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

by

A. Sheth and L. Leibowitz
Chemical Engineering Division

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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois 60439

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October 1975

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

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ABSTRACT

By the use of available low-temperature data for various thermophysical and transport properties for uranium and plutonium nitrides, values above the melting point of density, heat capacity, enthalpy, vapor pressure, thermal conductivity, and viscosity were estimated. Sets of recommended values have been prepared for the compounds UN, PuN, and $(U_{0.8}Pu_{0.2})N$.

INTRODUCTION

In order 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 possess better neutronic properties and higher thermal conductivities than oxide fuels. This permits operation of fuel pins at higher linear power ratings and also improves the flexibility of design of the reactor system.

In reactor safety analyses, data are needed on many thermophysical and transport properties of the materials used in reactors. These data are required for temperatures from those of normal operation to 6000°C and above. Although adequate data are available at lower temperatures, values for the region of interest to the safety analyst are sparse, and efforts are being made to extrapolate low-temperature data to higher temperatures. The compounds being considered are UC, UN, PuC, PuN, (U,Pu)C, and (U,Pu)N. This report describes only the work on nitride fuels. A companion report describing the work on carbide fuels,[1] is being issued.

The properties being extrapolated or estimated by us are heat capacity, enthalpy, density, vapor pressure, thermal conductivity, and viscosity. Alexander and co-workers[2] discussed emissivity of nitride fuels; since their recommended values were in a good agreement with our recommended values for carbides,[1] we have not discussed this property here.

To obtain self-consistent property values from low-temperatures to the liquid region, we selected low-temperature data from recent critical evaluations by Alexander and co-workers.[2] In the few cases where no data were available, estimates were made on the basis of similarity or various correlations between related compounds. The estimated or extrapolated data are presented in tables and as equations developed by standard regression techniques. No attempt was made to determine the optimum form of an equation. The estimated sets of properties values, along with the recommended low-temperature data used, are given below.

Density of Nitride Fuels

The density of molten ceramic fuel is an important parameter in safety analyses. No reliable data on the density of liquid nitride fuels are available, and therefore, we have used appropriate literature data on density of solid nitride fuels and the approach that was applied to carbide fuels to derive an equation giving the temperature dependency of the density of the liquid phase. Alexander *et al.*[2] recently reviewed the density and thermal expansion data of nitride fuels in the solid state. They gave values for average linear thermal expansion coefficients for UN, PuN, and $(U_{0.8}Pu_{0.2})N$ as a function of temperature. Using their data, values were estimated for α_s (average linear thermal expansion coefficient in solid) for UN, PuN, and $(U_{0.8}Pu_{0.2})N$, from room temperature to these compounds' melting points. Fee and Johnson[3] recently evaluated melting point data for candidate advanced fuels. In the present report, their recommended values are used as melting points for nitride fuels.

In the absence of a reliable value for the volume change on melting ($\Delta V/V_s$) for nitride fuels, we took the value of 15%, which was estimated for PuN by Alexander *et al.*[2] As was done with the carbide fuels on the basis of data for NaCl, the ratio of linear thermal expansion coefficients for the solid and liquid states was assumed to be 0.5. By use of this ratio, the average values for the thermal expansion coefficient (α_ℓ) of the liquid nitrides from the melting point to a higher temperature, T, were estimated. Alexander *et al.*[2] have reported the theoretical density of individual nitride fuel, which we assumed to be equal to the density at 298 K. In Table 1, values for α_s, α_ℓ, T_m, volume change on melting, and theoretical density for UN, PuN, and $(U_{0.8}Pu_{0.2})N$ are listed.

TABLE 1. Thermal Expansion of Nitride Fuels

Compound	α_s, cm/(cm)(K)	α_ℓ, cm/(cm)(K)	T_m, melting point, K	$\Delta V/V_s$, %	Theoretical Density at 25°C, g/cm^3
UN	10.8×10^{-6}	21.6×10^{-6}	3035	15	14.32
PuN	19.5×10^{-6}	39.0×10^{-6}	2843	15	14.24
$(U_{0.8}Pu_{0.2})N$	11.4×10^{-6}	22.8×10^{-6}	3053	15	14.30

To derive the liquid density of nitride fuels, the following equation was used:

$$\rho_L = \frac{\rho_{298}}{1.15[1 + 3(T_m - 298)\alpha_s][1 + 3(T - T_m)\alpha_\ell]} \qquad T \geq T_m \qquad (1)$$

where

ρ_L = liquid density g/cm^3

ρ_{298} = density at 25°C, g/cm^3

T = Temperature, K

By substitution of the values for α_s, T_m, α_ℓ, and ρ_{298} from Table 1 and a further simplification, the following equations were obtained:

$$\rho_L[\mathrm{UN}] = \frac{14.24}{1 + 8.07 \times 10^{-5}\ T} \qquad T \geqslant T_m \tag{2}$$

$$\rho_L[\mathrm{PuN}] = \frac{16.15}{1 + 1.75 \times 10^{-4}\ T} \qquad T \geqslant T_m \tag{3}$$

$$\rho_L[(\mathrm{U}_{0.8}\mathrm{Pu}_{0.2})\mathrm{N}] = \frac{14.36}{1 + 8.65 \times 10^{-5}\ T} \qquad T \geqslant T_m \tag{4}$$

The above equations represent our best estimates of the liquid densities of nitride fuels; however, they should be used cautiously to calculate liquid density at temperatures far above the melting point. The value for α_ℓ may change with temperature, and the method of calculation (assuming the ratio of α_s to α_ℓ to be equal to 0.5) may not be valid at higher temperatures. Moreover, in real situations the chemical compositions of nitride fuels may change, giving rise to unknown effects on density. Density for each of these nitrides is given in Fig. 1 as a function of temperature.

