The Viscosity and Thermal Conductivity Coefficients of Gaseous and Liquid Fluorine

H. J. M. Hanley² and R. Prydz³

Cryogenics Division, Institute for Basic Standards, National Bureau of Standards, Boulder, Colorado 80302

Tables of values for the viscosity and thermal conductivity of fluorine are presented in the range 70-300 K for pressures up to 200 atmospheres. Experimental results were reviewed but were judged to be unreliable. Accordingly, dilute gas values were determined from kinetic theory using the m-6-8 potential, and dense gas and liquid values were obtained from the modified Enskog theory. The critical point anomaly in the thermal conductivity coefficient is also discussed.

Key words: Critically evaluated data; fluorine; kinetic theory; modified Enskog theory; thermal conductivity; viscosity.

1. Introduction

Fluorine is so toxic and reactive that its physical properties are difficult to measure, but it is potentially an important cryogenic fluid and such properties are needed. In this paper we examine the transport properties of fluorine and present tables of the viscosity and thermal conductivity coefficients. Because, as we will discuss, the experimental data are generally unreliable, the tables cannot be regarded as definitive; nevertheless we believe they represent the best values one can obtain at this time.

2. Data

The experimental situation was first investigated, and it was apparent at once that the data available were scarce and scattered. A literature search carried out by the Cryogenic Data Center, National Bureau of Standards, Boulder, yielded the following experimental references applicable for temperatures less than 300 K: liquid viscosity [2],4 liquid thermal conductivity [19], dilute gas viscosity [4, 10], and dilute gas thermal conductivity [3]. We plot data from these references in figures 1 and 2.5

We evaluated the data as follows: the viscosities of Kanda, reference [10], cannot be considered reliable. From comparisons between the results of several properties measured by Kanda (PVT, dielectric constant,

surface tension) and more recent work [2, 14], we have concluded that the fluorine used by Kanda was impure. We also have to reject the thermal conductivity values

of reference [19]. The data were intended to be taken

close to the saturated liquid boundary but the pressures

reported at the various temperatures do not seem plau-

sible. Our opinion is that either the temperature control

in the experiment was inadequate, or hydrogen fluoride

was present in the fluorine. Based on an examination of

references [3] and [4], and our experience of the work of

Frank, we place an error estimate of five percent on the

data reported therein. It is difficult to judge the reliability of the data quoted in reference [2] since the experi-

mental procedure is not reported in sufficient detail.

Transport coefficients will be discussed in three sections corresponding to the dilute gas, the dense gas and liquid, and the region around the critical point. The experimental range covered is from 70 to 300 K for pressures up to 200 atmospheres.

This equation of state plays an essential role in our

predictive procedure.

3. Dilute Gas

Dilute gas coefficients form the basis for transport property calculations for the entire gas and liquid phases. In the absence of reliable data, the most practical way to determine them is to use kinetic theory.

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In summary, the experiments on the transport properties are clearly limited, and we considered it essentially impossible to base a correlation over a significant range of temperature and pressure on the available data. Accordingly, we decided to construct tables from predictive techniques only. This decision was also influenced by the fact that Prydz and Straty [14] have recently measured several equilibrium properties to a high degree of precision over a wide pressure and temperature range, from the triple point to 300 K for pressures up to 21 MN/m² (~200 atm). An equation of state was derived.

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To whom correspondence should be addressed.

Present address: Norsk Hydro a.s., Bygdsyalle 2, Oslo 2, Norway.

Numbers in brackets refer to literature references in the last section of this

paper.

⁵ Figures have been placed at the end of this paper.

3.1. Viscosity

The kinetic theory equation for the viscosity is [8]:

$$\eta_0 = \frac{5}{16} \frac{(\pi m k T)^{1/2}}{\pi \sigma^2 \Omega^{(2,2)*}},\tag{1}$$

where m is the weight of a molecule, k is Boltzmann's constant, and T the temperature in Kelvin. The quantity $\Omega^{(2,2)*}$ is a dimensionless collision integral which takes into account the dynamics of a binary collision and is characteristic of the intermolecular potential of the colliding molecules. For a given potential, $\Phi(r)$, with an energy parameter ϵ (defined as the value of $\Phi(r)$ at the maximum energy of attraction) $\Omega^{(2,2)*}$ can be determined as a function of reduced temperature T^* :

$$T^* = T/(\epsilon/k). \tag{2}$$

The parameter σ is a distance parameter, also characteristic of the intermolecular potential, which approximates an effective hard sphere diameter, and is the value of r when $\Phi(r)=0$. The specific relationship between $\Omega^{(2,2)*}$ and $\Phi(r)$ is as follows. We define a parameter g^* as the reduced relative kinetic energy of two colliding molecules: $g^* = \frac{1}{2} \mu g^2 / \epsilon$, where μ is the reduced mass and g the relative velocity. A parameter b is defined as the distance of one molecule from the direction of approach of another before collision.

If r is the intermolecular separation and r_c the distance when the molecules are closest, we can show that the angle of scatter, χ , after a collision is related to the potential by

$$\chi = \pi - 2b^* \int_{r_c *}^{\infty} dr^* / r^{*2} \left[1 - \frac{b^{*2}}{r^{*2}} - \frac{\Phi^*}{g^{*2}} \right]^{-1/2}, \quad (3)$$

where the variables are reduced according to the relations: $b^*=b/\sigma$, $r^*=r/\sigma$, $r^*_{\circ}=r_{\circ}/\sigma$, $\Phi^*=\Phi/\epsilon$. Integration of χ over all values of b^* produces the cross section, Q^* ,

$$Q^*(g^*) = 3 \int_0^\infty (1 - \cos^2 \chi) b^* db^*. \tag{4}$$

 $(Q^*$ is dimensionless and has been reduced by the corresponding value for molecules interacting with a hard sphere potential.) Finally, $\Omega^{(2,2)*}$ is obtained by integration of Q over all values of g^* ,

$$\Omega^{(2,2)*}(T^*) = \frac{1}{3T^{*4}} \int_0^\infty \exp\left(-g^{*2}/T^*\right) g^{*7} Q(g^*) dg^*.$$
 (5)

A full discussion on these equations is given in reference [9].

