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**LIQUID-PHASE COMPOSITIONS FROM VAPOR-PHASE ANALYSES:
VII. PRELIMINARY CALCULATIONS CONCERNING ARSENIC
TRIFLUORIDE (AsF_3) AND ARSENIC PENTAFLUORIDE (AsF_5)**

W. Davis, Jr.*
H. D. Cochran†

February 1990

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†Chemical Technology Division, Oak Ridge National Laboratory.

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Prepared by
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CONTENTS

	<u>Page</u>
LIST OF FIGURES	7
LIST OF TABLES	9
ABSTRACT	11
1. INTRODUCTION	13
2. THERMODYNAMIC PROPERTIES OF THE ARSENIC FLUORIDES	13
2.1 VAPOR PRESSURES OF THE ARSENIC FLUORIDES	13
2.1.1 Arsenic Trifluoride	13
2.1.2 Arsenic Pentafluoride	13
2.1.3 Extrapolation of Vapor-Pressure Data	15
2.2 CRITICAL TEMPERATURES AND PRESSURES	16
2.2.1 Comparisons of Critical Parameters	16
2.2.2 The Acentric Factor of Pitzer for Estimating T_c and P_c of AsF_5	17
2.3 STANDARD STATE FUGACITY AND HEAT CAPACITY EQUATIONS	21
3. RESULTS AND DISCUSSION	23
4. SUMMARY	29
REFERENCES	30
Appendix A. SUMMARY OF THERMODYNAMIC DATA	33
Appendix B. CALCULATED FUGACITY COEFFICIENTS AND LIQUID VOLUMES OF AsF_3 AND AsF_5	41
Appendix C. DATA OF JOHNSON AND OTEY ON LIQUID-VAPOR EQUILIBRIUM IN THE SYSTEM AsF_5 - UF_6	45

7/8

LIST OF FIGURES

<u>No.</u>		<u>Page</u>
1	Vapor pressures of AsF_3 and AsF_5 and their representations by Eqs. (1) and (2)	14
2	Comparison of calculated and observed concentrations of AsF_5 in UF_6	26
3	Distribution of F_2 , SiF_4 , AsF_3 , and AsF_5 between liquid and vapor phases of UF_6	28

LIST OF TABLES

<u>No.</u>		<u>Page</u>
1	Regression of vapor-pressure data on the Clapeyron equation (1)	16
2	Regression of vapor-pressure data on the Antoine equation (2)	17
3	Normal boiling temperatures and critical parameters of some trihalides	18
4	Normal boiling temperatures and critical parameters of some pentahalides	19
5	Estimates of critical temperatures and pressures of AsF_3 and AsF_5	20
6	Some thermodynamic properties of AsF_3 and AsF_5 . .	22
7	Experimental data ranges and calculated vapor-liquid equilibrium coefficients for various impurities in UF_6	24
8	Calculated vapor pressure of AsF_5	25
A.1	Vapor pressure data ranges and critical constants	36
B.1	Calculated fugacity coefficients and liquid volumes of AsF_3	43
B.2	Calculated fugacity coefficients and liquid volumes of AsF_5	44
C.1	Data of R. T. Johnson and M. G. Otey concerning properties of AsF_5 and its behavior in UF_6	47

11/12

ABSTRACT

Arsenic normally is not considered to be a contaminant. However, because arsenic was found in many cylinders of UF_6 , including in corrosion products, a study was performed of the distribution of the two arsenic fluorides, AsF_3 and AsF_5 , between liquid and vapor phases. The results of the study pertain to condensation or vaporization of liquid UF_6 . This study includes use of various experimental data plus many extrapolations necessitated by the meagerness of the experimental data. The results of this study provide additional support for the vapor-liquid equilibrium model of J. M. Prausnitz and his coworkers as a means of describing the distribution of various impurities between vapor and liquid phases of UF_6 . Thus, it is concluded that AsF_3 will tend to concentrate in the liquid phase but that the concentration of AsF_5 in the vapor phase will exceed its liquid-phase concentration by a factor of about 7.5, which is in agreement with experimental data. Because the weight of the liquid phase in a condensation operation may be in the range of thousands of times that of the vapor phase, most of any AsF_5 will be in the liquid phase in spite of this separation factor of 7.5. It may also be concluded that any arsenic fluorides fed into a uranium isotope separation plant will either travel with other low-molecular-weight gases or react with materials present in the plant.

1. INTRODUCTION

Arsenic is one of many chemical elements not mentioned in specifications concerning impurities in uranium hexafluoride (UF_6).^{1,2} However, in November 1988 some was found in corrosion materials that plugged a valve on a UF_6 cylinder obtained by Martin Marietta Energy Systems, Inc., from Canada's Eldorado Nuclear Ltd. in 1985.³ The source of the arsenic was reported to be one of Eldorado's suppliers of anhydrous hydrogen fluoride. According to the *Nuclear Fuel* report, "Enrichment officials have concluded that the arsenic somehow remains within the enrichment cascades and did not appear in the product."

Arsenic forms two fluorides — arsenic trifluoride (AsF_3) and arsenic pentafluoride (AsF_5) — whose thermodynamic properties have not been studied extensively. The purpose of this report is (1) to present some of these properties, (2) to describe the calculation of others, and (3) to obtain a first approximation of how these arsenic fluorides are distributed between liquid and vapor phases of systems containing primarily UF_6 . Calculations of these distributions are based on the mathematical models and computer programs of J. M. Prausnitz and his colleagues.⁴ Their adaptations and use with chemical systems involving distributions of impurities in UF_6 between liquid and vapor phases have been described extensively in seven previous reports in this series, including refs. 5 through 9.

2. THERMODYNAMIC PROPERTIES OF THE ARSENIC FLUORIDES

As indicated in previous discussions of the model of Prausnitz and his coworkers,⁴⁻⁹ calculating the distributions of AsF_3 and AsF_5 as impurities in UF_6 between liquid and vapor phases requires various types of data. These data include vapor pressures and densities of pure liquid AsF_3 and AsF_5 and heat capacities of their vapors. Various parameters of the Prausnitz model are then calculated to provide a basis for calculating vapor-liquid equilibrium distributions. Finally, if there are binary data on the distribution of the impurity between liquid and vapor phases, Universal Quasi Chemical (UNIQUAC) parameters can be calculated to improve the accuracies of the distribution calculations. Available data are scarce for both arsenic fluorides.

2.1. VAPOR PRESSURES OF THE ARSENIC FLUORIDES

2.1.1 Arsenic Trifluoride

Russell, Rundle, and Yost measured the heat capacity, heats of fusion and vaporization, and the vapor pressure of AsF_3 .¹⁰ Their 8 measurements of the vapor-pressures of liquid AsF_3 ranged from 28.9 to 137.5 mm Hg at temperatures of 265 to 292 K. These data are plotted in Fig. 1.

2.1.2 Arsenic Pentafluoride

Ruff, Menzel, and Plaut measured the vapor pressure of liquid AsF_5 at 6 temperatures ranging from -77.3 to -53.1°C (195.9 to 220.1 K);

Fig. 1. Vapor pressure of AsF_3 and AsF_5 and their representations by Eqs. (1) and (2). The vapor pressure of UF_6 is shown for comparison.

vapor pressures ranged from 180.2 to 747.9 mm Hg.¹¹ These authors reported the measured melting point to be -79.8°C (193.4 K) and the boiling point to be -52.8°C (220.4 K). Their vapor-pressure data are plotted in Fig. 1. For comparison, the vapor pressure of UF_6 is also plotted in this figure.

