = p/RT\rho$) of the unknown fluid "j" and a reference fluid "0" (for which an accurate, wide-ranging equation of state is available) are equal:

$$a_j^r\left(T_j, \rho_j\right) = a_0^r\left(T_0, \rho_0\right) \quad , \text{ and} \qquad Z_j\left(T_j, \rho_j\right) = Z_0\left(T_0, \rho_0\right) \quad .$$ (5, 6)

When combined with an expression for the ideal gas heat capacity (such as Eqs. 2 and 3), all other thermodynamic properties can be calculated. The reference fluid is evaluated at a "conformal" temperature and density:

$$T_0 = \frac{T_j}{f_j} = T_j \frac{T_0^{crit}}{T_j^{crit}\theta(T)} \quad , \text{ and} \qquad \rho_0 = \rho_j h_j = \rho_j \frac{\rho_0^{crit}}{\rho_j^{crit}}\phi(T) \quad .$$ (7, 8)

where the multipliers $1/f_j$ and h_j are termed reducing ratios; they are composed of ratios of the critical parameters. Simple corresponding states was developed for spherically symmetric molecules. The ECS model extends the method to other types of molecules by the introduction of the "shape factors" θ and ϕ. These shape factors are taken here to be functions only of temperature. (If data sufficient to fit the density dependence were available, it would be preferable to develop a full equation of state.) The shape factors are fitted to experimental data, typically vapor pressures and saturated liquid densities. The reference fluid is chosen to be chemically similar to the fluid of interest. The fluids represented with the ECS model are listed in Table 2.

Table 1—High-accuracy pure-fluid equations of state currently implemented in REFPROP 6

Fluid	Model	Source	Temperature (K)	Pressure (MPa)	Density (kg/m³)
				Limits of Application	
R11	Helmholtz	Jacobsen et al. (1992)	162.68 (T_{tp}) – 625	0 – 30	0 – 1768
R12	Helmholtz	Marx et al. (1992)	116.10 (T_{tp}) – 525	0 – 200	0 – 1830
R22	Helmholtz	Kamei et al. (1995)	115.73 (T_{tp}) – 550	0 – 60	0 – 1721
R32	MBWR	Outcalt and McLinden (1995)	136.34 (T_{tp}) – 500	0 – 60	0 – 1429
R113	Helmholtz	Marx et al. (1992)	236.93 (T_{tp}) – 525	0 – 200	0 – 1705
R123	MBWR	Younglove and McLinden (1994)	166 (T_{tp}) – 500	0 – 40	0 – 1774
R124	Helmholtz	de Vries et al. (1995)	100 – 400	0 – 40	0 – 1903
R125	MBWR	Outcalt and McLinden (1995)	172.52 (T_{tp}) – 500	0 – 60	0 – 1691
R134a	Helmholtz	Tillner-Roth and Baehr (1994)	169.85 (T_{tp}) – 460	0 – 70	0 – 1591
R143a	MBWR	Outcalt and McLinden (1994)	161.34 (T_{tp}) – 500	0 – 40	0 – 1329
R152a	MBWR	Outcalt and McLinden (1996)	154.56 (T_{tp}) – 500	0 – 60	0 – 1193
R170 (ethane)	MBWR	Younglove and Ely (1987)	90.35 (T_{tp}) – 600	0 – 70	0 – 664
R290 (propane)	MBWR	Younglove and Ely (1987)	85.87 (T_{tp}) – 600	0 – 100	0 – 752
R600 (butane)	MBWR	Younglove and Ely (1987)	134.86 (T_{tp}) – 500	0 – 70	0 – 754
R600a (isobutane)	MBWR	Younglove and Ely (1987)	113.55 (T_{tp}) – 600	0 – 35	0 – 745
R717 (ammonia)	Helmholtz	Tillner-Roth et al. (1993)	195.49 (T_{tp}) – 700	0 – 1000	0 – 900
R744 (CO_2)	MBWR	Ely et al. (1987)	216.58 (T_{tp}) – 500	0 – 40	0 – 1178

