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THE PHYSICAL PROPERTIES OF URANIUM HEXAFLUORIDE

A-753

I. Kirshenbaum

A study was made of all the available information on the physical properties of uranium hexafluoride. The data was evaluated and wherever possible thermodynamic correlations and tests for consistency were made. The results are given in this report. (auth)

Wherever both very good and preliminary values are available, only the good values are used. When, however, it is not possible from available information to decide which of several pieces of contradictory data is the best, all data are given. Some of the results given in this report are not available elsewhere. This is especially true of the vapor pressure equations and the thermodynamic data for the gas phase. When no data or perhaps only data at one temperature have been obtained experimentally, theoretical formulas for estimated values at all temperatures are given whenever possible. It must, however, be remembered that in some cases, especially in those referring to the liquid state, these equations are at best only first approximations.

Empirical equations have been fitted by statistical methods to experimental data wherever feasible. Graphs, tables, and equations are used as often as possible.

A bibliography of reports and papers on the physical properties of uranium hexafluoride is also included. This may be used by those who are interested in inspecting all the available data on any of the physical properties.

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Authorizing Official

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Date: July 1, 1943

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Thanks are owed to Drs. M. Kilpatrick, W. Libby and
E. Long for the many valuable suggestions and en-
lightening personal conferences.

Classification cancelled (or changed to UNCLASSIFIED)
by authority of Letter from sec. branch
dated 11-30-63
by R. Sullivan TIE, date 3-21-58

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I. Ideality of the Gas

Most experimental evidence indicates that for all practical purposes the gas may be considered ideal. The Berthelot correction for the entropy is less than 0.1 e.u. at 50°C (v. infra). Molecular weight determinations made by Haworth (1) and Simon (2) on both saturated and unsaturated vapors indicate that, within an experimental error of less than 1%, the vapor of the gas is ideal over a temperature range from 18° to 90°C and a pressure of 1.6 to 520 mm. The observed molecular weights are given in Table I.

TABLE I

<u>Temp Range</u> <u>t°C</u>	<u>Pressure Range</u> <u>mm Hg</u>	<u>Molecular Weight</u> <u>g/mol</u>
14-16	42-53	351 ± 4
18-90	1.6-18	352.2 ± 1.3
14.3-17.6	5.2-66	354 ± 3
50.2	523 (saturated vapor)	354)
	421 (unsaturated vapor)	352)
	393.7 (" ")	351) 352.3 ± 1 average

The theoretical molecular weight is 352.1.

The density of the gas at 49.2°C as a function of pressure has been determined by Haworth (3).

TABLE II

<u>ρ (gm/cc)</u>	<u>P (dynes/cm²)</u>
3.130 x 10 ⁻³	2.534 x 10 ⁵
3.627	2.862
4.278	3.479
5.295	4.246
6.482	5.072
7.240	5.717
8.119	6.315
9.016	7.027

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From this data it is found that $\left(\frac{\partial P}{\partial \rho}\right)_T = 49.2^\circ\text{C}$

$= 1.32 \pm 0.01 \times 10^{-8}$. This is in excellent agreement with the value 1.314×10^{-8} calculated from the perfect gas laws. There seems to be little doubt that for all practical purposes deviations from ideality may be neglected.

II. Spectroscopic Data

The Raman spectrum of the liquid has been studied by Duncan (4). Under the conditions of his experiment two Raman lines appeared: $\Delta\nu = 603$ and 228 cm^{-1} . Both were fairly strong and broad. The 603 cm^{-1} line was the more intense. If the assumption is made that the UF_6 molecule is a regular octahedron, three Raman lines are to be expected. The expected line may not appear because of lower intensity or because it is masked by the 603 cm^{-1} line which has a breadth of about 30 cm^{-1} . Molecular models of lower symmetry would give a larger number of Raman lines. Only a plane hexagon would give as small a number of lines as does the regular octahedron. It should be noted, however, that electron diffraction experiments (5) seem to indicate that uranium hexafluoride does not have a regular octahedral structure. The interference patterns can best be explained by the assumption of rhombic symmetry.

The infra-red spectrum of gaseous UF_6 has been studied by Turkevich (6). His results are summarized in Table III and Fig. 1. The other absorption peaks in the figure are due to impurities.

