**EQUATION OF STATE AND TRANSPORT
PROPERTIES OF URANIUM AND PLUTONIUM CARBIDES
IN THE LIQUID REGION**

by

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ABSTRACT

By the use of available low-temperature data for various thermophysical and transport properties for uranium and plutonium carbides, values above the melting point were estimated. Sets of recommended values have been prepared for the compounds UC, PuC, and (U,Pu)C. The properties that have been evaluated are density, heat capacity, enthalpy, vapor pressure, thermal conductivity, viscosity, and emissivity.

INTRODUCTION

To improve the performance of breeder reactors, fuels other than oxide fuels are being developed. The advanced fuels currently being considered are carbides and nitrides of uranium and plutonium. These fuels have better neutronic properties and higher thermal conductivities than oxide fuels. These properties will permit operation of fuel pins at higher linear power ratings and will also allow enough flexibility in design to optimize breeding ratio and minimize fabrication cost and fissile-material inventory.

Reactor safety analysis depends on the application of massive computer codes to predict the behavior of a system as a result of various hypothetical core disruptive accidents (HCDA). Involved in these calculations are many thermodynamic and transport properties of the materials employed in the reactor. These data are required for temperatures ranging from those of normal operation to as high as 6000°C. Although adequate data are available at lower temperatures, values for the temperature region of interest to the safety analyst are sparse, and, therefore, efforts are being made to extrapolate low-temperature data to these higher temperatures. The compounds being considered are UC, UN, PuC, PuN, (U,Pu)C, and (U,Pu)N. The properties that are being extrapolated or estimated by us are heat capacity, enthalpy, density, vapor pressure, thermal conductivity, viscosity, and emissivity. The present report describes only the work done on carbide fuels. A separate report is being prepared on the work done on nitride fuels.

To extrapolate various properties into the liquid region, the low-temperature data were selected from the recent critical evaluations made by scientists working at Argonne National Laboratory^{1,2} and Los Alamos Scientific Laboratory.^{3,4} By extrapolating data from lower to higher

TABLE 3. Recommended Values for Heat Capacity and Enthalpy of $(U_{0.8}Pu_{0.2})C$ below the Melting Point^a

Temp K	C_p° cal/(mol) (K)	$H_T^\circ - H_{298}^\circ$ cal/mol	Temp, K	C_p° cal/(mol) (K)	$H_T^\circ - H_{298}^\circ$ cal/mol
300	11.94	24	1600	16.02	18844
400	12.93	1274	1700	16.29	20459
500	13.44	2595	1800	16.56	22101
600	13.78	3957	1900 ^b	16.85	23772
700	14.03	5348	2000 ^b	17.15	25472
800	14.26	6762	2100 ^b	17.46	27202
900	14.46	8198	2200 ^b	17.79	28965
1000	14.67	9655	2300 ^b	18.13	30761
1100	14.87	11131	2400 ^b	18.49	32592
1200	15.08	12629	2500 ^b	18.85	34459
1300	15.30	14148	2600 ^b	19.23	36363
1400	15.53	15690	2700 ^b	19.63	38306
1500	15.77	17255	2758 ^b	19.86	39451

^aRecommended by Tetenbaum *et al.*¹

^bExtrapolation based on:

$$H_T^\circ - H_{298}^\circ = -4787 + 13.882T + 1.4283 \times 10^{-4}T^2 + 2.2886 \times 10^{-7}T^3 + \frac{188170}{T}$$

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

the solid state, one may need to apply different methods to extrapolate low-temperature data into the liquid region. Therefore, in the absence of adequate experimental information, we have applied the thermodynamic approach recommended by Winslow,⁹ to both compounds for the calculation of heat capacity and enthalpy values in the liquid region.

From basic thermodynamic considerations, one may derive the following equation:

$$C_p = C_v (1 + \gamma \beta T) \quad (5)$$

where

C_p = heat capacity at constant pressure

temperatures, sets of self-consistent values for the solid and liquid regions were obtained. In the few cases for which no data were available, estimates were made on the basis of similarity or various correlations between related compounds. To supplement the data that are presented in tabular form, best fitting equations were developed by applying regression techniques to standard forms of equations. No attempt was made to determine the optimum form of the equation. The estimated sets of properties values, along with the recommended low-temperature data that were used, are given below. The basis for choosing a method of extrapolation or estimation is also discussed.

Density of Carbide Fuels

The density of molten ceramic fuel is an important parameter in safety analyses. No reliable data on the density of liquid carbide fuels are available, and therefore we have made use of data on thermal expansion and density of solid carbides to derive an equation showing the temperature dependency of the density of the liquid carbides. Richards⁵ measured the thermal expansion coefficient in liquid UC, as well as the expansion accompanying the transition from solid to liquid. However, the chemical analyses that he performed after the experiments showed that the sample had a composition near UC₂. The values reported by him may therefore be suspect. Andrew³ has recently reviewed the density and thermal expansion data of carbide fuels in the solid state. He has given values for the average linear thermal expansion coefficients for UC, PuC_{0.85}, and (U_{0.95}Pu_{0.05})C_{0.98} as a function of temperature.

In deriving the temperature dependency of density of the liquid phase from low-temperature data, our approach was as follows: Andrew³ has given values for the average linear thermal expansion coefficient for UC_{solid} from room temperature to 2000°C. Extrapolating his data to the recommended melting point of UC of 2780 K (recommended by Fee and Johnson²), we obtained the average linear thermal expansion coefficient for the solid, α_s , from room temperature to the melting point; α_s was found to be equal to $1.38 \times 10^{-5}/K$. In the absence of a reliable value for the volume change on melting ($\Delta V/V_s$) for UC, we took the value of 26.06%, which is the value recommended for NaCl by Kirshenbaum *et al.*⁶ From the data on NaCl given by Janz,⁷ the ratio of linear thermal expansion coefficients for the solid and liquid states of NaCl was calculated to be 0.5. By using this ratio and the value of α_s given earlier, the average value for the thermal expansion coefficient of liquid UC (α_l) from the melting point to a higher temperature, T, was estimated as being equal to $2.60 \times 10^{-5}/K$. By using a value for the density of solid UC at 20°C of 13.61 g/cm³ (recommended by Andrew³), the following equation was derived to give the density of liquid UC:

$$\rho_L = \frac{13.61}{1.2606 [1 + 3(T_m - 293)\alpha_s] [1 + 3(T - T_m)\alpha_l]} \quad T \geq T_m \quad (1)$$

where ρ_L = liquid density, g/cm³

T_m = melting point, K

α_s = average linear thermal expansion coefficient for solid,
cm cm⁻¹ K⁻¹

α_l = average linear thermal expansion coefficient in liquid,
cm cm⁻¹ K⁻¹

T = temperature, K

Substitution of the values for α_s , T_m , and α_l for UC and a further simplification reduced the above equation to

$$\rho_L[\text{UC}] = \frac{12.57}{1 + 9.98 \times 10^{-5} T} \quad T \geq T_m \quad (2)$$

Equation 2 represents our best estimate of the density of liquid UC, but it should be used cautiously to calculate liquid density at temperatures much higher than the melting point. The value for α_l may change with temperature, and the method of calculation, *i.e.* by analogy with NaCl, may not be valid at higher temperatures; moreover, in real situations, the chemical composition of UC may change, giving rise to unknown effects on density.