Two additional vapor-pressure measurements were made available by M. G. Otey,¹² of the Paducah Gaseous Diffusion Plant, as follows: 16.6 atm on a cylinder pressure gauge (of unknown accuracy) at 23.8°C that he measured with a cylinder of liquid AsF_5 ; and a Material Safety Data Sheet of Ozark-Mahoning Co. specifying a pressure of 214 ± 10 psig (15.6 ± 0.7 atm absolute) at 25°C .

2.1.3 Extrapolation of Vapor-Pressure Data

As one step in estimating the critical temperatures and pressures of AsF_3 and AsF_5 , the vapor-pressure data of Russell, Rundle, and Yost¹⁰ and of Ruff, Menzel, and Plaut¹¹ were fitted to two approximate vapor-pressure equations:

$$(1) \ln[p^S] = \alpha + \beta/T \quad (1)$$

and

$$(2) \ln[p^S] = a + b/(c + t) \quad (2)$$

Equation (1), the Clapeyron equation, is the simplest function for describing vapor pressures at pressures much below the critical pressure. Equation (2) is the Antoine equation that has been used extensively in representing vapor-pressure data, again at pressures well below the critical pressure. In these equations

p^S = saturated-vapor pressure (mm Hg),

t = Celsius temperature ($^{\circ}\text{C}$),

T = Kelvin temperature (K),

α, β = adjustable constants obtained by fitting experimental vapor pressure-temperature data to Eq. (1)

a, b, c = adjustable constants obtained by fitting experimental vapor pressure-temperature data to Eq. (2).

Vapor pressure data were fitted to Eqs. (1) and (2) to generate numbers to aid in estimating critical temperatures and pressures of AsF_3 and AsF_5 . Results of fitting vapor pressure-temperature data to Eqs. (1) and (2) are also shown in Fig. 1. Summaries of results of analyses according to these equations are presented in Tables 1 and 2. The latter table contains estimates of the standard deviations of temperatures calculated to correspond to vapor pressures of 1, 2, and 3 atm. These deviations were calculated by standard statistical methods, as noted by Deming.¹³

Table 1. Regression of vapor-pressure data on the Clapeyron equation (1)^a

Component	α and $s(\alpha)$	β and $s(\beta)$ (K)	$s(\text{fit})$ (%)
AsF ₃	20.21666	-4.461890E+03	1.098
	0.10948	0.030495E+03	
AsF ₅	18.06308	-2.518734E+03	0.886
	0.09032	0.018830E+03	
Temperature, T (K), at which			
	<u>p^S = 1 atm</u>	<u>p^S = 2 atm</u>	<u>p^S = 3 atm</u>
AsF ₃	328.5	346.1	357.4
AsF ₅	220.4	234.6	243.8

^aIn the headings of the first two columns containing numeric values, s is the estimated standard deviation of the associated parameter; $s(\text{fit})$ in the third column of numbers is the estimated standard deviation of fit of the data to the model, namely the Clapeyron equation.

2.2 CRITICAL TEMPERATURES AND PRESSURES

Two parameters needed to apply the model of Prausnitz and his coworkers are critical temperature (T_C) and critical pressure (P_C). The absence of vapor pressure measurements at pressures in excess of 1 atm, particularly in the case of AsF₃, means that a large extrapolation of experimental data must be made to obtain any estimate of T_C and P_C . Estimation of critical parameters is based on three procedures: (1) comparisons with critical and boiling temperatures of compounds similar to AsF₃ and AsF₅; (2) by the acentric factor method of Pitzer and his coworkers,^{14,15} including the correlations of Lee and Kesler,¹⁶ as outlined by Reid, Prausnitz, and Poling;¹⁷ and (3) extrapolation of the reference-state fugacity equation.

2.2.1 Comparisons of Critical Parameters

Critical properties of several trihalides similar to AsF₃ are presented in Table 3; Table 4 lists these properties for several pentahalides. The first property used in this report is the ratio T_C/T_b (the ratio of critical temperature to boiling temperature). Data in these tables were taken from Reid, Prausnitz, and Poling,¹⁷ from Mathews,¹⁸ and from the *CRC Handbook of Chemistry and Physics*.¹⁹ Table 3 shows a ratio $T_C/T_b = 1.540$ for 10 trivalent halides; the standard deviation of this ratio is 0.081. Based on estimates of T_b for AsF₃ given in Tables 1 and 2 (i.e., $T_b = 331 \text{ K}$), T_C for AsF₃ is estimated to be in the

Table 2. Regression of vapor-pressure data on the Antoine equation (2)^a

Component	a and s(a)	b and s(b)	c and s(c) (°C)	s(fit) (%)
AsF ₃	6.27478E+00 0.36790E+00	7.92151E+02 1.31291E+02	1.72745E+02 0.14744E+02	0.53
AsF ₅	6.73863E+00 0.67296E+00	6.81093E+02 2.19974E+02	2.29231E+02 0.26300E+02	0.82
	Temperature, T, at which p ^S = 1 atm and s(T) (K)	Temperature, T, at which p ^S = 2 atm and s(T) (K)	Temperature, T, at which p ^S = 3 atm and s(T) (K)	
AsF ₃	333.8 1.2	356.5 1.4	372.0 1.4	
AsF ₅	220.5 1.4	235.4 1.6	245.4 1.7	

^aIn the headings of the first three columns containing numeric values, s is the estimated standard deviation of the associated parameter; s(fit) in the fourth column of numbers is the estimated standard deviation of fit of the data to the model, namely the Antoine equation.

range of 479 to 537 K with a single best estimate of 510 K. Similarly, on the basis of $T_c/T_b = 1.521$ (with a standard deviation of 0.046, see Table 4, for some pentahalides and the estimates of T_b for AsF₅ in Tables 1 and 2) T_c for AsF₅ is estimated to be in the range of 325 to 346 K, with a single best estimate of 335 K.

Critical pressures are also listed for a few trihalides and pentahalides in Tables 3 and 4. Critical pressures estimated for both AsF₃ and AsF₅ are listed in Table 5.

Note that the ratio T_c/T_b varies over only a small range for all 15 fluorides discussed in this series of reports, as shown in Table A.1 of Appendix A; in particular, the average value of this ratio is 1.562 and its standard deviation is 0.100. These chemicals have critical temperatures ranging from 144.3 K for F₂ to 515 K for AsF₃; T_c/T_b ranges from 1.39 for SiF₄ to 1.745 for Cl₂.

2.2.2 The Acentric Factor of Pitzer for Estimating T_c and P_c of AsF₅

An alternative method of estimating T_c and P_c involves use of the acentric factor, ω , of Pitzer^{14,15} in conjunction with the formulas of Lee and Kesler¹⁶ for two functions $f^{(0)}$ and $f^{(1)}$. These functions,

Table 3. Normal boiling temperatures and critical parameters of some trihalides^a

Component	Boiling temperature T_b (K)	Critical temperature T_c (K)	Critical pressure P_c (atm)	T_c/T_b
AsF ₃	(328.5 to 333.8) ^b	(479 to 537) ^c		
BF ₃	172	260.8	49.9	1.516
NF ₃	144.4	234.0	45.3	1.620
PF ₃	178	271.2	43.3	1.524
AlCl ₃	453	620	26.3	1.369
AsCl ₃	403	654		1.623
BCl ₃	285.8	455	38.7	1.592
PCl ₃	349.1	563		1.613
AlBr ₃	528	763	28.9	1.445
BBr ₃	364	581		1.596
AlI ₃	655	983		1.501
Overall average			38.7	1.540
Standard deviation			8.6	0.081

^aInformation taken from Appendix A of R. C. Reid, J. M. Prausnitz, and B. E. Poling, *The Properties of Gases and Liquids*, McGraw-Hill Book Co., Fourth Edition, 1987.

^bSee Tables 1 and 2.

