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# Investigation on CFC Diffusivity with an Improved Double Volume Method

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REFPROP: A Thermodynamic Properties Software Program  
for  
Refrigerants and Their Mixtures

by

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ABSTRACT:

This document introduces the computer software package, REFPROP [OSRD, 1990]. It describes the origin of the package, briefly describes the contents and output of the package, lists the data that have led to the equation of state parameters, and summarizes the precision of the fit of the correlation to the input data for R123.

I - INTRODUCTION

The software package, REFPROP, has been designed to produce tables of thermodynamic properties for 15 partially and fully halogenated chlorofluorocarbon refrigerants and 20 of their mixtures. As more data on both pure materials and mixtures become available, these data will be incorporated into the package.

REFPROP has its origins in a project begun at NBS (now NIST) in 1982 to produce an easily-used equation of state package to describe refrigerant mixtures. This work was motivated by an interest in the use of mixtures in domestic heat pumps [1985]. Historically, only azeotropic mixtures have ever been marketed, and then for specialized applications, such as R502, an azeotropic mixture of R22 and R115 used in supermarket open-case freezer systems. Azeotropes have the feature that their phase behavior is similar to pure materials; hence, these mixtures were treated as if they were pure materials. The use of mixtures, particularly where azeotropy was not a consideration, represented a major departure from the traditional materials' applications in the refrigeration industry. The Montreal Protocol, an international agreement to curtail and, eventually, to eliminate the use of several widely-used, fully-halogenated chlorofluorocarbons, in particular R11 and R12, gave further impetus to having tables of thermodynamic properties for a huge range of pure materials and their mixtures, particularly for screening applications. Many of these materials were represented in the literature by only a few data; hence, the detailed tabulations that existed for some of the major refrigerants, such as R22 [JAR, 1975], could not be produced despite the need to have such information for performance evaluation.

REFPROP was designed to provide thermodynamic property tables for design and comparison of different materials particularly where little experimental information exist. Section II of this paper describes the equation of state briefly and discusses its advantages and flaws. Section III presents a synopsis of the routines that generate the tables, and additional routines included with the package. Section IV reviews the data that were used to generate the equation of state parameters. Finally, Section V reviews ability of REFPROP to represent the properties of R123 as a representative example of the performance of the package.

II - THE EQUATION OF STATE

The thermodynamic properties tables in REFPROP are calculated using a modification of the hard sphere equation of state. Carnahan and Starling [1972] reported briefly on this modification in a review paper that characterized the equations of state that arose when their representation for the hard-sphere fluid was substituted into the many modifications of the van

der Waals equation of state. DeSantis et al. [1976] used this same form to describe the phase behavior of mixtures of simple fluids. This modification, referred to as the Carnahan-Starling-DeSantis (CSD) equation of state, given in equation 1, was chosen because it addressed needs in representing the

$$\frac{p V}{R T} = \frac{(1 + y + y^2 - y^3)}{(1 - y)^3} - \frac{a}{R T (V + b)} \quad [1]$$

$$\text{where } y = b / 4 V$$

properties of refrigerants. First, it was founded on a physical model, the fluid of hard spheres, that could be expected to describe the liquid state more correctly than equations using the traditional van der Waals term for the excluded molecular volume. Second, it was a closed mathematical form with few adjustable parameters. The physical foundation and this inherent simplicity have several advantages. The parameters are shown in equation 2; although

$$a = a_0 \exp(a_1 T + a_2 T^2) \quad [2a]$$

$$b = b_0 + b_1 T + b_2 T^2 \quad [2b]$$

they have a temperature dependence, they retain a physical interpretation. The equation can be used to correlate small sets of diverse thermodynamic information. Finally, the equation can be used with the assurance that a predicted property, while having limited accuracy because of its origin in a limited data set, will not be unphysical. The simplicity of the equation also leads to two other features: the retention of a physical meaning in the equation of state parameters allows it to be applied to mixtures in a straightforward manner as shown in equation 3; and the simplicity of its

$$a = a_1 x_1^2 + 2(1 - f_0)(a_1 a_2)^{1/2} x_1 x_2 + a_2 x_2^2 \quad [3a]$$

$$b = x_1 b_1 + x_2 b_2 \quad [3b]$$

topology lends a robustness to numerical solutions of its roots. An extended discussion of the use of this equation of state for refrigerants can be found in several publications by Morrison and McLinden [1985, 1986a].