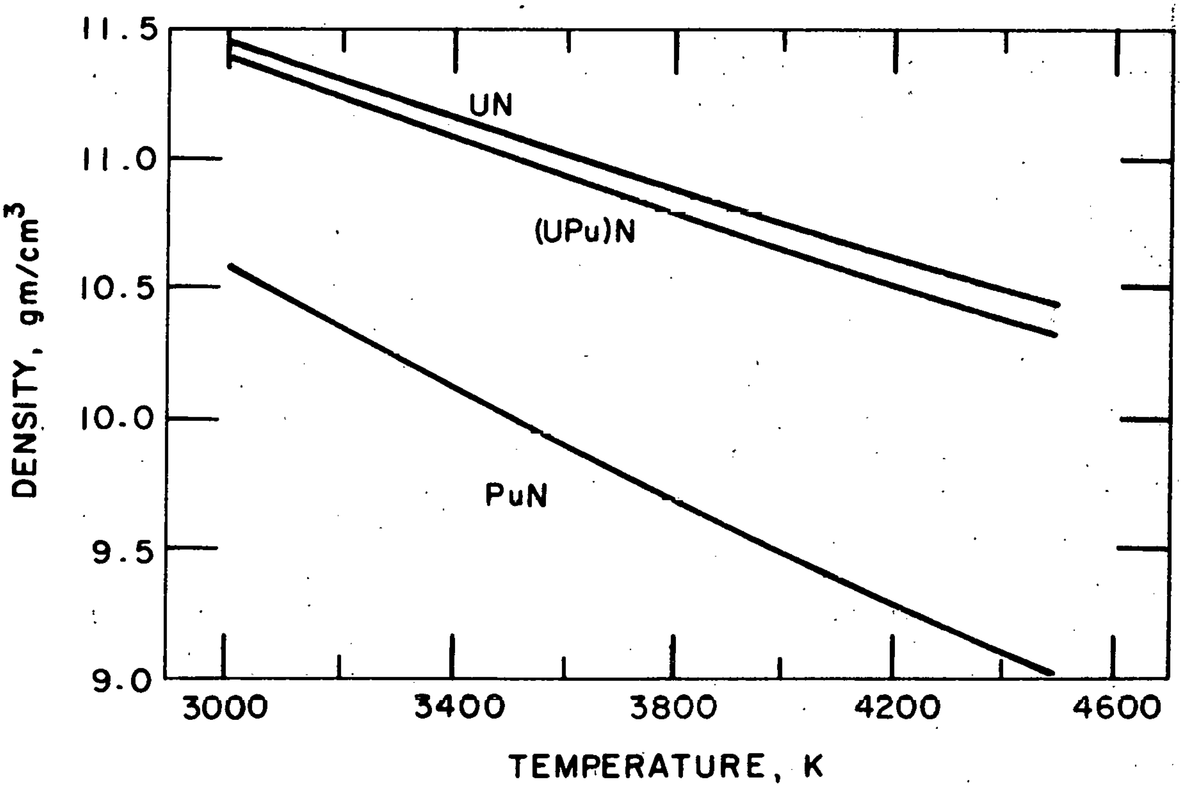

Fig. 1. Density of Nitride Fuels

Heat Capacity and Enthalpy of Nitride Fuels

The low-temperature heat capacity and enthalpy data for nitrides were taken from the report by Alexander *et al.*[2] 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.[3] To calculate heat capacity and enthalpy values up to the recommended melting points, we used the equations for C_p° for the solid region given by Alexander *et al.*[2] These equations may be inconsistent in that C_p° for mixed nitride is given as lower than both C_p° [UN] and C_p° [PuN], whereas it would be expected to be fairly close to the UN values. The differences are small, however, and for the present, we accept the values given in Ref. 2. The values for mixed nitride and for UN are based on measurements, and the PuN heat capacity values are estimated. Enthalpy values were calculated by integration, and the constraint was applied that $H_T^\circ - H_{298}^\circ$ is equal to zero at 298 K. The recommended low-temperature data with appropriate equations are presented here in Tables 2, 3, and 4 for UN, PuN, and $(U_{0.8}Pu_{0.2})N$, respectively.

To extrapolate the low-temperature enthalpy data above the melting points, corrections have to be made for the heats of fusion. A thermodynamic approach recommended by Winslow[4] was used to estimate liquid heat capacity. From basic thermodynamic considerations, one can derive the relation:

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

where

C_p = heat capacity at constant pressure

C_v = heat capacity at constant volume

γ = Grüneisen constant

β = volumetric expansion coefficient

T = Temperature

Winslow[5] has given the relationship between volumetric expansion coefficient, β, and defect density, temperature, and other lattice parameters for UO_2 and ThO_2. However, such information is not available for nitride fuels. Hence, it was decided to follow the same procedure as was used by us for carbide fuels, *viz.*, to calculate β from α_ℓ. Values of the Grüneisen constant, γ, 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 Winslow.[4] The value for the heat of fusion (ΔH_f) was estimated from the recommended melting point given by Fee and Johnson[3] and an estimated entropy of fusion equal to 4.2 eu, as was used for carbide fuels. Table 5 lists the values of γ, β, C_v, and ΔH_f used for nitride fuels in this study.

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, heat capacities increase very rapidly with increasing temperatures and

TABLE 2. Recommended Values for Heat Capacity and Enthalpy of UN 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	11.39	22	1800	16.44	21458
400	12.13	1202	1900	16.70	23115
500	12.62	2441	2000	16.97	24799
600	13.00	3722	2100	17.24	26509
700	13.34	5040	2200	17.51	28247
800	13.65	6390	2300	17.78	30011
900	13.95	7770	2400	18.04	31802
1000	14.24	9180	2500	18.31	33620
1100	14.52	10618	2600	18.58	35464
1200	14.80	12085	2700	18.84	37335
1300	15.08	13579	2800	19.11	39233
1400	15.35	15100	2900	19.38	41157
1500	15.62	16649	3000	19.64	43108
1600	15.90	18225	3035	19.74	43798
1700	16.17	19828			

[a] Recommended data of Alexander *et al.*[2]

Values are based on:

$$H_T^\circ - H_{298}^\circ = -3928.22 + 11.681T + 1.329 \times 10^{-3}T^2 + \frac{9.812 \times 10^4}{T}$$

$$C_p^\circ = 11.681 + 2.658 \times 10^{-3}T - \frac{9.812 \times 10^4}{T^2}$$

approach an infinite value at the critical temperature, T_c. The constant value of β used in Eq. 5 will give C_p values that are too low at temperatures close to the critical temperature. The critical temperatures for nitrides are not known; however, the same assumption was made as for carbides[1] ($T_c \cong 3.5T_m$), giving estimated critical temperatures for UN, PuN, and $(U_{0.8}Pu_{0.2})N$ of 10620, 9950, and 10690 K, respectively. Based on the above information, we used Eq. 5 and values given in Table 5 to calculate heat capacity and enthalpy data for UN (up to 8500 K), for PuN (up to 8000 K), and for $(U_{0.8}Pu_{0.2})N$ (up to 8600 K); these are given in Tables 6, 7, and 8, respectively.

TABLE 3. Recommended Values for Heat Capacity and Enthalpy of PuN 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	12.65	25	1700	16.15	20185
400	12.90	1303	1800	16.40	21813
500	13.15	2605	1900	16.65	23465
600	13.40	3933	2000	16.90	25143
700	13.65	5285	2100	17.15	26845
800	13.90	6663	2200	17.40	28573
900	14.15	8065	2300	17.65	30325
1000	14.40	9493	2400	17.90	32103
1100	14.65	10945	2500	18.15	33905
1200	14.90	12423	2600	18.40	35733
1300	15.15	13925	2700	18.65	37585
1400	15.40	15453	2800	18.90	39463
1500	15.65	17005	2843	19.01	40278
1600	15.90	18583			

[a] Recommended data of Alexander *et al.*[2]

Values are based on:

$$H_T^\circ - H_{298}^\circ = -3657.21 + 11.9T + 1.25 \times 10^{-3}T^2$$

$$C_p^\circ = 11.9 + 2.5 \times 10^{-3}T$$

By assuming ideal solution behavior, heat capacity data (and by integration of this, enthalpy data) for $(U_{0.8}Pu_{0.2})N$ were calculated, using data from Tables 6 and 7. The resulting values agreed within about ±5% with the values given in Table 8.

Vapor Pressure of Nitride Fuels

Consideration of the vapor pressure over nitride fuel material is complicated because of the various gaseous species that may exist over the solid and liquid. Moreover, Alexander *et al.*[2] report, "plutonium nitride can vaporize congruently while uranium nitride cannot; and this difference is caused by the much higher vapor pressure of plutonium as compared to uranium." The measurements in the low-temperature region are not complete, and considerable estimation and speculation are needed to derive vapor pressure values.[2] This makes high-temperature extrapolation and estimation particularly difficult.