3.2. Thermal Conductivity

The kinetic theory expression for a polyatomic gas used by us is the expression derived by Mason and

Monchick [13]:

$$\lambda_0 = \lambda'_0 + \rho D_0 c_v'' - \frac{2c_v''}{\pi Z} \left(\frac{5}{2} - \frac{\rho D_0}{\eta_0} \right) \eta_0, \tag{6}$$

where

$$\lambda'_0 = \frac{15}{4} \frac{k}{m} \eta_0. \tag{7}$$

In equation (6), c_v " is the internal ideal gas specific heat per molecule, Z the rotational collision number (defined as the number of collisions needed to relax the rotational energy to within 1/e of its equilibrium value, where e is the natural logarithm base), and ρD_0 is the product of the self-diffusion coefficient and the density, ρ , which can be obtained from the expression:

$$\rho D_0 = \frac{3}{8} \frac{(\pi m k T)^{1/2}}{\pi \sigma^2 \Omega^{(1,1)*}}.$$
 (8)

Here $\Omega^{(1,1)*}$ is the collision integral for diffusion, similar to equation (5).

3.3. The Intermolecular Potential Function, Φ (r)

It is apparent from equations (1–8) that, given c_r " and Z, the calculations for the viscosity and thermal conductivity coefficients require the function $\Phi(r)$ to be known. Unfortunately, obtaining $\Phi(r)$ for a fluid presents a problem: except for the very simplest systems, $\Phi(r)$ has to be based on a model of the intermolecular interaction and so uncertainty is inevitably introduced into kinetic theory or statistical mechanical calculations. Nevertheless, model functions are often all that one requires if they are employed carefully. For example, a recent function, proposed by Klein and Hanley [11], has been found to be very useful. The function is called an m-6–8 and has the form:

$$\begin{split} \Phi(r)/\epsilon &= \frac{1}{m-6} \left[6 + 2\gamma \right] \left(\frac{d}{r^*} \right)^m \\ &- \frac{1}{m-6} \left[m - \gamma (m-8) \right] \left(\frac{d}{r^*} \right)^6 - \gamma \left(\frac{d}{r^*} \right)^8, \quad (9) \end{split}$$

where $d=r_m/\sigma$. The potential given by equation (9) has four parameters; in addition to σ and ϵ , defined previously, the repulsion between molecules is represented by m while γ represents attraction due to the $1/r^{*8}$ term.

The m-6-8 has been tested by examining the relationship between experimental and theoretical properties of the simple gases such as the viscosity coefficient, given by equation (1) and the second virial coefficient, B. The second virial coefficient is given by the expression, for a monatomic gas,

$$B = \frac{2}{3}\pi N\sigma^3 \int_0^\infty r^{*3} \frac{d\Phi^*}{dr^*} \exp\left[-\Phi^*/T^*\right] dr^*, \quad (10)$$

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$$B = \frac{2}{3}\pi N\sigma^3 B^*(T^*),$$
 (11)

where N is Avogadros number. (Equations (10) and (11) should, strictly speaking, be modified for a polyatomic gas but that is not necessary here.) We have found that equation (9) can be used to correlate and predict the properties of simple non-polar polyatomic gases (oxygen, nitrogen, carbon dioxide, for example) to within about five percent of experiment.

3.4. Calculations for Fluorine

We need, therefore, to determine potential parameters for fluorine. We cannot do this from transport data so we obtained the parameters by fitting the second virial coefficients published by Prydz and Straty [14] as follows:

A set of reduced second virial coefficients, B^* of equation (11), are available as a function of T^* for several values of m and γ [12]. We do not have enough experimental information to determine a unique set of m, γ , σ and ϵ/k so we fixed m at 12 and γ at 2.0, based on our experience with other gases. We varied σ and ϵ/k until a best fit of the experimental second virials was obtained via equation (11). The parameters chosen were: m=12, $\gamma=2.0$, $\sigma=3.32$ Å (1 Å=10⁻¹⁰ m), and $\epsilon/k=138.0$ K, table 1.6

We also have tables of the collision integrals, equation (5), versus T^* . Hence, given the above parameters, $\Omega^{(2,2)*}$ was calculated at various temperatures for insertion into equations (1) and (6). The internal specific heat $c_{v}^{"}$ and the rotational collision number Z are also required. However, the former quantity has been determined by Straty based on previous NBS work [20]. The latter quantity can be estimated sufficiently well from the corresponding values for oxygen, nitrogen, and methane given in reference [5]. For these gases, the dimensionless Z varies between about 2.0 at T=100 Kand about 4.0 at T=300 K according to the linear equation (for this temperature range): Z=1.0+T/100.0. It was assumed that the equation held for fluorine. (We can verify that the contribution due to the last term on the right hand side of equation (6) is small, so Z is only required approximately.)

Having, then, values for c_v'' , Z, σ , and the collision integrals, the viscosity and thermal conductivity coefficients of dilute gaseous fluorine were calculated from equations (1) and (6) and tabulated in table 2. We judge the numbers to be accurate to within five percent based (a) on the possible uncertainty introduced when parameters obtained from the virial coefficients are used to calculate transport coefficients and (b) on the experimental error in the virials themselves.

4. Dense Gas and Liquid

As for the dilute gas, transport measurements will not be used to estimate the transport properties for the dense gas and liquid. Before discussing our prediction method, however, we introduce the transport coefficient excess functions. These functions are defined for the viscosity and thermal conductivity by the relations:

$$\Delta \eta = \eta(\rho, T) - \eta_0(T), \tag{12}$$

$$\Delta \lambda = \lambda(\rho, T) - \lambda_0(T), \tag{13}$$

where $\eta(\rho,T)$ and $\lambda(\rho,T)$ are the values of the coefficients at a particular density and temperature and $\eta_0(T)$ and $\lambda_0(T)$ are the dilute gas coefficients. The functions have been found to be a convenient way to represent transport coefficients over a wide range of temperatures and densities [1] because experiment indicates that they are generally a relatively weak function of temperature at constant density. That is, the temperature dependences of $\eta(\rho,T)$, and $\lambda(\rho,T)$ are apparently very close to the temperature dependences of the dilute gas coefficients. In fact, except for the light molecules, the temperature dependence can often be neglected, and it is therefore possible to compress a considerable amount of information on essentially a single curve of the excess function plotted versus density.

4.1. The Modified Enskog Theory (MET)

At this time no rigorous transport theory can be applied to fluorine, other than for the dilute gas. For reasons given in the appendix, we also reject the straightforward use of the law of corresponding states to obtain fluorine transport properties. The only procedure suitable to predict both the viscosity and thermal conductivity coefficients for this fluid over a wide experimental range is the semi-empirical modified Enskog theory (MET).

Since a full discussion on the MET has been presented in reference [6], it is not necessary to comment here on the theoretical background or the derivation of the expressions for the transport coefficients. However, the basic characteristic of the MET is especially relevant to this paper and should be stressed, viz., that transport coefficients in the dense gas and liquid can be determined by using only equation of state data and the dilute gas transport coefficients. The latter, in turn, can be calculated in principle via equations (1-8) with an intermolecular potential function obtained from the second virial coefficients. Thus, experimental transport data are not required.