The low-temperature data for PuC_{0.85} recommended by Andrew³ was used to estimate a value of α_s for PuC_{0.88}. In this estimate, the effect of stoichiometry was neglected. Between room temperature and the melting point of PuC of 1875 K (recommended by Fee and Johnson²), the value of α_s was estimated to be $1.13 \times 10^{-5}/\text{K}$. In the absence of information on the volume expansion on melting and the liquid thermal expansion coefficient, the same approach used for UC was adopted here. The value for α_l was calculated to be $2.26 \times 10^{-5}/\text{K}$ and the percentage volume increase on melting was assumed to be equal to 26.06%. By using the recommended³ room-temperature density of 13.55 g/cm³ for PuC_{0.90} and the above values of α_s , T_m , and α_l , the following equation was derived for the liquid density of PuC_{0.88}:

$$\rho_L[\text{PuC}_{0.88}] = \frac{11.69}{1 + 7.77 \times 10^{-5} T} \quad T \geq T_m \quad (3)$$

According to Andrew,³ the effect of stoichiometry, the slight addition (0.1 wt %) of Ni, and a small amount of the sesquicarbide phase do not change the thermal expansion coefficients of (U,Pu)C within the experimental error of 5%. On this basis, α_s for (U_{0.8}Pu_{0.2})C from room temperature to the melting point of 2758 K (recommended by Fee and Johnson²) was estimated from the low-temperature data for (U_{0.95}Pu_{0.05})C_{0.98} given by Andrew.³ This value is equal to $1.29 \times 10^{-5}/\text{K}$. For α_l and the percentage increase in volume on melting, the same assumptions made as those for UC and PuC_{0.88}. By using a value for the room temperature density of (U_{0.8}Pu_{0.2})C_{0.95} of 13.45 g/cm³, as recommended by Andrew,³ and the values of α_s , T_m , and α_l , the following equation was derived for the liquid density of (U_{0.8}Pu_{0.2})C:

$$\rho_L[(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}] = \frac{12.39}{1 + 9.84 \times 10^{-5} T} \quad T \geq T_m \quad (4)$$

In using Eqs. 3 and 4 for temperatures much above the melting point, the same cautionary statements apply as those made above for Eq. 2.

Heat Capacity and Enthalpy of Carbide Fuels

The low-temperature data for UC, $\text{PuC}_{0.88}$, and $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$ were taken from the report by Tetenbaum *et al.*¹ for the purpose of extrapolating into the liquid region. The data used were reported up to temperatures near the melting points recommended by Fee and Johnson.² In Tables 1-3, the heat capacity and enthalpy values for the three carbides are given up to the recommended melting points.

By using an ideal solution approach and the recommended^{1,8} heat capacity data for UC and $\text{PuC}_{0.88}$, the heat capacity of $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$ could be estimated. The resulting values would show a very steep increase in C_p° with increasing temperature. This behavior would be similar to that of $\text{PuC}_{0.88}$, probably resulting from substoichiometry and vacancy formation. However, since $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$ is a stoichiometric material one would expect it to behave much like UC where C_p° values did not show very steep rise with increasing temperature. Simply using the $\text{PuC}_{0.88}$ data, we believe, would give unrealistically high values. As an alternative approach, the following assumptions were made to calculate C_p° for the mixed carbide in solid state:

- (1) From $T = 300$ to 1000 K,
 $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}(\text{solid}) = \text{UC}(\text{solid}) + \text{PuC}_{0.88}(\text{solid})$

The calculations were made using the C_p° data of the IAEA panel for both UC (s) and $\text{PuC}_{0.88}$ (s).

- (2) From $T = 1900$ to 2700 K,
 $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}(\text{solid}) = \text{UC}(\text{solid}) + \text{PuC}_{0.88}(\text{liquid})$

The calculations were made using the C_p° data of the IAEA panel for UC (s). C_p° values for $\text{PuC}_{0.88}(\text{l})$ were calculated by a method due to Winslow^{9,10} (described in detail below).

The resulting heat capacity values for the stoichiometric mixed carbide in the range from 1900 – 2700 K were in a good agreement with the UC values in the same range. Because of this agreement, data calculated from the IAEA panel's recommended values for UC and $\text{PuC}_{0.88}$ in the range from 300 to 1000 K were connected by a smooth curve to the values in the 1900 – 2700 K region and a least-squares fit was made. The C_p° values calculated from the resulting equation did not show the steep rise with increasing temperature. The enthalpy values were then calculated from the fitted equation. The values calculated from these derived equations and the equations themselves are given in Table 3.

To extrapolate the low-temperature enthalpy data into the liquid region, a correction has to be made for the heat of fusion at the melting point. Based on the different behavior of C_p° data for UC and $\text{PuC}_{0.88}$ noted above for

TABLE 1. Recommended Values for Heat Capacity and Enthalpy of UC below the Melting Point^a

Temp, K	C _p ^o , cal/(mol)(K)	H _T ^o -H ₂₉₈ ^o , cal/mol
300	12.01	22
400	13.19	1290
500	13.76	2640
600	14.10	4034
700	14.36	5458
800	14.57	6904
900	14.76	8371
1000	14.95	9857
1100	15.15	11362
1200	15.36	12887
1300	15.58	14434
1400	15.82	16004
1500	16.07	17598
1600	16.34	19218
1700	16.62	20865
1800	16.93	22543
1900	17.25	24252
2000	17.59	25994
2100	17.96	27771
2200	18.34	29586
2300	18.74	31439
2400	19.16	33334
2500 ^b	19.60	35272
2600 ^b	20.06	37254
2700 ^b	20.53	39283
2780 ^b	20.93	40941

^aRecommended data of Tetenbaum *et al.*¹^bExtrapolation based on:

$$H_T^o - H_{298}^o = 5.174 \times 10^3 + 14.727T - 2.6733 \times 10^{-4}T^2 + 3.3304 \times 10^{-7}T^3$$

$$+ \frac{2.3797 \times 10^{-5}}{T} \quad (300 - 2418 \text{ K})$$

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

TABLE 2. Recommended Values for Heat Capacity and Enthalpy of $\text{PuC}_{0.88}$ below the Melting Point^a

Temp K	C_p° cal/(mol) (K)	$H_T^\circ - H_{298}^\circ$ cal/mol
300	11.42	21
400	11.86	1192
500	11.93	2382
600	12.02	3578
700	12.26	4790
800	12.74	6039
900	13.48	7349
1000	14.51	8746
1100	15.80	10259
1200	17.38	11915
1300	19.24	13744
1400	21.42	15775
1500	23.85	18036
1600 ^b	26.60	20557
1700 ^b	29.66	23367
1800 ^b	32.98	26496
1875 ^b	35.68	29070

^aRecommended data of Tetenbaum *et al.*¹

^bExtrapolation based on:

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

$$C_p^\circ = 18.607 - 1.8683 \times 10^{-2}T + 1.4842 \times 10^{-5}T^2 - \frac{2.6291 \times 10^5}{T^2}$$

C_v = heat capacity at constant volume

γ = Grüneisen constant (≈ 2)

β = volumetric expansion coefficient

T = temperature

Although Winslow¹⁰ has given the relationship between β and other lattice parameters for some compounds, information on the carbide fuels was not given. Hence, we decided to use the values listed in Table 4. Values of the Grüneisen constant for most materials lie between 1 and 3. We have therefore taken $\gamma = 2$. For C_v a value of $3R$ per atom was used as was done by Winslow⁹. From the earlier section on density, values of α_l were used to calculate β assuming a simple relation of volumetric expansion coefficient being three times greater than linear expansion coefficient.

TABLE 4. Values of Parameters to Calculate Heat Capacity at Very High Temperatures

Parameter	UC	PuC _{0.88}	U _{0.8} Pu _{0.2})C
γ	2	2	2
β	7.81×10^{-5}	6.78×10^{-5}	7.74×10^{-5}
C_v^a	6R	6R	6R

^aR is the gas constant

The value for the heat of fusion was estimated from the recommended melting point given by Fee and Johnson² and entropy of fusion equal to 4.2 eu used by Sheth and Leibowitz.¹¹ The calculated values for the heats of fusion of UC and PuC_{0.88} were 11.68 kcal/mol and 7.88 kcal/mol, respectively. The value of the heat of fusion for (U_{0.8}Pu_{0.2})C, calculated by linear interpolation, was equal to 10.92 kcal/mol.