Table 2—Fluids currently represented with the extended corresponding states (ECS) model in REFPROP 6

Fluid	Reference Fluid	Fluid	Reference Fluid	Fluid	Reference Fluid	Fluid	Reference Fluid
R13	R12	R115	R12	R142b	R22	RC270	propane
R14	R12	R116	R12	R227ea	R134a	RC318	propane
R23	R134a	R134	R134a	R236fa	R134a	R1270	propane
R114	R12	R141b	R22	R245ca	R134a	RE170	propane

Mixture Model

The thermodynamic properties of mixtures are calculated with a new model which was developed, in slightly different forms, independently by Tillner-Roth (1993) and Lemmon (1996). It applies mixing rules to the Helmholtz energy of the mixture components:

$$a_{mix} = \frac{A_{mix}}{RT} = \sum_{j=1}^{n} x_j \left(a_j^{id} + a_j^r \right) + x_j \ln x_j + \sum_{p=1}^{n-1} \sum_{q=p+1}^{n} x_p x_q F_{pq} a_{pq}^{excess} \quad . \tag{9}$$

This mixing formula may be applied directly to the Helmholtz equation of state. Application to the MBWR equation of state and the ideal gas heat capacity expression used with the MBWR and ECS models requires transformations:

$$a^r = \frac{1}{RT} \int_V^\infty (P - RT\rho) dV \quad , \text{ and} \tag{10}$$

$$a^{id} = \frac{h_{ref}}{RT} - \frac{s_{ref}}{R} - 1 + \ln\left(\frac{T\rho}{T_{ref}\rho_{ref}} \right) + \frac{1}{RT} \int_{T_{ref}}^{T} C_p^{id} dT - \frac{1}{R} \int_{T_{ref}}^{T} \frac{C_p^{id}}{T} dT \quad , \tag{11}$$

where h_{ref} and s_{ref} are an arbitrary reference enthalpy and entropy at the reference state specified by T_{ref} and ρ_{ref}.

The first summation in Eq. (9) represents the ideal solution; it consists of ideal gas (superscript id) and residual or real fluid (superscript r) terms for each of the pure fluids in the n component mixture. The $x_j \ln x_j$ terms arise from the entropy of mixing of ideal gases where x_j is the mole fraction of component j. The double summation accounts for the "excess" free energy or "departure" from ideal solution. The F_{pq} are generalizing parameters which relate the behavior of one binary pair with another; F_{pq} multiplies the a_{pq}^{excess} term(s), which are empirical functions fitted to experimental binary mixture data. The a^r and a_{pq}^{excess} functions in Eqs (9 and 10) are not evaluated at the temperature and density of the mixture T_{mix} and ρ_{mix} but, rather, at a reduced temperature and density τ and δ. These τ and δ are very much in the spirit of the conformal temperature and density of the ECS method and are a key innovation in this model. Several mixing rules for the reducing parameters are used, including

$$\tau = \frac{T^*}{T_{mix}}, \quad \text{with} \quad T^* = \sum_{p=1}^{n} \sum_{q=1}^{n} k_{T,pq} x_p x_q \left(T_p^{crit} T_q^{crit} \right)^{1/2}, \quad \text{and} \tag{12}$$

$$\delta = \frac{\rho_{mix}}{\rho^*}, \quad \text{with} \quad \frac{1}{\rho^*} = \sum_{p=1}^{n} \sum_{q=1}^{n} k_{V,pq} x_p x_q \frac{1}{8} \left[\left(V_p^{crit} \right)^{1/3} + \left(V_q^{crit} \right)^{1/3} \right]^3 \quad . \tag{13}$$

