27
1-23-75
25 NTIS

**A REVIEW OF THE THERMODYNAMICS OF THE U-C,
Pu-C, AND U-Pu-C SYSTEMS**

by

**M. Tetenbaum, A. Sheth,
and W. Olson**

MASTER

U of C-AUA-USERDA

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

**Prepared for the U. S. ENERGY RESEARCH
AND DEVELOPMENT ADMINISTRATION**

under Contract W-31-109-Eng-38

DISTRIBUTION OF THIS DOCUMENT UNLIMITED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

The facilities of Argonne National Laboratory are owned by the United States Government. Under the terms of a contract (W-31-109-Eng-38) between the U. S. Energy Research and Development Administration, Argonne Universities Association and The University of Chicago, the University employs the staff and operates the Laboratory in accordance with policies and programs formulated, approved and reviewed by the Association.

MEMBERS OF ARGONNE UNIVERSITIES ASSOCIATION

The University of Arizona	Kansas State University	The Ohio State University
Carnegie-Mellon University	The University of Kansas	Ohio University
Case Western Reserve University	Loyola University	The Pennsylvania State University
The University of Chicago	Marquette University	Purdue University
University of Cincinnati	Michigan State University	Saint Louis University
Illinois Institute of Technology	The University of Michigan	Southern Illinois University
University of Illinois	University of Minnesota	The University of Texas at Austin
Indiana University	University of Missouri	Washington University
Iowa State University	Northwestern University	Wayne State University
The University of Iowa	University of Notre Dame	The University of Wisconsin

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately-owned rights. Mention of commercial products, their manufacturers, or their suppliers in this publication does not imply or connote approval or disapproval of the product by Argonne National Laboratory or the U. S. Energy Research and Development Administration.

Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois 60439

A REVIEW OF THE THERMODYNAMICS OF THE U-C,
Pu-C, AND U-Pu-C SYSTEMS

by

M. Tetenbaum and A. Sheth

Chemical Engineering Division

W. Olson

Los Alamos Scientific Laboratory

NOTICE
This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Energy Research and Development Administration, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

June 1975

THIS PAGE
WAS INTENTIONALLY
LEFT BLANK

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	1
INTRODUCTION	1
URANIUM-CARBON SYSTEM	2
Uranium Monocarbide	2
Uranium Sesquicarbide	4
Vaporization Behavior of U-C System	4
PLUTONIUM-CARBON SYSTEM	4
Plutonium Monocarbide	4
Plutonium Sesquicarbide	12
Vaporization Behavior	14
URANIUM-PLUTONIUM-CARBON SYSTEM	14
Thermal Functions of Mixed Carbides	14
Vaporization Behavior of U-Pu-C System	18
SUMMARY AND RECOMMENDED RESEARCH	25
ACKNOWLEDGMENTS	25
REFERENCES	26

LIST OF FIGURES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Temperature Dependence of Uranium- and Total Carbon-Bearing Species over UC_x Compositions	9
2.	Temperature Dependency of Uranium- and Carbon-Bearing Species over Uranium Monocarbide	10
3.	Calculated Values for the Temperature Dependency of Plutonium- and Carbon-Bearing Species over $PuC_{0.88}$ and Pu_2C_3	15
4.	Calculated Values of Actinide and Carbon Pressures Above the Monocarbide-Sesquicarbide Region of the U-Pu-C System	21
5.	Calculated Values for the Temperature Dependence of Actinide- and Carbon-Bearing Species over Monocarbide Phase $U_{0.8}Pu_{0.2}C$	22
6.	Calculated Values for the Temperature Dependence of Actinide- and Carbon-Bearing Species over Sesquicarbide Phase $(U_{0.8}Pu_{0.2})_2C_3$	23
7.	Comparison of Actinide and Carbon Pressures Above UC, PuC, and $U_{0.8}Pu_{0.2}C$ Systems	24

LIST OF TABLES

<u>No.</u>	<u>Title</u>	<u>Page</u>
1.	Thermodynamic Properties of $UC_{1.0}$	3
2.	Thermodynamic Properties of U_2C_3	5
3.	Uranium and Carbon Activities and Free Energy of Formation at 2100 K	6
4.	Uranium and Carbon Activities and Free Energy of Formation at 2300 K	7
5.	Equations for Temperature Dependency of Uranium- and Carbon- Bearing Species over UC_x Compositions	8
6.	Thermodynamic Properties of $PuC_{0.88}$	11
7.	Thermodynamic Properties of Pu_2C_3	13
8.	Free Energies of Formation of Gaseous and Condensed Phases . . .	14
9.	Equations for Temperature Dependency of Plutonium- and Carbon- Bearing Species over Plutonium Monocarbide and Plutonium Sesquicarbide	16
10.	Calculated Values for Thermodynamic Properties of $(U_{0.8}Pu_{0.2})C$.	17
11.	Calculated Values for Thermodynamic Properties of $(U_{0.8}Pu_{0.2})_2C_3$	19
12.	Equations for Temperature Dependency of Actinide- and Carbon- Bearing Species over Monocarbide and Sesquicarbide Phases in the U-Pu-C System	20
13.	Comparison of Experimental and Calculated $Pu(g)$ Pressure Above $U_{0.8}Pu_{0.2}C$	25

A REVIEW OF THE THERMODYNAMICS OF THE U-C, Pu-C, AND U-Pu-C SYSTEMS

by

M. Tetenbaum, A. Sheth, and M. Olson

ABSTRACT

Thermodynamic properties such as enthalpy, heat capacity, entropy, heat and free energy of formation, and vaporization behavior are presented for the U-C, Pu-C, and U-Pu-C systems. These properties are of interest to scientists and engineers involved in the expanding field of advanced fuel LMFBR systems. The information on these systems has been derived largely from the discussions of the IAEA Panel on the Assessment of Thermodynamic Properties of the U-C, Pu-C, and U-Pu-C Systems, which were held in Grenoble, France, May 6-8, 1974.

INTRODUCTION

The advantages of actinide carbide over actinide oxide nuclear fuels have been reasonably well documented in the literature. Carbide fuels have a higher fissile density and higher thermal conductivity than oxide nuclear fuels. The neutronic properties are such that higher rod power and higher breeding rates are to be expected. On the other hand, the disadvantages of carbide fuels are that they have a lower chemical stability than oxide fuels and are much more difficult to fabricate, particularly if one wants to minimize oxygen and nitrogen contamination.