Usually, as temperature increases, C_p values initially increase slowly, but, at a value of the reduced temperature ($T_r = T/T_c$) equal to about 0.8, they increase very rapidly with increasing temperature and reach an infinite value at the critical temperature. The equation suggested by Winslow and used by us show similar behavior if a proper variation of β with temperature was included. Because we used a constant value of β the heat capacity values will be too low at temperatures close to the critical temperature. However, critical temperatures, T_c , for carbides are not known. From the values given by Kirshenbaum *et al.*,⁶ one can calculate that the average ratio of melting point to critical temperature for alkali halides is equal to about 3.5. Using this ratio, we estimated the critical temperatures for UC, PuC_{0.88}, and (U_{0.8}Pu_{0.2})C to be 9750, 6550, and 9653 K, respectively. Based on the above information and using equation 5 and values given in Table 4, we calculated the heat capacity and enthalpy data for UC (up to 7800 K) and PuC_{0.88} (up to 5200 K); these values are given in Tables 5 and 6, respectively.

TABLE 5. Estimated Values of Heat Capacity and Enthalpy of UC in Liquid Region.^a

Temp, K	C _p ^o , cal/(mol)(K)	H _T ^o -H ₂₉₈ ^o , cal/mol	Temp, K	C _p ^o , cal/(mol)(K)	H _T ^o -H ₂₉₈ ^o , cal/mol	Temp, K	C _p ^o , cal/(mol)(K)	H _T ^o -H ₂₉₈ ^o , cal/mol
2780	17.10	52621	4500	21.31	84792	6200	23.47	122005
2800	17.14	52963	4600	20.49	86832	6300	23.66	124361
2900	17.33	54687	4700	20.68	88891	6400	23.85	126737
3000	17.51	56429	4800	20.87	90968	6500	24.03	129130
3100	17.70	58189	4900	21.05	93064	6600	24.22	131543
3200	17.88	59968	5000	21.24	95178	6700	24.41	133974
3300	18.07	61766	5100	21.42	97311	6800	24.59	136424
3400	18.26	63582	5200	21.61	99463	6900	24.78	138892
3500	18.44	65417	5300	21.80	101633	7000	24.96	141379
3600	18.63	67271	5400	21.93	103822	7100	25.15	143885
3700	18.82	69143	5500	22.17	106030	7200	25.34	146409
3800	19.00	71034	5600	22.36	108256	7300	25.52	148952
3900	19.19	72944	5700	22.54	110501	7400	25.71	151514
4000	19.38	74872	5800	22.73	112764	7500	25.90	154094
4100	19.56	76819	5900	22.91	115047	7600	26.08	156693
4200	19.75	78784	6000	23.10	117347	7700	26.27	159310
4300	19.93	80768	6100	23.29	119667	7800	26.45	161946
4400	20.12	82771						

^aValues are based on:

$$C_p^o = 11.9232 + 1.8629 \times 10^{-3}T$$

$$H_T^o - H_{298}^o = 12275.9 + 11.9232T + 9.3145 \times 10^{-4}T^2$$

TABLE 6. Estimated Values of Heat Capacity and Enthalpy of $\text{PuC}_{0.88}$ in Liquid Region.^a

Temp, K	C_p° , cal/(mol) (K)	$H_T^\circ - H_{298}^\circ$, cal/mol	Temp, K	C_p° , cal/(mol) (K)	$H_T^\circ - H_{298}^\circ$, cal/mol	Temp, K	C_p° , cal/(mol) (K)	$H_T^\circ - H_{298}^\circ$, cal/mol
1875	14.96	36950	3100	16.94	56483	4300	18.88	77969
1900	15.00	37324	3200	17.10	58154	4400	19.04	79865
2000	15.16	38832	3300	17.26	59902	4500	19.20	81776
2100	15.32	40356	3400	17.42	61636	4600	19.36	83704
2200	15.48	41896	3500	17.58	63386	4700	19.52	85649
2300	15.64	43452	3600	17.74	65152	4800	19.68	87609
2400	15.80	45024	3700	17.91	66935	4900	19.85	89585
2500	15.97	46612	3800	18.07	68733	5000	20.01	91578
2600	16.13	48217	3900	18.23	70548	5100	20.17	93587
2700	16.29	49838	4000	18.39	72379	5200	20.33	95612
2800	16.45	51475	4100	18.55	74226			
2900	16.61	53128	4200	18.71	76090			
3000	16.77	54797						

^aValues are based on:

$$C_p^\circ = 11.9232 + 1.6168 \times 10^{-3}T$$

$$H_T^\circ - H_{298}^\circ = 11752 + 11.9232T + 3.084 \times 10^{-4}T^2$$

Liquid heat capacity values calculated in this way show a very large decrease in heat capacity on melting for $\text{PuC}_{0.88}$ (from 35.68 to 14.96) as compared with UC (which decreases from 20.93 to 17.10). Clearly, further analysis of the $\text{PuC}_{0.88}$ case is warranted. In the absence of additional experimental information, the values of C_p° and $H_T^\circ - H_{298}^\circ$ reported here should be used with due caution.

By assuming an ideal solution behavior, heat capacity (and by integration, enthalpy) data for $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$ were calculated using data from Tables 5 and 6. Other calculations were made in which values for the mixed carbide given in Table 4 were used in applying Winslow's equation (e.g. equation 5). The values for heat capacity and enthalpy obtained by using Eq. 5 agreed within $\pm 1\%$ with the values obtained by assuming an ideal solution behavior. The values calculated from Winslow's equation are given in Table 7.

Vapor Pressure of Carbide Fuels

Consideration of the vapor pressure over carbide fuel material is complicated because of the various gaseous species that may exist over the solid and liquid. Moreover, available measurements in the low-temperature region are not complete and consistent, which makes high-temperature extrapolation and estimation very difficult. The well-known law of corresponding states implies that substances are characterized by their thermodynamic critical properties: critical temperature, T_c , critical density, ρ_c , (or critical volume, V_c), critical pressure, P_c , and critical compressibility factor, Z_c ; and that materials under the same reduced conditions of temperature, pressure, and volume will have similar behavior. This approximate relation has been used by several investigators first to estimate the critical properties of reactor fuel materials and then to apply the generalized vapor-pressure correlations given by Hougen, Watson, and Ragatz.¹² Miller¹³ has given a detailed report on various empirical and semi-theoretical approaches that have been used in the literature for estimation purposes. In general, the low-temperature vapor-pressure equations are obtained by fitting experimental data determined by effusion, transpiration, or mass-spectrometric methods. These data are thus sensitive to the details of stoichiometry, system pressure and the materials of containment used for the experiment. For reactor safety analysts, these equations must often be extrapolated six to nine orders of magnitude or higher in pressure than that experimentally measured. Such extrapolations are often reasonably accurate for common materials, but, at higher temperatures, effects such as dissociation, excited states, and ionization would be expected to complicate behavior for reactor fuel materials. Moreover, in using low-temperature vapor-pressure data to estimate high-temperature values, there is the additional danger that vapor species which may be important at high temperatures will not be adequately represented by the lower temperature data. The empirical approach that we have taken is to extrapolate equations derived from basic thermodynamic data for the pressures of individual vapor species and to sum the pressures to give the total pressure above the condensed phase. In making these extrapolations above the melting point, an adjustment is required for the heat of fusion. Following a suggestion of P. Blackburn,¹⁴ we have corrected each partial pressure equation by a multiple of the heat of fusion

TABLE 7. Estimated Values of Heat Capacity and Enthalpy of $(U_{0.8}Pu_{0.2})C$ in Liquid Region.^a

Temp, K	C_p° cal/(mol) (K)	$H_T^\circ - H_{298}^\circ$ cal/mol	Temp, K	C_p° cal/(mol) (K)	$H_T^\circ - H_{298}^\circ$ cal/mol	Temp, K	C_p° cal/(mol) (K)	$H_T^\circ - H_{298}^\circ$ cal/mol
2758	17.01	50371	4500	20.23	82809	6200	23.37	119865
2800	17.09	51087	4600	20.41	84841	6300	23.55	122211
2900	17.28	52806	4700	20.60	86892	6400	23.74	124576
3000	17.46	54542	4800	20.78	88961	6500	23.92	126958
3100	17.65	56298	4900	20.97	91048	6600	24.11	129360
3200	17.83	58071	5000	21.15	93154	6700	24.29	131779
3300	18.01	59863	5100	21.34	95279	6800	24.47	134217
3400	18.20	61674	5200	21.52	97422	6900	24.65	136674
3500	18.38	63503	5300	21.71	99583	7000	24.84	139149
3600	18.57	65351	5400	21.89	101763	7100	25.03	141643
3700	18.75	67217	5500	22.08	103961	7200	25.21	144155
3800	18.94	69101	5600	22.26	106178	7300	25.40	146685
3900	19.12	71004	5700	22.44	108413	7400	25.58	149234
4000	19.31	72925	5800	22.63	110666	7500	25.77	151801
4100	19.49	74865	5900	22.81	112938	7600	25.95	154387
4200	19.68	76824	6000	23.00	115229	7700	26.14	156992
4300	19.86	78800	6100	23.18	117538	7800	26.32	159614
4400	20.04	80796						

^aValues are based on:

$$C_p^\circ = 11.9232 + 1.8457 \times 10^{-3}T$$

$$H_T^\circ - H_{298}^\circ = 10467.1 + 11.9232T + 9.2285 \times 10^{-4}T^2$$

(ΔH_f), depending on the number of condensed-phase molecules that are required to form the gaseous molecule, *e.g.*, ΔH_f for U(g), $2 \Delta H_f$ for C₂(g), $3 \Delta H_f$ for C₃(g).