TABLE III

<u>cm⁻¹</u>	<u>ρ</u>	<u>Intensity</u>
623	16.05	100
675	14.82	10
713-719	14.03-13.91	1/2
825	12.12	2
850	11.68	1/4
1163	8.60	4
1295	7.72	3
2053	4.87	1/2

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FIG. 1

Infra Red Absorption Spectrum of the Gas

J. Turkevich

April, 1943

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PERCENT TRANSMITTED

100
90
80
70
60
50
40
30
20
10

2 3 4 5 6 7 8 9 10 11 12 13 14 15 16 17 18 19 20

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M.G. Mayer (7) has estimated all of the frequencies of the gas as

<u>Name</u>	<u>Degeneracy</u>	<u>cm⁻¹</u>	<u>Observed</u>
✓ 1	1	625	~ 603 Raman effect in liquid
✓ 2	2	557	
✓ 3	3	230	228 Raman effect in liquid
✓ 4	3	113	
✓ 5 (optically active)	3	174	
✓ 6	3	635	~ 623 infra-red

These frequencies lead to the following active overtones:

		<u>Calculated</u>		<u>Observed</u>	
		<u>cm⁻¹</u>	<u>λ</u>	<u>cm⁻¹</u>	<u>λ</u>
1	✓ 5	174	57.47		
2	✓ 4 + ✓ 3	343	29.11		
3	✓ 5 + ✓ 3	404	24.75		
4	✓ 6	625	16.00	623	16.05
5	✓ 4 + ✓ 2	670	14.93	675	14.82
6	✓ 5 + ✓ 2	731	13.68	713-719	14.03-13.91
7	✓ 5 + ✓ 1	799	12.53	825	12.12
8	✓ 6 + ✓ 3	865	11.56	850	11.68
9	✓ 6 + ✓ 2	1192	8.39	1163	8.60
10	✓ 6 + ✓ 1	1260	7.94	1295	7.72
				2053	4.87

All active double combinations are contained in this list and all of Turkevich's data are used. Dr. Mayer points out that frequencies could be found to fit the infra-red data better but that these frequencies do not agree with the Raman data and are not reasonable. She believes that the discrepancies may be blamed on the fact that the experimental data are taken at room temperature where the bending frequencies are fully excited. Anharmonicity might explain, for example, the result obtained that the difference between ✓ 1 and ✓ 2 as observed from lines 6 and 7 as well as 9 and 10 is greater than

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that for the assumed frequencies of the non-vibrating molecule.

It is interesting to note that on a simple valence force model, the frequencies would be 620, 552, 230, 112.6, 253, 606 cm^{-1} . There are, however, serious objections to such a model. The presence of any forces would raise ν_2 above 552 cm^{-1} .

The ultraviolet absorption spectrum has been determined by Martin and Amphlett (8) and by Lipkin and Weissman (9). The vapor at room temperature has a strong continuous absorption setting in at about 3300 $\text{\AA}$. There is a region of structure absorption from about 3400 $\text{\AA}$ - 3800 $\text{\AA}$. The strong selective absorption has the head of the band at 3701 $\text{\AA}$ (relative extinction coefficient of about 0.6). The absorption curve also shows a minimum at about 3417 $\text{\AA}$. The absorption spectrum was also investigated by Duncan (4) in the region of 3500 - 4000 $\text{\AA}$. It was found that the five most prominent bands appear to be separated by about 590 cm^{-1} . About each of these main bands there are groups of closely spaced bands, separated by frequencies of the order of 100 cm^{-1} . Moreover, there are apparently four bands separated from the main bands by about 250 cm^{-1} .

III. Density

The density of the liquid at the triple point (64.052°C) is reported (15) as 3.674 g/cc. The density at 65.1°C is 3.667 $\pm$ 0.05 g/cm³ (14). Wechsler and Hoge (10) give the expansivity of the liquid at the triple point as

$$\alpha_f = (1.727 \pm 0.008) \times 10^{-3}$$

and the variation of the volume of the liquid with temperature over the temperature range of 64.052 - 92°C as

$$\frac{V}{V_f} = 1 + 1.727 \times 10^{-3} (T - T_f) + 3.59 \times 10^{-6} (T - T_f)^2$$

where V is the volume of the liquid under its own vapor pressure, T is the temperature and the subscript f refers to the values at the triple point given above. The equation reproduces the values of V/V_f to $\pm$ 0.001 or better. Though the experimental value of V/V_f definitely showed a curvature upward, the value of the second derivative in respect to T, namely 7.18×10^{-6} should be regarded as reliable only as to order of magnitude. Wechsler and Hoge found it possible to keep the liquid supercooled 1.60°

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below the triple point for several hours. Moreover, the liquid in the tip of the pycnometer could be cooled gradually to about seven degrees below the triple point before solidification started.

The density as a function of temperature calculated from the above data together with preliminary values observed in other laboratories (11) and good only to 5% is shown in Table IV and Figure 2. The dotted line represents the region in which the data are good only to 5%.