TABLE 4. Recommended Values for Heat Capacity and Enthalpy of $(U_{0.8}Pu_{0.2})N$ 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	11.62	23	1800	15.52	20378
400	11.88	1198	1900	15.78	21943
500	12.14	2399	2000	16.04	23534
600	12.40	3626	2100	16.30	25151
700	12.66	4879	2200	16.56	26794
800	12.92	6158	2300	16.82	28463
900	13.18	7463	2400	17.08	30158
1000	13.44	8794	2500	17.34	31879
1100	13.70	10151	2600	17.60	33626
1200	13.96	11534	2700	17.86	35399
1300	14.22	12943	2800	18.12	37198
1400	14.48	14378	2900	18.38	39023
1500	14.74	15839	3000	18.64	40874
1600	15.00	17326	3053	18.78	41866
1700	15.26	18839			

[a] Recommended data of Alexander *et al.*[2]

Values are based on:

$$H_T^\circ - H_{298}^\circ = -3345.8 + 10.84T + 1.3 \times 10^{-3}T^2$$

$$C_p^\circ = 10.84 + 2.6 \times 10^{-3}T$$

The well-known law of corresponding states has been used to estimate high-temperature thermodynamic properties. This has been used by several investigators to estimate first the critical properties of reactor fuel materials and then their vapor pressures by using the generalized vapor pressure correlations given by Hougen, Watson, and Ragatz.[6] Miller[7] has given a detailed report on various empirical and semitheoretical approaches reported in the literature for estimation of vapor pressures.

Low-temperature vapor pressure data are very sensitive to the details of stoichiometry, pressure, and the materials of containment used for the experiment. For reactor safety analyses, these data must often be extrapolated six to nine orders of magnitude or higher in pressure than that experimentally measured. In such extrapolations, there is always a danger that vapor species which may be important at high temperatures

TABLE 5. Values of Parameters to Calculate Heat Capacity and Enthalpy at Very High Temperatures

Parameter	UN	PuN	$(U_{0.8}Pu_{0.2})N$
γ	2	2	2
β	6.48×10^{-5}	1.71×10^{-4}	6.84×10^{-5}
C_v[a]	6R	6R	6R
ΔH_f, kcal/mol	12.75	11.94	12.59

[a] R is the gas constant

may not be adequately represented by the lower temperature data. The approach that we have taken is to derive from basic thermodynamic data, equations for the pressures of individual vapor species, to extrapolate the equations, and to sum the pressures to give the total pressure above the condensed phase. For extrapolations above the melting point, adjustment is required for the heat of fusion. Following a suggestion of Blackburn,[8] we have corrected each partial pressure equation by a multiple of the heat of fusion (ΔH_f). The multiple depends on the number of condensed-phase molecules required to form the gaseous molecule, *e.g.*, ΔH_f for U(g), $2\Delta H_f$ for N_2(g), $2\Delta H_f$ for Pu_2(g), etc. This method was also applied to carbide fuels and gave reasonable results. Since the partial pressure of each species is extrapolated, equal consideration is given to each species. In this way, species less dominant at lower temperatures are given proper weight at higher temperatures.

In the recent critical evaluation of low-temperature vapor pressure data of UN, Alexander *et al.*[2] reported that the evaporation coefficient of uranium, α_V, lies between 0.3 and 1.0 and that the evaporation coefficient for nitrogen is much lower (being of the order of 0.01). Because of these values, Alexander and co-workers[2] showed that while UN appears to vaporize congruently at temperatures to 2000 K, this congruency may be the result of kinetics, not thermodynamics. Therefore, for practical purposes, vaporization of UN can be considered incongruent vaporization by preferential loss of nitrogen to form a two-phase system comprising nitrogen-saturated liquid uranium and uranium-saturated nonstoichiometric uranium mononitride. In such a case, increasing the temperature of a stoichiometric UN sample is certain to yield substoichiometric UN due to incongruent vaporization. Study of the phase equilibrium of UN is not yet complete, and beyond the melting point (or decomposition temperature) of UN, the system is not well understood. It was decided by us to use the data of Gingerich,[9] who investigated, by the Knudsen effusion technique in combination with a mass spectrometer, the two-phase region of U(ℓ) + UN(s). He expressed the overall vaporization reaction of uranium-saturated uranium mononitride

TABLE 6. Estimated Values of Heat Capacity and Enthalpy of UN in Liquid Region

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
3035	16.61	56548	4900	19.50	90218	6800	22.43	130047
3100	16.71	57631	5000	19.65	92175	6900	22.59	132298
3200	16.87	59310	5100	19.80	94148	7000	22.74	134564
3300	17.02	61005	5200	19.96	96136	7100	22.89	136846
3400	17.18	62715	5300	20.11	98140	7200	23.05	139143
3500	17.33	64440	5400	20.27	100159	7300	23.20	141456
3600	17.49	66181	5500	20.42	102193	7400	23.36	143784
3700	17.64	67937	5600	20.58	104243	7500	23.51	146127
3800	17.80	69709	5700	20.73	106309	7600	23.67	148486
3900	17.95	71496	5800	20.89	108389	7700	23.82	150861
4000	18.10	73299	5900	21.04	110486	7800	23.98	153250
4100	18.26	75117	6000	21.19	112597	7900	24.13	155656
4200	18.41	76951	6100	21.35	114724	8000	24.29	158076
4300	18.57	78800	6200	21.50	116867	8100	24.44	160513
4400	18.72	80664	6300	21.66	119025	8200	24.59	162964
4500	18.88	82544	6400	21.81	121199	8300	24.75	165431
4600	19.03	84439	6500	21.97	123388	8400	24.90	167914
4700	19.19	86350	6600	22.12	125592	8500	25.06	170412
4800	19.34	88277	6700	22.28	127812			

Values are based on:

$$C_p^\circ = 11.9232 + 1.5452 \times 10^{-3}T$$

$$H_T^\circ - H_{298}^\circ = 13244.5 + 11.9232T + 7.726 \times 10^{-4}T^2$$

TABLE 7. Estimated Values of Heat Capacity and Enthalpy of PuN in Liquid Region

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
2843	19.86	52218	4600	24.76	91410	6400	29.78	140493
2900	20.01	53354	4700	25.04	93900	6500	30.06	143485
3000	20.29	55370	4800	25.32	96417	6600	30.34	146504
3100	20.57	57413	4900	25.59	98963	6700	30.62	149552
3200	20.85	59484	5000	25.87	101536	6800	30.90	152628
3300	21.13	61583	5100	26.15	104137	6900	31.17	155731
3400	21.41	63710	5200	26.43	106766	7000	31.45	158862
3500	21.69	65865	5300	26.71	109424	7100	31.73	162022
3600	21.97	68048	5400	26.99	112109	7200	32.01	165209
3700	22.25	70258	5500	27.27	114821	7300	32.29	168424
3800	22.53	72497	5600	27.55	117562	7400	32.57	171667
3900	22.80	74763	5700	27.83	120331	7500	32.85	174938
4000	23.08	77058	5800	28.11	123127	7600	32.13	178237
4100	23.36	79380	5900	28.38	125952	7700	33.41	181563
4200	23.64	81730	6000	28.66	128804	7800	33.69	184918
4300	23.92	84108	6100	28.94	131685	7900	33.96	188300
4400	24.20	86514	6200	29.22	134593	8000	34.24	191711
4500	24.48	88948	6300	29.50	137529			