In the recent critical evaluation of low-temperature vapor-pressure data of UC, Tetenbaum *et al.*¹ assumed the existence of monatomic uranium species and UC₂(g), C₁(g), C₂(g), and C₃(g). In the present work, we have added C₄(g). Tetenbaum and coworkers derived equations giving the temperature dependence of the pressures of the various species. They were based on the carbon and uranium activity values given in the IAEA panel assessment⁸ of the U-C system. Necessary thermodynamic property values were also obtained from the IAEA panel's assessment.⁸ The equations with the values calculated therefrom are given in Table 8. The contribution of the C₄(g) species to the pressure was calculated on the basis of the following equations:

$$4 \text{ UC(s)} = 4 \text{ U(g)} + \text{C}_4(\text{g}) \quad (6)$$

$$4\Delta G_f^\circ(\text{U, g}) + \Delta G_f^\circ(\text{C}_4, \text{g}) - 4\Delta G_f^\circ(\text{UC, s}) = -RT \ln (P_U^4 P_{C_4}) \quad (7)$$

By using the free energy of formation values for UC(s) and U(g) from the report of Tetenbaum and coworkers¹ and for C₄(g) from the JANAF tables, the following equation was derived for partial pressure of C₄(g) over UC(s)

$$\log P_{C_4} = 12.23 - \frac{63800}{T} \quad (8)$$

Values calculated from Eq. 8 are given along with the recommended low-temperature data of Tetenbaum and coworkers in Table 8.

By fitting the standard form of the vapor pressure versus temperature relation to the total pressure data given in Table 8, the following equation was obtained:

$$\log P_T = 1.3681 - \frac{26993}{T} + 1.0995 \log T \quad (9)$$

(P_T in atm, T in K)

Beyond the melting point, one must correct the equation representing the vapor pressure for the heat of fusion in order to extrapolate total pressure over UC into the liquid region. As mentioned earlier, according to P. Blackburn¹⁴, a reasonable approach is to correct the slope of the partial pressure curve of each species by an appropriate multiple of the heat of fusion of UC at the melting point and then to use the corrected equation to extrapolate beyond the melting point. The corrected equations for the liquid region are as follows:

$$\log P_U = 4.58 - \frac{25400}{T} \quad (10)$$

$$\log P_{UC_2} = 6.45 - \frac{36100}{T} \quad (11)$$

$$\log P_{C_1} = 7.44 - \frac{37500}{T} \quad (12)$$

TABLE 8. Low-Temperature Vapor Pressure Data of UC

Temp, K	P_U^a , atm	$P_{C_1}^a$, atm	$P_{C_2}^a$, atm	$P_{C_3}^a$, atm	P_{C_4} , atm	$P_{UC_2}^a$, atm	P_T^b = Total Pressure, atm
2000	3.16×10^{-9}	2.04×10^{-12}	8.51×10^{-15}	6.31×10^{-15}	2.25×10^{-20}	4.90×10^{-13}	3.16×10^{-9}
2100	1.47×10^{-8}	1.84×10^{-11}	1.21×10^{-13}	9.11×10^{-14}	7.41×10^{-19}	4.69×10^{-12}	1.47×10^{-8}
2200	5.93×10^{-8}	1.36×10^{-10}	1.35×10^{-12}	1.03×10^{-12}	1.78×10^{-17}	3.65×10^{-11}	5.94×10^{-8}
2300	2.12×10^{-7}	8.42×10^{-10}	1.22×10^{-11}	9.46×10^{-12}	3.23×10^{-16}	2.38×10^{-10}	2.13×10^{-7}
2400	6.81×10^{-7}	4.43×10^{-9}	9.19×10^{-11}	7.22×10^{-11}	4.62×10^{-15}	1.33×10^{-9}	6.87×10^{-7}
2500	2.00×10^{-6}	2.09×10^{-8}	5.89×10^{-10}	4.68×10^{-10}	5.34×10^{-14}	6.46×10^{-9}	2.02×10^{-6}

^aValues are calculated based on the following equations given by Tetenbaum and coworkers¹:

$$\log P_U = 5.5 - \frac{28000}{T}$$

$$\log P_{C_1} = 8.36 - \frac{40100}{T}$$

$$\log P_{C_2} = 10.13 - \frac{48400}{T}$$

$$\log P_{C_3} = 10.15 - \frac{48700}{T}$$

$$\log P_{UC_2} = 8.29 - \frac{41200}{T}$$

$$^b P_T = P_U + P_{UC_2} + P_{C_1} + P_{C_2} + P_{C_3} + P_{C_4}$$

$$\log P_{C_2} = 8.29 - \frac{43300}{R} \quad (13)$$

$$\log P_{C_3} = 7.40 - \frac{41000}{T} \quad (14)$$

$$\log P_{C_4} = 8.56 - \frac{53600}{T} \quad (15)$$

Using these equations and the relation

$$\text{Total pressure} = P_T = P_U + P_{C_1} + P_{C_2} + P_{C_3} + P_{C_4} + P_{UC_2} \quad (16)$$

we calculated values for various partial pressures and total pressure as a function of temperature. These are given in Table 9.

Fitting the standard form of the vapor pressure versus temperature relation to the total pressure data given in Table 9, one obtains the following equation:

$$\log P_T = -24.0089 - \frac{18132}{T} + 7.5425 \log T \quad (17)$$

(P_T in atm, T in K)

From the values given in Table 9, the normal boiling point of UC was calculated to be 4814 K.

Because of the limited amount of data available on the vaporization behavior of well-characterized U-Pu-C compositions, calculations were made by Tetenbaum and coworkers¹ to estimate the total pressure of actinide and carbon-bearing species over the U-Pu-C system. Those calculations were based on the equations given by Potter *et al.*^{5, 15, 16} Necessary thermodynamic property values were obtained either from the IAEA Panel's assessment or JANAF tables. In a manner similar to that for UC, Tetenbaum and coworkers assumed the existence of Pu(g), U(g), UC₂(g), C₁(g), C₂(g), and C₃(g) carbon species. Because C₄(g) is expected to make a significant contribution at very high temperatures, we have added this to the list. The pertinent derived equations for C₄(g) are as follows:

$$9[UC_{1.5}, s]_{PuC_{1.5}} = C_4(g) + 8[UC, s]_{PuC} \quad (18)$$

$$8\Delta G_f^\circ(UC, s) + \Delta G_f^\circ(C_4, g) - 8\Delta G_f^\circ(UC_{1.5}, s) = -RT \ln \frac{P_{C_4} [UC]^8}{[UC_{1.5}]^8} \quad (19)$$

When the two-phase region is expressed by $U_{1-x_1}Pu_{x_1}C - U_{1-x_2}Pu_{x_2}C_{1.5}$, the above equation reduces to

$$RT \ln P_{C_4} = -8\Delta G_f^\circ(UC, s) - \Delta G_f^\circ(C_4, g) + 8\Delta G_f^\circ(UC_{1.5}, s) - 8RT \ln (1-x_1) + 8RT \ln (1-x_2) \quad (20)$$