If only limited vapor-liquid equilibrium (VLE) data are available the a_{pq}^{excess} term is taken to be zero, and only the $k_{T,pq}$ and/or $k_{V,pq}$ parameters are fitted. The $k_{T,pq}$ parameter is most closely associated with bubble point pressures, and it is necessary to reproduce azeotropic behavior. The $k_{V,pq}$ parameter is associated with volume changes on mixing. (Ternary and higher order mixtures are modeled in terms of their constituent binary pairs: $k_{T,pq} = 1$ and $k_{V,pq} = 1$ for $p = q$.) If extensive data, including single-phase pressure-volume-temperature (PVT) and heat capacity data, are available, the a_{pq}^{excess} function can be determined. The F_{pq} parameter is used (either alone or in combination with $k_{T,pq}$ and $k_{V,pq}$) to generalize the detailed mixture behavior described by the a_{pq}^{excess} function to other, similar, binary pairs. Lemmon (1996) has determined an a_{pq}^{excess} function based on data for 28 binary pairs of hydrocarbons, inorganics, and HFC's (but using different mixing rules than Eqs. 12 and 13).

This "mixture Helmholtz model" provides a number of advantages. By applying mixing rules to the Helmholtz energy of the mixture components, it allows the use of high-accuracy equations of state for the components, and the properties of the mixture will reduce exactly to the pure components as the composition approaches a mole fraction of 1. Different components in a mixture may be modeled with different forms; for example, a MBWR equation may be mixed with a Helmholtz equation of state. If the components are modeled with the ECS method, this mixture model allows the use of a different reference fluid for each component. The mixture is modeled in a fundamental way, and thus the departure function is a relatively small contribution to the total Helmholtz energy for most refrigerant mixtures. The great flexibility of the adjustable parameters in this model allows an accurate representation of a wide variety of mixtures, provided sufficient experimental data are available.

TRANSPORT PROPERTY MODELS

Pure Fluid Models

The transport properties of viscosity and thermal conductivity are modeled with the residual concept. In this representation, the property χ (representing either viscosity η or thermal conductivity λ) is composed of three contributions:

$$\chi = \chi^{id}(T) + \Delta\chi^{r}(T,\rho) + \Delta\chi^{c}(T,\rho) \ . \tag{14}$$

χ^{id} is a dilute gas term which is a function only of temperature, and $\Delta\chi^{r}$ is a residual term accounting for the behavior at higher densities, including liquid densities. $\Delta\chi^{r}$ is primarily a function of density, but it may also be a function of temperature and density. The thermal conductivity approaches infinity at the critical point, and this critical enhancement is expressed by $\Delta\chi^{c}$. This term is significant for thermal conductivity even quite far from the critical point. For viscosity the enhancement is small except extremely close to the critical point and may be safely ignored in all practical applications. A variety of fluid-specific correlations for viscosity and thermal conductivity, based on the residual concept, have been implemented in the database.

Where fluid-specific correlations are not available, the transport properties are modeled with the extended corresponding states method of Klein et al. (1996). This method is a modification of the ECS model of Huber et al. (1992a, 1992b) and shares many of the concepts of the ECS model for the thermodynamic properties described above. The dilute gas term is modeled using kinetic theory (Hirschfelder et al. 1954). The residual term is scaled to the properties of a reference fluid:

$$\Delta\chi_j^r(T,\rho) = \Delta\chi_0^r(T_0,\rho_0) f_j^{1/2} h_j^{-2/3} \left[\frac{M_j}{M_0}\right]^{1/2} , \tag{15}$$

·where M is molar mass. The determination of the reducing ratios involves an iterative procedure. Eqs. (5) and (6) are iterated to find the f_j and h_j satisfying those two conformal relations. The conformal temperature T_0 is defined in Eq. (7). The conformal density ρ_0 in the residual term (but not the critical enhancement) is further modified from that in the thermodynamic ECS method (Eq. 8) by the introduction of a third shape factor Ψ:

$$\rho_0 = \rho_j \frac{\rho_0^{crit}}{\rho_j^{crit}} \phi(T)\Psi(\rho) \ . \tag{16}$$

The shape factor Ψ is a linear function of density and is determined by a least-squares fit to experimental data. Separate Ψ functions are determined for viscosity and thermal conductivity.