A knowledge of the thermodynamic properties of actinide carbide systems is important in understanding and predicting the behavior of carbide fuels under various reaction conditions.

The object of this report is to present, in a concise manner, the most recent thermodynamic properties of the U-C, Pu-C, and U-Pu-C systems. The information on these systems has been derived largely from the discussions of the IAEA Panel on the Assessment of Thermodynamic Properties of the U-C, Pu-C, and U-Pu-C Systems, which were held in Grenoble, France, May 6-8, 1974. The assessments of particular importance to this report are those made by the following Panel members: E. K. Storms (LASL) and R. J. Ackermann (ANL) on the U-C system, M. Rand (AERE, UK) on the Pu-C system, and P. Potter (AERE, UK) on the U-Pu-C system. The chairman of the Panel was F. L. Oetting (IAEA, Vienna). All values presented by the Panel are to be considered preliminary at this time. The thermodynamic analyses upon which these values are based will be presented in a forthcoming IAEA publication.

The thermodynamic information assembled in this review should be of interest to scientists and engineers involved in the expanding field of advanced fuel LMFBR systems.

URANIUM-CARBON SYSTEM

Uranium Monocarbide

The IAEA Panel¹ values for the thermal functions of uranium monocarbide are given in Table 1. These values are based on low-temperature heat-capacity measurements of Westrum *et al.*^{2,3} and Farr *et al.*⁴ and high-temperature drop-calorimetric measurements of Harrington and Rowe⁵ (273-1477 K), Oetting, Navratil, and Storms⁶ (298-1672 K), and Levinson^{7,8} (1287-2481 K). The following equations were derived:

Enthalpy

$$H_T^\circ - H_{298}^\circ = -5.1/41 \times 10^3 + 14.727 T - 2.6733 \times 10^{-4} T^2 \\ + 3.3304 \times 10^{-7} T^3 + \frac{2.3797 \times 10^5}{T} \quad (298-2418 \text{ K})$$

Heat Capacity

$$C_p^\circ = 14.727 - 5.3466 \times 10^{-4} T + 9.9912 \times 10^{-7} T^2 - \frac{2.3797 \times 10^5}{T^2}$$

Entropy

$$S_T^\circ = -70.976 + 14.727 \ln T - 5.3466 \times 10^{-4} T + 4.9956 \times 10^{-7} T^2 + \frac{1.1899 \times 10^5}{T^2}$$

Free energy of formation values were calculated from the free energy functions of uranium monocarbide, those of uranium^{9,10} and graphite,¹¹ and the heat of formation of uranium monocarbide at 298 K by means of the expression:

$$\Delta G_{f,T}^\circ = \Delta H_{f,298}^\circ + T \Delta \left(\frac{G_T^\circ - H_{298}^\circ}{T} \right)$$

where

$$\frac{G_T^\circ - H_{298}^\circ}{T} = -S_T^\circ + \frac{H_T^\circ - H_{298}^\circ}{T}$$

The IAEA Panel value for the heat of formation of UC is $\Delta H_{f,298}^\circ = -23.40$ kcal/mol, based on combustion measurements of Storms and Huber⁹ and Huber and Holley.¹² Values for the heat of formation of UC as a function of temperature can be calculated from the expression

$$\Delta H_{f,T}^\circ = \Delta H_{f,298}^\circ + \Delta (H_T^\circ - H_{298}^\circ)$$

TABLE 1. Thermodynamic Properties of UC_{1.0}

T, K	$(H_T^\circ - H_{298}^\circ)$ cal/mol	C_p° cal/mol, K	S_T° cal/mol, K	$-(G_T^\circ - H_{298}^\circ)/T$ cal/mol, K	$-\Delta G_f^\circ$ kcal/mol	$-\Delta H_f^\circ$ kcal/mol
298	0	11.98	14.15	14.15	23.63	23.40
300	22	12.01	14.22	14.15	23.63	23.39
400	1290	13.19	17.86	14.64	23.77	23.03
500	2640	13.76	20.87	15.59	23.99	22.72
600	4034	14.10	23.41	16.69	24.27	22.51
700	5458	14.36	25.60	17.81	24.57	22.41
800	6904	14.57	27.54	18.91	24.88	22.40
900	8371	14.76	29.26	19.97	25.19	22.45
1000	9857	14.95	30.83	20.98	25.45	23.22
1100	11362	15.15	32.26	21.94	25.61	24.35
1200	12887	15.36	33.59	22.85	25.73	24.38
1300	14434	15.58	34.83	23.73	25.84	24.40
1400	16004	15.82	35.99	24.56	25.95	24.41
1500	17598	16.07	37.09	25.36	25.95	26.00
1600	19218	16.34	38.14	26.13	25.94	26.11
1700	20865	16.62	39.13	26.86	25.93	26.21
1800	22543	16.93	40.09	27.57	25.91	26.28
1900	24252	17.25	41.02	28.26	25.88	26.33
2000	25994	17.59	41.91	28.92	25.86	26.35
2100	27771	17.96	42.78	29.56	25.84	26.34
2200	29586	18.34	43.62	30.18	25.81	26.30
2300	31439	18.74	44.45	30.78	25.79	26.22
2400	33334	19.16	45.25	31.37	25.78	26.10
2500 ^a	35272	19.60	46.04	31.94	25.77	25.94
2600 ^a	37254	20.06	46.82	32.50	25.76	25.75
2700 ^a	39283	20.53	47.59	33.04	25.77	25.51
2800 ^a	41362	21.03	48.34	33.57	25.79	25.22
2900 ^a	43491	21.55	49.09	34.10	25.81	24.89
3000 ^a	45672	22.09	49.83	34.61	25.85	24.50

^a Extrapolated

Uranium Sesquicarbide

The IAEA Panel values for the thermal functions of uranium sesquicarbide are given in Table 2. These values are based on low temperature heat capacity measurements of Farr *et al.*¹³ and Andon *et al.*¹⁴ and the high temperature drop-calorimetric measurements of Oetting, Navratil, and Storms.⁶ The following equations were derived:

Enthalpy

$$H_T^\circ - H_{298}^\circ = -12642 + 36.003 T - 5.7208 \times 10^{-3} T^2 + 3.295 \times 10^{-6} T^3 \\ + \frac{6.9432 \times 10^5}{T} \quad (298-1667 \text{ K})$$

Heat Capacity

$$C_p^\circ = 36.003 - 1.1442 \times 10^{-2} T + 9.885 \times 10^{-6} T^2 - \frac{6.9432 \times 10^5}{T^2}$$

Entropy

$$S_T^\circ = -173.12 + 36.003 \ln T - 1.1442 \times 10^{-2} T + 4.9425 \times 10^{-6} T^2 + \frac{3.4716 \times 10^5}{T^2}$$

Based on combustion measurements of Huber, Holley, and Witteman,¹⁵ the IAEA Panel value for the heat of formation of U_2C_3 is $\Delta H_{f,298}^\circ = -43.30$ kcal/mol. Calculated values for the free energies and heat of formation of U_2C_3 at higher temperatures are given in Table 2.

Vaporization Behavior of U-C System

Equations have been derived for the temperature dependence of monatomic uranium species, $UC_2(g)$, $C_1(g)$, $C_2(g)$, and $C_3(g)$ carbon species, and total pressure $\Sigma[U(g) + UC_2(g) + C_1(g) + C_2(g) + C_3(g)]$ over UC_x compositions in the single phase region of the U-C system. These equations are based on carbon and uranium activity values (Table 3 and 4) from the IAEA Panel assessment of the U-C system. The values of carbon activity, which were derived from a Gibbs-Duhem treatment of Storms^{16,17} adjusted uranium activity values, are in good agreement with measurements of carbon activity by Tetenbaum and Hunt.¹⁸ The results are summarized in Table 5 and Figs. 1 and 2. Over the composition range $C/M = 0.92-1.10$, uranium is the predominant gaseous species, and $C_1(g)$ the predominant carbon species. For compositions having a C/U ratio of approximately 0.92-1.00, $C_1(g) > UC_2(g) > C_2(g) > C_3(g)$. For compositions ranging from $C/U = 1.05$ to 1.10, $C_1(g) > C_3(g) \approx UC_2(g) > C_2(g)$.

PLUTONIUM-CARBON SYSTEM

Plutonium Monocarbide

The IAEA Panel values for the thermal functions of plutonium monocarbide are given in Table 6. These values are based on low-temperature heat-capacity measurements of Sutcliffe¹⁹ and the high-temperature drop-calorimetric

TABLE 2. Thermodynamic Properties of U_2C_3

T, K	$(H_T^\circ - H_{298}^\circ)$ cal/mol	C_p° cal/mol, K	S_T° cal/mol, K	$-(G_T^\circ - H_{298}^\circ)/T$ cal/mol, K	$-\Delta G_f^\circ$ kcal/mol	$-\Delta H_f^\circ$ kcal/mol
298	0	25.66	32.93	32.93	44.74	43.30
300	47	25.75	33.09	32.93	44.75	43.29
400	2791	28.67	40.96	33.99	45.34	42.60
500	5730	29.98	47.52	36.06	46.09	42.06
600	8769	30.77	53.06	38.44	46.93	41.78
700	11879	31.42	57.85	40.88	47.80	41.74
800	15054	32.09	62.09	43.27	48.66	41.88
900	18300	32.86	65.91	45.58	49.50	42.16
1000	21630	33.75	69.42	47.79	50.21	43.85
1100	25056	34.80	72.68	49.90	50.73	46.21
1200	28596	36.03	75.76	51.93	51.14	46.32
1300	32267	37.42	78.70	53.88	51.54	46.33
1400	36087	39.01	81.53	55.75	51.94	46.23
1500	40074	40.77	84.28	57.56	52.14	49.17
1600	44248	42.73	86.97	59.32	52.34	49.05
1700 ^a	48627	44.88	89.63	61.02	52.55	48.74
1800 ^a	53230	47.22	92.26	62.69	52.79	48.23
1900 ^a	58077	49.76	94.88	64.31	53.06	47.49
2000 ^a	63188	52.49	97.50	65.90	53.38	46.51
2100 ^a	68581	55.41	100.13	67.47	53.76	45.25

^a Extrapolated

TABLE 3. Uranium and Carbon Activities and Free Energy of Formation at 2100 K

C/U	Uranium Activity	Carbon Activity	$-\Delta G_f^\circ$ kcal/mol	
1.88	1.49E-03	1.00E-00	27.15	UC ₂
1.86	1.70E-03	9.32E-01	27.14	
1.84	1.91E-03	8.76E-01	27.14	
1.82	2.14E-03	8.23E-01	23.12	
1.80	2.37E-03	7.78E-01	27.10	
1.78	2.59E-03	7.40E-01	27.08	
1.76	2.81E-03	7.07E-01	27.05	
1.74	2.99E-03	6.83E-01	27.02	
1.71	3.30E-03	6.45E-01	26.97	
1.10	3.30E-03	6.45E-01	25.86	UC
1.08	3.96E-03	5.46E-01	25.81	
1.06	5.05E-03	4.34E-01	25.75	
1.04	6.37E-03	3.48E-01	25.67	
1.03	7.69E-03	2.90E-01	25.63	
1.02	9.45E-03	2.37E-01	25.57	
1.01	1.25E-02	1.80E-01	25.51	
1.00	3.19E-02	7.10E-02	25.41	
0.99	7.69E-02	2.93E-02	25.29	
0.98	1.05E-01	2.13E-02	25.13	
0.97	1.32E-01	1.69E-02	24.97	
0.96	1.65E-01	1.42E-02	24.79	
0.94	2.20E-01	9.90E-03	24.42	
0.92	3.08E-01	6.90E-03	24.02	
0.25	3.08E-01	6.90E-03	10.11	liquid