^cBased on $T_c/T_b = (1.540 \pm 0.081)$

described by Eqs. (2-3.1) and (7-2.6) through (7-2.8) in ref. 17, must be used with pressures in atmospheres. They are involved in a description of the variation of the reduced vapor pressure with reduced temperature. The formulation is as follows.

$$\ln [p^s_r] = f^{(0)}(T_r) + \omega f^{(1)}(T_r) , \quad (3)$$

where

$$f^{(0)}(T_r) = 5.92714 - 6.09648/T_r - 1.28862 \ln T_r + 0.169347 T_r^6 , \quad (4)$$

and

$$f^{(1)}(T_r) = 15.2518 - 15.6875/T_r - 13.4721 \ln T_r + 0.43577 T_r^6 . \quad (5)$$

Table 4. Normal boiling temperatures and critical parameters of some pentahalides

Component	Boiling temperature ^a T_b (K)	Critical temperature ^b T_c (K)	Critical pressure ^b P_c (atm)	T_c/T_b
AsF ₅	(220.4 to 220.5) ^c	(325 to 346) ^d		
NbF ₅	509	737	62	1.448
MoCl ₅	541	850		1.571
NbCl ₅	527	807		1.531
TaCl ₅	515	767		1.489
TaBr ₅	622	973		1.564
Overall average				1.521
Standard deviation				0.046

^aSee R. C. Reid, J. M. Prausnitz, and B. E. Poling, *The Properties of Gases and Liquids*, McGraw-Hill Book Co., Fourth Edition, 1987.

^bSee J. F. Mathews, "The Critical Constants of Inorganic Substances," *Chem. Rev.* 72,71 (1972).

^cSee Tables 1 and 2.

^dBased on $T_c/T_b = (1.521 \pm 0.046)$

In these equations

T = temperature (K),

T_c = critical temperature (K),

T_r = reduced temperature (T/T_c),

ω = Pitzer acentric factor,

p^s = saturated-vapor pressure (atm),

P_c = critical pressure (atm),

p_r^s = reduced saturated-vapor pressure (p^s/P_c).

Equation (3) can be recast by use of the definition of p_r^s , namely

$$\ln [p_r^s] = \ln [p^s] - \ln P_c, \quad (6)$$

from which we obtain

$$\ln [p^s] = [f^{(0)}(T, T_c) + \omega f^{(1)}(T, T_c)] + \ln P_c, \quad (7)$$

where $f^{(0)}(T_r)$ and $f^{(1)}(T_r)$ are expressed in terms of T and T_c rather than $T_r (=T/T_c)$. This equation can readily be described for use in the

Table 5. Estimates of critical temperatures and pressures of AsF₃ and AsF₅^a

Estimated Temperature (K)	Estimated Pressure (atm)	Authors	Method	Additional data used
<u>AsF₃</u>				
570	50	Russell, Rundle, and Yost	Corresponding states	None
479 to 537	38.7	This report	Comparison of T _C to T _b	See Table 3
508.2	53.1	This report	Acentric factor of Pitzer	1, 2, 3, atm extrapolation of Clapeyron and Antoine equations plus estimates T _C = 515 K and P _C = 40 atm
(515) ^b	50.8	This report	Vapor pressure-fugacity-molar volume Eq. (8)	As above
<u>AsF₅</u>				
325 to 346	--	This report	Comparison of T _C /T _b	See Table 4
347	50	This report	Acentric factor	None
322.5	29.9	This report	Acentric factor	1, 2, 3 atm extrapolations of Clapeyron and Antoine equations and estimated P _C = 40 atm, T _C = 335 K
312.2	22.8	This report	Acentric factor	As above but including measurements of pressure in tank of liquid AsF ₅
345.7	45.4	This report	Acentric factor	As above with P _C = 35 atm, T _C = 335 K
330.1	33.8	This report	Acentric factor	1, 2, 3 atm extrapolations of Clapeyron and Antoine equations plus measurements of pressure in tank of liquid AsF
(335) ^b	28.7	This report	Vapor pressure-fugacity-molar volume Eq. (8)	As above

^aIn all cases, original experimental data of Russell, Rundle, and Yost, for AsF₃, and of Ruff, Menzel, and Plaut, for AsF₅, were used.

^bInput value.

Marquardt nonlinear least squares program;²⁰ T_c and P_c (actually $\ln P_c$) are the parameters to be calculated. The functions $f^{(0)}$ and $f^{(1)}$ are evaluated according to the procedure recommended by Reid, Prausnitz, and Poling [their Eq. (2-3.4)]. Reasonable estimates of T_c and P_c — based only on the experimental data of Ruff, Menzel, and Plaut,¹¹ and with the additional data from Otey¹² — were obtained for AsF_5 , as shown in Table 5. However, the standard deviations are large.

2.3 STANDARD STATE FUGACITY AND HEAT CAPACITY EQUATIONS

The analyses in Sects. 2.1 and 2.2 were performed to evaluate T_c and P_c , which are needed to determine the parameters C_i of the reference state fugacity equation (8).

$$\ln[p^S] = C_1 + C_2/T + C_3T + C_4\ln(T) + C_5T^2 - \ln(\phi^S) + p^{SL}/(RT) \quad (8)$$

In this equation

- p^S = the experimental saturated-vapor pressure, bar (the pressure unit used in the model of Prausnitz et al.),
- T = the absolute temperature (K),
- v^L = the volume of the saturated liquid (mL/g·mole),
- ϕ^S = the saturated-vapor fugacity coefficient,
- C_i = adjustable constants obtained by fitting experimental vapor-pressure data to Eq. (8),
- R = the gas constant.

Equation (8) was used as an additional method of calculating P_c ; this method requires that the critical temperature be specified as input data. The results of this calculation are presented in Table 5. The selected values of P_c and T_c and other pertinent parameters for calculating liquid-vapor equilibria are listed in Table 6. These parameters, their use, and methods of calculation are discussed by Prausnitz and his colleagues⁴ and by Cochran and Davis.⁵ The five parameters C_i of Eq. (8), evaluated by means of vapor-pressure data and the parameters of Table 6, constitute the second of three lines of data needed for each pure chemical in the library of thermodynamic properties. This library is listed as file LIBRARY.NEW in Exhibit A.1 of Appendix A.

Heat capacity parameters of the ideal-gas state of each chemical species are also required for completion of the library of thermodynamic properties, as shown in Eq. (9).

$$C_p^{ig} = B_1 + B_2/T + B_3T + B_4\ln T \quad (9)$$

where:

- C_p^{ig} = the heat capacity of the ideal-state gas (J/K/g·mol),
- B_i = adjustable constants obtained by fitting experimental heat-capacity data to Eq. (9).

Table 6. Some thermodynamic properties of AsF₃ and AsF₅

	AsF ₃	AsF ₅
Molecular weight, g/g•mole	131.917	169.914
Critical temperature, K	515	335
Critical pressure, atm	40	40
Critical pressure, bar	40.5	40.5
Normal boiling temperature, K	328.5 to 333.8	220.5
Dipole moment, D	2.815	0.0
Van der Waals' volume, V _W , cm ³ /g•mole	30.04	38.85
Van der Waals' area, A _W × 10 ⁻⁸ , cm ² /g•mole	4.53	7.25
UNIQUAC r ^a	1.98	2.56
UNIQUAC q ^a	1.81	2.90
Rackett parameter, Z _{RA} , Å	0.3264	0.3082
Radius of gyration, Å	2.528	2.395

^aThis is a dimensionless parameter of the pure chemical component. It should not be confused with the parameters Δu_{12} and Δu_{21} , which pertain to the liquid phase and which can be determined only from binary liquid-vapor equilibrium data, as noted in refs. 4 and 5.