By using the information provided by Tetenbaum and coworkers¹ on the free energies of formation and the values of x_1 and x_2 , and using ΔG_f° for C₄(g)

TABLE 9. Estimated Vapor Pressure Data of UC

Temp, K	P _U , atm	P _{C₁} , atm	P _{C₂} , atm	P _{C₃} , atm	P _{C₄} , atm	P _{UC₂} , atm	P _T = Total Pressure atm
2800	3.12x10 ⁻⁵	1.08x10 ⁻⁶	6.78x10 ⁻⁸	5.46x10 ⁻⁸	2.71x10 ⁻¹¹	3.65x10 ⁻⁷	3.27x10 ⁻⁵
3000	1.26x10 ⁻⁴	8.43x10 ⁻⁶	7.28x10 ⁻⁷	5.18x10 ⁻⁷	5.11x10 ⁻¹⁰	2.64x10 ⁻⁶	1.38x10 ⁻⁴
3500	2.05x10 ⁻³	5.18x10 ⁻⁴	8.39x10 ⁻⁵	4.67x10 ⁻⁵	1.81x10 ⁻⁷	1.38x10 ⁻⁴	2.80x10 ⁻³
4000	1.66x10 ⁻²	1.13x10 ⁻²	2.95x10 ⁻³	1.36x10 ⁻³	1.48x10 ⁻⁵	2.69x10 ⁻³	3.50x10 ⁻²
4500	8.45x10 ⁻²	0.13	4.70x10 ⁻²	1.88x10 ⁻²	4.55x10 ⁻⁴	2.71x10 ⁻²	0.30
5000	0.31	0.86	0.43	0.15	7.04x10 ⁻³	0.17	1.93
5500	0.90	4.12	2.64	0.86	6.63x10 ⁻²	0.78	9.36
6000	2.19	15.27	11.96	3.59	0.43	2.74	36.18
6500	4.64	46.26	42.93	12.05	2.09	7.95	115.92
7000	8.84	119.63	128.39	34.05	8.08	19.82	318.80
7500	15.44	272.53	331.77	83.74	26.15	43.74	773.38
8000	25.17	560.15	761.42	184.05	73.06	87.43	1691.3
8500	38.72	1057.8	1584.8	368.73	180.88	161.08	3392.0
9000	56.79	1861.2	3040.4	683.84	404.91	277.30	6324.5

from the JANAF tables, the following equation was obtained:

$$\log P_{C_4} = 10.16 - \frac{52200}{T} \quad (21)$$

Values calculated from Eq. 21, along with the recommended low-temperature data by Tetenbaum and coworkers,¹ are given in Table 10.

By fitting the standard form of vapor pressure versus temperature relation to the total pressure data given in Table 10, the following equation was obtained.

$$\log P_T = 2.9716 - \frac{18940}{T} + 0.17958 \log T \quad (22)$$

(P_T in atm, T in K)

Using the equations given above and following a similar approach to that which was used for UC to correct for the heat of fusion, one obtains the following equations in the liquid region:

$$\log P_{Pu} = 2.77 - \frac{16700}{T} \quad (23)$$

$$\log P_U = 5.05 - \frac{28500}{T} \quad (24)$$

$$\log P_{C_1} = 7.26 - \frac{35500}{T} \quad (25)$$

$$\log P_{C_2} = 8.01 - \frac{39400}{T} \quad (26)$$

$$\log P_{C_3} = 6.98 - \frac{35200}{T} \quad (27)$$

$$\log P_{C_4} = 6.70 - \frac{42700}{T} \quad (28)$$

$$\log P_{UC_2} = 6.58 - \frac{35000}{T} \quad (29)$$

Using the above equations along with an equation similar to equation 16, we calculated values for various partial pressure as a function of temperature. These are listed in Table 11.

Fitting the standard form of the vapor pressure versus temperature relation to the total pressure data given in Table 11, one obtains the following equation:

$$\log P_T = -38.8409 - \frac{6157.4}{T} + 10.985 \log T \quad (30)$$

(P_T in atm, T in K)

From the values given in Table 11, the normal boiling point of $(U_{0.8}Pu_{0.2})C$ was calculated to be 4558 K.

As in the case of the mixed carbide, measurements for vapor pressure of $PuC_{0.88}$ are not complete. Tetenbaum and coworkers¹ calculated plutonium and carbon pressures over the monocarbide phase from the appropriate values of

TABLE 10. Low-Temperature Vapor Pressure Data of (U_{0.8}Pu_{0.2})C

Temp, K	P _U ^a , atm	P _{Pu} ^a , atm	P _{C₁} ^a , atm	P _{C₂} ^a , atm	P _{C₃} ^a , atm	P _{C₄} , atm	P _{UC₂} ^a , atm	P _T ^b = Total Pressure, atm
1500	2.19x10 ⁻¹⁵	8.21x10 ⁻¹⁰	7.30x10 ⁻¹⁸	1.88x10 ⁻²⁰	2.07x10 ⁻¹⁹	2.18x10 ⁻²⁵	5.76x10 ⁻¹⁹	8.21x10 ⁻¹⁰
1600	4.23x10 ⁻¹⁴	5.12x10 ⁻⁹	2.77x10 ⁻¹⁶	1.31x10 ⁻¹⁸	1.21x10 ⁻¹⁷	3.27x10 ⁻²³	2.63x10 ⁻¹⁷	5.12x10 ⁻⁹
1700	5.77x10 ⁻¹³	2.58x10 ⁻⁸	6.84x10 ⁻¹⁵	5.51x10 ⁻¹⁷	4.37x10 ⁻¹⁶	2.72x10 ⁻²¹	7.65x10 ⁻¹⁶	2.58x10 ⁻⁸
1800	5.88x10 ⁻¹²	1.08x10 ⁻⁷	1.18x10 ⁻¹³	1.53x10 ⁻¹⁵	1.06x10 ⁻¹⁴	1.39x10 ⁻¹⁹	1.53x10 ⁻¹⁴	1.08x10 ⁻⁷
1900	4.70x10 ⁻¹¹	3.91x10 ⁻⁷	1.52x10 ⁻¹²	3.01x10 ⁻¹⁴	1.84x10 ⁻¹³	4.66x10 ⁻¹⁸	2.24x10 ⁻¹³	3.91x10 ⁻⁷
2000	3.05x10 ⁻¹⁰	1.24x10 ⁻⁶	1.51x10 ⁻¹¹	4.38x10 ⁻¹³	2.40x10 ⁻¹²	1.10x10 ⁻¹⁶	2.50x10 ⁻¹²	1.24x10 ⁻⁶
2100	1.66x10 ⁻⁹	3.54x10 ⁻⁶	1.21x10 ⁻¹⁰	4.95x10 ⁻¹²	2.46x10 ⁻¹¹	1.93x10 ⁻¹⁵	2.22x10 ⁻¹¹	3.54x10 ⁻⁶
2200	7.72x10 ⁻⁹	9.16x10 ⁻⁶	7.96x10 ⁻¹⁰	4.48x10 ⁻¹¹	2.03x10 ⁻¹⁰	2.61x10 ⁻¹⁴	1.61x10 ⁻¹⁰	9.17x10 ⁻⁶
2300	3.15x10 ⁻⁸	2.18x10 ⁻⁵	4.47x10 ⁻⁹	3.35x10 ⁻¹⁰	1.40x10 ⁻⁹	2.81x10 ⁻¹³	9.87x10 ⁻¹⁰	2.19x10 ⁻⁵
2400	1.14x10 ⁻⁷	4.84x10 ⁻⁵	2.17x10 ⁻⁸	2.12x10 ⁻⁹	8.20x10 ⁻⁹	2.49x10 ⁻¹²	5.20x10 ⁻⁹	4.85x10 ⁻⁵
2500	3.73x10 ⁻⁷	1.01x10 ⁻⁴	9.29x10 ⁻⁸	1.15x10 ⁻⁸	4.17x10 ⁻⁸	1.84x10 ⁻¹¹	2.40x10 ⁻⁸	1.01x10 ⁻⁴