Values are based on:

$$C_p^\circ = 11.9232 + 2.79 \times 10^{-3}T$$

$$H_T^\circ - H_{298}^\circ = 7045 + 11.9232T + 1.395 \times 10^{-3}T^2$$

TABLE 8. Estimated Values of Heat Capacity and Enthalpy of $(U_{0.8}Pu_{0.2})N$ in Liquid Region

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
3053	16.90	54456	4900	19.92	88458	6800	23.02	129242
3100	16.98	55252	5000	20.08	90458	6900	23.18	131551
3200	17.14	56958	5100	20.24	92474	7000	23.34	133877
3300	17.31	58681	5200	20.41	94506	7100	23.50	136219
3400	17.47	60420	5300	20.57	96555	7200	23.67	138578
3500	17.63	62175	5400	20.73	98620	7300	23.83	140953
3600	17.80	63946	5500	20.89	100701	7400	23.99	143344
3700	17.96	65734	5600	21.06	102798	7500	24.16	145752
3800	18.12	67538	5700	21.22	104912	7600	24.32	148175
3900	18.28	69358	5800	21.38	107043	7700	24.48	150615
4000	18.45	71194	5900	21.55	109189	7800	24.65	153072
4100	18.61	73047	6000	21.71	111352	7900	24.81	155545
4200	18.77	74917	6100	21.87	113531	8000	24.97	158034
4300	18.94	76802	6200	22.04	115726	8100	25.14	160539
4400	19.10	78704	6300	22.20	117938	8200	25.30	163061
4500	19.26	80622	6400	22.36	120166	8300	25.46	165599
4600	19.43	82557	6500	22.53	122411	8400	25.62	168153
4700	19.59	84507	6600	22.69	124671	8500	25.79	170724
4800	19.75	86475	6700	22.85	126948	8600	25.95	173310

Values are based on:

$$C_p^\circ = 11.9232 + 1.6311 \times 10^{-3}T$$

$$H_T^\circ - H_{298}^\circ = 10452.9 + 11.9232T + 8.1555 \times 10^{-4}T^2$$

by the expression

$$UN[UN_y(s)] = U(g) + 0.5N_2(g) \tag{6}$$

where y is a parameter designating the temperature dependence of the uranium-saturated solid UN phase which is in equilibrium with nitrogen-saturated liquid uranium. The corresponding partial reaction for the formation of 0.5 mole of $N_2(g)$ was expressed by the equation

$$UN[UN_y(s)] = UN[UN_h(\ell)] + 0.5N_2(g) \tag{7}$$

and the partial reaction for the formation of 1 mole of U(g) was given as:

$$U[UN_h(\ell)] = U(g) \tag{8}$$

In Eqs. 7 and 8, $UN_h(\ell)$ represented nitrogen-containing liquid uranium that is in equilibrium with nitrogen-deficient UN phase, where h is a temperature-dependent term. At lower temperatures, the reaction represented by Eq. 7 is dominant over the reaction represented by Eq. 8 and there is a continuous shift in the N/U atomic ratio toward lower values as vaporization proceeds. From his experimental work, Gingerich[9] gave the following equations representing partial pressures of U and N_2 in the two-phase region of $U(\ell) + UN(s)$:

$$\log P_U \text{ (atm)} = 5.825 - \frac{26854}{T} \tag{9}$$

$$\log P_{N_2} \text{ (atm)} = 8.904 - \frac{30464}{T} \tag{10}$$

In a number of experiments at Battelle,[10] gaseous UN was observed experimentally. In the absence of precise measurement of UN(g) species, Alexander and co-workers[2] made some estimates and suggested the following vapor pressure equation:

$$\log P_{UN} \text{ (atm)} = 8.39 - \frac{38200}{T} \tag{11}$$

A series of experiments were directed toward the observation of gaseous U_2 at Battelle Columbus Laboratory.[2] At no time was the dimer observed, even with uranium pressures as high as 10^{-4} atm in an effusion cell mass spectrometer. Based on this, Alexander *et al.*[2] suggested that gaseous U_2 is of little concern, even in safety analysis work at very high temperature.

From Eqs. 9, 10, and 11, the total pressure, P_T, was determined by extrapolating Gingerich's data up to the melting point of UN and summing all partial pressures (*i.e.*, $P_T = P_U + P_{N_2} + P_{UN}$) to obtain low-temperature (1900-3000 K) vapor pressure data. 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 yields the following equation:

$$\log P_T = 6.4351 - \frac{29695}{T} + 0.6386 \log T \tag{12}$$

(P_T in atm, T in K)

TABLE 9. Low-Temperature Vapor Pressure of UN

Temp, K	P_U, atm	P_{N_2}, atm	P_{UN}, atm	P_T = Total Pressure, atm
1900	$4.91x10^{-9}$	$7.42x10^{-8}$	$1.93x10^{-12}$	$7.91x10^{-8}$
2000	$2.50x10^{-8}$	$4.70x10^{-7}$	$1.95x10^{-11}$	$4.95x10^{-7}$
2100	$1.09x10^{-7}$	$2.50x10^{-6}$	$1.58x10^{-10}$	$2.61x10^{-6}$
2200	$4.16x10^{-7}$	$1.14x10^{-5}$	$1.06x10^{-9}$	$1.18x10^{-5}$
2300	$1.41x10^{-6}$	$4.56x10^{-5}$	$6.04x10^{-9}$	$4.70x10^{-5}$
2400	$4.32x10^{-6}$	$1.62x10^{-4}$	$2.97x10^{-8}$	$1.67x10^{-4}$
2500	$1.21x10^{-5}$	$5.23x10^{-4}$	$1.29x10^{-7}$	$5.35x10^{-4}$
2600	$3.14x10^{-5}$	$1.54x10^{-3}$	$4.99x10^{-7}$	$1.57x10^{-3}$
2700	$7.57x10^{-5}$	$4.18x10^{-3}$	$1.75x10^{-6}$	$4.26x10^{-3}$
2800	$1.72x10^{-4}$	$1.06x10^{-2}$	$5.59x10^{-6}$	$1.08x10^{-2}$
2900	$3.67x10^{-4}$	$2.51x10^{-2}$	$1.65x10^{-5}$	$2.55x10^{-2}$
3000	$7.48x10^{-4}$	$5.62x10^{-2}$	$4.54x10^{-5}$	$5.69x10^{-2}$

In the absence of low-temperature data for single-phase UN, the data from the two-phase study of Gingerich[9] were extrapolated to the liquid region, and represent a higher limit. Since with increasing temperature, the N/U atom ratio shifts towards lower values, substoichiometric UN will have lower vapor pressures than that calculated by mere extrapolation. In the absence of any information on how and to what extent this substoichiometry affects the results, it is recommended that the following data (the extrapolation to the liquid region) should be used with care.

At temperatures above the melting point, one must correct the equation representing the vapor pressure for the heat of fusion in order to extrapolate total pressure over UN into the liquid region. As mentioned above, according to Blackburn,[8] 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 UN 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:

$$\log P_U = 4.91 - \frac{24100}{T} \qquad (13)$$

$$\log P_{N_2} = 7.07 - \frac{24900}{T} \qquad (14)$$

$$\log P_{UN} = 7.47 - \frac{35400}{T} \qquad (15)$$

By use of the above equations and the relation

$$\text{Total pressure} = P_T = P_U + P_{N_2} + P_{UN} \tag{16}$$

we calculated values for various partial pressures and total pressures as a function of temperature. These are given in Table 10.