Mixture Model

The viscosity of a mixture is calculated with the extended corresponding states method of Klein et al. (1996):

$$\eta(T,\rho,x) = \eta^{id}(T,x) + \Delta\eta_0(T/f_x,\rho h_x)F_\eta + \Delta\eta^*(\rho,x) \ , \quad \text{with} \quad F_\eta = f_x^{1/2} h_x^{-2/3} g_x^{1/2} \ . \tag{17, 18}$$

The low-density contribution η^{id} is calculated with kinetic theory (Hirschfelder et al. 1954) and $\Delta\eta^*$ is an Enskog-theory hard-sphere correction for size and mass differences given by Ely (1981). The reducing ratios f_x and h_x are determined by applying Eqs. (5) and (6) to the mixture. The "mass shape factor" for the mixture, g_x, is determined by mixing rules applied to the component mass shape factors, defined as

$$g_j^{1/2} = \frac{\Delta\eta_j(T_j,\rho_j)}{\Delta\eta_0(T/f_j,\rho h_j)} f_j^{-1/2} h_j^{2/3} \ . \tag{19}$$

The residual viscosity of component j in Eq. (19) is evaluated, using either a fluid-specific correlation or the pure-fluid ECS model described above, at the conformal conditions

$$T_j = \frac{T_{mix} f_j}{f_x} \quad \text{and} \qquad\qquad \rho_j = \frac{\rho_{mix} h_x}{h_j} \ . \tag{20, 21}$$

This formulation yields the pure-fluid values exactly as the composition approaches a mole fraction of 1. A analogous method is used for thermal conductivity. For the transport properties, R134a is used as the reference fluid for all mixture components.

PROPERTY SUBROUTINES

The property models described above are implemented as a suite of FORTRAN subroutines. These routines have been completely rewritten from earlier versions of REFPROP. Source code is provided with the database so that users may link the property routines with their own application. ("Translation" routines will allow applications written using the subroutine calls

of earlier versions of REFPROP to use the new subroutines.) The routines are written in ANSI-standard FORTRAN 77 and are compatible with FORTRAN 90. They are written in a structured format, are internally documented with extensive comments, and have been tested on a variety of compilers.

The fluid or mixture of interest is specified with a (required) call to the subroutine "SETUP." This routine reads the coefficients to the NIST-recommended models for that fluid. Alternative property models and/or nonstandard reference states may be specified by calls to additional (optional) setup routines. Routines are provided to calculate thermodynamic and transport properties and surface tension at a given (T, ρ, x) state. Iterative routines provide saturation properties at a specified (T, x) or (P, x) state. Flash calculations calculate single- or two-phase states at specified (P, h, x), (P, T, x), etc.

The routines mentioned above are independent of the model. Underlying these routines are sets of "core" routines for each of the models implemented in the database. Each such set is highly modular and is contained in a separate file. Coefficients needed for a particular model are stored in common blocks. but these commons are referenced only by routines in the same file. These sets of subroutines, thus, resemble "units" in the Pascal language with clearly demarcated "interface" and "local" declarations. This structure is intended to simplify the addition of future models to the database and will make such additions almost totally transparent to the user.

Numerical coefficients to the property models are stored in text files. There is one file per fluid and one file containing coefficients for the mixture departure functions. These files are read (once) upon the call to SETUP. Fluids can be added to or deleted from the database without recompilation.

USER INTERFACE

A graphical user interface has been developed for REFPROP 6 independent of the code implementing the algorithms. The interface provides a convenient means to calculate and display thermodynamic and transport properties. It is written for the Windows™ operating system. (Mention of commercial products is to fully specify the database and does not constitute endorsement by NIST or imply that they are necessarily the best suited for the purpose.) The interface is written in Pascal; it accesses the FORTRAN property subroutines via a dynamic link library. Screen shots of the interface program are shown in Figures 1 and 2. The program is controlled through the use of the following pull-down menus (visible at the top of Figure 1):

File provides commands to save and print generated tables and plots. Individual items or entire sessions with multiple windows may be saved or recalled. The standard "print setup" and "quit" commands are also present.

The Edit menu provides copy and paste commands which allow selected data to be exchanged with other applications.

The Options menu provides commands for selecting the unit system, properties of interest, and the reference state. These options may be stored for recall at a later time. A user-defined set of preferences is loaded upon program startup.