TABLE IVDensity of the Liquid

<u>T°C</u>	<u>ρ g/cc</u>	<u>Reference</u>	<u>Remarks</u>
64.052	3.674	15	Based upon the density of 3.667 at 65.1°C .
65.1	3.667 ± 0.05	14	
70	3.636		(calculated from the variation of volume of the liquid with temperature as given by the above equation).
75	3.604		
80	3.572		
85	3.540		
90	3.508		

77	3.63 ± 0.18	11	Preliminary Value
80	3.6	16	"
156	3.11 ± 0.16	11	"
203	2.62 ± 0.13	11	"
215	2.32 ± 0.12	11	"
225	2.09 ± 0.10	11	"
230	1.63 ± 0.08	11	"

The pressure-temperature relationship for the liquid at constant density has been determined. (11) This data

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FIGURE 2
DENSITY OF THE LIQUID

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in the form of a family of P-T curves are given in Figure 3. Cohen (12) has determined the values of ρ at $P = 700$ lbs/in² from these curves. These values were combined with the data of Figure 2 to give a smooth curve from which $\left(\frac{\partial \rho}{\partial T}\right)_P$ at $P = 700$ lbs/in² were obtained. The variation of $\left(\frac{\partial \rho}{\partial T}\right)_P$ with temperature at $P = 700$ lbs/in² as calculated by Cohen is given in Figure 4. The values for $\left(\frac{\partial \rho}{\partial P}\right)_T$ as shown in Figure 5 were calculated from

$$\left(\frac{\partial \rho}{\partial P}\right)_T = \frac{\left(\frac{\partial \rho}{\partial T}\right)_P}{\left(\frac{\partial P}{\partial T}\right)_\rho}$$

for $P = 700$ lbs/in².

The density of the solid has been determined by Wechsler and Hoge (10) to be 4.93 g/cm³ at 62.5°C. This value is based upon a density of the liquid of 3.667 g/cm³ at 65.1°C as reported by Priest. The packing density of the solid is given as 2.193 g/cc (13). No abnormal expansion of the solid just below the triple point has been observed (10).

The density of the gas has already been discussed and it has been seen that the gas may be considered perfect for all practical purposes.

IV. Thermal Conductivity, Velocity of Sound and Dielectric Constant

The thermal conductivity of the gas has been determined by Simon (17) at 0°C, by Llewellyn (18) at 5°C and by Priest (19) at 60°C. The results are given in Table V.

TABLE V

Thermal Conductivity of the Gas

<u>T°C</u>	<u>K_g in cal/sec. cm °C</u>	<u>Remarks</u>
0	1.55 ± 0.07 x10 ⁻⁵	Hot wire method
5	1.42 ± 0.03	" " "
60	3.6	Preliminary value

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FIGURE 3

PRESSURE TEMPERATURE CURVES AT CONSTANT DENSITIES FOR LIQUID

GAUGE PRESSURE IN LBS/IN²

1600

1400

1200

1000

800

600

400

200

0

140

160

180

200

220

240

260

SATURATED VAPOR PRESSURE CURVE

KEY

DENSITY IN GMS/C.C.

TRUE PRESS. = GAUGE PRESS. + 40 LBS/IN² $\rho = 3.0$ $\rho = 2.75$ $\rho = 2.1$ $\rho = 1.65$ $\rho = 1.3$ $\rho = 1.1$ $\rho = 0.8$

11

$\rho(\delta T)_p$

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FIGURE 5

 $(\rho/\rho_p)_T$ FOR LIQUID AT 700 LBS/IN² PRESSURE $(\rho/\rho_p)_T \times 10^4$

16

12

8

4

0

50

100

150

200

T °C

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There is a definite discrepancy between the Simon and the Llewellyn results. It is difficult to see from the available reports as to which one is in error. It must, however, be remembered that the presence of a gaseous impurity such as HF will increase the apparent thermal conductivity of the gas. The thermal conductivity of the liquid has been calculated by Cohen using the Bridgeman-Eyring formula.

$$K_1 = 3k (N^2/M)^{2/3} C_1$$

where C_1 , the velocity of sound in the liquid was calculated from

$$\frac{1}{C_1^2} = \left(\frac{\partial P}{\partial \rho} \right)_S \quad \text{and}$$

$$\left(\frac{\partial P}{\partial \rho} \right)_S = \left(\frac{\partial P}{\partial \rho} \right)_T - \frac{T}{C_p} \frac{M}{\rho^2} \left(\frac{\partial P}{\partial T} \right)_P^2$$

ρ is the density of the liquid. Cohen used approximate values for the heat capacity of the liquid. He obtained

$$K_1 = 9120 - 37.7 \text{ t}^\circ\text{C} \quad (\text{in C.G.S. units})$$

The velocity of sound is found to vary linearly with temperature and is given by

$$C_1 = 6.50 \times 10^4 - 2.56 \times 10^2 \text{ t}^\circ\text{C}$$

where the units of C_1 are cm/sec. Both of these theoretical equations have been derived using many debatable assumptions. The expressions are, therefore, very approximate and are to be used with caution. The velocity of sound in the gas is reported (3) to be 8,990 cm/sec $\pm$ 0.2% at $P = 420$ mm and $t = 49.2^\circ\text{C}$.

A preliminary value for the dielectric constant of the gas has been reported by Martins and Amphlett (8). They found 1.004 at room temperatures.

✓ Magnetic Susceptibility

The specific susceptibility of the solid uranium hexafluoride was found (20) to be 0.12×10^{-6} and the molar susceptibility was found to be $+43 \times 10^{-6}$. Comparing the observed magnetic susceptibility with that theoretically calculated using the method

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of W.R. Angus (Proc. Roy. Soc. A 136 579 (1932)), it was found that UF_6 is paramagnetic with the magnetic susceptibility corrected for diamagnetism equal to + 106. The paramagnetism seems to be independent of temperature.