TABLE 10. Estimated Vapor Pressure of UN

Temp, K	P_U, atm	P_{N_2}, atm	P_{UN}, atm	P_T = Total Pressure, atm
3100	1.39×10^{-3}	0.11	1.12×10^{-4}	0.11
3500	1.07×10^{-2}	0.90	2.26×10^{-3}	0.92
4000	7.76×10^{-2}	7.00	4.15×10^{-2}	7.12
4500	0.36	34.39	0.40	35.15
5000	1.24	122.89	2.45	126.58
5500	3.40	348.39	10.79	362.58
6000	7.86	830.23	37.13	875.22
6500	16.00	1731.1	105.61	1852.7
7000	29.43	3249.7	258.74	3537.9
7500	49.88	5609.2	562.51	6221.6
8000	79.16	9043.3	1109.8	10232.0

The equation correlating total vapor pressure over UN with temperature in the liquid region is:

$$\log P_T = 4.9968 - \frac{24048}{T} + 0.51735 \log T \tag{17}$$

(P_T in atm, T in K)

From the values given in Table 10, the boiling point at standard conditions of UN was calculated to be 3520 K.

In their low-temperature evaluation of nitride fuels, Alexander and co-workers[2] reported that plutonium nitride can vaporize congruently, which required that the number of moles of plutonium vaporizing per unit time be exactly twice the molar rate of nitrogen vaporizing. Numerically, this means that $P_{Pu}/P_{N_2} = 5.8$. By the use of free energy of formation data for various species from the literature and the above constraint, partial pressures of plutonium and nitrogen were calculated. These partial pressures agreed with those calculated from the equations recommended by Sheth and Leibowitz[11] within a factor of two. Since the equations recommended by Sheth and Leibowitz[11] were arrived at by fitting the standard equations to the experimental data of Kent,[12] Battelle Memorial Institute,[13,14] and Rand,[15] it was decided to use the equations of Sheth and Leibowitz for extrapolation into the liquid region. For the partial pressures of plutonium and nitrogen these equations are as follows:

$$\log P_{Pu} = 5.9863 - \frac{21056}{T} \tag{18}$$

$$\log P_{N_2} = 5.2125 - \frac{20967}{T} \tag{19}$$

Alexander *et al.*[2] indicated that partial pressure of PuN(g) is of more significance than partial pressure of UN(g) at elevated temperatures. From their estimate of standard dissociation energy (D_0°) for PuN, they recommended the following equation for the partial pressure of PuN(g):

$$\log P_{PuN} = 8.30 - \frac{30600}{T} \tag{20}$$

Alexander and co-workers[2] also estimated the standard dissociation energy, D_0°, for Pu_2. According to them, at the melting point of PuN, the dimer concentration could reach a few percent of the pressure of monomeric plutonium. Hence, for equation-of-state calculations, they recommended the inclusion of Pu_2. From basic thermodynamic principles and estimated values for pertinent thermodynamic parameters, the following equation was derived for partial pressure of Pu_2 below the melting point of PuN:

$$\log P_{Pu_2} = 7.27 - \frac{30300}{T} \tag{21}$$

By the use of Eqs. 18, 19, 20, and 21, the partial pressures of various species over PuN were calculated from 1600 to 2800 K. Total pressure, P_T, was obtained by summing all partial pressure terms. For temperatures below the melting point, Table 11 gives partial pressures of various species, as well as total pressure over PuN.

By fitting the standard form of the vapor pressure versus temperature relation to the total pressure data given in Table 11, the following equation was obtained:

$$\log P_T = 2.7541 - \frac{20329}{T} + 0.89151 \log T \tag{22}$$

(P_T in atm, T in K)

By using Eqs. 18-21 and following a similar approach to that used for UN to correct for the heat of fusion, one obtains the following equations in the liquid region:

$$\log P_{Pu} = 5.07 - \frac{18400}{T} \tag{23}$$

$$\log P_{N_2} = 3.38 - \frac{15700}{T} \tag{24}$$

$$\log P_{PuN} = 7.38 - \frac{28000}{T} \tag{25}$$

$$\log P_{Pu_2} = 5.44 - \frac{25100}{T} \tag{26}$$

TABLE 11. Low-Temperature Vapor Pressure of PuN

Temp, K	P_{Pu}, atm	P_{N_2}, atm	P_{PuN}, atm	P_{Pu_2}, atm	P_T = Total Pressure, atm
1600	6.70×10^{-8}	1.28×10^{-8}	1.50×10^{-11}	2.05×10^{-12}	7.99×10^{-8}
1700	3.99×10^{-7}	7.57×10^{-8}	2.00×10^{-10}	2.68×10^{-11}	4.74×10^{-7}
1800	1.94×10^{-6}	3.67×10^{-7}	2.00×10^{-9}	2.62×10^{-10}	2.31×10^{-6}
1900	8.02×10^{-6}	1.50×10^{-6}	1.57×10^{-8}	2.02×10^{-9}	9.54×10^{-6}
2000	2.87×10^{-5}	5.36×10^{-6}	1.00×10^{-7}	1.27×10^{-8}	3.42×10^{-5}
2100	9.11×10^{-5}	1.69×10^{-5}	5.35×10^{-7}	6.71×10^{-8}	1.09×10^{-4}
2200	2.60×10^{-4}	4.81×10^{-5}	2.46×10^{-6}	3.04×10^{-7}	3.11×10^{-4}
2300	6.79×10^{-4}	1.25×10^{-4}	9.90×10^{-6}	1.21×10^{-6}	8.14×10^{-4}
2400	1.63×10^{-3}	2.99×10^{-4}	3.55×10^{-5}	4.29×10^{-6}	1.97×10^{-3}
2500	3.66×10^{-3}	6.69×10^{-4}	1.15×10^{-4}	1.37×10^{-5}	4.46×10^{-3}
2600	7.72×10^{-3}	1.41×10^{-3}	3.39×10^{-4}	4.02×10^{-5}	9.51×10^{-3}
2700	1.54×10^{-2}	2.80×10^{-3}	9.26×10^{-4}	1.09×10^{-4}	1.92×10^{-2}
2800	2.93×10^{-2}	5.30×10^{-3}	2.35×10^{-3}	2.74×10^{-4}	3.72×10^{-2}

By the use of the above equations along with an equation similar to Eq. 16, partial pressures were estimated for various species as a function of temperature. These are listed in Table 12. For the data given in Table 12, the correlation between total pressure and temperature at higher temperatures was obtained as:

$$\log P_T = -9.7188 - \frac{15234}{T} + 3.984 \log T \qquad (27)$$

(P_T in atm, T in K)

From the values given in Table 12, the normal boiling point of PuN was calculated to be 3509 K.

Measurements of the vapor pressure of $(U_{0.8}Pu_{0.2})N$ are not complete. Alexander and Ogden[16] performed weight loss effusion measurements on various compositions from 80 mol% PuN to 20 mol% PuN. At each composition, they report that the systems appeared to behave ideally. Later work,[17] including high-temperature mass spectrometry, indicated that UN-PuN but not U-Pu could be treated ideally. Sheth and Leibowitz[11] fitted standard equations to the experimental data of Alexander and Ogden,[16] which are as follows:

$$\log P_U = 7.6619 - \frac{30416}{T} \qquad (28)$$

$$\log P_{Pu} = 5.2505 - \frac{20893}{T} \qquad (29)$$

$$\log P_{N_2} = 4.5797 - \frac{21089}{T} \qquad (30)$$

According to Alexander and co-workers,[2] vaporization of $(U_{0.8}Pu_{0.2})N$ is characterized mainly by a preferential loss of PuN as elemental plutonium and molecular nitrogen. They did not believe that any species other than U, Pu, and N_2 is present. They considered it possible that UPu(g) is present; however, their estimates indicated that less UPu would be present than Pu_2. Hence, in this study, for mixed nitrides, only U, Pu, and N_2 species are considered.