^aValues are calculated based on the following equations given by Tetenbaum and Coworkers¹:

$$\log P_{Pu} = 3.63 - \frac{19100}{T}$$

$$\log P_U = 5.92 - \frac{30900}{T}$$

$$\log P_{C_1} = 8.13 - \frac{37900}{T}$$

$$\log P_{C_2} = 9.75 - \frac{44200}{T}$$

$$\log P_{C_3} = 9.58 - \frac{42400}{T}$$

$$\log P_{UC_2} = 8.31 - \frac{39800}{T}$$

$$P_T = P_{Pu} + P_U + P_{C_1} + P_{C_2} + P_{C_3} + P_{C_4} + P_{UC_2}$$

TABLE 11. Estimated Vapor Pressure Data of $(U_{0.8}Pu_{0.2})C$

Temp, K	P_{Pu} , atm	P_{Pu} , atm	P_{C1} , atm	P_{C2} , atm	P_{C3} , atm	P_{C4} , atm	P_{UC2} , atm	P_T = Total Pressure, atm
2800	7.61×10^{-6}	6.42×10^{-4}	3.79×10^{-6}	8.53×10^{-7}	2.50×10^{-6}	2.83×10^{-9}	1.15×10^{-9}	6.58×10^{-4}
3000	3.63×10^{-5}	1.60×10^{-3}	2.66×10^{-5}	7.41×10^{-6}	1.73×10^{-5}	2.94×10^{-8}	7.85×10^{-6}	1.70×10^{-3}
3500	8.24×10^{-4}	9.99×10^{-3}	1.30×10^{-3}	5.60×10^{-4}	8.21×10^{-4}	3.17×10^{-6}	3.66×10^{-4}	1.39×10^{-2}
4000	8.58×10^{-3}	3.94×10^{-2}	2.42×10^{-2}	1.43×10^{-2}	1.49×10^{-2}	1.06×10^{-4}	6.54×10^{-3}	0.11
4500	5.30×10^{-2}	0.11	0.23	0.18	0.14	1.62×10^{-3}	6.15×10^{-2}	0.79
5000	0.23	0.27	1.44	1.34	0.86	1.44×10^{-2}	0.37	4.52
5500	0.75	0.54	6.37	7.00	3.76	8.61×10^{-2}	1.60	20.11
6000	2.03	0.97	21.98	27.69	12.85	0.38	5.44	71.35
6500	4.70	1.59	62.70	88.70	36.35	1.35	15.32	210.70
7000	9.66	2.42	153.96	240.59	88.65	3.96	37.19	536.44
7500	18.05	3.50	335.40	571.26	191.96	10.11	80.21	1210.5
8000	31.17	4.82	662.90	1217.4	377.40	22.92	157.14	2473.8
8500	50.49	6.39	1209.3	2373.6	685.27	47.22	284.45	4656.7
9000	77.50	8.21	2063.4	4296.8	1164.5	89.76	482.03	8182.2

the free energies of formation for the condensed phase, Pu(g) , $\text{C}_1(\text{g})$, $\text{C}_2(\text{g})$, and $\text{C}_3(\text{g})$. Their recommended data with equations for temperature dependency of plutonium- and carbon-bearing species are given in Table 12.

TABLE 12. Low-Temperature Vapor Pressure Data of $\text{PuC}_{0.88}$ ^a

Temp, K	P_{Pu} , atm	P_{C_1} , atm	P_{C_2} , atm	P_{C_3} , atm	P_{T} = Total Pressure, atm
1000	2.10×10^{-14}	3.97×10^{-33}	2.82×10^{-40}	$< 10^{-40}$	2.10×10^{-14}
1100	7.87×10^{-13}	1.80×10^{-29}	7.58×10^{-36}	6.20×10^{-37}	7.87×10^{-13}
1200	1.61×10^{-11}	2.01×10^{-26}	3.72×10^{-32}	3.41×10^{-33}	1.61×10^{-11}
1300	2.08×10^{-10}	7.63×10^{-24}	4.93×10^{-29}	4.99×10^{-30}	2.08×10^{-10}
1400	1.86×10^{-9}	1.24×10^{-21}	2.34×10^{-26}	2.58×10^{-27}	1.86×10^{-9}
1500	1.24×10^{-8}	1.02×10^{-19}	4.89×10^{-24}	5.78×10^{-25}	1.24×10^{-8}
1600	6.55×10^{-8}	4.84×10^{-18}	5.24×10^{-22}	6.60×10^{-23}	6.55×10^{-8}
1700	2.84×10^{-7}	1.46×10^{-16}	3.24×10^{-20}	4.31×10^{-21}	2.84×10^{-7}
1800	1.05×10^{-6}	3.01×10^{-15}	1.27×10^{-18}	1.77×10^{-19}	1.05×10^{-6}

^aValues are based on:

$$\log P_{\text{Pu}} = 3.64 - \frac{17300}{T}$$

$$\log P_{\text{C}_1} = 7.83 - \frac{40200}{T}$$

$$\log P_{\text{C}_2} = 9.17 - \frac{48700}{T}$$

$$\log P_{\text{C}_3} = 8.68 - \frac{49400}{T}$$

Looking at the phase diagram of the Pu-C system given in Fig. 1, one realizes that in the absence of experimental work on single phase plutonium monocarbide, the vaporization behavior of PuC will be better approximated by the average properties calculated from the adjacent two-phase regions, i.e. $\text{Pu(l)} + \text{PuC}$ and $\text{PuC} + \text{Pu}_2\text{C}_3$. In the case of UC this problem did not arise since enough experimental work was done on the vaporization behavior of single phase UC. The carbon and uranium activities in single-phase UC are very well known. The low-temperature vapor-pressure data of Table 12, given by Tetenbaum *et al.*,¹ represented the boundary of PuC adjacent to the $\text{Pu(l)} + \text{PuC}$ side of the PuC single-phase region. If the approach that was used for UC and $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$ were to be applied to the data of Table 12, one would get unreasonably low values for various partial-pressure and total-pressure terms. We decided, therefore, that more reasonable values would be obtained by utilizing the properties of the two adjacent two phase regions.

Fig. 1. Phase Diagram of the Pu-C System.⁸

From the report of Tetenbaum *et al.*,¹ equations were also available for carbon pressures representing the two phase region corresponding to $\text{PuC} + \text{Pu}_2\text{C}_3$. Using this information in conjunction with the data of Table 12, equations were derived for the average carbon pressures in the single phase PuC region, which are as follows:

$$\log P_{C_1} = 8.63 - \frac{17300}{T} \quad (31)$$

$$\log P_{C_2} = 11.00 - \frac{48400}{T} \quad (32)$$

$$\log P_{C_3} = 11.60 - \frac{48600}{T} \quad (33)$$

Partial pressure of plutonium corresponding to these average carbon pressures was calculated based on the following equations:

$$0.88 \Delta G_f^\circ(\text{C}_1, \text{g}) + \Delta G_f^\circ(\text{Pu}, \text{g}) - \Delta G_f^\circ(\text{PuC}_{0.88}, \text{s}) = -RT \ln (P_{\text{Pu}} P_{\text{C}_1}^{0.88}) \quad (35)$$

By using the free energy of formation values for various species reported by Tetenbaum *et al.*,¹ and the C_1 pressure calculated with Eq. 31, the following equation was calculated

$$\log P_{Pu} = 3.88 - \frac{17300}{T} \quad (36)$$

With the use of equations 31, 32, 33, and 36, low-temperature pressures were recalculated; these are given in Table 13.