The MET equations are:

Viscosity:

$$\eta = \eta_0 b \rho \left[\frac{1}{b \rho \chi} + 0.800 + 0.761 b \rho \chi \right],$$
(14)

Thermal Conductivity:

$$\lambda = \lambda_0' b_\rho \left[\frac{1}{b_{\rho \chi}} + 1.20 + 0.755 b_{\rho \chi} \right] + \frac{\lambda_0''}{\chi},$$
 (15)

⁶ Tables have been placed at the end of this paper.

where η_0 , λ_0' , and λ_0'' are dilute gas transport coefficients. The viscosity η_0 is given by equation (1), λ_0' by equation (7), and $\lambda_0'' = \rho D_0 c_v''$ from equation (6). The term $b\rho\chi$ is a function of the equation of state variables, pressure (P), temperature (T), and volume (V):

$$b\rho\chi = T\frac{\partial}{\partial T}\left(\frac{PV}{RT}\right)_{V} + \frac{PV}{RT} - 1, \tag{16}$$

with ρ the density and R the gas constant. To find b we write PV/RT as a virial expansion:

$$\frac{PV}{RT} = 1 + B\rho + C\rho^2 + \dots, \tag{17}$$

where B and C are the second and third virial coefficients. Substituting equation (17) into equation (16) we have

$$b\rho\chi = \left(B + \frac{TdB}{dT}\right)\rho + \left(C + \frac{TdC}{dT}\right)\rho^2 + \dots \quad (18)$$

But, in order that equations (14) and (15) approach the correct limiting values as $\rho \rightarrow 0$, we require $\chi \rightarrow 1$ as $\rho \rightarrow 0$. Hence,

$$b = B + \frac{dB}{dT}. (19)$$

Equations (16) and (19) also allow χ to be found for inclusion in equation (15).

We have determined MET transport coefficients for many fluids from the appropriate equations of state and intermolecular potential functions [6]. Figures 3 and 4 illustrate selected comparisons of the MET calculations against experiment for argon, nitrogen, oxygen, and methane. The results are shown in the excess function format, equations (12) and (13). The temperature dependence of both the theoretical and the experimental excess functions has been neglected, which is justified for temperatures not exceeding about 300 K.

A conclusion from figures 3 and 4—substantiated by results for other fluids (H₂, He, Ne, CO₂ [6, 7])—is that the representation of experiment by the MET is good up to the critical density, ρ_c , and reasonable up to densities of about $2\rho_c$. By reasonable, we mean that an agreement of around 10 percent between experiment and theory is achieved. Consequently, returning to fluorine, we have every reason to assume that a straight-forward prediction of the transport properties of fluorine by the MET from the equation of state of reference [14] would be adequate for densities up to $\sim 2\rho_c$. Our objective is to produce values from the triple point to 300 K for pressures up to 200 atmospheres; therefore the MET values would be adequate for temperatures above 160 K. For temperatures and pressures corresponding to densities above the upper limit, we must expect the MET calculations to be in error. However, an inspection of figures 3 and 4 indicates that the pattern of deviations between theory and experiment can vary from one fluid to another and

it is not obvious what kind of deviation would be observed for fluorine if data were available. Fortunately, we think this problem can be overcome. In our previous work we attempted to clarify why, in a macroscopic sense, deviations between MET and experiment apparently do not follow a consistent pattern, and our conclusions can be applied to fluorine. In reference [6] we investigated the density dependence of the excess function at constant temperature, $(\partial \Delta X/\partial \rho)_T$, the temperature dependence at constant density, $(\partial \Delta X/\partial T)_p$, and the variation of the transport coefficients along the saturated liquid boundary, $(dX/dT)_{\text{sat.}}$, where $X=\eta$, λ . These last derivatives were most convenient to work with, and because the behavior of the transport coefficients at saturation is indicative of their behavior in the liquid as a whole, they gave a great deal of information. The MET expression for $(d\eta/dT)_{\text{sat}}$ is written here to illustrate the procedure. From equation (14), a dimensionless equation can be derived:

$$\frac{T}{\eta} \left(\frac{d\eta}{dT} \right)_{\text{sat.}} = \frac{Td\eta_0}{\eta_0 dT} + \frac{Tdb}{bdT} + \frac{T}{\rho} \left(\frac{d\rho}{dT} \right)_{\text{sat.}}$$

$$- \frac{T}{\eta} \left[\frac{1}{f^2} - 0.761 \right] \cdot L, \qquad (20)$$

where L is given by

$$L = \left(\frac{\partial f}{\partial T}\right)_{\rho} + \left(\frac{\partial f}{\partial \rho}\right)_{T} \left(\frac{d\rho}{dT}\right)_{\text{sat}}.$$
 (21)

To shorten the notation, we have written, $f \equiv b\rho\chi$ (given by equation (16)),

$$[\]_{\eta} = \left(\frac{1}{f} + 0.8 + 0.761f\right).$$

By substituting experimental values for several fluids into each of the dimensionless terms of equation (20) (and into the corresponding terms for the thermal conductivity equation), and by plotting these terms against a dimensionless temperature, T/T_c , with T_c the critical temperature, it was possible to compare MET results for the different fluids in detail. Actually, the comparison turned out to be relatively simple because the differences showed up essentially only in the term involving the second virial coefficient, T(db/dT)/b. Figure 5 illustrates a plot of this dimensionless derivative against T/T_c for oxygen, methane, argon, and nitrogen.

We have avoided applying the law of corresponding states directly but one could still hope that a restricted form of correspondence between dense fluids might occur. For instance, it is possible that fluids which have a similar behavior in some dimensionless variable (such as T(db/dT)/b) as a function of reduced temperature and density will show similar behavior in their transport coefficients. This may well be so because a direct correlation between the behavior of T(db/dT)/b and the deviations between the MET and experiment seems to exist.

For example, the MET predictions for argon and nitrogen are too low for viscosity but too high for thermal conductivity, whereas the predictions for oxygen and methane are too high for both coefficients. Inspection of figure 5 reveals that the values for T(db/dT)/b are similar for argon and nitrogen, that is, they follow about the same curve when plotted against T/T_c . Such values, however, are substantially different from those for oxygen and methane which are in turn, quite similar. In other words, plots of T(db/dT)/b against T/T_c for the four fluids seem to fall into two groups and can be associated with a given MET prediction of experiment. Observations with other fluids not discussed in detail here, hydrogen, helium, and neon for example, reinforce this.