TABLE 12. Estimated Vapor Pressure of PuN

Temp, K	P_{Pu}, atm	P_{N_2}, atm	P_{PuN}, atm	P_{Pu_2}, atm	P_T = Total Pressure, atm
2900	5.10×10^{-2}	8.84×10^{-3}	5.37×10^{-3}	5.96×10^{-4}	6.58×10^{-2}
3000	8.31×10^{-2}	1.34×10^{-2}	1.13×10^{-2}	1.16×10^{-3}	0.11
3500	0.63	7.54×10^{-2}	0.24	1.82×10^{-2}	0.96
4000	2.86	0.28	2.42	0.14	5.71
4500	9.32	0.75	14.52	0.72	25.31
5000	23.94	1.69	60.83	2.59	89.05
5500	51.83	3.26	196.35	7.41	258.85
6000	98.65	5.65	521.35	17.80	643.46
6500	170.06	8.99	1191.2	37.37	1407.6
7000	271.22	13.40	2418.7	70.55	2773.9
7500	406.44	18.92	4468.6	122.39	5016.3
8000	579.06	25.60	7645.8	198.18	8448.7

By use of Eqs. 28-30 and a similar approach to that used for UN and PuN to correct for heat of fusion, calculations were made to estimate partial pressures of various species over $(U_{0.8}Pu_{0.2})N$ in the liquid region. In comparison with UN and PuN, calculations indicated a much higher uranium partial pressure and a much lower plutonium partial pressure in the mixed nitride. If UN and PuN in mixed nitride behave ideally, even in the liquid region, partial pressures of uranium and plutonium should be of the same order or similar to those in pure UN and PuN. Since low-temperature experimental work is not complete, it was decided not to extrapolate Alexander and Ogden's[16] results into the liquid region. It was assumed that UN and PuN behave ideally in U-Pu-N, and total pressure over $(U_{0.8}Pu_{0.2})N$ was estimated, using the following equation:

$$P_T[(U_{0.8}Pu_{0.2})N] \approx 0.8\ P_T[UN] + 0.2\ P_T[PuN] \tag{31}$$

In Table 13, total pressure calculated using Eq. 31 is compared with total pressure obtained from Alexander and Ogden's[16] experimental data. From Table 13, one can see that the data based on the assumption of ideal behavior between UN and PuN agree very well with the experimental data of Alexander and Ogden.[16]

TABLE 13. Low-Temperature Vapor Pressure of $(U_{0.8}Pu_{0.2})N$

Temp, K	Total Pressure, atm (Calculated from ideal solution assumption)	Total Pressure, atm (obtained from Ref. 16)
1600	1.60×10^{-8}	1.75×10^{-8}
1700	9.57×10^{-8}	-
1800	4.71×10^{-7}	5.98×10^{-7}
1900	1.97×10^{-6}	-
2000	7.24×10^{-6}	7.84×10^{-6}
2100	2.38×10^{-5}	-
2200	7.17×10^{-5}	6.87×10^{-5}
2300	2.01×10^{-4}	-
2400	5.28×10^{-4}	3.72×10^{-4}
2500	1.32×10^{-3}	-
2600	3.16×10^{-3}	-
2700	7.25×10^{-3}	-
2800	1.60×10^{-2}	-

The calculated total pressure data of Table 13 were correlated with the temperature by the following relation:

$$\log P_T = -29.84 - \frac{13597}{T} + 9.5388 \log T \tag{32}$$

(P_T in atm, T in K)

Similarly, by use of Eq. 31 and the recommended liquid region data for UN and PuN given earlier, estimates of total pressure over $(U_{0.8}Pu_{0.2})N$ in the liquid region were made. These estimated values are in good agreement with the similar values for UN and PuN and are given in Table 14.

TABLE 14. Estimated Vapor Pressure of $(U_{0.8}Pu_{0.2})N$ in the Liquid Region

Temp, K	P_T = Total Pressure, atm	Temp, K	P_T = Total Pressure atm
3100	0.12	6000	828.87
3500	0.93	6500	1763.7
4000	6.83	7000	3385.1
4500	33.18	7500	5980.5
5000	119.07	8000	9875.5
5500	341.83		

Fitting the standard form of the vapor pressure versus temperature relation to the total pressure data of Table 14 gives the following equation:

$$\log P_T = 3.0073 - \frac{22886}{T} + 0.98591 \log T \qquad (33)$$

(P_T in atm, T in K).

From the values given in Table 14, at a total pressure of 1 atm, the boiling point of $(U_{0.8}Pu_{0.2})N$ was calculated to be 3519 K.

In Fig. 2, comparison is made between our calculated values for the total pressures of UN, PuN, and $(U_{0.8}Pu_{0.2})N$ as a function of inverse temperature in the entire temperature range.

Thermal Conductivity of Nitride Fuels

As do carbide fuels, nitride fuels have a NaCl-type crystal structure, and therefore, display many metallic characteristics. At high temperatures, heat transfer is mainly electronic, obeying the Wiedmann-Franz law; that is, thermal conductivity is proportional to the product of temperature and electrical conductivity. At low temperature, a large phonon component will also be present. Alexander *et al.*[2] recently made a critical evaluation of the low-temperature thermal conductivity of nitride fuels. The same approach as was used for carbide fuels,[1] was used here to extrapolate their data beyond the melting point. This approach was an empirical rule proposed by Turnbull,[18] which states that the ratio of thermal conductivity of liquid to that of solid at the melting point is about 0.86 ± 0.13. In the absence of any knowledge on how thermal conductivity of nitrides would vary as a function of temperature beyond the melting point, a constant value should be used for liquid thermal conductivity, as was done for carbide fuels.