The advantages of this equation of state lies in its simplicity; its shortcomings are also rooted in that same feature. Morrison and McLinden [1985b] have shown that saturation data alone - liquid and vapor density and vapor pressure - produce nearly the optimal function for correlating all the thermodynamic properties. They have also shown that the equation does not represent the region near the critical point well, a feature of all so-called "classical" equations of state, and that the liquid densities deviate in a systematic way at high pressures, tending to be too high at a given pressure. Neither of these deficiencies affects the application to refrigeration cycles, however. Typically, one must resort to significantly more complicated functions to eliminate these problems. The data sets for many of the materials included in REFPROP do not justify correlation with functions with more complexity than the CSD equation of state. Inevitably, materials, for which there are large sets of data, will be represented by complex equations of state. Nevertheless, the significance of a simpler representation, such as the one found in REFPROP, is that all the materials of interest, ones hardly studied as well as ones well-studied, are represented on an "even footing" and can be legitimately compared.

### III - SOFTWARE AND ROUTINES

The equation of state and its extension to mixtures described in Section II has been implemented as a FORTRAN program and subroutines capable of running on a personal computer. The calculation of the equilibrium thermodynamic properties of the fluid from the equation of state are made through a series

of subroutines as described in NBS Technical Note 1226, along with the main program and three subroutines which allow the user, through a dialog process, to specify the desired calculations and to set up the parameters and calls to the property subroutines. All calculations are in double precision to insure enough accuracy with a word length of 32 bits.

A brief description of the interactive subroutines that the user encounters and their functions are as follows:

Subroutine Choice sets up the system of units desired by the user. The user has the opportunity to choose a complete set of units with a single input from a selection of commonly used systems, including SI, the customary engineering system, and a system used often by chemists. The user may also choose the units for temperature, energy, volume, and amount separately. The assumed units for composition will be mass fractions if mass units are chosen for the volume and mole fractions if molar units are chosen for the volume. This subroutine, after the choices have been made, establishes the proper conversion factors and the proper unit names to be used in the table headings, etc.

Subroutine Dialog presents the list of fluids for which parameters are available and asks the user to choose the fluid(s) of interest. Calculations could be made for a pure fluid or a binary mixture in the first version of REFPROP; the user now has the option of entering equation of state parameters for a material not included in the package library as one component in the calculation and for calculating the properties of mixtures with as many as five components. The mixture interaction parameter,  $f_0$ , has been calculated for a number of two-component mixtures from experimental information; these combinations are listed. The program operates on the assertion that there will be only vapor and single-phase liquid solutions to the equation of state. More complex phase behavior that might be encountered with lubricants has not been incorporated into the phase equilibrium solution algorithm. The user may supply the value of a mixture interaction parameter when a mixture is chosen for which there is not a previously stored value.

Subroutine Dialog2 is now called to query the user as to the type of calculations and independent variables to be desired. The available choices are tables at constant 1) temperature, 2) pressure, 3) volume, 4) entropy, 5) enthalpy, or dew-point and bubble-point values at increments of 6) temperature or 7) pressure. When an option from choices 1 through 5 is chosen, the user is asked to enter the value of the constant quantity and then, as in all the options, the range of the other independent variable (temperature or pressure) as the initial and final values and the increment. An additional feature, offered when the running variable is pressure, is to allow pressure to increase "exponentially" with table entries at 1, 2, and 5 times powers of ten within the specified range. Table entries in the region just below the critical pressure, where many thermodynamic properties vary rapidly, will be more closely spaced when the exponential mode is chosen. In all the choices, when a phase boundary is crossed, a line is inserted containing the values of the properties at the phase boundary. When calculations are for a mixture, the composition of the coexisting phase at the phase boundary is also printed. Should the values of the independent variables specify a state point within the two-phase region, the isobaric heat capacity and the speed of sound are not calculated as they are undefined in the two-phase region; instead, the value of the quality, the fraction of the fluid in the vapor phase, is printed.

In addition to the three user interactive subroutines described above, the FORTRAN source code for the following functions and subroutines are also included in this package for the user who wishes to implement this model as part of his own design package:

BLOCK BDESC contains the equation-of-state parameters for each of the refrigerants included in the library.

SUBROUTINE ESPAR (IOPT,T,X,A,B) calculates the temperature-dependent values of "a" and "b" for the desired mixture as a function of T and x.

SUBROUTINE ECONST (I1,I2,F0,F1) accesses the equation of state parameters for refrigerants identified by the index numbers I1 and I2, and the value of the mixture interaction parameter, F0, for the chosen mixture (F1 has not been implemented at present) and makes them available to other routines. Reference values for the enthalpy and the entropy are also calculated at this point.