4.2. Application to Fluorine

MET values for the viscosity and thermal conductivity coefficients of fluorine were determined from equations (14) and (15) using the fluorine equation of state [14]. The derivative T(db/dT)/b was also computed as a function of T/T_c for the liquid. Plotting this derivative in figure 5, one notices the similarity with oxygen or methane. We will, therefore, assume that the MET representation of fluorine would be similar to the MET representation of oxygen and methane. Further, we assume that the fluorine prediction would deviate by the same amount as observed for methane. Accordingly, the MET viscosities and thermal conductivities for fluorine were expressed in the excess function format and the curves lowered by a percentage consistent with the methane deviation pattern ~10-30 percent, the difference increasing with density. The MET values and scaled adjusted values for fluorine are shown in figure 6. It should be noted that the scaling adjustment affects the transport coefficients significantly only at densities greater than $\sim 2\rho_c$.

5. Critical Region

The adjusted viscosity as shown in figure 6 is effectively our final result for that coefficient, but further calculations are required before the thermal conductivity coefficients can be tabulated. It is now recognized that this latter coefficient exhibits an anomalous rise in the critical region and approaches infinity at the critical point. While the phenomenon cannot at present be incorporated into any systematic theory, such as the MET, it has been studied separately by several authors. In particular, Sengers and Keyes [18], have an expression for the critical excess conductivity close to the critical point (ρ_c, T_c) . Nevertheless, calculations of the excess conductivity away from the critical point present some problems. A very elementary problem, for example, is to decide how far from the critical point the excess is significant.

The procedure adopted by us—which must be regarded

as entirely preliminary—is based on a computation of the critical point excess for oxygen proposed by Roder [15]. It is based on the fact that the specific heat at constant pressure, C(p), approaches infinity at the critical point. Extending equation (13), one can write:

$$\Delta \lambda = \lambda(\rho, T) - \lambda_0(T) + \lambda_c(\rho, T), \tag{13a}$$

where $\lambda_c(\rho, T)$ is the critical point excess thermal conductivity at a given density and temperature. Consider a critical excess specific heat $C_c(p)$ which has the property of approaching infinity at ρ_c , T_c and zero far from ρ_c , T_c . One can then show λ_c is related to this quantity by the equation (15)

$$\lambda_c = KC_c(p)^{m'}, \tag{22}$$

where K is a scaling constant, and m' is a function of ρ and T which varies between 1.0 far from the critical point, and 0.6 at the critical point. When m'=0.6, equation (22) approximates the result of Sengers and Keyes. The detailed form of m' is complicated; for temperatures along the critical isochore it is given by the relation:

$$\ln m' = a + b \left\{ \ln \left| \frac{T - T_c}{T_c} \right| \right\} \tag{23}$$

where a and b are constants; for temperatures along other isochores, m' varies in a pattern illustrated in figure 7. In this figure, T_1 , T_2 , T_3 are in the range $T_c < T_\chi < T'$ with $\chi = 1$, 2, 3, and T' = an arbitrary temperature of magnitude $\sim 1.2 T_c$. Also shown as a dashed curve is an isotherm representing temperatures less than T_c , i.e., $T'' < T_4 < T_c$ where T'' is a temperature of magnitude of $\sim 0.8 T_c$ (see reference [15]).

For fluorine, we have no way of measuring the required constants K, a, and b, or the details of the curves corresponding to figure 7, but we have established that fluorine transport coefficients roughly correspond with those for oxygen or methane. Consequently, parameters for fluorine were estimated from the recent similar calculation for oxygen [15], but using the fluorine equation of state with the result that K=0.0108, a=0.205614, b=.0910835, T'=175.9 K, and T''=113.0 K. The functions for the curves similar to figure 7 at various temperatures are available as a computer routine. The critical point excess thermal conductivity coefficients were thus generated for several temperatures and added to the excess thermal conductivity previously determined. The total excess curve is shown as figure 8.

6. Dense Gas and Liquid Tables

The viscosity and thermal conductivity tables were generated from the excess function curves using the equation of state to convert from density to pressure. The results are given in tables 3 and 4. We also include for convenience table 5 which gives the transport coeffi-

cients at the saturated liquid and vapor boundaries. Units chosen for the tables are: temperature in Kelvin, pressure in atmospheres (1 atm = 0.101325 MN/m²), viscosity in g/cm·s, and thermal conductivity in mW/cm·K. We remark, however, that these tables have been converted to other sets of units and will be published shortly in an NBS Technical Note. We place an error estimate on the numerical values of about 10-20 percent, worse in the critical region for thermal conductivity (~50 percent). The error estimate is based on the uncertainties in the MET known for other fluids plus the uncertainty in the dilute gas values discussed previously.

7. Comparison With Experiment

We have decided that, while the available experimental transport data are too scattered or too imprecise to form a basis for the construction of tables, theoretical calculations permit acceptable tables to be generated. Since transport data have effectively been eliminated on experimental grounds, a comparison between theory and these experiments throws no light on the reliability of the predicted tables. Nonetheless, as a matter of interest, we plot calculated viscosity and thermal conductivity coefficients (solid lines) along with the experimental points in figures 1 and 2. It turns out that agreement between prediction and experiment is generally close.

8. Conclusion

Tables for the viscosity and thermal conductivity coefficients of fluorine have been generated without recourse to transport property data. The tables are believed to be as good as possible at the present time, but are not to be regarded as authoritative. We place an error estimate of five percent on the values in the dilute gas tables and 10–20 percent on the values associated with pressures above five atmospheres.

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Appendix—Corresponding States

We think it necessary to comment on the law of corresponding states which is a convenient correlating tool for many properties of fluids and obviously comes to mind in our case: it would be quite straightforward if one could obtain the transport properties of fluorine given the properties of another fluid. In fact, this approach has been followed in the past [9, 16, 17]. Unfortunately, we can demonstrate that it does not work very well. A typical corresponding states diagram for the

viscosity of oxygen, nitrogen, methane, and argon is sketched in figure 9. Experimental saturated liquid viscosities were reduced via potential parameters and the temperature by equation (1). Since the reduced viscosities do not fall on a common curve, the law of corresponding states is not obeyed. While this can be explained as a failure of the law to apply to polyatomic molecules, the important conclusion to be drawn from the figure is that one has no indication how fluorine would behave. One could equally assume fluorine to be like nitrogen, say, or like oxygen. (The fluids shown in figure 9 could be made to fall on a common curve by incorporating extra parameters into the reducing equations. Invariably, then, knowledge of any extra parameters comes from the data themselves, and here we have assumed that no reliable fluorine data exist.)