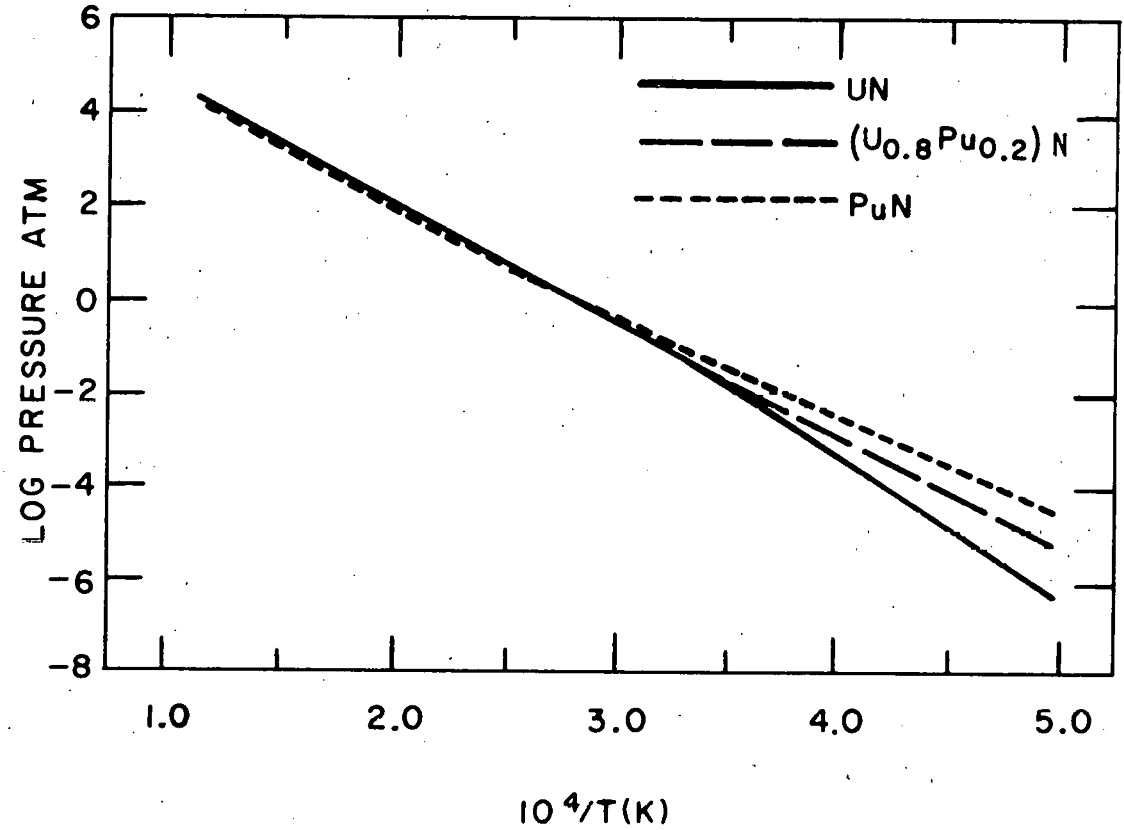

Fig. 2. Vapor Pressure of Nitride Fuels

Alexander and co-workers'[2] recommendations for thermal conductivity values of solid nitride fuels are given in Figs. 3-5 as a function of temperature. Since their data did not extend up to the recommended melting points of nitride fuels, we extrapolated their low-temperature data to determine thermal conductivity of solid nitride fuels at the melting point. Using this information and the empirical rule of Turnbull, we calculated thermal conductivity of liquid nitride fuels. Table 15 gives the thermal conductivity of nitride fuels, where K_s represents thermal conductivity in the solid state, K_ℓ represents thermal conductivity in the liquid state, and T_m represents the melting point.

Viscosity of Nitride Fuels

Viscosity data for molten fuel materials are vary scarce. From experimental measurements, Tsai and Olander[19] had reported a viscosity of 9 ± 1 cp for UO_2 at the melting point. Recently, Woodley[19A] measured viscosity of molten UO_2 with an oscillating cup viscometer and reported a median viscosity of 4.2 cp. Using empirical rules, Chasanov[20] recommended a mean value of about 5 cp at the melting point for mixed-carbide fuel. Sheth and Leibowitz[1] used empirical rules given by Bird, Stewart, and Lightfoot[21] and Andradae[22] to calculate viscosity of carbides of uranium and plutonium at the melting

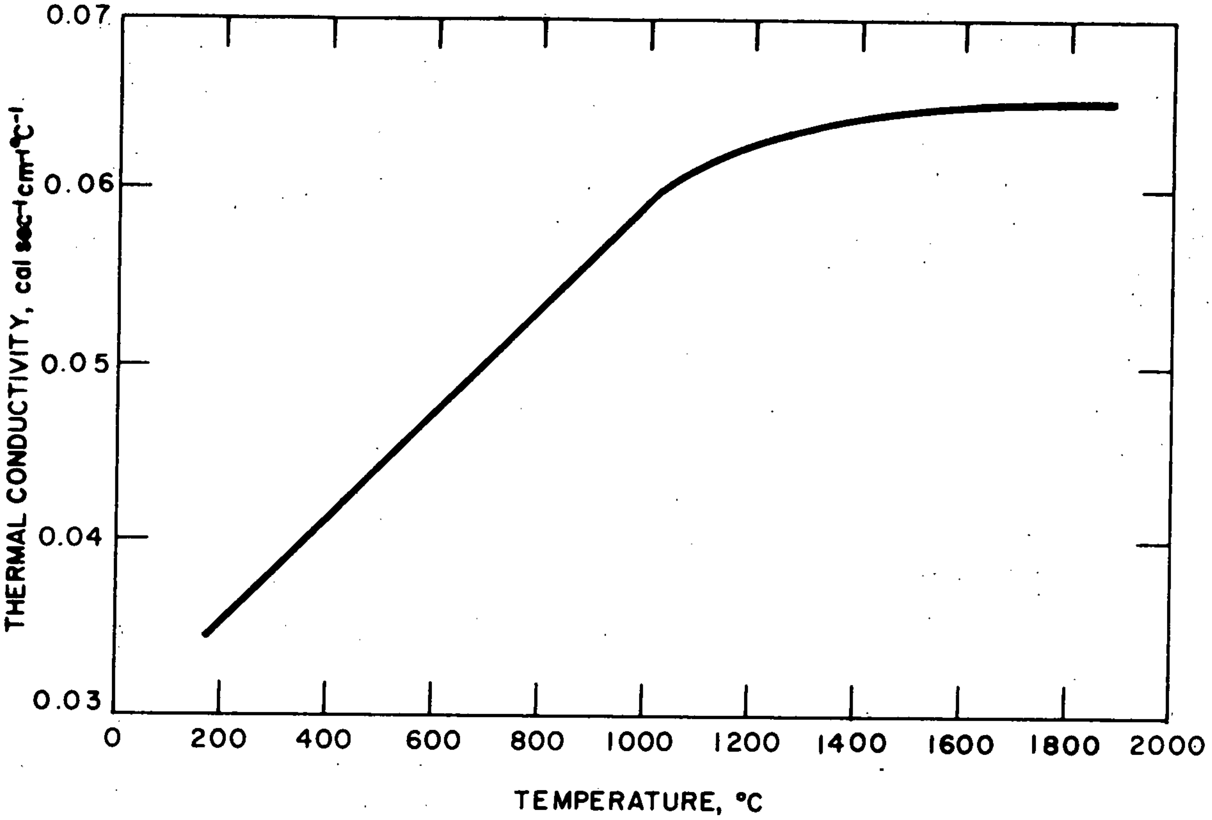

Fig. 3. Thermal Conductivity of UN[2] (Below Melting Point)

point. The calculated and recommended values by Sheth and Leibowitz[1] agreed satisfactorily with Tsai and Olander's value for molten UO_2 and Chasanov's value for mixed carbide. Hence, it was decided to use Sheth and Leibowitz's approach to calculate the viscosity of nitride fuels.