$$p^\circ(\text{uranium}) = 4.55 \times 10^{-7} \text{ atm}$$

TABLE 4. Uranium and Carbon Activities and Free Energy of Formation at 2300 K

C/U	Uranium Activity	Carbon Activity	$-\Delta G_f^\circ$ kcal/mol	
1.90	2.09E-03	1.00E-00	28.21	UC ₂
1.88	2.66E-03	8.80E-01	28.20	
1.86	3.04E-03	8.20E-01	29.19	
1.84	3.42E-03	7.69E-01	28.16	
1.82	3.70E-03	7.36E-01	28.14	
1.80	4.02E-03	7.03E-01	28.11	
1.78	4.33E-03	6.75E-01	28.07	
1.76	4.55E-03	6.56E-01	28.04	
1.74	4.74E-03	6.40E-01	28.00	
1.72	4.97E-03	6.23E-01	27.96	
1.70	5.16E-03	6.10E-01	27.91	
1.65	5.54E-03	5.85E-01	27.79	
1.60	5.84E-03	5.66E-01	27.67	
1.55	6.03E-03	5.54E-01	27.53	
1.50	6.07E-03	5.52E-01	27.40	
1.45	6.09E-03	5.51E-01	27.26	
1.41	6.11E-03	5.50E-01	27.15	
1.14	6.11E-03	5.50E-01	26.42	UC
1.12	7.12E-03	4.80E-01	26.35	
1.10	8.35E-03	4.16E-01	26.28	
1.08	9.68E-03	3.63E-01	26.19	
1.06	1.18E-02	3.03E-01	26.09	
1.05	1.31E-02	2.73E-01	26.04	
1.04	1.47E-02	2.45E-01	25.98	
1.03	1.65E-02	2.19E-01	25.91	
1.02	1.94E-02	1.87E-01	25.84	
1.01	2.31E-02	1.57E-01	25.75	
1.00	3.98E-02	9.15E-02	25.66	
0.99	7.40E-02	4.91E-02	25.55	
0.98	9.49E-02	3.82E-02	25.39	
0.97	1.18E-01	3.06E-02	25.24	
0.96	1.39E-01	2.58E-02	25.07	
0.95	1.59E-01	2.23E-02	24.90	
0.94	1.82E-01	1.94E-02	24.73	
0.92	2.28E-01	1.52E-02	24.35	
0.90	2.85E-01	1.19E-02	23.96	
0.38	2.85E-01	1.19E-02	13.43	liquid

$$p^\circ(\text{uranium}) = 5.27 \times 10^{-6} \text{ atm}$$

TABLE 5. Equations for Temperature Dependency of Uranium- and Carbon-Bearing Species over UC_x Compositions

$$\log p \text{ (atm)} = - \frac{A \times 10^4}{T} + B \quad (2000-2500 \text{ K})$$

C/U	U(g)		UC ₂ (g)		C ₁ (g)		C ₂ (g)		C ₃ (g)		C _T (g)		p _T (g)	
	A	B	A	B	A	B	A	B	A	B	A	B	A	B
0.92	2.25	3.88	4.70	10.02	4.56	9.97	5.99	13.54	6.57	15.18	4.56	9.97	2.25	3.88
0.94	2.37	4.29	4.53	9.38	4.42	9.46	5.67	12.40	6.18	13.84	4.42	9.45	2.37	4.29
0.96	2.45	4.51	4.48	9.30	4.35	9.32	5.53	12.05	6.06	13.75	4.55	9.30	2.45	4.51
0.98	2.46	4.41	4.47	9.41	4.35	9.44	5.58	12.56	5.91	13.50	4.33	9.35	2.46	4.41
0.99	2.53	4.58	4.39	9.20	4.27	9.26	5.39	11.99	5.68	12.92	4.31	9.32	2.53	4.58
1.00	2.80	5.50	4.12	8.29	4.01	8.36	4.84	10.13	4.87	10.15	4.02	8.43	2.80	5.52
1.01	3.21	7.06	3.72	6.78	3.60	6.79	4.04	7.09	3.66	5.59	3.61	6.85	3.21	7.08
1.02	3.32	7.45	3.10	4.18	3.45	6.24	3.79	6.17	3.33	4.37	3.38	5.91	3.34	7.56
1.04	3.45	7.87	3.48	5.97	3.38	6.09	3.55	5.41	2.94	3.14	3.34	5.98	3.45	7.87
1.06	3.46	7.84	3.48	6.02	3.36	6.04	3.50	5.34	2.92	3.24	3.30	5.85	3.47	7.90
1.08	3.51	7.94	3.85	7.89	3.31	5.93	3.45	5.33	2.79	2.99	3.22	5.65	3.46	7.78
1.10	3.54	8.05	3.41	5.84	3.28	5.82	3.40	5.16	2.70	2.69	3.16	5.41	3.50	7.91

Fig. 1. Temperature Dependence of Uranium- and Total Carbon-Bearing Species over UC_x Compositions

Fig. 2. Temperature Dependency of Uranium- and Carbon-Bearing Species over Uranium Monocarbide

TABLE 6. Thermodynamic Properties of $\text{PuC}_{0.88}$

T, K	$(H_T^\circ - H_{298}^\circ)$ cal/mol	C_p° cal/mol, K	S_T° cal/mol, K	$-(G_T^\circ - H_{298}^\circ)/T$ cal/mol, K	$-\Delta G_f^\circ$ kcal/mol	$-\Delta H_f^\circ$
298	0	11.40	17.38	17.38	11.40	11.40
300	21	11.42	17.45	17.38	12.45	11.40
400	1192	11.86	20.82	17.83	12.79	12.09
500	2382	11.93	23.47	18.71	12.98	12.14
600	3578	12.02	25.65	19.69	13.15	12.33
700	4790	12.26	27.51	20.67	13.27	12.38
800	6039	12.74	29.18	21.63	13.36	12.81
900	7349	13.48	30.74	22.56	13.43	13.48
1000	8746	14.51	32.19	23.45	13.43	13.48
1100	10259	15.80	33.63	24.31	13.43	13.46
1200	11915	17.38	35.09	25.14	13.43	13.27
1300	13744	19.24	36.54	25.96	13.48	12.93
1400	15775	21.42	38.05	26.77	13.53	12.40
1500	18036	23.85	39.60	27.58	13.65	11.66
1600 ^a	20557	26.60	41.23	28.37	13.79	10.66
1700 ^a	23367	29.66	42.93	29.18	14.05	9.37
1800 ^a	26496	32.98	44.72	30.00	14.36	7.77

^aExtrapolated

measurements of Kruger and Savage²⁰ and Oetting.²¹ The thermodynamic equations derived are as follows:

Enthalpy

$$H_T^\circ - H_{298}^\circ = -5728.5 + 18.607 T - 9.3415 \times 10^{-3} T^2 + 4.9473 \times 10^{-6} T^3 + \frac{2.6291 \times 10^5}{T}$$

Heat Capacity

$$C_p^\circ = 18.607 - 1.0683 \times 10^{-2} T + 1.4842 \times 10^{-5} T^2 - \frac{2.6291 \times 10^5}{T^2}$$

Entropy

$$S_T^\circ = -85.078 + 18.607 \ln T - 1.8683 \times 10^{-2} T + 7.421 \times 10^{-6} T^2 + \frac{1.3146 \times 10^5}{T^2}$$

The value for the heat of formation of $\text{PuC}_{0.88}$ is $\Delta H_{f,298}^\circ = -11.4$ kcal/mol. This value, chosen by the IAEA Panel on the basis of combustion measurements of Johnson *et al.*,²² is consistent with the emf results of Campbell, Kent, and Leary,²³ and Marcon's²⁴ vaporization data if the carbon rich phase boundary is $\text{PuC}_{0.88}$ at or near the peritectic temperature of 1873 K. This value, however, does not take into consideration three vaporization measurements of the $\text{PuC-Pu}_2\text{C}_3$ diphasic system that are in very good agreement, namely, those of Campbell, Kent, and Leary²³ (mass spectrometric), Harris *et al.*²⁵ (transpiration), and Olson and Mulford²⁶ (mass spectrometric and mass effusion).

Calculated Values for the heat of formation and free energy of formation are listed in Table 6.

Plutonium Sesquicarbide

The IAEA Panel values for the thermal functions of plutonium sesquicarbide are given in Table 7. The values at 298 K are based on low-temperature heat-capacity measurements of Danan.²⁷ The thermal functions at higher temperatures are based on drop-calorimetric measurements of Oetting²¹ between 395-1495 K. These data were used to obtain the following equations:

Enthalpy

$$H_T^\circ - H_{298}^\circ = -10639.0 + 31.722 T - 2.3928 \times 10^{-3} T^2 + 2.7934 \times 10^{-6} T^3 + \frac{3.9475 \times 10^5}{T}$$

Heat Capacity

$$C_p^\circ = 31.722 - 4.7856 \times 10^{-3} T + 8.3803 \times 10^{-6} T^2 - \frac{3.9475 \times 10^5}{T^2}$$

TABLE 7. Thermodynamic Properties of Pu_2C_3

T, K	$(\text{H}_T^\circ - \text{H}_{298}^\circ)$ cal/mol	C_p° cal/mol, K	S_T° cal/mol, K	$-(\text{G}_T^\circ - \text{H}_{298}^\circ)/T$ cal/mol, K	$-\Delta\text{G}_f^\circ$ kcal/mol	$-\Delta\text{H}_f^\circ$ kcal/mol
298	0	26.60	32.10	32.10	39.68	39.68
300	49	26.65	32.26	32.10	40.03	39.68
400	2828	28.68	40.24	33.17	40.18	40.94
500	5758	29.85	46.78	35.26	40.01	40.89
600	8790	30.77	52.30	37.65	39.84	41.09
700	11912	31.67	57.11	40.10	39.65	40.99
800	15127	32.64	61.41	42.50	39.39	41.73
900	18444	33.72	65.31	44.82	39.13	41.52
1000	21874	34.92	68.92	47.05	38.72	42.97
1100	25433	36.27	72.31	49.20	38.27	43.00
1200	29134	37.77	75.53	51.26	37.88	42.93
1300	32993	39.43	78.62	53.24	37.43	42.73
1400	37025	41.25	81.61	55.16	37.07	42.40
1500	41248	43.22	84.52	57.02	36.71	41.90
1600	45676	45.37	87.38	58.83	36.35	41.23
1700 ^a	50326	47.67	90.20	60.59	36.11	40.34
1800 ^a	55215	50.14	92.99	62.32	35.90	39.25
1900 ^a	60359	52.77	95.77	64.00	35.66	37.91
2000 ^a	65775	55.57	98.55	65.66	35.66	36.33
2100 ^a	71479	58.54	101.33	67.29	35.66	34.82
2200 ^a	77489	61.67	104.13	68.90	35.80	32.31
2285 ^a	82848	64.46	106.52	70.27	35.97	30.23

^aExtrapolated

Entropy

$$S_T^\circ = -149.79 + 31.722 \ln T - 4.7856 \times 10^{-3} T + 4.1902 \times 10^{-6} T^2 + \frac{1.9738 \times 10^5}{T^2}$$

The value for the heat of formation of Pu_2C_3 suggested by the IAEA Panel is $\Delta H_f^\circ, 298 = -39.7$ kcal/mol. This value is based on an assessment of the combustion measurements of Johnson *et al.*²⁸ and a composite of vaporization data of Campbell *et al.*²³ Marcon,²⁴ and Olson and Mulford.²⁴ Calculated values for the heat of formation and free energy of formation at higher temperatures are listed in Table 7.

Vaporization Behavior

Plutonium and carbon pressures over monocarbide and sesquicarbide phases were calculated from the appropriate values for the free energies of formation of the condensed phases (Tables 6, 7, and 8) and those for Pu(g) , $\text{C}_1(\text{g})$, $\text{C}_2(\text{g})$, and $\text{C}_3(\text{g})$ (Table 8). Equations for the temperature dependency of plutonium- and carbon-bearing species are given in Table 9 and plotted in Fig. 3. It is apparent that Pu(g) is the predominant species above the monocarbide and sesquicarbide phases.

TABLE 8. Free Energies of Formation of Gaseous and Condensed Phases

$$\Delta G_f^\circ = A + BT \text{ (kcal/mol)}$$

Species	A	B	Ref.
UC(s)	-26260	0.20	1
UC _{1.5} (s)	-24035	-1.33	1
PuC(s)	-11503	-1.50	1
PuC _{1.5} (s)	-18920	0.49	1
U(g)	117500	-27.0	1
UC ₂ (g)	153500	-38.6	1
Pu(g)	78300	-21.0	35
C ₁ (g)	170940	-37.52	11
C ₂ (g)	197354	-45.27	11
C ₃ (g)	186568	-44.84	11

URANIUM-PLUTONIUM-CARBON SYSTEM

Thermal Functions of Mixed Carbides

Information on the thermodynamic properties of $(\text{U,Pu})\text{C}$ and $(\text{U,Pu})_2\text{C}_3$, based on measurements, is entirely lacking in the literature.