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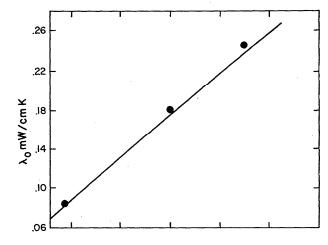
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Table 1. Physical parameters for fluorine [14]

Molecular Weight = 37.9968 Critical Temperature, $T_c = 144.31$ K Critical Density, $\rho_c = 0.574$ g/cm³ Critical Pressure, $P_c = 51.47$ atm Normal Boiling Point Temperature = 84.950 K Triple Point Temperature = 53.481 K m-6-8 Potential Function Parameters: m = 12, $\gamma = 2$, $\sigma = 3.32$ Å $(3.32 \times 10^{-10} \text{ m})$, $\epsilon/k = 138$ K

TABLE 2. Dilute gas transport coefficients for fluorine

T K	$10^3\eta_0$ g/cm s	λ ₀ mW/cm K	T K	$10^3 \eta_0$ g/cm s	mW/cm K
70	0.059	0.062	190	0.153	0.164
80	0.067	0.070	200	0.161	0.172
90	0.075	0.079	210	0.168	0.180
100	0.083	0.087	220	0.175	0.189
110	0.091	0.096	230	0.182	0.197
120	0.099	0.104	240	0.189	0.206
130	0.107	0.113	250	0.195	0.214
140	0.115	0.121	260	0.202	0.223
150	0.123	0.130	270	0.208	0.231
160	0.131	0.138	280	0.215	0.239
170	0.138	0.147	290	0.211	0.248
180	0.146	0.155	300	0.227	0.256



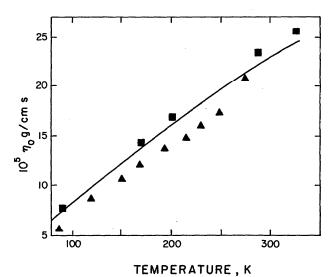
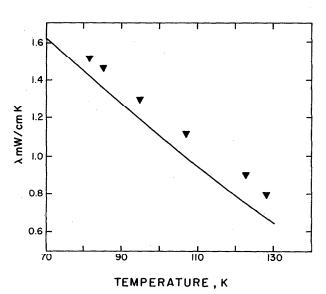


FIGURE 1. Upper drawing: Dilute gas thermal conductivity coefficients due to Frank [3]. Lower drawing: Dilute gas viscosity coefficients from Frank [4], squares, and Kanda [10], triangles. The solid curves are our calculated values.



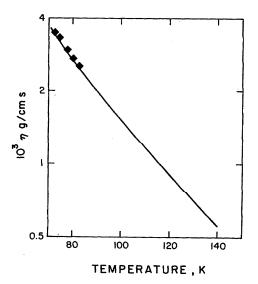


FIGURE 2. Upper drawing: Thermal conductivity coefficients for the saturated liquid from reference [19]. Lower drawing: Viscosity coefficients for the saturated liquid from reference [2]. Solid curves are our calculated values.

Table 3. Viscosity of fluorine as a function of pressure (atmospheres) and temperature (Kelvin). Units: g/cm s.

φ. /²	P,atm	S.	10	15	20	25	30	3.5	0 *	4.5	50	55
	3		3	ľ	ľ	3	;			'		1
75	3.216	3.000	3.840	3.262	3.279	3,895	3,314	3,330	3,948	3,366	3.985	4.003
80	73	2.742	. 75	77	. ~	80	8	2.831	4	9 4	8.4	, ~
85	.07	2.338	35	3	m	.39	. 40	2.418	3		4.0	
06	• 0.7	2.019	• 03	• 0 4	Ξ,	• 06	.08	2.092	10	11	.12	7
95	.08	1.750	• 76	.77	۲.	• 79	.80	1.820	. 83	1.843	. 85	8
100	• 08	1.521	53	.54	ı.	• 56	• 57	1.587	• 59	1.609	• 62	9
105	0.0	760.0	34	33	m.	.37	.38	1,393	3.	1,413	• 45	4
110	60	0.098	. 19	23.	oi o	-22	. 23	1.246	52	1,261	• 26	ď
115	60,	0.102	٠ ا	.0.		60.	10	1.109	=	1.127	13	7
120	ц.	0.106	91.	7:	δ.	96	97	0.985	66	1.005	. 01	•
123	2 -	0.110	4 +	0.118	4 -			U . 853	0.875	0.886	98.	יי יי
7 T	1 -			1 4	•		- 1	0 - 1 - 2 - 2	0 1	200	•	•
140	11	0.121	12	12		13	41.	1.155	-	1.586	3	ي .
145	12	0.125	12	12	! =	.13	4	0.153	15	192	23	
150	.12	0.129	.12	۲.	ᅻ.	14	41.	0.152	16	0.181	.20	. ~
155	.12	0.133	•13	7	٦.	.14	.14	0.153	16	0.175	19	~
160	.13	0.137	.13	۲.	7	• 14	•14	0.154	.16	0.171	.18	ď
165	•13	0.140	• 14	٠.	7	• 15	.15	0.155	• 16	0.170	.18	7
170	• 14	0.144	77.	∹	٦.	.15	.15	0.158	.16	0.171	• 17	٦.
175	14	0.148	14	ᅻ.	ᅻ.	•15	.15	0.160	16	0.172	.17	₹
180	4	0.151	.15	ᅻ .	₩.	16	• 16	0.163	.16	0.174	. 18	₹
185	15	1.155	15	₽.	┥.	91.	• 16	0.156	16	0.175	18	7
190	.15	0.159	919	ᅻ.	ᠳ ੶	16	17	0.170	. 17	0.177	. 18	ᅻ.
250		701.0	9.	: '	٠.	7.	. 1.	# I T + D	1,	0.179	9	٠,
002	10	156	10	0.157		.17	.17	0.178	•	0.181	81.	ᅻ.
210	01.	173	1.		• •	, t.	9 7	10 10 10 10 10 10 10 10 10 10 10 10 10 1	9 4	201.0	9 4	; ፣
770	9 7	110		- •	-		01.	0.100	2 4	0.186	4,	፣ ነ
220	17	180	13	! ~	: -		• •	0.1.0	9 5	100	ָר קלי	•
225	17	0.183	18	١.	٦,	18	61.	0.195		195		
230	.18	0.186	18	4	7	.19	19	0.198	19	0.198	202	. 5
235	.18	0.190	13	ᅻ.	ᅻ	•19	• 19	0.201	.20	0.202	. 20	ď
240	• 19	1.193	13	ᅻ.	7	• 19	• 20	0.204	20	0.205	.20	ď
242	•19	1.196	.19	٠.	₹.	•20	• 20	0.207	20	0.209	.20	ď
250	19	0.200	.20	٠,	Ç,	•20	• 20	0.210	7	0.213	5	ď
252	20	0.203	• 20	'n	Š	.20	• 20	0.213	21	0.216	21	٠
260	• 20	0.206	.20	ผู	o.	20	.21	0.215	27	0.219	.21	ç
265	50	9.208	-21	ď	ď	• 21	•21	0.218	22	0.222	. 22	ď
270	20	0.212	7	Ņ	2	.21	.21	0.221	22	0.225	. 22	ç
272	27.	6.216	42.5	'n	Ŋ,	22.	.22	0.223	22	0.228	22	ď
200	7.5		,,	ď	'n	'n	, c	9229	200	0.231	30	'n
200		777	V ('n	'n	7 6	7 0	0.228	23	0.234	23	Ÿ
162	2 0	0000	200	•	ů	,,,	27.	152.0	36	0.53.0	Si c	Ņ
262	• •	0.224	9 6	• •	• •		9 6	100	3	0.550	, t	'n
)	J •	1	3	•	J	9	9	0 0	7	0+70	*	ä