TABLE 13. Low-Temperature Vapor Pressure Data of $PuC_{0.88}$ (Calculated using average values derived from $Pu(l) + PuC$ and $PuC + Pu_2C_3$ phases)

Temp, K	P_{Pu} , atm	P_{C_1} , atm	P_{C_2} , atm	P_{C_3} , atm	P_T =Total Pressure atm
1000	3.66×10^{-14}	4.44×10^{-32}	4.38×10^{-38}	9.84×10^{-38}	3.66×10^{-14}
1100	1.37×10^{-12}	1.92×10^{-28}	1.09×10^{-33}	2.58×10^{-33}	1.37×10^{-12}
1200	2.81×10^{-11}	2.05×10^{-25}	5.02×10^{-30}	1.24×10^{-29}	2.81×10^{-11}
1300	3.63×10^{-10}	7.50×10^{-23}	6.31×10^{-27}	1.62×10^{-26}	3.63×10^{-10}
1400	3.25×10^{-9}	1.18×10^{-20}	2.86×10^{-24}	7.60×10^{-24}	3.25×10^{-9}
1500	2.17×10^{-8}	9.46×10^{-19}	5.75×10^{-22}	1.57×10^{-21}	2.17×10^{-8}
1600	1.14×10^{-7}	4.39×10^{-17}	5.95×10^{-20}	1.66×10^{-19}	1.14×10^{-7}
1700	4.95×10^{-7}	1.29×10^{-15}	3.57×10^{-18}	1.02×10^{-17}	4.95×10^{-7}
1800	1.82×10^{-6}	2.62×10^{-14}	1.36×10^{-16}	3.95×10^{-16}	1.82×10^{-6}

Comparing Tables 12 and 13, one can see that the partial pressure of Pu and the total pressure have changed by less than a factor of 2, but the partial pressures of the carbon species have increased by factors of 10 to 1000. Fitting the standard form of vapor pressure versus temperature relation to P_T values of Table 13, one obtains

$$\log P_T = 3.8819 - \frac{17319}{T} + 1.7284 \times 10^{-7} \log T \quad (37)$$

Using equations 31, 32, 33, and 36, and following the approach that was used for UC and $(U_{0.8}Pu_{0.2})C$ to correct for heat of fusion, one obtains the following equations in the liquid region.

$$\log P_{Pu} = 2.96 - \frac{15600}{T} \quad (38)$$

$$\log P_{C_1} = 7.71 - \frac{38300}{T} \quad (39)$$

$$\log P_{C_2} = 9.16 - \frac{44900}{T} \quad (40)$$

$$\log P_{C_3} = 8.84 - \frac{43400}{T} \quad (41)$$

With the above equations, various partial pressures and the total pressure of $\text{PuC}_{0.88}$ were calculated; these are given in Table 14.

The correlation of P_T with temperature was calculated as:

$$\log P_T = -65.0395 + \frac{4484.9}{T} + 17.505 \log T \quad (42)$$

TABLE 14. Estimated Vapor Pressure Data of $\text{PuC}_{0.88}$

Temp, K	$P_{\text{Pu}},$ atm	$P_{\text{C}_1},$ atm	$P_{\text{C}_2},$ atm	$P_{\text{C}_3},$ atm	P_T =Total Pressure atm
1900	5.68×10^{-6}	3.76×10^{-13}	3.33×10^{-15}	9.57×10^{-15}	5.68×10^{-6}
2000	1.46×10^{-5}	3.82×10^{-12}	5.06×10^{-14}	1.33×10^{-13}	1.46×10^{-5}
2500	5.31×10^{-4}	2.56×10^{-8}	1.57×10^{-9}	2.94×10^{-9}	5.31×10^{-4}
3000	5.81×10^{-3}	9.11×10^{-6}	1.55×10^{-6}	2.31×10^{-6}	5.83×10^{-3}
3500	3.22×10^{-2}	6.05×10^{-4}	2.13×10^{-4}	2.71×10^{-4}	3.32×10^{-2}
4000	0.12	1.41×10^{-2}	8.55×10^{-3}	9.64×10^{-3}	0.15
4500	0.31	0.16	0.15	0.16	0.78
5000	0.70	1.15	1.50	1.43	4.79
5500	1.34	5.71	9.86	8.83	25.74
6000	2.31	21.71	47.24	40.18	111.45
6500	3.66	67.19	177.88	144.85	393.58
7000	5.44	176.91	554.17	434.80	1171.3
7500	7.65	409.42	1483.8	1127.2	3028.0
8000	10.33	853.15	3512.6	2594.2	6970.2

From the data given in Table 14, the boiling point of $\text{PuC}_{0.88}$ was calculated to be 4564 K.

Thermal Conductivity of Carbide Fuels

Uranium carbide and similar NaCl-type compounds have many metallic characteristics. At high temperatures, heat transfer is mainly electronic and obeys the Wiedmann-Franz law, that is, the thermal conductivity is proportional to the product of temperature and electrical conductivity. At low temperature, a large phonon component will also be present. Lewis and Kerrisk⁴ have recently made a critical evaluation of the low-temperature thermal conductivity of carbide fuels. Their data were extrapolated beyond the melting point, using the approach described below.

No data on the thermal conductivity of molten carbide fuels are available. According to Turnbull,¹⁷ molten salts just above the melting point are generally regarded as having a quasi-crystalline structure, which indicates that heat is transferred mainly by electronic and vibrational mechanisms. Translational or diffusional motion is not important as is evidenced by the high ratio of thermal to mass diffusivity for liquids, which varies from about 70 to 100. For molten salts, the Lorenz ratio (thermal conductivity/electrical conductivity $\times T$) is not constant and is several hundred times the value for metals. Many empirical correlations are available in the literature to estimate the thermal conductivity of liquids. Gambill¹⁸ has compiled a few of these correlations; however, the values calculated by these empirical equations differ from the thermal conductivity of solid carbide fuels (in the vicinity of melting point) by a factor of 100 to 1000. Because of the inadequacy of these equations for application at or near the melting point, an empirical rule proposed by Turnbull¹⁷ for fused salt was used. According to this rule, the ratio of thermal conductivity of liquid to that of solid at the melting point is about 0.86 ± 0.13 . Beyond the melting point, the thermal conductivity of liquids, in general, show varied behavior. Often it either increases first and then levels off or it decreases continuously. In the case of carbide fuels, the behavior of thermal conductivity above melting is difficult to predict. In the absence of specific information, a constant value is recommended for liquid thermal conductivity. The low-temperature data for 100% theoretically dense UC, PuC, and $(U_{0.8}Pu_{0.2})C$ were calculated from the equations given by Lewis and Kerrisk⁴. These are given in Tables 15-17. These data and the empirical approach mentioned above was used to calculate values for the liquid thermal conductivity; these are given in Table 18.

Viscosity of Carbide Fuels

Viscosity data for molten fuel materials are very scarce. Tsai and Olander¹⁹ reported the viscosity of UO_2 at the melting point to be 8 ± 1 centipoise. Using several empirical rules and corresponding-state and Eyring-type calculations, Chasanov²⁰ recommended a mean value of about 5 centipoise at the melting point of mixed-carbide fuel. Bird, Stewart, and Lightfoot²¹ presented the following empirical equation for the viscosity of liquids:

$$\mu \sim \frac{Nh}{V} \exp (3.8 T_b/T) \quad (43)$$

where μ = viscosity, poise $[g/(cm)(sec)]$

N = Avogadro's number, $(6.023 \times 10^{23} \text{ molecules/mol})$

V = Molar volume at temperature T , (cc/mol)

T_b = normal boiling point, (K)

T = temperature, (K)

h = Planck's constant, $(6.624 \times 10^{-27} \text{ erg-sec or } g \cdot cm^2/sec)$

Andrade's²² equation, based on the quasi-crystalline structure of liquids, states that at the melting point:

$$\mu = 5.1 \times 10^{-4} (MT_m)^{1/2} / V_m^{2/3} \quad (44)$$

TABLE 15. Thermal Conductivity of 100% Theoretically Dense UC^a

Temp, °C	k cal/(sec)(cm)(°C)	Temp, °C	k cal/(sec)(cm)(°C)
300	0.0505	1500	0.0536
400	0.0503	1600	0.0539
500	0.0504	1700	0.0543
600	0.0506	1800	0.0547
700	0.0510	1900	0.0550
800	0.0511	2000	0.0554
900	0.0515	2100	0.0557
1000	0.0518	2200	0.0561
1100	0.0522	2300	0.0564
1200	0.0525	2400 ^b	0.0568
1300	0.0529	2507 ^b	0.0572
1400	0.0532		