Values of B_i were obtained using the Marquardt nonlinear least squares program²⁰ and the C_p^{ig} versus temperature tabulations of O'Hare.²¹ These and other thermodynamic parameters for AsF₃ and AsF₅ are also listed in Appendix A in the file LIBRARY.NEW. On the second page of this exhibit, AsF₃ and AsF₅ are shown as components 28 and 29 respectively. The first of the three lines allocated each of these components lists its name, critical temperature in K, critical pressure in bars, the Rackett parameter and radius of gyration in angstrom units, the dipole moment in Debyes, the dimensionless UNIQUAC r and q, and the dimensionless UNIQUAC q*, which is normally set equal to q except for solutions containing water and alcohols, as discussed in Chap. 4 of Prausnitz et al.⁴ These parameters are listed in Table 6. As noted previously, the second line of data for each contains the parameters C_i of Eq. (8), and the third line contains the parameters B_i of Eq. (9).

The Rackett parameter, which is used to estimate saturated-liquid molar volumes as described in refs. 4 and 5, is the only additional

calculation discussed here. That for AsF_3 was calculated as the critical compressibility; that for AsF_5 was calculated from the two measurements of liquid density reported by Ruff, Menzel, and Plaut.¹¹ Liquid volumes and fugacity coefficients derived from these Rackett parameters and the associated vapor-pressure data are presented in Appendix B.

3. RESULTS AND DISCUSSION

Two objectives of this report are (1) to evaluate the nature of the very practical problem of the distribution of arsenic fluoride impurities between liquid and vapor phases of UF_6 and (2) to test further the applicability of the mathematical model of Prausnitz and his coworkers concerning liquid-vapor equilibrium in systems of low-concentration impurities in UF_6 . Vapor pressure data for AsF_3 have been measured only at pressures below 138 mm Hg, and there are no data on vapor-liquid equilibrium in the system AsF_3 - UF_6 . Fortunately, there are methods available whereby the critical temperature and pressure of AsF_3 can be estimated and the vapor pressure estimated for temperatures above the maximum at which this was measured. In spite of the need for large extrapolations of available data, Fig. 1 shows that the vapor pressure of AsF_3 is nearly equal to that of UF_6 , from which we might guess that the vapor/liquid equilibrium constant $[K(v/l)]$ is approximately 1. On the basis of the extrapolations, a value $K(v/l) \approx 0.7$ is shown in Table 7. As usual, $K(v/l)$ is defined as $[y(\text{impurity})/y(\text{UF}_6)]$ in the vapor phase divided by $[x(\text{impurity})/x(\text{UF}_6)]$ in the liquid phase, where x and y are mole fractions in liquid and vapor phases respectively. Therefore we conclude that AsF_3 will tend to concentrate in the liquid phase of UF_6 in either a UF_6 -condensing or UF_6 -vaporizing operation if the operation is conducted under (nearly) equilibrium conditions.

Calculations pertaining to AsF_5 are not subject to as large uncertainties as those of AsF_3 for several reasons. First, vapor pressures measured by Ruff, Menzel, and Plaut¹¹ extend nearly up to 1 atm. Second, Johnson and Otey measured the distribution of AsF_5 between vapor and liquid phases of UF_6 .²² Third, Otey¹² advised us of two measurements of the pressure in cylinders of liquid AsF_5 .

Regression of Eq. (8) was performed with several combinations of data, all of which included the measurements of Ruff, Menzel, and Plaut plus the values of temperature calculated by extrapolating the Clapeyron Eq. (1) (see Table 1) and the Antoine Eq. (2) (see Table 2). To these were added either the AsF_5 -cylinder-pressure measurements or critical pressure estimates of 35, 40, and 45 atm at 335 and 340 K. The brief parametric study represented by the 6 sets of calculations was chosen to provide a span of 5 atm less than and 5 atm greater than the "best guess" critical pressure of 40 atm shown in Table 6. Similarly, the 340 K was chosen to be 5 K above the best guess critical temperature of 335 shown in Table 6. In summary, all 6 sets of conditions gave essentially the same results: an estimated standard deviation of fit of $\ln[p^S]$ of Eq. (8) equal to ~ 0.0176 , which corresponds to an estimated standard deviation of pressure, p^S , less than 2%. The values of C_i , of course, did depend on the input estimates of P_c and T_c . On the basis of this parametric study, the estimated critical pressure of 40 atm and critical temperature of 335 K listed in Table 6 remain our best estimates and we disregard the measurements of AsF_5 cylinder pressure at ambient temperature.

Table 7. Experimental data ranges and calculated vapor-liquid equilibrium coefficients for various impurities in UF₆

Impurity in UF ₆	Ranges of experimental data				Range of calculational variables		UNIQUAC parameters		Range of K(v/l)
	Temperature (K)	Pressure (bar)	Composition (mole fraction)		Temperature (K)	Pressure (bar)	Δu_{12}	Δu_{21}	
			Liquid	Vapor			(cm ³ ·bar/mol)	(cm ³ ·bar/mol)	
F ₂	337 - 358	1.93 - 3.21	0.66E-03 - 1.51E-03	0.128 - 0.224	337 - 358	1.9 - 3.2	0 -11.765	0 4212.2	96 - 158 99 - 330
Cl ₂	None	None	None	None	339.9	1.6	0	0	8.3
ClF	None	None	None	None	339.9	1.6	0	0	73.6
ClF ₃	298 - 353	1.5 - 3.1	0.01 - 0.86	0.10 - 0.935	298 - 353	1.5 - 3.1	0 -488	0 958	0.964 - 15.5 1.275 - 9.85
ClO ₂ F	None	None	None	None	339.9	1.6	0	0	128
SiF ₄	374 - 376	4.5 - 5.2	1.9E-06 - 1.42E-04	1.67E-05 - 9.45E-03	374 - 376	4.5 - 5.2	0 76.59	0 -28500	21.1 - 23.4 54.5 - 152
CF ₄	None	None	None	None	339.9	1.6	0	0	71
C ₂ F ₆	None	None	None	None	339.9	1.6	0	0	27
Freon-114 (CClF ₂ CClF ₂)	304 - 359	2.61 - 3.47	0.024 - 0.952	0.094 - 0.983	304 - 359	2.61 - 3.47	0 13.84	0 -30.35	1.04 - 4.14 1.05 - 3.78
CO ₂	None	None	None	None	344	2.38	0	0	22.6
SF ₆	None	None	None	None	344	2.38	0	0	23.4 - 23.5
ClO ₂ F	None	None	None	None	344	2.38	0	0	16.4 - 16.6
AsF ₃	None	None	None	None	375	4.4 - 4.6	0	0	0.72 - 0.73
AsF ₅	375	4.4 - 4.6	6.2E-06 - 8.1E-04	4.1E-05 - 7.5E-03	375	4.4 - 4.6	0 230	0 -230	11.5 - 11.7 7.42 - 7.48

The results of these analyses are presented in the file LIBRARY.NEW, Appendix A. The parameters C_i of Eq. (8) plus fugacity coefficients and molar volumes shown in Table B.2 were used to calculate saturated vapor pressures of AsF_5 at temperatures ranging from 0 to 35°C. Results are listed in Table 8, where values may be compared with those mentioned by Otey, namely his measurement 16.6 atm at 23.8°C and 15.6 ± 0.7 atm at 25°C, from the Material Safety Data Sheet of Ozark-Mahoning.¹² It is apparent that there is agreement between the "best estimate" pressure of Table 8 and the values specified by Ozark-Mahoning at 25°C.

Table 8. Calculated vapor pressure of AsF_5

Temperature (°C)	p^s (atm)	
	Best estimate ^a	Range ^b
0.0	8.01	7.97 - 8.10
5.0	9.17	9.22 - 9.45
10.0	10.65	10.73 - 10.97
15.0	12.15	12.04 - 12.62
20.0	13.77	13.61 - 14.46
25.0	15.51	15.27 - 16.49
30.0	17.35	17.02 - 18.73
35.0	19.29	18.84 - 21.21

^aBased on $P_C = 40$ atm and $T_C = 335$ K.