9	65	2.0	8.0	06	100	0 ₹ ₹	120	130	153	175	200
	4.040	4.059	960.4	4.133	4.170	4.218	4 - 245	4.279	4.353	4.446	4 + 539
75 3.415	3.433	3.450	3.485	3,525	3.560	3.595	3.631	3.666	3.736	3.822	3.912
_	2.924	2.940	2.972	3.005	3.038	3.071	3.163	3.133	3.198	3.279	3.359
	2.502	2.516	2.544	2.573	2.662	2.631	2.659	2.694	2.752	2.824	2.896
_	2.164	2.176	2.199	2.223	2.249	2,275	2.302	2.328	2.382	5.445	2.512
	1.889	1.901	1.924	1.948	1.971	1.994	2.017	2.040	2.085	2.142	2.197
	1.652	1.663	1.685	797.	1.729	14/21	7.775	1.735	1.857	1.892	1.04
	1.453	1.464	1.483	1.583	1.524	10°0'	1.504	1.585	1.625	1.676	1.72
	40.00	1.503	1.321	1.00 200 100 100 100 100 100 100 100 100 1	1.355	1000	7.659	1.409	1.440	1.492	40.00
	1.104	7,11.7	1.130	7.4	7.67	7 + 7 + 7		7.75	900	7.040	200
	1.043	T • C 20	1.07	1.000	1.163	10101	0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	1.124	1.100 1.100	1.660	1.203
	0.93	0.00		0.070	0000	210.0	9 0 0 0	470	100	1.001	1 -
-	0.735	0.746	1754	0.781	1,799	1 . A + 6	300	0.853	98.4	0.931	200
	0.652	0.663	0.685	0.708	0.728	0.746	8.762	0.277	0.807	1.847	88.0
	0.567	0.587	0.617	0.641	0.659	0.677	0.696	0.714	747-0	0.780	0.81
	0.382	0.466	0.537	0.573	0.600	0.622	0.641	0.657	0.688	0.727	0.76
	0.264	0.309	0.415	0.491	0.535	0.566	0.588	0.608	0.642	0.676	0.71
	0.234	0.259	0.322	0.392	164.0	0.512	0.535	0.560	0.599	0.637	99.0
	0.221	0.238	0.281	0.331	0.382	0.434	0.476	0.509	255*0	0.599	0.634
	0.214	0.227	0.259	0.298	0.338	0.378	614.0	0.457	0.514	0.564	9.0
	0.211	0.221	0.246	0.277	0.311	0.344	0.377	0.410	0.473	0.529	19.57
_	0.208	0.217	0.238	0.263	0.291	0.321	645.0	0.377	0.434	0 4 95	. 5
_	0.206	0.215	0.233	9.554	0.278	105.0	0.330	0.354	0.403	0.463	0.51
	192.0	0.213	0.230	/ #Z • N	0.258	162.0	6.62.5	10.53	0.380	454.0	3
	0.203	212.0	0.228	0.243	0.261	162.0	5 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	0.324	E . 35.3	0.411	200
	0.00	0.611	122.0	0.74		# 070 # 0	9 00 00 00 00 00 00 00 00 00 00 00 00 00	0.313	0.50 200 200	0.000	3
	702.0	0.210	0.225	200	200	0.055	20.00	202	7 7 7	44.0	17.0
_	02.0	0.212	2,00	2000	1000	2000	0.277	200.0	2000	0000	7 M
	0.207	0.212	0.224	0.237	0.249	0.261	0.274	0.287	0.316	0.352	0.38
	0.209	0.213	0.225	0.237	0.249	0.260	0.271	0.284	0.311	0.345	0.37
	0.211	0.215	0.225	0.237	0.249	0.259	0.270	0.282	0.307	0.339	0.36
	0.213	0.217	0.226	0.238	0.249	0.259	0.269	0.280	0.303	0.334	0.35
_	0.215	0.219	0.227	0.238	0.249	0.259	0.269	0.279	0.301	0.330	0.35
	0.218	0.221	0.228	0.238	0.249	0.260	0.268	0.278	0.298	0.326	0.35
	0.220	0.223	0.230	0.239	0.250	0.260	0.269	0.277	0.297	0.323	9.34
	0.222	0.225	0.232	0.240	0.250	0.260	0.269	0.278	0.296	0.320	934
	0.224	0.228	0.234	0.241	0.251	0.261	0.270	0.278	0.295	0.318	9.34
	0.226	0.238	0.236	0.243	0.252	0.261	0.270	0.278	0.294	0.317	10.34
	0.229	0.232	0.238	0.244	0.253	0.262	0.271	0.279	0.294	0.316	33
	0.231	0.234	0.240	0.246	0.254	0.263	0.272	0.280	0.294	0.315	133
_	0.234	0.236	0.242	0.248	0.255	0.264	0.272	0.281	0.295	0.314	0.33
	0.236	0.239	0.245	0.00 0.00 0.00 0.00 0.00 0.00 0.00 0.0	0.255	0.265	0.273	0.281	0.295	415.0	0.333
	U.Z.S.Y	0.241	742.0	262.0	U • 758	002.0	4 72 0	797.0	0.62.0	U. 514	.00.1
_	c		5	t t c	0000	1	100	*	1		

TABLE 4. Thermal conductivity of fluorine as a function of pressure (atmospheres) and temperature (Kelvin): Units: mW/cm K.