Fig. 3. Calculated Values for the Temperature Dependency of Plutonium- and Carbon-Bearing Species over $\text{PuC}_{0.88}$ and Pu_2C_3

TABLE 9. Equations for Temperature Dependency of Plutonium- and Carbon-Bearing Species over Plutonium Monocarbide and Plutonium Sesquicarbide

$$\log p(\text{atm}) = - \frac{A \times 10^4}{T} + B \quad (1200-1800 \text{ K})$$

	PuC _{0.88}		Pu ₂ C ₃	
	A	B	A	B
Pu(g)	1.73	3.64	2.12	4.69
C ₁ (g)	4.02	7.83	4.00	8.90
C ₂ (g)	4.87	9.17	4.84	11.29
C ₃ (g)	4.94	8.68	4.86	11.90
C _{Total} (g)	4.02	7.83	4.00	8.91
P _{Total} (g)	1.73	3.64	2.12	4.69

Thermodynamic property values for U_{0.8}Pu_{0.2}C calculated assuming ideal solution and IAEA Panel values for UC and PuC_{0.88} show a rapid rise in heat capacity with increasing temperature in the temperature range 1000-1900 K. This increase is probably related to the presence of vacancies in the crystal structure of carbon-deficient PuC_{0.88} compared with that of stoichiometric UC. However, heat capacity values for PuC_{0.88} liquid, estimated using the method of Winslow²⁹ and discussed by Sheth and Leibowitz,³⁰ do not show abnormally high values probably because of the disappearance of vacancies in the liquid state; moreover, these values are consistent with the magnitude and trend in C_p values for stoichiometric uranium monocarbide above 1900 K. Therefore, heat capacity values for U_{0.8}Pu_{0.2}C were calculated assuming ideal solution between UC and PuC_{0.88} above the melting point of PuC_{0.88} and drawing a smooth curve connecting the values between 1000 and 1900 K. The thermodynamic equations derived for the monocarbide U_{0.8}Pu_{0.2}C over the temperature range 298-1900 K are:

Enthalpy

$$H_T^\circ - H_{298}^\circ = -4787 + 13.882 T + 1.4283 \times 10^{-4} T^2 + 2.2866 \times 10^{-7} T^3 + \frac{188170}{T}$$

Heat Capacity

$$C_p^\circ = 13.882 + 2.8565 \times 10^{-4} T + 6.8597 \times 10^{-7} T^2 - \frac{188170}{T}$$

Entropy

$$S_T^\circ = -65.443 + 13.882 \ln T + 2.8565 \times 10^{-4} T + 3.4299 \times 10^{-7} T^2 + \frac{94085}{T^2}$$

Calculated values for these and other thermodynamic properties are given in Table 10.

TABLE 10. Calculated Values for Thermodynamic Properties of $(U_{0.8}Pu_{0.2})C$

T, K	$(H_T^\circ - H_{298}^\circ)$ cal/mol	C_p° cal/mol, K	S_T° cal/mol, K	$-(G_T^\circ - H_{298}^\circ)/T$ cal/mol, K	$-\Delta G_f^\circ$ kcal/mol	$-\Delta H_f^\circ$ kcal/mol
298	0	11.86	14.80	14.80	21.18	21.00
300	24	11.94	14.90	14.82	21.39	20.99
400	1274	12.93	18.49	15.30	21.57	20.84
500	2595	13.44	21.43	16.24	21.79	20.60
600	3957	13.78	23.92	17.32	22.04	20.48
700	5348	14.03	26.06	18.42	22.31	20.40
800	6762	14.26	27.95	19.50	22.58	20.48
900	8198	14.46	29.64	20.53	22.84	20.66
1000	9655	14.67	31.17	21.52	23.05	21.27
1100	11131	14.87	32.58	22.46	23.17	22.17
1200	12629	15.08	33.88	23.36	23.27	22.16
1300	14148	15.30	35.10	24.22	23.37	22.11
1400	15690	15.53	36.24	25.04	23.46	22.01
1500	17255	15.77	37.32	25.82	23.49	23.13
1600	18844	16.02	38.35	26.57	23.51	23.02
1700	20459	16.29	39.33	27.29	23.55	22.84
1800	22101	16.56	40.27	27.99	23.60	22.58

Thermodynamic property values for $(U_{0.8}Pu_{0.2})_2C_3$ were calculated assuming ideal solution and IAEA Panel thermal function values for U_2C_3 and Pu_2C_3 . The results are summarized in Table 11. The thermodynamic equations derived for the sesquicarbide $(U_{0.8}Pu_{0.2})_2C_3$ are:

Enthalpy

$$H_T^\circ - H_{298}^\circ = -12238.0 + 35.15 T - 5.058 \times 10^{-3} T^2 \\ + 3.1954 \times 10^{-6} T^3 + \frac{6.3423 \times 10^5}{T}$$

Heat Capacity

$$C_p^\circ = 35.15 + 1.0116 \times 10^{-2} T + 9.5863 \times 10^{-6} T^2 - \frac{6.3423 \times 10^5}{T^2}$$

Entropy

$$S_T^\circ = -168.47 + 35.15 \ln T - 1.0116 \times 10^{-2} T + 4.7933 \times 10^{-6} T^2 + \frac{3.1712 \times 10^5}{T^2}$$

Vaporization Behavior of U-Pu-C System

Because of the very limited data available on the vaporization behavior of well characterized U-Pu-C compositions, calculations have been made to estimate the total pressure of actinide- and carbon-bearing species over the U-Pu-C system. These calculations are based on the equations given by Potter *et al.*^{1, 31-33}

The equations defining the terminal compositions of the $U_{1-x_1}Pu_{x_1}C-U_{1-x_2}Pu_{x_2}C_{1.5}$ diphasic region in the ternary U-Pu-C system is given by

$$\left[\Delta G_f^\circ(UC_{1.5}, s) - \Delta G_f^\circ(UC, s) \right] - \left[\Delta G_f^\circ(PuC_{1.5}, s) - \Delta G_f^\circ(PuC, s) \right] = RT \ln \left(\frac{1-x_1}{x_1} \right) \left(\frac{x_2}{1-x_2} \right)$$

The calculations predict that the sesquicarbide phase will be richer in plutonium than the monocarbide. This has been verified experimentally;³¹ however, the magnitude of the segregation is less than the calculated values.