The pure fluid or mixture of interest is specified with commands in the Substance menu. Most of the refrigerant mixtures of current commercial interest (those having an ASHRAE R400 or R500-series designation) are predefined. In addition, new mixtures can be specified and saved by combining up to five of the pure components listed in Tables 1 and 2.
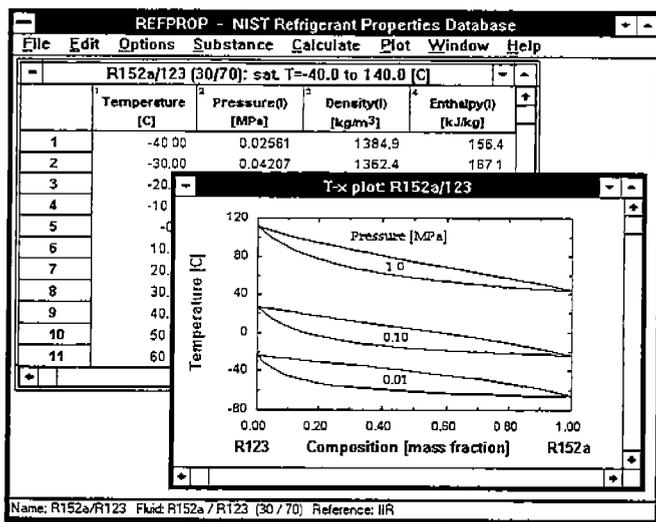
Figure 1. Screen shot of interface showing a typical data table and temperature-composition diagram.
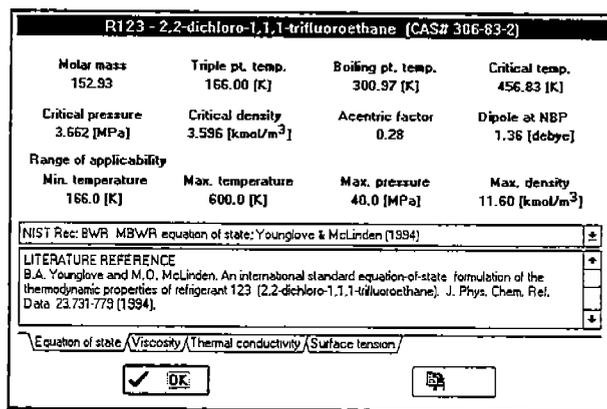
Figure 2. Pure-component information screen available from the interface.

The <u>Calculate</u> menu initiates the calculations that generate a property table. Each property selected for display is shown in a separate column of the table. Two types of tables are provided. The first type, such as that shown in Figure 1, provides properties at saturation or with a property (such as temperature or pressure) held constant with another selected property varying over a specified range. The second type allows the user to select the independent variables. Values of the independent variables may then be entered with the keyboard, read from a file, or pasted from another application.

The <u>Plot</u> menu provides publication-quality x-y plots of any variables appearing in a table. In addition, temperature-entropy, pressure-enthalpy, temperature-composition and pressure-composition diagrams may be generated automatically. Controls are provided to modify the plot size, axis scaling, plot symbols, line type, legend, and other plot features.

Each table or plot appears in a separate window and can be accessed, resized, or retitled with commands in the <u>Window</u> menu. The number of windows is limited only by available memory.

A complete online-help system can be accessed through the <u>Help</u> menu.

A status line at the bottom of the screen displays the currently specified mixture, composition, and reference state. Clicking on the status line will call up a screen for each of the components providing documentation for fluid constants, the source of the models, and their range of applicability, as shown in Figure 2.

CONCLUSIONS

The new REFPROP 6 database implements a variety of high-accuracy models for the thermodynamic and transport properties of refrigerants and their mixtures. These models are implemented as a suite of FORTRAN 77 subroutines written in a modular fashion; this structure will facilitate the incorporation of additional fluids and future models. A graphical user interface provides a convenient means of accessing the models and producing tables and plots of any specified mixture.

ACKNOWLEDGEMENTS

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