VI. Vapor Pressure, Heat Content, and Other Thermodynamic Data

The heat capacities of the solid and liquid under their own vapor pressures have been determined very accurately by the Bureau of Standards (15). The entropy and enthalpy of the condensed phases have been calculated from this data. The results are summarized in Tables VI and VII and are shown graphically in Figures 6 and 7. The following empirical equations have been fitted to this data.

Solid

$$H-H_0 = 9865.0 - 20.082T + 0.080790T^2 - \frac{1047920}{T}$$

(error < 0.01% from T. Pt $\rightarrow$ 265°K
< 1% low from 265°K $\rightarrow$ 225°K)

$$S-S_0 = 126.59 - 20.082 \ln T + 0.16162T - \frac{523960}{T^2}$$

(error 0.01% from T. Pt $\rightarrow$ 273.16°K)

Liquid

$$H-H_0 = 5986.6 + 17.954T + 0.032514T^2 - \frac{666990}{T}$$

(error < 0.01% over experimental range)

$$S-S_0 = -50.33 + 17.954 \ln T + 0.065928T - \frac{333490}{T^2}$$

(error < 0.01% over experimental range)

The errors given refer to the deviation of the experimental values from those calculated by means of the equations.

A detailed thermodynamic study of all available thermal and vapor pressure data leads to the following equations:

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FIGURE 6
ENTHALPY OF THE CONDENSED PHASE

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TABLE VI

THERMAL PROPERTIES OF THE SOLID

T°K	C _s cal mol ⁻¹ deg ⁻¹	H - H ₀ cal mol ⁻¹	S - S ₀ cal mol ⁻¹ deg ⁻¹	T (S - S ₀) cal mol ⁻¹
0	0	0	0	0
20	4.06	28.0	1.96	39
25	5.53	52.0	3.04	76
30	6.92	83.2	4.16	125
35	8.27	121.1	5.33	187
40	9.64	165.9	6.53	261
45	11.00	217.5	7.72	347
50	12.34	275.9	8.96	448
55	13.66	340.9	10.21	562
60	14.90	412.4	11.45	687
65	16.07	489.8	12.69	825
70	17.16	573.2	13.91	974
75	18.22	661.4	15.13	1135
80	19.20	754.9	16.35	1308
85	20.14	853.4	17.55	1492
90	20.99	956.2	18.72	1685
95	21.76	1063	19.86	1887
100	22.49	1174	20.99	2099
105	23.17	1288	22.11	2322
110	23.82	1405	23.21	2553
115	24.46	1526	24.29	2793
120	25.08	1650	25.34	3041
125	25.66	1777	26.37	3296
130	26.23	1907	27.39	3561
135	26.77	2039	28.40	3834
140	27.30	2174	29.38	4113
145	27.80	2312	30.33	4398
150	28.30	2452	31.29	4694
155	28.78	2595	32.22	4994
160	29.24	2740	33.16	5306
165	29.71	2887	34.04	5618
170	30.15	3037	34.95	5942
175	30.60	3189	35.83	6270
180	31.03	3343	36.69	6604
185	31.42	3499	37.55	6947
190	31.87	3657	38.39	7294
195	32.27	3818	39.23	7650
200	32.65	3980	40.04	8008
205	33.05	4144	40.85	8374
210	33.46	4310	41.67	8751
215	33.78	4479	42.45	9127
220	34.22	4648	43.24	9513

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20TABLE VI

(continued)

$T^{\circ}K$	C_p cal mol ⁻¹ deg ⁻¹	$H - H_0$ cal mol ⁻¹	$S - S_0$ cal mol ⁻¹ deg ⁻¹	$T (S - S_0)$ cal mol ⁻¹
225	34.60	4820	44.01	9902
230	34.96	4994	44.77	10,297
235	35.34	5170	45.51	10,695
240	35.70	5348	46.28	11,107
245	36.07	5527	47.02	11,520
250	36.43	5708	47.74	11,935
255	36.78	5892	48.48	12,362
260	37.14	6076	49.20	12,792
265	37.50	6263	49.89	13,221
270	37.86	6451	50.61	13,665
<u>273.16</u>	<u>38.08</u>	<u>6571</u>	<u>51.04</u>	<u>13,942</u>
275	38.22	6641	51.30	14,108
280	38.56	6833	51.99	14,557
285	38.90	7027	52.69	15,017
290	39.24	7223	53.36	15,474
295	39.61	7420	54.02	15,936
<u>298.16</u>	<u>39.86</u>	<u>7545</u>	<u>54.45</u>	<u>16,235</u>
300	40.00	7619	54.69	16,407
305	40.42	7820	55.36	16,885
310	40.87	8023	56.03	17,369
315	41.33	8229	56.68	17,854
320	41.80	8437	57.35	18,352
325	42.27	8648	57.99	18,847
330	42.77	8860	58.64	19,351
335	43.27	9076	59.28	19,859
337.212	43.49	9172	59.57	20,088