^bBased on estimated P_C ranging from 35 to 45 atm and T_C equal to 335 to 340 K.

As noted above, Johnson and Otey measured concentrations of AsF_5 in liquid and vapor phases of UF_6 at 216°F. Because the data of Johnson and Otey are not publicly available, they are listed in Appendix C with their approval.²² Figure 2 shows the concentration of AsF_5 in liquid UF_6 calculated from the vapor-phase concentration of AsF_5 plotted against the measured concentration in the liquid phase. The open circles of this figure are based only on the properties of pure AsF_5 and UF_6 ; i.e., the UNIQUAC parameters Δu_{12} and Δu_{21} are both set to zero. It is apparent that the calculated concentrations of AsF_5 are all less than measured concentrations. However, when we set $\Delta u_{12} = 230$ K and $\Delta u_{21} = -230$ K, concentrations of AsF_5 in the liquid phase calculated from vapor-phase concentrations are in excellent agreement with measured concentrations. We emphasize again the fact that the parameters of the reference-state fugacity Eq. (8) are based on extrapolated vapor pressures and that the liquid-vapor equilibrium data of Johnson and Otey (Appendix C)²² are not adequate to permit nonlinear least-squares estimation of Δu_{ij} . Thus, it is not possible to present estimates of the standard deviations of Δu_{ij} or of the fit of the data to the model of Prausnitz and his coworkers.

An additional uncertainty concerning the estimation of the Δu_{ij} arises from the fact that AsF_5 is reported by Gut and Gautschi to be a quite strong fluoride ion acceptor.²³ Additionally, these authors state

Fig. 2. Comparison of calculated and observed concentrations of AsF₅ in UF₆. Data from Johnson and Otey (ref. 22). The line corresponds to the calculated concentration of AsF₅ in the liquid phase being equal to the measured value.

that the particle $\text{As}_2\text{F}_{11}^-$ appears as intermediate during the formation of AsF_6^- . Therefore, the concentration of HF in any solution containing AsF_5 in UF_6 may have a significant effect on the distribution of AsF_5 between liquid and vapor phases. In their work Johnson and Otey used UF_6 from a cylinder that had been flashed several times to remove impurities, including HF, that are more volatile than UF_6 . The vapor pressure of the resulting material at 23.8°C was found to be very close to that of pure UF_6 listed by DeWitt.^{2*} However, trace quantities of HF may have been present.¹²

Association and solvation between any two chemical species, such as HF and AsF_5 , or between molecules of the same specie, such as HF with HF, are represented by the parameters η_{ij} , shown as starting on the second page of Exhibit A.1 of Appendix A. Prausnitz and his coworkers⁴ present such parameters for 92 fluids in their Appendix C-4. To date no values of η_{ij} have been determined for interactions between any pair of potential impurities in UF_6 , nor for association of any individual impurity with UF_6 ; hence, the effects of η_{ij} on systems involving UF_6 as a solvent have not been evaluated. When binary equilibrium data are regressed to estimate UNIQUAC parameters and values of η_{ij} are set to zero, the error in ignoring the effects of association is accommodated, to some degree, in the UNIQUAC parameters. In Exhibit A.1 the nonzero values of η_{ij} are taken from Prausnitz et al.³ except for the estimated 2.00 for HF-HF interactions. It may be noted that the η_{ij} are read by SUBROUTINE PRDTA2(ISTP) and used in calculations of virial coefficients in SUBROUTINE BIJS (N, ID, KEY, T, ERB) and SUBROUTINE BIJS2 (KEY, T) as described in Appendix D of ref. 4.

Figure 3 provides a comparison of the separation factors of several impurities from UF_6 , namely, F_2 , SiF_4 , AsF_5 , and AsF_3 . For F_2 the experimental data⁶ show $K(v/l)$ to range from 100 to 300. As noted on Fig. 3 for SiF_4 , $K(v/l) = 78.80$, the standard deviation of this value being 29.74, or about 38%. A visual comparison of the scatter of the AsF_5 data with the SiF_4 data suggests that the standard deviation of the value $K(v/l) = 7.5$ for AsF_5 is less than 30% of 7.5.

The separation factor for AsF_3 is shown as 0.72 in Fig. 3. No estimate of the accuracy of this value can be given because of the large extrapolations of vapor pressure involved in its calculation and because of the absence of experimental data on liquid-vapor equilibrium in the system AsF_3 - UF_6 .

As indicated in Sect. 2.2, there is much uncertainty in the estimates of P_c and T_c . The value 15.6 atm pressure listed on the Material Safety Data Sheet of Ozark-Mahoning is most consistent with critical parameters of 335 K and 40 atm. However, the uncertainty, ± 0.7 atm, stated for the 15.6 atm, places all calculated values within the range of uncertainty, as seen in Table 8. The data of Johnson and Otey on the distribution of AsF_5 between vapor and liquid phases of UF_6 (Table C.1) also provide an additional method for estimating P_c when T_c is specified as 335 K for AsF_5 . Thus, the vapor-pressure data plus values obtained by extrapolating Eqs. (1) and (2) to 1, 2, and 3 atm were combined with an assumed $P_c = 35, 40, \text{ or } 45$ atm. Distribution coefficients, $K(v/l)$ subsequently calculated from the resulting coefficients of Eq. (8), produced the following results with $\Delta u_{ij} = 0$: (1) with P_c assumed to be

Fig. 3. Distribution of F_2 , SiF_4 , AsF_3 , and AsF_5 between liquid and vapor phases of UF_6 .

35 atm, $K(v/l) \approx 4.2$; (2) with P_c assumed to be 40 atm, $K(v/l) = 11.3$ to 11.6. When $\Delta u_{12} = 230$ and $\Delta u_{21} = -230$ cm³ bar/mol, the value of $K(v/l)$ is reduced to the range 7.22 to 7.37; this range includes the average of the values calculated from the data of Johnson and Otey,²² Appendix C, namely 7.3 ± 1.5 . The chosen values of T_c and P_c are consistent with available data on pure AsF₅ and UF₆, and on the available binary system data; thus they are useful to the extent that HF does not grossly affect the system.

4. SUMMARY

In support of studies to determine the distribution of arsenic fluorides between vapor and liquid phases in operations consisting of condensing UF₆ into, or vaporizing UF₆ from, a cylinder, various thermodynamic properties of AsF₃ and AsF₅ have been assembled. Some of the data are from experimental data and some from calculations and extrapolations according to several procedures. Experimental data include low-pressure measurements of vapor pressure and, in the case of AsF₅, concentrations of AsF₅ in liquid and vapor phases under equilibrium conditions.²⁰ Among the various properties estimated from the meager data are critical temperature (T_c) and critical pressure (P_c); various other properties are estimated for inclusion in the library of thermodynamic data required by the model of Prausnitz and his coworkers to describe liquid-vapor equilibria. Data on AsF₅ provide additional support for this model and its potential use in calculating distributions of impurities between vapor and liquid phases of UF₆.

Arsenic trifluoride is found to concentrate in the liquid-UF₆ phase; the concentration of AsF₅ in the vapor phase will be about 7.5 times its concentration in the liquid phase. Unless the concentration of AsF₅ is large enough to prevent condensation of UF₆ in a condensation operation, most of any AsF₅ present will report to the liquid phase because its weight will be so much greater than that of the vapor phase. Therefore, AsF₅, in addition to AsF₃, will be available to cause corrosion.