For plutonium vaporization

$$RT \ln p(Pu, g) = 3\Delta G_f^\circ(PuC, s) - 2\Delta G_f^\circ(PuC_{1.5}, s) - \Delta G_f^\circ(Pu, g) - 2RT \ln x_2 + 3RT \ln x_1$$

For uranium vaporization

$$RT \ln p(U, g) = 3\Delta G_f^\circ(UC, s) - 2\Delta G_f^\circ(UC_{1.5}, s) - \Delta G_f^\circ(U, g) \\ - 2RT \ln (1-x_2) + 3RT \ln (1-x_1)$$

TABLE 11. Calculated Values for Thermodynamic Properties of $(U_{0.8}Pu_{0.2})_2C_3$

T, K	$(H_T^\circ - H_{298}^\circ)$ cal/mol	C_p° cal/mol, K	S_T° cal/mol, K	$-(G_T^\circ - H_{298}^\circ)/T$ cal/mol, K	$-\Delta G_f^\circ$ kcal/mol	$-\Delta H_f^\circ$ kcal/mol
298	0	25.85	32.76	32.76	43.73	42.58
300	52	25.93	32.94	32.76	43.81	42.57
400	2803	28.67	40.83	33.83	44.31	42.27
500	5740	29.95	47.38	35.90	44.87	41.83
600	8778	30.77	52.92	38.29	45.51	41.64
700	11891	31.47	57.72	40.73	46.17	41.59
800	15074	32.20	61.96	43.12	46.81	41.85
900	18334	33.03	65.80	45.43	47.43	42.03
1000	21684	33.99	69.33	47.65	47.91	43.68
1100	25136	35.10	72.62	49.77	48.24	45.57
1200	28709	36.38	75.73	51.81	48.49	45.64
1300	32417	37.83	78.70	53.76	48.72	45.61
1400	36280	39.45	81.56	55.65	48.97	45.46
1500	40314	41.26	84.34	57.47	49.05	47.72
1600	44538	43.26	87.07	59.23	49.14	47.49
1700	48971	45.44	89.75	60.95	49.26	47.06
1800	53632	47.81	92.42	62.62	49.41	46.43
1900	58539	50.36	95.07	64.26	49.58	45.57
2000	63710	53.11	97.72	65.87	49.84	44.47
2100	69166	46.04	100.38	67.45	50.14	43.17

For $\text{UC}_2(\text{g})$ vaporization

$$\begin{aligned} RT \ln p(\text{UC}_2, \text{g}) &= 2\Delta G_f^\circ(\text{UC}_{1.5}, \text{s}) - \Delta G_f^\circ(\text{UC}, \text{s}) - \Delta G_f^\circ(\text{UC}_2, \text{g}) \\ &+ 2RT \ln (1 - x_2) - RT \ln (1 - x_1) \end{aligned}$$

The carbon activity is given by

$$RT \ln a_c = 2\Delta G_f^\circ(\text{UC}_{1.5}, \text{s}) - 2\Delta G_f^\circ(\text{UC}, \text{s}) + 2RT \ln (1 - x_2) - 2RT \ln (1 - x_1)$$

Equations for the free energies of formation of the gaseous species and the condensed phases are given in Table 8. Typical results calculated for 1800 K are shown in Fig. 4. Equations for the temperature dependency of actinide and carbon gaseous species above the monocarbide phase (at the lower phase boundary) and sesquicarbide phase (upper phase boundary), where $\text{Pu}/(\text{U} + \text{Pu}) = 0.2$, are given in Table 12 and plotted in Figs. 5 and 6. Figures 5 and 6 show that, in the temperature range approximately 1800–2500 K, $\text{Pu}(\text{g})$ is the predominant species above the MC-MC_{1.5} system. This is consistent with the loss of plutonium observed by many investigators when working with plutonium-bearing materials at high temperatures. The $\text{Pu}(\text{g})$ pressures estimated from the results reported by Ohse³⁴ above the monocarbide phase are compared with those obtained via calculation in Table 13. The agreement between the measured and calculated results is reasonable. The calculated pressures at the lower phase boundary of the MC-MC_{1.5} system represent a lower limit of the plutonium pressure above the monocarbide phase.

A comparison of actinide and carbon partial pressures above UC, $\text{PuC}_{0.88}$, and $\text{U}_{0.8}\text{Pu}_{0.2}\text{C}$ monocarbide phase compositions is shown in Fig. 7.

TABLE 12. Equations for Temperature Dependency of Actinide- and Carbon-Bearing Species over Monocarbide and Sesquicarbide Phases in the U-Pu-C System

$$\log p(\text{atm}) = - \frac{A \times 10^4}{T} + B \quad (1500\text{--}2500 \text{ K})$$

	$\text{U}_{0.8}\text{Pu}_{0.2}\text{C}_{1.00}$		$(\text{U}_{0.8}\text{Pu}_{0.2})_2\text{C}_3$	
	A	B	A	B
$\text{Pu}(\text{g})$	1.91	3.63	2.20	4.78
$\text{U}(\text{g})$	3.09	5.92	3.16	6.23
$\text{UC}_2(\text{g})$	3.98	8.31	3.82	7.65
$\text{C}_1(\text{g})$	3.79	8.13	3.70	7.74
$\text{C}_2(\text{g})$	4.42	9.75	4.24	8.97
$\text{C}_3(\text{g})$	4.24	9.58	3.96	8.41
$\text{P}_{\text{C}_T}(\text{g})$	3.85	8.54	3.78	8.27
$\text{P}_T(\text{g})$	1.91	3.64	2.20	4.78

Fig. 4. Calculated Values of Actinide and Carbon Pressures Above the Monocarbide-Sesquicarbide Region of the U-Pu-C System

Fig. 5. Calculated Values for the Temperature Dependence of Actinide- and Carbon-Bearing Species over Monocarbide Phase $\text{U}_{0.8}\text{Pu}_{0.2}\text{C}$