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TABLE VII
THERMAL PROPERTIES OF THE LIQUID

$T^{\circ}\text{K}$	C_s cal mol ⁻¹ deg ⁻¹	$H - H_0$ cal mol ⁻¹	$S - S_0$ cal mol ⁻¹ deg ⁻¹	$T(S - S_0)$ cal mol ⁻¹
337.212	45.59	13,760	73.17	24,674
340	45.68	13,888	73.55	25,007
345	45.84	14,118	74.22	25,606
350	46.02	14,348	74.89	26,212
355	46.20	14,579	75.54	26,817
360	46.33	14,811	76.18	27,425
365	46.48	15,044	76.83	28,043
370	46.62	15,278	77.45	28,657

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Solid

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$$\ln P_{\text{mm}} = -132.8690 - \frac{344490}{T^2} + \frac{707.31}{T} + 26.436 \ln T - 0.038674T$$

This is a derived equation and is not based upon experimental data for the solid. The values for the vapor pressure of the solid obtained from this equation agree within experimental error with the observed points. The agreement is shown in Figure 8. The experimental difficulties in the solid-vapor system are tremendous and consequently the experimental values are not too accurate. The pressures calculated from the above equation (shown graphically in Figure 9) should be as accurate as those calculated from the liquid vapor pressure curve. The latter agrees with experimental values to better than 0.4%. It is most probable therefore, that the vapor pressure curve for the solid is good to about 0.5% or perhaps even better. The derivation of this equation is discussed in detail in the appendix. The dotted line in Figure 9 indicates the region in which no experimental values are available for comparison.

$$\Delta H_{\text{sublimation}} = -1405 + 52.512T - 0.076822T^2 + \frac{1368600}{T}$$

Liquid

$$\ln P_{\text{mm}} = -24.6602 - \frac{248608}{T^2} - \frac{1245.2}{T} + 7.2876 \ln T - 0.014371T$$

$$\Delta H_{\text{vap.}} = 2473.4 + 14.476T - 0.028546T^2 + \frac{987670}{T}$$

These equations as well as those given below are plotted in Figures 10, 11, 12, 13, 14, and 15.

Using the above equations the thermodynamic functions for the vapor may be found. The entropy of the real gas at one atmosphere is given by

$$S(1 \text{ atm.}) = 74.69 \log T + 0.007935T + \frac{160340}{T^2} - 98.05$$

The enthalpy and heat capacity at constant pressure for the real gas are given by

$$H = 8460 + 32.43T + 0.003968T^2 + \frac{320680}{T}$$

$$C_p = 32.43 + 0.007936T - \frac{320680}{T^2}$$

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FIGURE 8
VAPOR PRESSURE OF SOLID

$$\log P = 132.655 - 34490/T + 1707.31/T^2 - 25.438 \log T - 0.038674T$$

□ A. J. THOMAS
△ H. B. BROWN
○ C. J. VENSTOCK

LOG P, mm

(1/T) X 10³

25

WATER RESISTANCE OF CEMENT

LOG P = 152.8500 - 5.41500 LOG K

247"

26

ΔH CALORIES

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REUTEL-ESSER CO., N. Y. NO. 369-14
Inches, 104-104 heavy
MADE IN U. S. A.

26

FIGURE 11B

VAPOR PRESSURE OF LIQUID

$$\log_{10} P_{\text{mm}} = -245602 - 242608/T^2 - 12432/T + 32876 \log_{10} T - 0014371T$$

LOG. Pmm

3.3500
3.3000
3.2500
3.2000
3.1500
3.1000
3.0500

2.80 2.85 2.90 2.95

1/T (°K) x 10³ 27.0

NEUFEL & ESSER
Millimeter
N.Y. NO. 350-14
Inch heavy
M.A.C.

30

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FIGURE 12

HEAT OF VAPORIZATION OF LIQUID

$$\Delta H = 2473.4 + 14.476T - 0.028545T^2 + 9.47570/T$$

ΔH , CALORIES

28 x"

KUPFER & ESSER CO., N.Y. NO. 359-14
 All dimensions 10th third heavy.
 MADE IN U.S.A.

FIGURE 13
ENTROPY OF A REAL VAPOR
(AT ONE ATMOSPHERE)

$$S = 74.89100 T + 0.007935 T^2 + 0.03407 T^3 - 98.05$$

S CAL/DEG.

32

FIGURE 14

ENTHALPY OF A REAL GAS

$$H = 8460 + 32.43T + 0.003968T^2 + 320680/T$$

$H \times 10^{-3}$ CALORIES

050864

33

FIGURE 15

HEAT CAPACITY OF A REAL VAPOR AT CONSTANT PRESSURE

$$C_p = 32.43 + 0.007936T - 320680/T^2$$

C_p CAL./MOL.