SUBROUTINE CRITX (X,TC,PC,VC) estimates the value of the critical point predicted by the equation of state for the chosen mixture and composition.

FUNCTION PVT(T,V,X) performs the basic equation of state calculation of pressure as a function of T, V, and x.

SUBROUTINE HCVCP (IOPT,T,V,X,H,CV,CP,SND) calculates values of the enthalpy, heat capacities, and speed of sound as functions of T, V, and x. IOPT allows the calculation of only H if desired.

FUNCTION ENTROP T,V,X) calculates the value of the entropy as a function of T, V, and x.

SUBROUTINE VIT (T,P,A,B,V,LLIQ,LCON) performs the iteration to obtain the volume when the temperature and pressure are given. The values of the equation of state parameters "a," and "b" must be supplied. LLIQ and LCON are logical variables: set LLIQ = .TRUE. for a liquid point and = .FALSE. for a vapor point. LCON is set = .TRUE. by the subroutine if the iteration has converged.

SUBROUTINE BUBLT (T,XL,XV,PSL,VL,VV,LPHASE,LCRIT) and SUBROUTINE BUBLP (P,XL,XV,TSL,VL,VV,LPASE,LCRIT) determine, through an iteration to find the equality in chemical potentials in each phase, the dew- and bubble-points of the specified mixture.

SUBROUTINE HPIN (H,P,TSAT,HSAT,VSAT,T,V,LLIQ) is also included for convenience, performing the iteration necessary to find the temperature and volume corresponding to a specified enthalpy, pressure, and composition.

SUBROUTINE SPIN (S,P,X,TSAT,VSAT,T,V,LLIQ) operates in a way similar to HPIN with the specification of the entropy rather than enthalpy.

#### IV. DATA SOURCES

This Section reviews the data that were used to evaluate the six equation of state parameters associated with the CSD equation of state and the additional three parameters for the ideal gas heat capacity. The heat capacity data are necessary to generate a complete thermodynamic surface; without these values, only states at the same temperature can be compared with one another.

The minimum set of information necessary to evaluate the coefficients for "a" and "b" is saturation information. As previously mentioned, these data alone produce nearly the optimal fit for the CSD equation of state. Since the location of the saturation boundary is so important a feature in cycle design, even where more extensive data exist, the saturation data were weighted heavily. The parameters were chosen to optimize the fit to the data. For example, when saturation data alone were used, values of "a" and "b" were determined to give the best fit to liquid and vapor densities and the vapor pressure. In principle, only two of these three quantities are actually necessary to satisfy the thermodynamic requirements of equating the pressure and the Gibbs free energy. When larger sets of data were available, values of "a" and "b" were determined that optimized the fit to the data by means of a non-linear regression routine from the IMSL Statistical Library [1937].

The values of the ideal gas heat capacities came from a variety of sources. In nearly all cases, the values were calculated from spectroscopic information. The ideal gas heat capacities were fit to the following function:

$$C_p / R = c_0 + c_1 T + c_2 T^2 \quad [4]$$

Although the value of the heat capacity has a more complicated functional form, the quadratic fit was always found to be accurate to 0.01% in the prescribed range of application.

Mixtures are characterized by the quantity  $f_0$ . The optimal value of this quantity was chosen to be consistent with the data that exist. In most cases, there were only bubble pressure/liquid composition data. In a few cases, there were also other information such as liquid densities, and the composition and densities of the coexistent vapor phase. Morrison and McLinden [1986b] have shown the value of  $f_0$  to be clearly composition dependent; that feature of the behavior has not been implemented in REFPROP. Evidence for variation with temperature is sketchy at best, even for the most extensive sets of mixture data; the values of  $f_0$  were then assumed to be temperature independent. Other properties, such as virial coefficients for the mixture or the location of an azeotropic composition were also used to determine  $f_0$ .

The following is a listing of the data sources and the character of those data that were correlated with the CSD equation of state:

#### PURE MATERIALS

- R11: ASHRAE (1980).  
Ideal gas heat capacity: JANAF (1970).
- R12: ASHRAE (1980).  
Ideal gas heat capacity: JANAF (1970).
- R13: ASHRAE (1980).  
Ideal gas heat capacity: JANAF (1970).
- R13B1: ASHRAE (1980).  
Ideal gas heat capacity: JANAF (1970).
- R14: ASHRAE (1980).  
Ideal gas heat capacity: JANAF (1970).
- R22: Vapor pressure: A. V. Kletskii (1964); R. Kohlen, H. Kratzke, and S. Muller (1985).  
Liquid and vapor volumes: R. Kohlen, H. Kratzke, and S. Muller (1985); A. Kumagai and H. Iwasaki (1978).  
Enthalpy of vaporization and liquid heat capacity: E. F. Neilson and D. White (1957).  
Vapor heat capacity: G. Ernst and J. Busser (1970); K. Bier, G. Ernst, and G. Maurer (1974).  
Ideal gas heat capacity: JANAF (1970).
- R23: ASHRAE (1980).  
Ideal gas heat capacity: JANAF (1970).
- R113: Vapor pressure, liquid densities, vapor phase p-v-T, and critical temperature: M. J. Mastroianni, R. F. Stahl, and P. N. Sheldon (1978).  
Ideal gas heat capacity: Thermodynamics Research Center Data Project (1986).
- R114: Saturation densities, liquid and vapor, and pressures: W. H. Mears et al. (1955).  
Ideal gas heat capacity: Thermodynamics Research Center Data Project (1986).
- R123: Saturation densities, liquid and vapor, and pressures, critical pressure, temperature, and density: L. A. Weber and J. M. H. Levelt Sengers (1989).  
Ideal gas heat capacity: Thermodynamic Research Center Data Project (1986).
- R124: Saturation densities, liquid and vapor, and pressure: H. Kubota et al. (1988).  
Ideal gas heat capacity: R. Bender, K. Bier, and G. Maurer (1980).
- R134: Saturation densities, pressures, critical pressure, critical temperature, critical density: D. K. Ward, G. Morrison (1989).  
Ideal gas heat capacity from spectroscopic data: Klabo, P. and J. R. Nielsen (1960).60).
- R134a: Saturation densities, liquid and vapor, and pressures: R. S. Basu and D. P. Wilson (1989); D. K. Ward and G. Morrison (1989).

- Saturation densities, vapor, and vapor pressure: L. A. Weber (1989).  
 Critical temperature, pressure, and density: D. K. Ward and G. Morrison (1989).  
 Ideal gas heat capacity: S. S. Chen, A. S. Rogers, J. Chao, R. C. Wilhoit, and B. J. Zwolinski (1975).
- R142b: Saturation densities, liquid and vapor, and pressure: W. H. Mears, et al. (1956).  
 Ideal gas heat capacity: Thermodynamic Research Center Data Project (1985).
- R152a: Saturation densities, liquid and vapor, and pressure: W. H. Mears, et al. (1956). Y. Higashi, et al. (1987).  
 Compressed liquid: A. Iso and M. Uematsu (1989).  
 Ideal gas heat capacity: S. S. Chen, A. S. Rogers, J. Chao, R. C. Wilhoit, and B. J. Zwolinski (1975).

MIXTURES: for all mixtures, the refrigerant with the lower numerical designation is listed first.

- R11 + R12: bubble point properties. N. D. Loi (1983).  
 R11 + R22: bubble point properties. M. Meskel-Leavre, D. Richon, and H. Renon (1982).  
 R12 + R13: bubble point properties. J. Mollerup and A. Fredenslund (1976).  
 R12 + R13B1: vapor-liquid equilibria. E. G. Wright (1985).  
 R12 + R22: composition and normal boiling temperature. B. J. Eiseman (1957).  
 R12 + R114: bubble point properties. M. Kuver (1983).  
 R12 + R142b: vapor-liquid equilibria. V. A. Nikol'skii, O. V. Baklan, and N. I. Artemenko (1983).  
 R12 + R152a: azeotrope composition. W. A. Pennington (1952).  
 R13 + R14: vapor-liquid equilibria. W. L. Kubic and F. P. Stein (1981).  
 R13 + R23: bubble point properties. F. P. Stein and P. C. Proust (1971).  
 R13B1 + R22: bubble point properties. A. P. Kuznetsov (1972).  
 R13B1 + R114: bubble point properties. Kuver (1986).  
 R13B1 + R152a: bubble point properties. G. Morrison and M. O. McLinden (1985).  
 R14 + R23: vapor-liquid equilibria. A. Piacentini and F. P. Stein (1967).  
 R22 + R114: bubble point properties. G. Hackstein (1976); A. Valtz, S. Laugier, and H. Richon (1987).  
 R22 + R142b: bubble point properties. A. Valtz, S. Laugier, and H. Richon (1987).  
 R134 + R134a: liquid density and composition and bubble point pressure. D. K. Ward and G. Morrison (1989).  
 R134 + R152a: liquid density and composition and bubble point pressure. D. K. Ward and G. Morrison (1989).  
 R134a + R152a: liquid density and composition and bubble point pressure. D. K. Ward and G. Morrison (1989).