Fig. 6. Calculated Values for the Temperature Dependence of Actinide- and Carbon-Bearing Species over Sesquicarbide Phase $(\text{U}_{0.8}\text{Pu}_{0.2})_2\text{C}_3$

Fig. 7. Comparison of Actinide and Carbon Pressures Above UC, PuC, and $\text{U}_{0.8}\text{Pu}_{0.2}\text{C}$ Systems

TABLE 13. Comparison of Experimental and Calculated
Pu(g) Pressures Above $U_{0.8}Pu_{0.2}C$

T, K	-log p, atm	
	Experimental	Calc. (Lower Phase Boundary, MC-MC _{1.5})
1800	6.01	6.98
2000	4.99	5.92
2200	4.15	5.05
2400	3.46	4.33
2500	3.15	4.01

SUMMARY AND RECOMMENDED RESEARCH

The thermodynamic properties of the U-C system are reasonably well established. More thermal function data, particularly at high temperatures and on well-characterized samples, are needed to define the heat capacity of plutonium monocarbide and plutonium sesquicarbide. Carbon and plutonium activity data above the monocarbide and sesquicarbide homogeneity regions of the phase diagram are needed, again on well-defined compositions. Additional phase studies are needed to establish phase boundaries in this system.

Hardly any thermodynamic information exists for the U-Pu-C system, and the areas for research in this ternary system are numerous. In addition to phase equilibria and thermal function data, actinide and carbon activity measurements over well-defined compositions in the ternary U-Pu-C system would be particularly valuable.

ACKNOWLEDGMENTS

The authors are indebted to Dr. R. J. Ackermann for pertinent discussions on actinide carbide systems and the IAEA Panel for permission to quote preliminary values from their assessment. We are also indebted to Dr. C. E. Johnson for the difficult task of coordinating various groups involved in current property assessments of advanced fuel systems.

REFERENCES

1. R. J. Ackermann, P. Potter, M. Rand, and E. K. Storms, IAEA Panel Assessment of the Thermodynamic Properties of the U-C, Pu-C, and U-Pu-C Systems, Grenoble, France, May 6-8, 1974, to be published.
2. E. F. Westrum, Y. Takahashi, and N. D. Stout, J. Phys. Chem. 69, 1520 (1965).
3. E. F. Westrum, E. Suite, and H. K. Lonsdale, *Thermophysical Properties at Extreme Temperatures and Pressures*, S. Gratch, Ed., p. 156, Amer. Soc. Mech. Engrs. (1965).
4. J. D. Farr, E. J. Huber, E. L. Head, and C. E. Holley, Jr., J. Phys. Chem. 63, 1455 (1959).
5. L. C. Harrington and G. H. Rowe, *Carbides in Nuclear Energy*, Vol. I, L. E. Russell, Ed., p. 342, Macmillan, New York.
6. F. L. Oetting, J. D. Navratil, and E. K. Storms, J. Nucl. Mater. 45, 271 (1973).
7. L. S. Levinson, J. Chem. Phys. 40, 3584 (1964).
8. L. S. Levinson, *Carbides in Nuclear Energy*, Vol. I, L. E. Russell, Ed., p. 429, Macmillan, New York (1964).
9. E. J. Storms and E. J. Huber, J. Nucl. Mater. 23, 19 (1967).
10. H. P. Stephens, High Temp. Sci. 6, 156 (1974).
11. H. L. Schick, *Thermodynamics of Certain Refractory Compounds*, Vol. II, Academic Press, New York (1966).
12. E. J. Huber and C. E. Holley, J. Chem. Thermodyn. 1, 267 (1969).
13. J. D. Farr, W. G. Wittman, P. L. Stone, and E. F. Westrum, *Thermophysical Properties at Extreme Temperatures and Pressures*, S. Gratch, Ed., p. 162, Amer. Soc. Mech. Engrs. (1965).
14. R. J. L. Andon, J. F. Counsell, J. F. Martin, and H. J. Hedger, Trans. Faraday Soc. 60, 1030 (1964).
15. E. J. Huber, C. E. Holley, and W. G. Wittman, J. Chem. Thermodyn. 1, 579 (1969).
16. E. K. Storms, *Thermodynamics*, Vol. I, IAEA, Vienna, p. 309 (1966).
17. E. K. Storms, *The Refractory Carbides*, Chapter XI, pp. 205-213, Academic Press, New York (1967).
18. M. Tettenbaum and P. D. Hunt, J. Nucl. Mater. 40, 104 (1971).
19. P. Sutcliffe, UKAEA Report AERE-R-7172 (1972).

20. O. L. Kruger and H. Savage, J. Chem. Phys. 40, 3324 (1964).
21. F. L. Oetting, private communication to Rand (1974).
22. G. K. Johnson, E. H. VanDeventer, O. L. Kruger, and W. N. Hubbard, J. Chem. Thermodyn. 2, 617 (1970).
23. G. M. Campbell, R. A. Kent, J. A. Leary, *Plutonium 1970*, p. 781.
24. J. P. Marcon, J. Inorg. Nucl. Chem. 32, 2581 (1970).
25. P. S. Harris, B. A. Phillips, M. H. Rand, and M. Tetenbaum, AERE-R-5353 (1967).
26. W. M. Olson and R. N. R. Mulford, *Thermodynamics of Nuclear Materials*, IAEA, Vienna, p. 467 (1968).
27. J. Danan, J. Nucl. Mater. 45, 339 (1972/1973).
28. G. K. Johnson, E. H. VanDeventer, O. L. Kruger, and W. N. Hubbard, J. Chem. Thermodyn. 6, 219 (1974).
29. G. Winslow, Argonne National Laboratory, private communication.
30. A. Sheth and L. Leibowitz, "Equation of State and Transport Properties for Actinide Carbides in the Liquid Region," ANL-AFP-11, to be published.
31. P. E. Potter and W. G. Roberts, UKAEA Report AERE-R-5661 (1968).
32. P. G. Mardon and P. E. Potter, UKAEA Report AERE-R-6514 (1970).
33. P. E. Potter, UKAEA Report AERE-R-5922.
34. R. W. Ohse, Euratom Progress Report No. 15, January-June 1973, p. 22.
35. R. J. Ackermann and E. G. Rauh, J. Chem. Thermodyn. 7, 211 (1975).