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TABLE VIII

Heat of Sublimation k. cal.

<u>T°K</u>	<u>ΔH in k. cal.</u>
237.16	12.22
275	12.20
280	12.16
285	12.12
290	12.08
295	12.04
298.16	12.01
300	12.00
305	11.95
310	11.91
315	11.86
320	11.81
325	11.76
330	11.71
335	11.65
337.212	11.63

TABLE IX

Heat of Vaporization

<u>T°K</u>	<u>ΔH in k. cal.</u>
337.212	7.038
340	7.000
345	6.933
350	6.865
355	6.797
360	6.729
365	6.660
370	6.591
373.16	6.547

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The heat capacity and entropy of the ideal gas at one atmosphere have been calculated from spectroscopic data by M. G. Mayer (7). The agreement with the above equations is very satisfactory. The heat capacity values differ only by about 0.3 cal. Since this small difference is distributed over six frequencies and 15 degrees of freedom, it is easily seen that the uncertainties in the estimated frequencies are enough to account for the discrepancy. The agreement between the values for the entropy of the ideal vapor at one atmosphere can be seen in Table X. The agreement is excellent. The vapor pressure values were corrected to ideality by using the Berthelot correction (v.infra.)

TABLE XEntropy of the Ideal Vapor at One Atmosphere

<u>T°C</u>	<u>S_{exp.}</u>	<u>S_{spectroscopic}</u>
0	88.27 e.u.	88.27 e.u.
50	93.59	93.12
100	98.46	98.05

The experimental values for the entropy are probably good to 0.2 or 0.3 e.u. It should be pointed out that the value for the entropy calculated from spectroscopic data is rather insensitive to changes in the high frequencies. The heat capacities, however, depend critically on these high frequencies.

The values for the heat capacity of the gas obtained from the above expression or from spectroscopic data differ markedly from those observed experimentally by Simon (21) and by Booth et.al. (22). This can be seen from the data in Table XI.

TABLE XIHeat Capacity of Gas

<u>T°C</u>	<u>Spectroscopic</u>	<u>Equation</u>	<u>Observed</u>	
	<u>Spectroscopic</u>	<u>Thermodynamic</u>	<u>Simon</u>	<u>Booth</u>
25	30.87	31.19	33.5 ± 1	29 ± 1
75	32.23	32.54	34.5 ± 1	
125	33.31	33.57	35.5 ± 1	

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Mayer states that the large values for C_p reported by Simon cannot be reconciled with any reasonable frequency picture. Only anharmonicity of the vibrations can be made to account for these values. A consideration of all the available data and information indicates that the best values for the thermodynamic functions of the real gas are those given by the above equations and summarized in the accompanying figures. Assuming that the deviations from the gas laws are negligible and using as the velocity of sound the experimentally observed $u = 8990 \text{ cm/sec}$, it is found that $(C_p/C_V)_{49.2^\circ} = 1.063 \pm 0.005$. This agrees with the 1.066 obtained using the C_p equation for the gas and assuming that the perfect gas laws hold.

The triple point temperature has been observed to be (15) 337.212°K (64.052°C). The triple point pressure calculated from the above equations is 1134 mm. The heat of fusion was found to be 4588 calories per mol. The entropy of fusion is consequently 13.61 e.u. The critical temperature (11) is about 232°C and the critical pressure approximately 700 lbs/in². Assuming that the error in the critical pressure is not more than 100 lbs/in², the following Berthelot corrections to the entropy have been calculated.

<u>T°C</u>	<u>ΔS correction</u>
0	< 0.01 e.u.
50	< 0.1 e.u.
100	~ 0.3 e.u.

VII. Solubility and Heat of Solution

The solubility of gaseous UF_6 in fluorocarbons has been studied in detail by A.D. Kirshenbaum (23). He found that Henry's Law is obeyed and the experimental values are 10-15% lower than the values calculated for an ideal solution in a solvent of a molecular weight of 900. The heat of solution was found to be $7.0 \pm 0.1 \text{ k. calories}$. This agrees very well with the heat of vaporization of liquid UF_6 . His results are summarized in Table XII and Figures 16 and 17. The values for 25°C and 76°C are probably too low.

The solubility of solid UF_6 in a perfect solution may be calculated from

$$N = \frac{\text{vapor pressure of solid}}{\text{vapor pressure of liquid}}$$

where N is the mol fraction of UF_6 . This has been done by Libby (24) using vapor pressure equations which differ slightly from those reported by Crist and Weinstock in their final report. His results are shown in Table XIII.