^aRecommended by Lewis *et al.*⁴^bExtrapolation based on:

$$k = 0.0519 - 7.266 \times 10^{-6}T + 8.628 \times 10^{-9}T^2 ; \quad 50 < T^{\circ} \text{ C} < 700$$

$$k = 0.0483 + 3.537 \times 10^{-6}T ; \quad 700 < T^{\circ} \text{ C} \leq 2300$$

TABLE 16. Thermal Conductivity of 100% Theoretically Dense PuC.^a

Temp, °C	k, cal/(sec)(cm)(°C)	Temp, °C	k, cal/(sec)(cm)(°C)
300	0.0175	1000	0.0368
400	0.0185	1100	0.0419
500	0.0201	1200	0.0475
600	0.0223	1300	0.0537
700	0.0251	1400 ^b	0.0605
800	0.0284	1500 ^b	0.0679
900	0.0323	1602 ^b	0.0759

^aRecommended by Lewis *et al.*⁴^bExtrapolation based on:

$$k = 0.0178 - 9.656 \times 10^{-6}T + 2.868 \times 10^{-8}T^2 ; \quad 100 \leq T^{\circ} \text{ C} \leq 1300$$

TABLE 17. Thermal Conductivity of 100% Theoretically Dense (U_{0.8}Pu_{0.2})C.

Temp, °C	k, cal/(sec)(cm)(°C)	Temp, °C	k, cal/(sec)(cm)(°C)
300	0.0395	1500	0.0516
400	0.0395	1600	0.0523
500	0.0399	1700	0.0529
600	0.0414	1800	0.0534
700	0.0429	1900	0.0538
800	0.0443	2000	0.0542
900	0.0456	2100	0.0544
1000	0.0468	2200	0.0545
1100	0.0480	2300	0.0545
1200	0.0490	2400 ^b	0.0545
1300	0.0500	2485 ^b	0.0545
1400	0.0508		

^aRecommended by Lewis *et al.*⁴

^bExtrapolation based on:

$$k = 0.0418 - 1.3504 \times 10^{-5}T + 1.946 \times 10^{-8}T^2 \quad ; \quad 50 \leq T^{\circ}C \leq 500$$

$$k = 0.0305 + 2.082 \times 10^{-5}T - 4.493 \times 10^{-9}T^2 \quad ; \quad 500 \leq T^{\circ}C \leq 2300$$

TABLE 18. Thermal Conductivity of Carbide Fuels in the Liquid Region. (100% T.D.)^a

Compound	k _s @ T _m cal/(sec)(cm)(°C)	k _l @ T _m cal/(sec)(cm)(°C)
UC	0.0572	0.049
PuC	0.0759	0.065
(U _{0.8} Pu _{0.2})C	0.0545	0.047

^aIn this table, k_s represents the thermal conductivity of the solid state; k_l represents the thermal conductivity of the liquid state; and T_m represents the melting point.

where M = molecular weight

T_m = melting point, (K)

V_m = molar volume, (cc/mol)

Usually viscosity is related to the inverse of temperature by a logarithmic relation; however, in the present case, the use of a constant is recommended for the liquid region until more experimental data are available. By means of Eqs. 43 and 44 and other necessary data from this report, viscosity values were calculated at the melting point. These are listed in Table 19, along with the recommended values.

TABLE 19. Viscosity of Carbide Fuels

Compound	μ_m	μ_m	μ_m
	(centipoise) Calcd. from Eq. 43	(centipoise) Calcd. from Eq. 44	(centipoise) Recommended
UC	11.3	4.9	8
(U _{0.8} Pu _{0.2})C	8.3	4.9	7
PuC _{0.88}	169.6	4.1	4

The value of viscosity for PuC_{0.88}, calculated from Eq. 43, is unreasonably high. This may be due to the lower melting point of PuC_{0.88} in comparison with those of other carbides. Therefore, the use of this particular value is not recommended.

Emissivity of Carbide Fuels

In analyzing the interchange of radiant energy among various components of a reactor, their emission, reflection, and absorption characteristics must be known. In specifying the radiation properties, various degrees of detail are required, depending upon the nature of the application. For instance, the calculation of the radiant energy emitted into the entire hemispherical space above a surface element requires knowledge of the hemispherical emittance. Furthermore, depending on whether the calculation is to be performed over all wavelengths or in a specific wavelength range, either the total or the spectral (monochromatic) emittance is correspondingly required. The magnitude, the angular distribution, and the wavelength dependence of the radiation properties are all very sensitive to the condition of the surface. Surface condition includes such factors as roughness, oxide layers, and physical and chemical contamination. In the case of dielectric materials, the grain structure of the material may also affect the radiation properties. The qualitative description of a surface by terms such as smooth, polished, rough, and oxidized is generally insufficient to permit an accurate specification of the radiation properties.

Emissivity measurements on UC have been made by Grossman²³, and DeConinck²⁴. Grossman measured the normal spectral ($\epsilon_{0.65\mu\text{m}}$) as well as total normal emissivity (ϵ_t) of UC, whereas DeConinck²⁴ measured only the normal spectral emissivity. To evaluate the reliability of Grossman and DeConinck's data, we compare their data with the experimental data available on refractory carbides such as niobium, hafnium, tantalum, titanium, tungsten, vanadium, and zirconium. The data for total normal emittance for refractory carbides were taken from a report prepared by Southern Research Institute.²⁵ The data for normal spectral emittance (0.65 μm) for refractory carbides were taken from the compilation of Touloukian and DeWitt.²⁶ In Fig. 2, total normal emissivity is presented as a function of temperature.

Fig. 2. Total Normal Emittance of Refractory Carbides.

In Fig. 3, normal spectral emissivity is presented as a function of temperature. The total as well as spectral emissivity data for refractory carbides fell within a narrow bands and therefore, in Figs. 2 and 3 only the band is shown. The equations representing the mean value are (using the respective units given in Ref. 25 and 26):

$$\epsilon_t = -0.6403 + 1.5906 \times 10^{-3}t - 5.5607 \times 10^{-7}t^2 + 5.5741 \times 10^{-11}t^3$$

800°F < t < 4800°F; Average error = $\pm 40\%$ (45)

$$\epsilon_{0.65\mu\text{m}} = 0.5209 + 3.737 \times 10^{-4}T - 1.6747 \times 10^{-7}T^2 + 1.6901 \times 10^{-11}T^3$$

1000 K < T < 4200 K; Average error = $\pm 21\%$ (46)

Fig. 3. Normal Spectral Emittance of Refractory Carbides.

Grossman's²³ and DeConinck's²⁴ data are also shown in Figs. 2 and 3. Their values are lower than the lower limits given by Eqs. 45 and 46. Unreported measurements by D. Stahl²⁷ of the total emissivity of UC also indicated that higher values (about 0.8) were correct. In considering these uncertainties we tentatively recommend for UC, $\text{PuC}_{0.88}$, and $(\text{U}_{0.8}\text{Pu}_{0.2})\text{C}$, the emissivity values measured on UC by Grossman. It should be borne in mind, however, that these emissivity values may be too low. The recommended values are as follows:

$$\epsilon_t = 0.42 \pm 0.02 \quad ; \quad 1250 \text{ K} < T < 1980 \text{ K} \quad (47)$$

$$\epsilon_{0.65 \mu\text{m}} = 0.539 - 2 \times 10^{-5}T \quad ; \quad 1150 \text{ K} < T < 1890 \text{ K} \quad (48)$$

Conclusions

This report presents a recommended set of various thermophysical and transport property values for candidate advanced carbide fuels, which may reasonably be used until more experimental or theoretical data are available. Whenever possible, standard estimation procedures were used to estimate the values. In some cases, analogy with compounds of better known behavior was used to estimate the necessary values. In some cases, the estimated data were fitted to standard equations with little or no attempt made to optimize the form of equations. In making estimates of various property values, attempts were made to preserve internal consistency as well as consistency with other properties and also with the low-temperature data (that is, data

below the melting point). Even then, care should be exercised in using these data. This work has revealed a significant gap in the information for carbides in the liquid region. Both experimental work and additional analyses are needed to rectify this situation.

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