As is well known, uranium isotope separation processes are based on differences in masses between ²³⁵UF₆ and ²³⁸UF₆. The ²³⁵U travels toward the top of the separation cascade while ²³⁸U travels toward the bottom, thereby producing separation of the uranium isotopes. So-called light gases of molecular weight significantly less than the 352 g UF₆/mole, such as AsF₃ and AsF₅, with molecular weights of 131.917 and 169.914 g/mole, respectively (Table 6), will thus travel up the cascade. In particular, the separation of these light gases from UF₆ is much more extensive than the separation of ²³⁵UF₆ from ²³⁸UF₆. In practice the light gases are separated from UF₆ in a "purge cascade." Therefore, AsF₃ and AsF₅ will become part of the purge-gas stream (along with nitrogen, oxygen, and other low-molecular weight gases) unless these arsenic fluorides are consumed by chemical reaction. The arsenic fluorides will not otherwise remain in the plant. There are many discussions of isotope separation processes such as that in Benedict and Pigford.²⁵

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APPENDIX A
SUMMARY OF THERMODYNAMIC DATA

Appendix A
SUMMARY OF THERMODYNAMIC DATA

This appendix lists in Table A.1 some thermodynamic data for the 15 fluorides studied in this series of reports and as Exhibit A.1, the library file LIBRARY.NEW. This file contains the library of thermodynamic properties associated with the study of UF_6 and some of its potential impurities, as well as some other chemicals that are included as being of possible use. The authors extracted the data concerning fluorine compounds from the literature or, in some cases, calculated these data.

Table A.1. Vapor pressure data ranges and critical constants

Chemical	Temperature range of data (K)	Pressure range of data (bar)	Critical ^a temperature T _c (K)	Critical ^a pressure P _c (bars)	Boiling temperature T _b (K)	Temperature ratio T _c /T _b
UF ₆ ^b	337.35 - 503.35	1.527 - 46.1	504.3	46.1	324.98	1.552
F ₂ ^c	54.01 - 143.49	0.003 - 50.36	144.3	52.15	84.95	1.699
HF ^d	273.15 - 377.56	0.486 - 11.75	461.0	64.85	292.67	1.575
Cl ₂ ^e	162.7 - 235.2	0.003 - 0.845	417.0	77.1	239.1	1.744
Cl ₂ ^f	172.12 - 240.05	0.014 - 1.058	417.2	77.1	239.1	1.745
ClF ^g	128.15 - 172.35	0.011 - 1.013	259.0	40.5	183.0	1.42
ClF ₃ ^h	226.18 - 302.7	0.039 - 2.041	427.6	42.62	284.91	1.501
ClO ₂ F ⁱ	152.81 - 362.92	0.004 - 48.90	368.4	53.7	228.5	1.612
ClO ₂ F ₂ ^j	164.11 - 228.67	0.014 - 1.121	368.4	53.7	228.5	1.612
ClO ₂ F ₂ ^k	161.15 - 229.45	0.011 - 1.208	368.4	53.7	228.5	1.612
SiF ₄ ^{m,n}	183.85 - 258.88	1.887 - 37.10	259.0	40.5	187.0	1.39
CF ₄ ^p	89.77 - 145.59	0.001 - 1.047	227.5	37.42	145.12	1.568
C ₂ F ₆ ^q	188.20 - 292.59	6.679 - 30.106	293.0	30.4	194.88	1.503
Freon-114 ^r	290.6 - 415.2	1.69 - 30.61	419.0	32.6	276.9	1.513
SF ₆ ^s	273.0 - 318.7	12.65 - 46.18	318.7	37.6	205.0	1.55
ClO ₂ F ₂ ^{t,u}	195.2 - 266.9	0.01 - 0.99	419.1	64.6	268.0	1.56
AsF ₃ ^v	265 - 291.9	0.038 - 0.183	515.0	40.0	331.0	1.54
AsF ₅ ^w	195.8 - 220.0	0.24 - 0.997	335.0	40.0	220.5	1.52
Average ^x						1.562
Standard deviation						0.100

^aSee W. Davis, Jr., and H. D. Cochran, *Modeling Vapor-Liquid Equilibrium of UF₆ and Common Impurities*, K/ITP-171, Martin Marietta Energy Systems, Inc., Oak Ridge Gaseous Diffusion Plant, March 1989.

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^xThe ratio T_c/T_b for ClO₂F is counted only once.

Exhibit A.1 File LIBRARY.NEW

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$ THESE ARE THE THERMODYNAMIC PROPERTIES OF THE PURE COMPONENTS
$J-***NM1(J)**--NM2(J)--+TC(J)+**PC(J)*--A(J)--+RD(J)+*DM(J)-RU(J)+QU(J)-QP(J)
$
$                                     RADIUS
$                                     OF
$                                     DIPOL
$                                     UNQUAC
$                                     ZRA GYRATION MOM R Q Q'
1HYDROGEN 43.60 20.80 0.0000 0.3710 0.000 0.000 0.000 0.000
$ THESE ARE THE PARAMETERS OF THE FUGACITY-COEFFICIENT EQUATION
1 2.00670E+01 -3.28480E+02 0.00000E+00 -2.59800E+00 0.00000E+00
$ THESE ARE THE PARAMETERS OF THE HEAT-CAPACITY EQUATION
1 9.45050E+01 -2.73890E+03 1.07070E-02 -1.04780E+01
2NITROGEN 126.26 33.99 0.0000 0.5470 0.000 0.000 0.000 0.000
2 2.33200E+01 -9.51240E+02 0.00000E+00 -2.59800E+00 0.00000E+00
2 6.97480E+01 -8.70720E+02 1.56370E-02 -7.43770E+00
3OXYGEN 154.76 50.82 0.0000 0.6040 0.000 0.000 0.000 0.000
3 2.42510E+01 -1.16600E+03 0.00000E+00 -2.59800E+00 0.00000E+00
3 -1.68200E+01 1.71130E+03 2.29060E-03 6.97710E+00
4CARBON MONOXIDE 132.93 34.99 0.0000 0.5580 0.130 0.000 0.000 0.000
4 2.34830E+01 -1.00150E+03 0.00000E+00 -2.59800E+00 0.00000E+00
4 6.01740E+01 -5.46420E+02 1.49300E-02 -5.90960E+00
5ARGON 150.71 48.65 0.0000 1.0760 0.000 0.000 0.000 0.000
5 2.41390E+01 -1.13540E+03 0.00000E+00 -2.59800E+00 0.00000E+00
5 2.07900E+01 0.00000E+00 0.00000E+00 0.00000E+00
6METHANE 190.58 41.91 0.0000 1.1230 0.000 0.000 0.000 0.000
6 2.45990E+01 -1.43580E+03 0.00000E+00 -2.59800E+00 0.00000E+00
6 -1.86040E+02 8.14260E+03 2.45590E-02 3.28410E+01
7CARBON TETRACHLORIDE 556.30 45.57 0.2721 3.4580 0.000 3.330 2.820 2.820
7 3.88950E+01 -5.36280E+03 -5.77840E-03 -3.74900E+00 3.16630E-06
7 3.13890E+01 -5.94940E+03 -1.22070E-02 1.33380E+01
8CHLOROFORM 536.54 54.74 0.2748 3.1780 1.020 2.700 2.340 2.340
8 1.74240E+02 -8.14000E+03 6.59750E-02 -2.90110E+01 -3.00010E-05
8 -2.84240E+02 5.21180E+03 -4.57330E-02 6.07030E+01
9DICHLOROMETHANE 510.37 60.67 0.2620 2.4320 1.540 2.140 1.920 1.920
9 -3.05090E+02 3.17440E+03 -1.59500E-01 5.89530E+01 6.22520E-05
9 -3.84350E+02 9.94580E+03 -4.18030E-02 7.27280E+01
10URANIUM HEXAFLUORIDE 505.70 46.60 0.2741 2.3246 0.000 3.270 3.120 3.120
10 -2.61208E+02 1.85089E+03 -1.48012E-01 5.13377E+01 6.31756E-05
10 1.35690E+02 3.32040E-02 0.00000E+00 -1.34170E+06
11FLUORINE 144.31 52.15 0.0000 0.7140 0.000 0.000 0.000 0.000
11 2.40900E+01 -1.08720E+03 0.00000E+00 -2.59800E+00 0.00000E+00
11 2.69280E+01 1.71800E-02 -6.63410E-06 4.18610E+03
12HYDROGEN FLUORIDE 461.00 64.80 0.1451 0.2006 1.736 0.623 0.746 0.746
12 -3.84676E+02 5.91874E+03 -1.94731E-01 7.29272E+01 8.49052E-05
12 2.74320E+01 2.22660E-03 5.86260E-07 9.19770E+04
13CARBON DIOXIDE 241.00 53.80 0.2000 0.9920 0.180 1.320 1.280 1.280
13 6.32080E+00 -2.64230E+03 -3.93220E-02 2.73470E+00 2.67180E-05
13 -1.09190E+02 2.46630E+03 -9.75310E-03 2.47400E+01
14CHLORINE 417.00 77.00 0.2768 0.9873 0.000 1.570 1.450 1.450
14 2.12829E+01 -3.21025E+03 -1.34546E-02 -9.45042E-01 9.20980E-06
14 3.14990E+01 1.07260E-02 -4.44590E-06 -3.40860E+04
15CHLORINE MONOFLUORID 259.00 55.70 0.0000 1.2484 0.881 0.000 0.000 0.000
15 2.53627E+01 -1.95131E+03 0.00000E+00 -2.59800E+00 0.00000E+00
15 2.82890E+01 1.52480E-02 -6.18560E-06 -7.88310E+03
16CHLORINE TRIFLUORIDE 427.60 42.62 0.1958 2.3066 0.554 2.830 1.480 1.480
16 3.12929E+01 -4.61117E+03 -6.23739E-03 -2.33201E+00 -2.05938E-06
16 5.28360E+01 5.15760E-02 -2.21740E-05 -2.14650E+05
17PERCHLORYL FLUORIDE 368.40 53.70 0.2787 2.0411 0.023 2.090 1.930 1.930
17 1.16041E+02 -4.68596E+03 5.67884E-02 -1.96287E+01 -3.49162E-05
17 3.99170E+01 1.03770E-01 -4.18050E-05 -1.54260E+05
18SILICON TETRAFLUORID 259.10 37.20 0.0000 2.7291 0.000 0.000 0.000 0.000
18 2.52755E+01 -1.95131E+03 0.00000E+00 -2.59800E+00 0.00000E+00