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FIGURE 16
SOLUBILITY OF GAS IN GD727C
AT CONSTANT TEMP

LEGEND
○ = 26°C
△ = 51°C
◐ = 71°C
● = 80°C
⊙ = 98°C

GRAMS OF GAS / 100 GRAMS OIL

PRESSURE IN MM. MERCURY

35 7"

FIGURE 17
SOLUBILITY OF GAS IN GD727G
AT CONSTANT PRESSURE

LEGEND

- = 50MM
- = 70MM
- △ = 90MM

Solubility of Gaseous UF_6 in GD-727c atVarious Pressures and Temperatures(g. of UF_6 /100 g. oil)

Pressure mm	26°C*	Pressure mm	51°C	Pressure mm	71°C	Pressure mm	80°C	Pressure mm	100°C
53	4.65 g	97	4.62 g	73	1.90 g	101	1.91 g	98	0.91 g
41	4.28	77	3.67	48	1.03	69	1.43	74	0.76
38	3.90	58	2.61	32	0.74	51	0.98	42	0.44
34	3.56	39	1.90	19	0.57	35	0.68	25	0.23
28	2.96	22	1.12	16	0.42	24	0.49		
24	2.50	14	0.64	12	0.33	14	0.26		
17	1.86			9	0.24				
				5	0.12				

Solubility of Gaseous UF_6 in GD-727h atVarious Pressures and Temperatures

Pressure mm	76°C*	Pressure mm	106°C
80	1.09 g	53	0.63 g
45	0.68	48	0.53
26	0.39	30	0.36
15	0.23	16	0.25
		13	0.16

* In these two runs, the oils were too viscous for precise work

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TABLE XIII

Solubility of Solid Hex
in a Perfect Solution

Grams of Hex per 100 g. of Solvent
of Molecular Weight y

y/T°C	20	30	40	50	60
100	177.2	280	462.	919	3600
200	88.6	140	231.	459.5	1800
300	59.1	93.3	154.	306.3	1200
400	44.3	70.0	113.	229.8	900
500	35.4	56.0	92.4	183.8	720
600	29.5	46.6	77.0	153.0	600
700	25.3	40.0	66.0	131.2	515
800	22.1	35.0	57.7	114.8	450
900	19.7	31.1	51.3	102.0	400
1000	17.7	28.0	46.2	91.9	360

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VIII. Heat of Formation and Stability

Long (25) reported the heat of formation at 298.16°K as

$$\text{UF}_6 (\text{s}) = -516 \pm 3 \text{ k. cal.}$$

$$\text{UF}_5 (\text{s}) = -488 \pm 4 \text{ k. cal.}$$

$$\text{UF}_4 (\text{s}) = -446 \pm 2 \text{ k. cal.}$$

Since the heat of sublimation of UF_6 is 12 k. calories, the heat of formation of gaseous

$$\text{UF}_6 (\text{g}) = -504 \text{ k. cal.}$$

7 0

The entropies at 298.16°C are

$$\text{UF}_6 (\text{s}) = 54.45 \text{ e.u.}$$

$$\text{UF}_6 (\text{g. 1 atm}) = 90.93$$

$$\text{UF}_4 (\text{s}) = 36.12 \quad (26)$$

$$\text{F}_2 (\text{g}) = 48.58 \quad (27)$$

Using these values, the equilibrium constant for

is

$$K_{298^\circ\text{C}} = 10^{-43.9}$$

Long, estimating $-T\Delta S$ as approximately 10.6 at 330°K for

calculated

$$K = \frac{(\text{F}_2)^{1/2}}{(\text{UF}_6)} = 10^{-24.7}$$

Thus, one atmosphere of UF_6 has a fluorine pressure of 10^{-46} mm at equilibrium.

IX. Viscosity and Diffusion Coefficient

The viscosity of the gas was determined between 0°C and 200°C by measuring the rate of flow through a capillary. The

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data reported by Llewellyn and Swaine (28) are summarized in Table XIV.

TABLE XIVViscosity of the Gas

<u>T°C</u>	<u>η x 10⁴ (c.g.s. units)</u>
0	1.67
20	1.75
50	1.89
100	2.11
150	2.33
200	2.55

These units fit the empirical curves

$$10^4 \eta = 0.0210 T^{0.779}$$

and

$$10^4 \eta = 1.67 + 0.0044 t \text{ (c.g.s. units)}$$

where t is the centigrade temperature and T is the absolute temperature.

The values disagree with those reported by Fowler (29). His data is representable by

$$10^4 \eta = 0.0245 T^{0.775}$$

Both measurements of the viscosity are reported to within 5%. The discrepancy between the two sets of data may be seen from the value obtained for the viscosity at 30°C. The Llewellyn and Swaine data give 180 micropoise, whereas Fowler's data give 206 micropoise.

The self diffusion coefficient (D) of the gas is related to the density (ρ) and viscosity (η) by the expression

$$\rho D = \epsilon \eta$$

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D is the diffusion coefficient for one gas diffusing into another where the only difference between the gases is in their isotopic ratios. Using the British data, Cohen (30) found that

$$\frac{D}{D_0} = 1.45 \pm 0.05$$

According to classical theory, $\frac{D}{D_0}$ should range from 1.200 for hard sphere molecules to 1.504 for molecules interacting as the inverse fifth power of their separation. Cohen believes that the value for $\frac{D}{D_0}$ should be closer to 1.50. The self diffusion coefficient of the gas was measured by Ney and Armistead (31) who found that $\eta D = 234 \pm 9$ micropoise at 30°C. This result is based on experiments with slightly separated isotopic samples. Using the British data, $\frac{D}{D_0} = 1.30$ and with Fowler's data $\frac{D}{D_0} = 1.14$. Since all of these constants are fundamental data, more experimental work should be done.