Note: The light shaded areas indicate values near the critical point which are uncertain. The heavy shaded areas indicate values close to the critical point which are extremely uncertain.

		2	10•U	0.02	0.62	0.00	9 .6 6	f 00 t	45 • U	0.06	N • 66
62	62	. 62	9.	.63	Y.	1.635	33	1.540	1.642	1.645	1.64
5 1.54	R.	•54	.54	55	55	1.556	55	1.561	1.564	1.567	1.569
1.45	46	949	•46	•46	. 47	1.475	7	1.481	1.484	1.487	1.490
5 0.07	34	.37	.38	38	.38	1.390	33	1.397	1.400	1.403	1.407
0.08	28	•29	•29	.30	• 30	1.307	× 1	1.314	1.318	1.321	1.325
5 0.08	20	.20	.21	.21	• 22	1.224	22	1.231	1,235	1.239	1.243
60.0	11	12	.12	• 13	• 13	1.140	<u> </u>	1.149	1,153	1.158	1,162
60 • 0 - 2	6	.13	40.	40.	. 05	1.057	9	1.067	1.071	1.076	1.081
0.09	70	.95	9.5	• 96	96	0.973	37	186.0	066.0	0.995	1.00(
5 0.10	1.0	.10	.87	.88	89	968.0	8	206.0	0,912	0.918	0.92
0.10	1	.11	12	.83	81	0.818	ž	0.832	0.838	0.844	0.85
5 0.11	1	11	.12	24.	• 72	0.738	0.746	0.754	0.762	0.769	0.77
0 0.11	. 12	•12	.12	.13	. 21	0.663	~	0.530	0.688	0.695	0,70
5 0.11	. 12	-12	.12	.14	• 18	0.297		0.510	0,621	0.630	0.63
0 0.12	12	.12	.13	•14	, 17	0.228	**	0.740	0.692	0.559	0.57
5 0.12	. 13	13	,13	.15	. 16	0.204	-	0.387	959*0	959";	1/2/2
0 0.13	.13	7	.14	.15	.17	0.193	۳.	0.296	1050	0.575	0.96
5 0.13	1.4	.14	41.	.16	.17	0.189	X.	16210	0.314	0.440	0.53
0 0.14	#	41.	.15	.15	. 17	0.186	₩.	0.235	0,272	0.322	0.39
5 0.14	.15	.15	.15	.15	.17	0.185	Ħ	1221	0.248	0.281	0.32
0 0.149	0.153	0.155	0.156	0.163	0.172	0.177	0.167	0.202	0.221	0.243	0.271
5 0.15	• 15	.15	.15	•16	. 17	0.174		161.0	0.205	0.221	0,24
0 0.15	16	.16	•16	•16	. 17	0.173	2	0.177	0.183	0 • 1 90	0.197
5 0.16	.15	•16	• 16	• 16	. 17	0.177	17	0.180	0.185	0.191	0.19
0 0.16	15	•17	.17	.17	. 17	0.181	8	0.183	0.188	0.193	0.19
5 0.17	.17	• 17	•17	•17	. 18	0.185	<u>~</u>	0.185	0.190	0.196	0.20
0 0.17	.17	•17	.17	13	. 18	0.189	2	0.1.00	0.193	0.198	0.23
5 0.17	13	138	138	• 18	• 18	0.193	5	0.193	0.196	0.200	0.50
0 0.18	13	.18	• 18	13	• 13	0.196	2	961.0	0.199	0.203	0.20
5 0.18	13	19	13	•19	• 19	0.200	S	0.202	0.203	0.206	0.21
0 0.19	13	£.	61.	19	. 19	0.204	5	0.206	0.206	0.209	0.21
5 0.19	£	.20	.20	.20	200	0.207	굶	0.211	0.210	0.212	0.21
0 0.19	23	.20	.20	• 20	• 20	0.211	컶	0.215	0.214	0.216	0.21
5 0.20	.20	.20	•20	• 20	• 21	0.214	컶	0.219	0.219	0.220	0.22
0.50	2	•21	2	2	•2	0.218	2	0.223	0.223	0.223	0.22
5 0.21	.21	•21	•21	. 21	. 21	0.221	2	0.227	0.227	0.227	0.22
0 0.21	.21	225	•22	.22	• 22	0.224	22	0.231	0.232	0.231	0.23
5 0.22	.22	525	225	.22	• 22	0.228	2	0.235	0.236	0.235	0.23
0 0.22	.22	525	.23	.22	. 23	0.232	7	0.239	0.240	0.240	0.24
5 0.22	.23	23	.23	.23	.23	0.236	7	0.243	0.244	0.244	0.24
0 0.23	. 23	5	-23	.23	• 23	0.240	4	0.247	0.248	0.248	0.24
5 0.23	. 23	.24	•24	•24	• 24	0.244	7	0.251	0.252	0.253	0.25
0 0.24	\$	24	• 24	24	. 24	0.248	5	0.254	0.256	0.257	0.256
5 0.24	÷.	٠ ت	35	55	. 25	0.251	5	0.258	0.260	0.261	0.26
0 0.24	.25	.25	25	.25	.25	0.255	5	0.262	0.264	0.265	0.26
	1										
			25		25	0.259	29	0.265	0.268	D-269	0.27

TABLE 4. Thermal conductivity of fluorine as a function of pressure (atmospheres) and temperature (Kelvin): Units: mW/cm K.—Continued
Note: The light shaded areas indicate values near the critical point which are uncertain. The heavy shaded areas indicate values close to the critical point which are extremely uncertain.