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Exhibit A.1 File LIBRARY.NEW (continued)

18	5.48400E+01	1.05650E-01	-6.17890E-05	-6.90820E+05				
19	1,2-DICHLOROETHANE	560.93	53.71	0.2660	2.8510	0.000	2.880	2.520 2.520
19	-2.46030E+02	2.94260E+03	-7.11430E-02	4.47810E+01				0.00000E+00
19	-4.22180E+02	1.09500E+04	-2.13400E-02	8.27130E+01				
20	CARBON TETRAFLUORIDE	227.60	37.40	0.0000	2.6350	0.000	0.000	0.000 0.000
20	2.49745E+01	-1.71399E+03	0.00000E+00	-2.59800E+00				0.00000E+00
20	3.35880E+01	1.11550E-05	-4.44510E-05	-1.04390E+05				
21	HEXAFLUOROETHANE	292.90	31.30	0.0000	3.3797	0.000	0.000	0.000 0.000
21	2.56757E+01	-2.20671E+03	0.00000E+00	-2.59800E+00				0.00000E+00
21	6.05340E+01	2.15020E-01	-1.19030E-04	-6.83620E+05				
22	HYDROGEN CHLORIDE	324.54	82.60	0.2625	0.2990	1.070	1.000	1.000 1.000
22	3.73830E+01	-2.80160E+03	-1.66250E-03	-4.26520E+00				3.12100E-06
22	5.71390E+01	-6.61650E+02	9.71000E-03	-5.03060E+00				
23	WATER	647.37	221.20	0.2380	0.6150	1.830	0.920	1.400 1.400
23	5.70420E+01	-7.00480E+03	5.58880E-03	-6.66890E+00				-8.50540E-07
23	2.55730E+01	8.97860E+02	1.37050E-02	1.58250E-01				
24	AMMONIA	405.54	112.80	0.2465	0.8530	1.470	1.000	1.000 1.000
24	1.64110E+01	-3.53190E+03	-1.85220E-02	3.97730E-01				9.91780E-06
24	-8.75470E+01	4.09850E+03	1.17200E-02	1.85830E+01				
25	CCLF2CCLF2	418.9	32.2	0.2752	3.6751	0.000	2.71	2.02 2.02
25	-1.45035E+03	2.86732E+04	-7.81991E-01	2.73014E+02				3.65584E-04
25	2.25341E+03	-7.70866E+04	5.79899E-01	-3.60174E+02				
26	SF6	318.73	37.1	0.2810	2.8224	0.000	2.95	3.47 3.47
26	3.52579E+02	-1.17910E+04	5.42954E-02	-5.79553E+01				4.27463E-05
26	-6.21483E+02	8.50873E+03	-1.15725E-01	1.26945E+02				
27	CHLORYL FLUORIDE	419.1	63.8	0.2913	2.2763	1.722	1.99	1.39 1.39
27	-5.14507E+03	7.44906E+04	-4.67032E+00	1.05054E+03				3.41429E-03
27	-2.28327E+02	3.80743E+03	-4.45152E-02	5.00910E+01				
28	ARSENIC TRIFLUORIDE	515.	40.5	0.3264	2.528	0.000	1.98	1.81 1.81
28	-5.40895E+03	1.09240E+05	-2.96601E+00	1.01828E+03				1.39211E-03
28	-1.38228E+02	6.20575E+02	-4.23906E-02	3.74514E+01				
29	ARSENIC PENTAFLUORIDE	335.	40.5	0.3082	2.395	0.000	2.56	2.90 2.90
29	-1.51884E+02	-3.73073E+02	-1.54414E-01	3.38961E+01				9.67849E-05
29	-3.08916E+02	1.63980E+03	-8.28884E-02	7.44728E+01				

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\$ THESE ARE THE BINARY ASSOCIATION PARAMETERS, ETA

1	H2	0.00							
2	N2	0.00	N2	0.00					
3	O2	0.00	N2	0.00	O2	0.00			
4	CO	0.00	N2	0.00	O2	0.00	CO	0.00	
5	AR	0.00	N2	0.00	O2	0.00	CO	0.00	AR
6	CH4	0.00	N2	0.00	O2	0.00	CO	0.00	AR
7	CCL4	0.00	N2	0.00	O2	0.00	CO	0.00	AR
			CCL4	0.00					
8	CHCL3	0.00	N2	0.00	O2	0.00	CO	0.00	AR
			CCL4	0.00	CHCL3	0.00			
9	CH2CL2	0.00	N2	0.00	O2	0.00	CO	0.00	AR
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	
10	UF6	0.00	N2	0.00	O2	0.00	CO	0.00	AR
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6
11	F2	0.00	N2	0.00	O2	0.00	CO	0.00	AR
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6
12	HF	0.00	N2	0.00	O2	0.00	CO	0.00	AR
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6
			HF	2.00					
13	CO2	0.00	N2	0.00	O2	0.00	CO	0.00	AR
			CCL4	0.00	CHCL3	0.10	CH2CL2	0.00	UF6
			HF	0.00	CO2	0.16			
14	CL2	0.00	N2	0.00	O2	0.00	CO	0.00	AR
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6