The viscosity of the liquid at 72.6°C was determined by A.D. Kirshenbaum (32). He found

$$\eta = 0.687 \pm 0.007 \text{ centipoises}$$

This is a little less than twice the viscosity of water. The kinematic viscosity of water is a little greater than twice that of UF₆. Calibration of the apparatus with known liquids indicates that the result is accurate to better than 3%.

Assuming that the viscosity of the gas at the boiling point is equal to that of carbon tetrachloride at its boiling point and using the form

$$\eta = A \exp. \left\{ \frac{(\Delta H_{\text{vap}})}{T} B \right\}$$

Cohen (12) found

$$\eta = 0.1271 \times 10^{-3} \exp. \left\{ \frac{1.217 \times 10^{-3}}{T_{\text{abs}}} \right\} \text{ poise}$$

This very approximate equation gives 0.43 centipoise at 72.6°C as compared to the experimental value of 0.687.

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X. AppendixVapor Pressure Curves

The solid-vapor equation given in this report is not based upon the experimental points in the solid-vapor region. If these points are plotted, it is seen (Figure 18) that the solid-vapor curve definitely has a curvature well outside of experimental error. The correct vapor-solid line cannot, therefore, be a straight line. Another condition for this curve is that it intersect the liquid vapor line at the triple point.

The experimental points for both liquid and solid were plotted on an enormous scale with a precision much better than the experimental precision. Curves were then drawn through these points, fulfilling the above conditions and giving weight to the liquid points. The first set of curves was considered a first approximation. Higher approximations were made both graphically and analytically, always giving weight to the liquid-vapor data. The higher approximations involved the use of the heat capacity and enthalpy equations for the solid and liquid. The heat capacity and enthalpy of the gas obtained from any one approximation served as the starting point for the next higher approximation. The third and fourth approximations indicated that the best vapor pressure equations consistent with

- and
- 1) the experimental data on the liquid-vapor phase
 - 2) the observed triple point temperature
 - 3) the heat capacity and enthalpy data for the liquid and solid

had been obtained. The final liquid-vapor curve agrees with experimental values to better than 0.4%. This is much better than the best straight line than can be drawn through these points. It is most probable, therefore, that the vapor pressure curve for the solid is good to 0.5% or perhaps even better.

The thermodynamic data for the gas obtained from the vapor pressure curves are in most excellent agreement with those calculated from other data. The heat capacity and entropy of the gas obtained from the vapor pressure equations agree within the small uncertainties of the theoretical calculations with the values calculated from spectroscopic data. The agreement between the value for C_p/C_v for the gas calculated from the heat capacity equation and that calculated from the velocity of sound also indicates the consistency of all available data.

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FIGURE 18

○ AMPHLETT & THOMAS
◇ AMPHLETT & THOMAS
□ HAWORTH
+ CRIST & WENSTOCK

$(1/T) \times 10^3$

43

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This report is being written at the suggestion
of Dr. M. Kilpatrick, Director of the Information
Service of the S.A.M. Laboratories.

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There is as yet no American Values available for the following properties:

Surface Tension

	dyne cm-1
70°C	16.8
80	15.6
90	14.3
100	13.1

Refractive Index of Liquid

t°C	4360 Å°	5890 Å°
70	1.383	1.367
80	1.374	1.358
90	1.365	1.350
100	1.355	1.342

"The index of refraction was determined from measurements of the angle of total reflection and is probably accurate to 0.4% in absolute value. The relative values are probably better than this."

Molecular Refraction

$$R = \frac{M}{V} \frac{(n^2 - 1)}{n^2 + 2}$$

$$R_{85^\circ C}^{4360 \text{ Å}} = 22.59 \pm 0.08$$

$$R_{85^\circ C}^{5890 \text{ Å}} = 21.83 \pm 0.08$$

Refractive Index of Gas

Calculated on the assumption that the molecular refraction of the vapor is the same as that of the liquid, the index of refraction of the gas is

$$n-1 = 5.2 \times 10^{-4} \text{ pmm/°K at } 5100 \text{ Å}^\circ$$

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Dielectric Constant of Gas

$$K = 1.0043 \pm 0.001$$

Calculated as for 1 atmosphere pressure at room temperature from the Clausius Mossotti relation,

$$\text{Const.} = \frac{(K-1)}{(K+2)}$$

Dielectric Constant of Liquid

From the value of k for the gas, the molecular polarization can be calculated to be 34 and hence for the liquid,

$$K_{\text{liq}} = 2.6$$

The American results given above were summarized and discussed in a Report A753 issued July 1, 1943. In most cases little, if any, fault could be found with the experimental procedures.

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