#### V - DATA REPRESENTATION

As indicated in the previous section, the extent and variety of data depend upon the material. In this section, the properties of R123, as represented by REFPROP, will be examined. The values of the equation of state parameters were determined exclusively from the saturation properties listed in the bibliography between 273 K and 423 K. The agreement between the saturation curve correlation and the saturation properties are shown in Figure 1. Since the determination of those quantities, a large collection of p-V-T information on this material has become available; indeed, the amount of information is so large that the thermodynamic properties have been correlated to a complex, multi-parameter equation of state [McLinden et al., 1989].

Figure 2 shows the difference between the calculated density at a given temperature and pressure and the experimental density. The densities in the liquid phase were measured by Morrison and Ward [1990] from 278 K to 368 K to a pressure of 3800 kPa. The densities in the vapor phase were derived from Burnett measurements made by Weber [1990]. Those measurements range from 358 to 453 K and from 330 to 1960 kPa. The densities in the vapor phase are systematically below the measured values; whereas the opposite is true in the

liquid phase. The figure shows, however, that below 1200 kPa the equation of state represents the properties of R123 to better than +/- 0.5%. The deviations at higher pressures are the result of the poor representation of the critical point, particularly for the vapor, and the systematic over prediction of the compressibility by the CSD equation of state in the liquid phase. Figure 1 does show that with the few parameters the CSD equation has, it quantitatively represents the properties in the range of operation.

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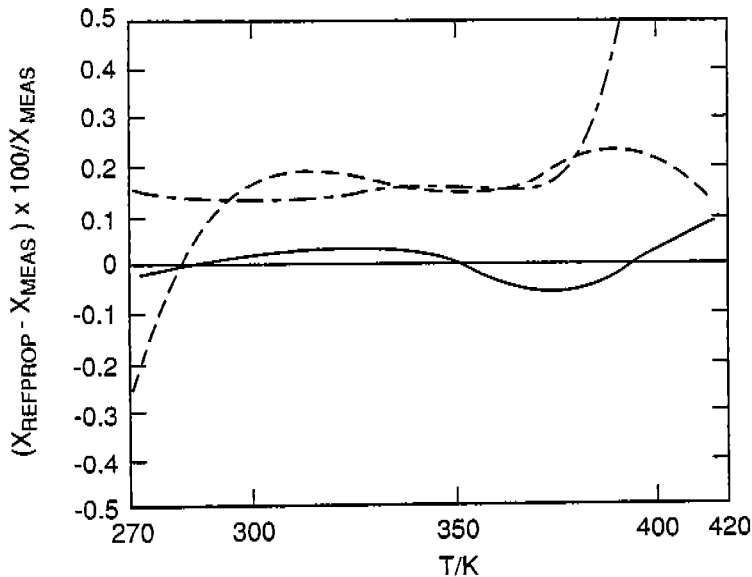


Figure 1: The deviation of the correlated saturation properties from the measured properties for R123. Solid line - saturated liquid density; dashed line - vapor pressure; double dashed line - saturated vapor volume.

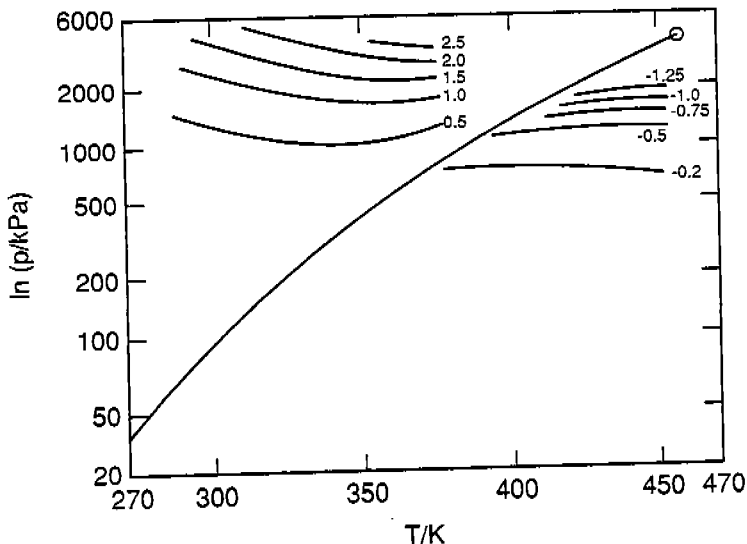


Figure 2: The deviation (in per cent) of the CSD predicted density from the measured density for R123.