Exhibit A.1 File LIBRARY.NEW (continued)

		HF	0.00	CO2	0.00	CL2	0.00			
15	CLF	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
16	CLF3	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
17	CL03F	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
			CL03F	0.00						0.00
18	SIF4	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00				0.00
19	CH4CL2	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00	CH4CL2	0.00		0.00
20	CF4	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00
21	C2F6	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00
22	HCL	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00
			HCL	0.00						0.00
23	H2O	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.10	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.30	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00
			HCL	1.38	H2O	1.70				0.00
24	NH3	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.30	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.20	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00
			HCL	2.20	H2O	0.20	NH3	0.00		0.00
25	F-114	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00
			HCL	0.00	H2O	0.00	NH3	0.00	F-114	0.00
26	SF6	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00
			HCL	0.00	H2O	0.00	NH3	0.00	F-114	0.00
27	CL02F	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00
			HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00
			CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00
			HCL	0.00	H2O	0.00	NH3	0.00	F-114	0.00
			CL02F	0.00						0.00
28	ASF3	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
			CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00

Exhibit A.1 File LIBRARY.NEW (continued)

	HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00	CLF3	0.00
	CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00	C2F6	0.00
	HCL	0.00	H2O	0.00	NH3	0.00	F-114	0.00	SF6	0.00
	CL02F	0.00	ASF3	0.00						
29	ASF5	0.00	N2	0.00	O2	0.00	CO	0.00	AR	0.00
	CCL4	0.00	CHCL3	0.00	CH2CL2	0.00	UF6	0.00	F2	0.00
	HF	0.00	CO2	0.00	CL2	0.00	CLF	0.00	CLF3	0.00
	CL03F	0.00	SIF4	0.00	CH4CL2	0.00	CF4	0.00	C2F6	0.00
	HCL	0.00	H2O	0.00	NH3	0.00	F-114	0.00	SF6	0.00
	CL02F	0.00	ASF3	0.00	ASF5	0.00				

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 \$ THESE ARE THE UNIQUAC BINARY INTERACTION PARAMETERS

1	23	9.97	-	880.00	H2 IN WATER
1	24	0.68		1344.00	H2 IN AMMONIA
2	10	1.507	-	36.85	N2 IN UF6
2	23	3.76		913.00	N2 IN WATER
2	24	3.35		2098.00	N2 IN AMMONIA
3	10	5.349		1864.	O2 IN UF6
3	7	14.74		38.19	CHCL3 IN CCL4
11	10	11.765		4212.2	F2 IN UF6
13	10	0.		0.	CO2 IN UF6
16	10	488.03		958.0	CLF3 IN UF6
18	10	76.59	-	2.85E+04	SIF4 IN UF6
25	10	13.84	-	30.35	FREON-114 IN UF6
26	10	0.		0.	SF6 IN UF6
27	10	0.		0.	CL02F IN UF6
28	10	0.		0.	ASF3 IN UF6
29	10	230.	-	230.	ASF5 IN UF6

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APPENDIX B
CALCULATED FUGACITY COEFFICIENTS AND
LIQUID VOLUMES OF AsF_3 AND AsF_5

Appendix B
CALCULATED FUGACITY COEFFICIENTS AND
LIQUID VOLUMES OF AsF_3 AND AsF_5

The model of Prausnitz and his coworkers calculates the fugacity coefficient and liquid volume of each pure material from the Rackett parameter and from vapor pressure-temperature data. These properties are listed in Table B.1 for AsF_3 and in Table B.2 for AsF_5 .

Table B.1. Calculated fugacity coefficients and liquid volumes of AsF_3^a

Temperature (K)	Pressure (bars)	Fugacity coefficient	Liquid volume (mL/mole)
265.02	3.85E-02	0.9960	138.70
268.23	4.82E-02	0.9953	139.17
272.06	6.09E-02	0.9944	139.73
275.27	7.36E-02	0.9935	140.21
280.18	9.89E-02	0.9919	140.96
288.05	1.50E-01	0.9896	142.19
291.82	1.83E-01	0.9873	142.79
291.88	1.83E-01	0.9873	142.80
328.50	1.01E+00	0.9565	149.23
333.80	1.01E+00	0.9591	150.25
346.10	2.03E+00	0.9298	152.76
356.50	2.03E+00	0.9371	155.01
357.40	3.04E+00	0.9080	155.21
372.00	3.04E+00	0.9203	158.65
515.00	4.05E+01	0.7148	172.22

^aSource of vapor pressure-temperature data: H. Russell, Jr., R. E. Rundle, and D. M. Yost, "The Heat Capacity, Heats of Fusion and Vaporization, Vapor Pressure and Entropy of Arsenic Trifluoride," *J. Am Chem. Soc.* 63, 2825 (1941).

Table B.2. Calculated fugacity coefficients and liquid volumes of AsF_5^a

Temperature (K)	Pressure (bars)	Fugacity coefficient	Liquid volume (mL/mole)
195.85	2.40E-01	0.9873	96.98
201.85	3.55E-01	0.9832	98.00
207.95	5.15E-01	0.9780	99.18
211.98	6.50E-01	0.9739	99.99
216.25	8.06E-01	0.9698	100.89
220.05	9.97E-01	0.9648	101.71
220.50	1.01E+00	0.9645	101.81
235.40	2.03E+00	0.9431	105.31
245.40	3.04E+00	0.9258	107.95
220.40	1.01E+00	0.9644	101.79
234.60	2.03E+00	0.9425	105.11
243.80	3.04E+00	0.9244	107.51
298.15	1.69E+00	0.7918	115.38
298.15	1.48E+00	0.8156	115.38

^aSource of vapor pressure-temperature data: O. Ruff, W. Menzel, and H. Plaut, "Die Darstellung Dampfdrucke und Dichten des BF_3 , AsF_5 , and BrF_3 ," *Z. anorg. allgem. Chemie*, 206, 59(1932).

APPENDIX C
DATA OF JOHNSON AND OTEY ON LIQUID-VAPOR
EQUILIBRIUM IN THE SYSTEM $\text{AsF}_5\text{-UF}_6$

Appendix C
DATA OF JOHNSON AND OTEY ON LIQUID-VAPOR
EQUILIBRIUM IN THE SYSTEM $\text{AsF}_5\text{-UF}_6$

Johnson and Otey have obtained and reported data on the distribution of AsF_5 between vapor and liquid UF_6 systems; however, their data are not readily available. For this reason we list in Table B.1 their data and the concentrations in mole fractions obtained from the data.

Table C.1. Data of R. T. Johnson and M. G. Otey concerning properties of AsF_5 and its behavior in UF_6^a

Total pressure (mm Hg)	Reported composition (ppm) ^b		Calculated composition (mole fraction $\times 10^6$)		Calculated distribution coefficient $K(v/l)$
	Liquid phase	Vapor phase	Liquid phase	Vapor phase	
3288	3	20	6.2	41	6.6
3308	7	52	14.5	108	7.4
3339	29	120	60	249	4.2
3345	65	465	135	963	7.1
3342	110	930	230	1927	8.4
3376	170	1450	350	3000	8.6
3376	260	2100	540	4350	8.1
3394	310	1900	640	3940	6.2
3417	390	3620	810	7500	9.3
Average					7.3
Standard deviation					1.5

^aTemperature = 216°F (102°C).

^bReported concentration on an arsenic-to-uranium weight ratio.

Source: R. J. Johnson and M. G. Otey, *Properties of AsF_5 and its Behavior in UF_6* , Paducah Gaseous Diffusion Plant, personal communication, July 1989.

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