<u>/</u>) (0.67	80.08	⊃ • • •	1001	110.0	120.0	150.0	150.0	175.0	0.002
			1.658	1.563	99		I wa		89	69	
75 1.572	1.574	1.577	1.582	1.587	1.592	1.597	1.502	1.607	1.616	1.628	1.639
₩		•	1.504	1.509	53	•	rv.	•	.54	r.	•
-	.41	•	1.422	1.429	. F.	•	-+	•	94.	4.8	•
- i	3	•	1.342	1.349	10	•	m		38	0 7	•
-		•	1,262	1.269	.27	•	Λŀ	•	31	32	•
- -		•	1.182	1.191	19	•	NI.	•	23	25	•
-		•	1.104	1,112	.12	•	-	•	16	1.8	
- í		•	1.025	1.035	• 04	•	~	•	.08	11	•
_		•	0.949	0.959	σ,	•	m	•	.01	• 0 4	•
_		•	0.880	0.891	. 90	•	ന	•	94	- 64	
-		•	0.810	0.822	. 83	•	ထ	•	.88	.91	•
_		•	0.740	0.754	• 76	•	~	•	.82	85	
_		•	0.678	0.691	۲.	•	~	•	• 76	• 79	•
		•	0.622	0.638	. 65	•	ω.	•	.71	74	•
		•	0.567	0.587	. 60		w	•	. 56	.69	
	164.5	1.619	0.820	0.529	.55	•	ın		.62	.65	•
		1,518		0.931	330	•	ın	•	59	61	
۳		111.0		981.1	***	- 335	ŧ.c		55	5.8	
6	677.0	183.0	55/:0	616.6	***	8888	80 N N	3333	51	55	
•	0.348	33.00	0.510			3333	887.9	Ͼ.	8 7 .	522	
c	0.291	888 E	0.394				æv.			649	
_	7	8 •	0.240		8 •	8 .		8 .	63	74.	
_	0.212	0.220	0.236	0.256	0.276) M		36	7 7 7	
_	2		0.234		•		m		37	.42	
_	2		0.233		•		· N		35	70	
-	.21	•	0.233		•	•	· (C)	•	34	.38	•
	0.216	•	0.233		•	•	N		333	.37	
6	0.218	•	0.234		•	•	N	•	.32	35	
_	0,220	•	0.235		•	•	N	•	.32	.35	
-	22	•	0.236		•	•	N	•	31	34	•
_	0.225	•	0.238			•	.c∪	•	.31	34	•
_	22	•	0.240			•	N	•	31	.33	
<u>.</u>	23		0.242		•	•	N	•	31	333	•
6	23		0.245		•		C)		30	33	
-	23		0.247		•	•	•	•	30	33	•
_	6		0.250						2	7	•
-	2		000		•	•	ıΛ	•		M (
_	0.245		0.255		•		ın		M.	(A)	
_	76		0.258				ľ			Pr	
_	0.252		0.261		. 27	•	IN		31	333	
_	25		0.264		. 27	•	•		31	10	•
0	25		0.267		28	•	~		31	33	
-	25		0.270		28	•		•	31	33	•
_	25	•	0.274				N		31	53	•
_	27		0.277		2		. M	•	M	P.	
											١

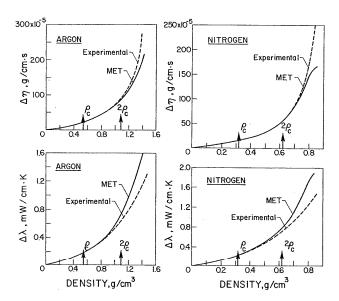


FIGURE 3. Modified Enskog Theory (MET) determination of the transport coefficients of nitrogen and argon compared to experiment.

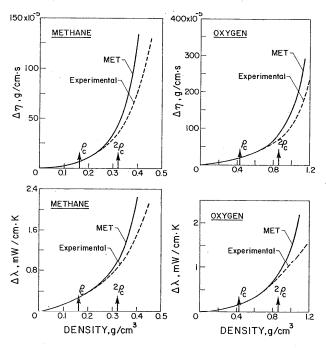


FIGURE 4. MET determination of the transport coefficients of oxygen and methane compared to experiment.

Table 5. Values of the transport coefficients at the saturated liquid and vapor boundaries

	Satura	ted liquid	Satura	ted vapor
T K	$10^3 \eta$ g/cm s	λ mW/cm K	$10^3 \eta$ g/cm s	mW/cm K
70	3.801	1.621	0.060	0.062
80	2.729	1.456	0.069	0.072
90	2.011	1.285	0.079	0.083
100	1.520	1.116	0.089	0.094
110	1.195	0.949	0.099	0.102
120	0.945	0.798	0.116	0.121
130	0.739	0.657	0.137	0.294
140	0.573	1.046	0.203	1.546

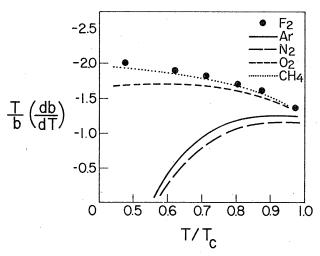


FIGURE 5. Plots of the dimensionless derivative T(db/dT)/b, versus T/T_c for fluorine, argon, nitrogen, oxygen, and methane. Note that fluorine appears to correspond to methane and oxygen.

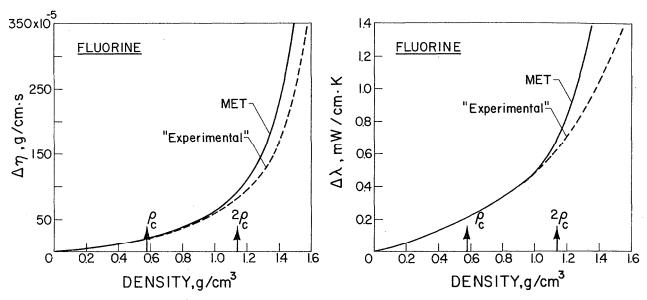


FIGURE 6. Excess function estimates of the transport properties of fluorine, excluding the critical point anomaly in thermal conductivity. The curves have been adjusted above a density of $\sim 2\rho_c$. See text.

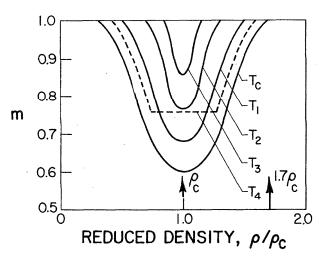


FIGURE 7. Sketch of the variation of the index m' as a function of density and temperature. See equation (23).

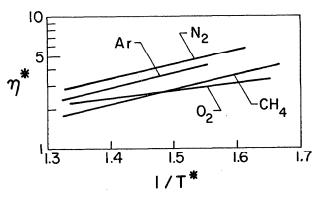


FIGURE 9. Typical variation of the reduced saturated liquid viscosity versus reduced temperature for several fluids. $\eta^* = \eta \sigma^2 / \sqrt{m\epsilon}$ where m is the molecular mass. Values of σ and ϵ are the Lennard-Jones values taken from reference [8].

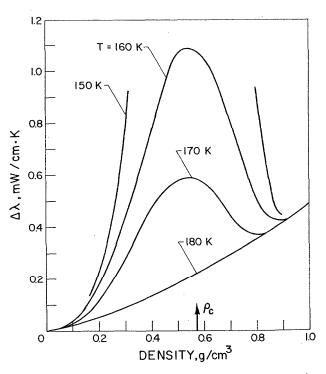


FIGURE 8. Variation of the thermal conductivity coefficient in the critical region.