Bird, Stewart, and Lightfoot[21] presented the following empirical equation for viscosity of liquids:

$$\mu \approx \frac{Nh}{V} \exp\,(3.8\ T_b/T) \tag{34}$$

where

μ = viscosity, poise [g/(cm)(sec)]

N = Avogadro's number, (6.023×10^{23} molecules/mol)

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

T_b = normal boiling point, (K)

T = temperature, (K)

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

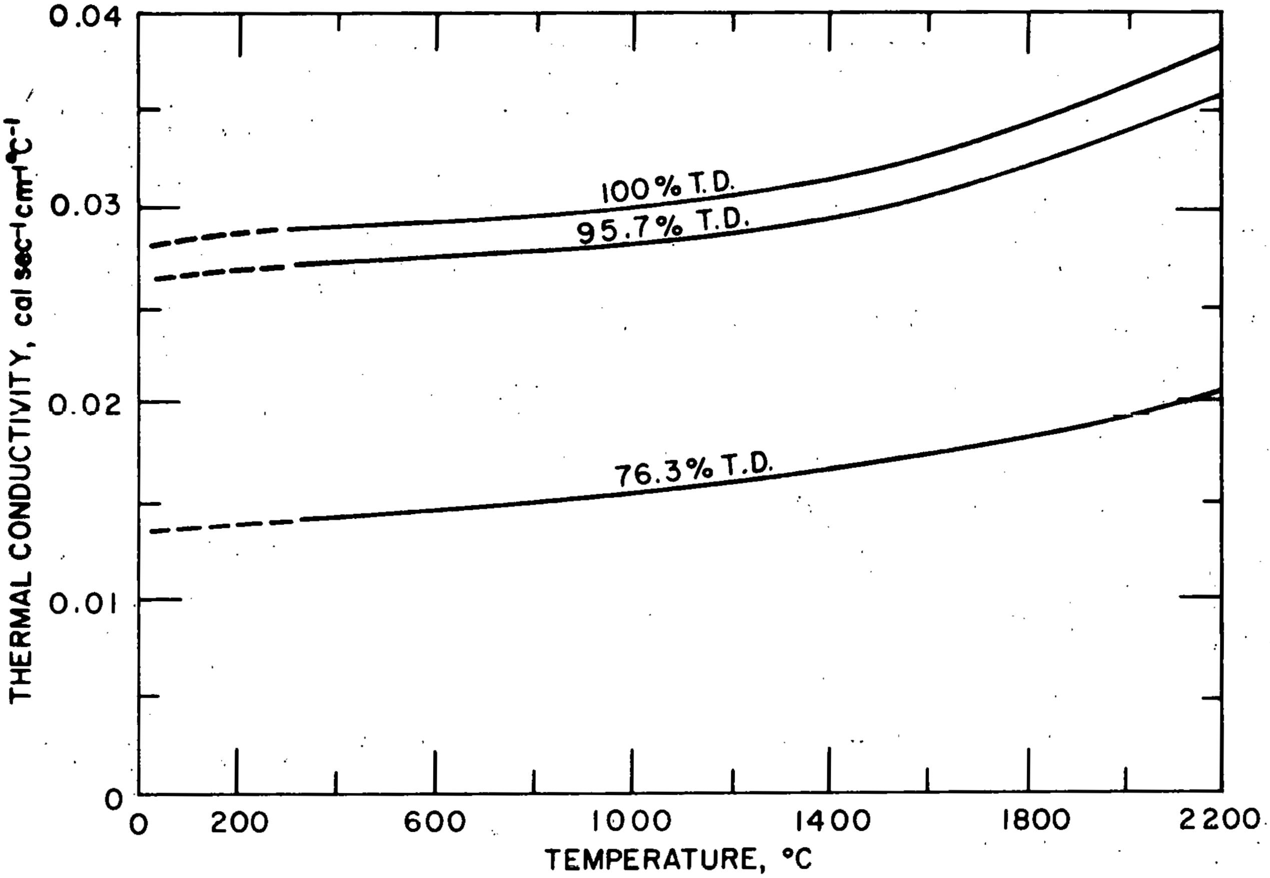

Fig. 4. Thermal Conductivity of PuN as a Function of Temperature and Density[2] (Below Melting Point)

Andradae's[22] equation, based on the quasi-crystalline structure of liquids, states that at the melting point:

$$\mu = 5.1 \times 10^{-4} \, (MT_m)^{1/2} / V_m^{\,2/3} \qquad (35)$$

where

M = molecular weight

T_m = melting point, (K)

V_m = molar volume, (cc/mol)

Usually, viscosity is related to an inverse of temperature by a logarithmic relation; however, in the present case the use of a constant value is recommended for the liquid region until more experimental data are available. By means of Eqs. 34 and 35 and other necessary data from this report, viscosity values at the melting point were calculated. These are listed in Table 16, along with the recommended values.

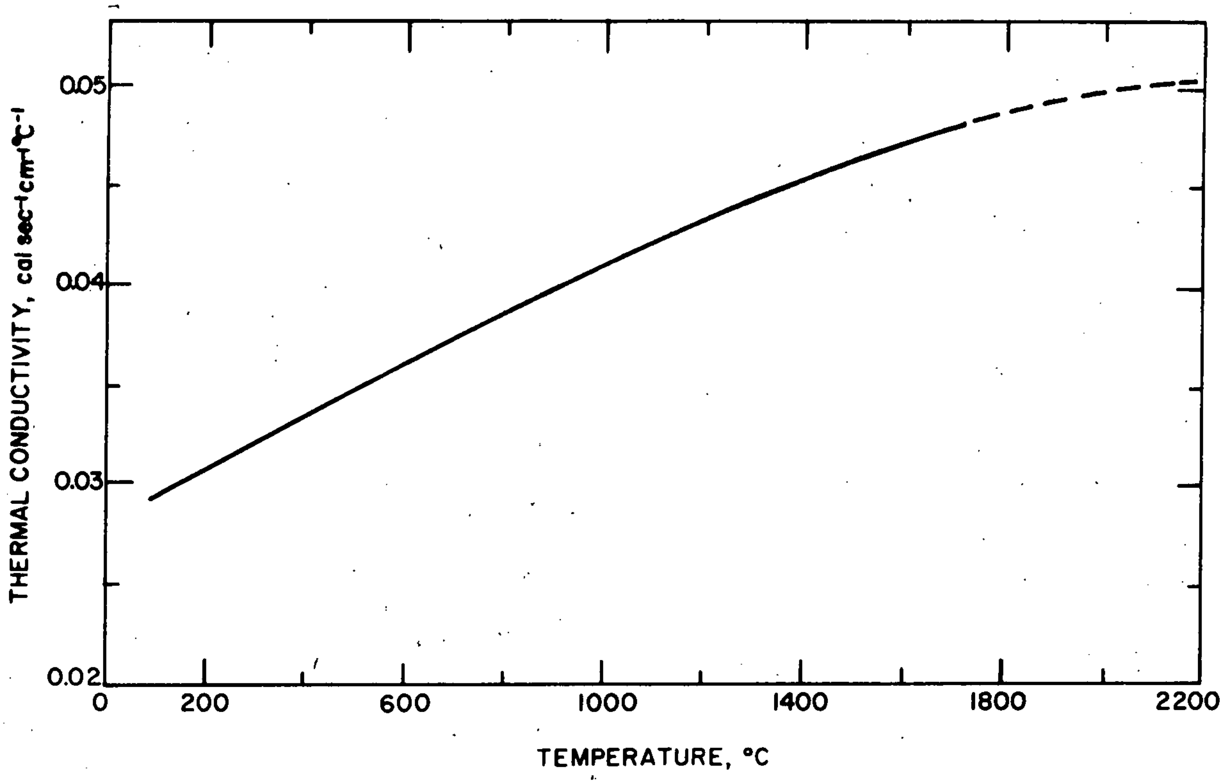

Fig. 5. Thermal Conductivity of $(U_{0.8}Pu_{0.2})N$ of 93% Theoretical Density[2] (Below Melting Point)

Conclusions

This report presents a set of preliminary results of various thermophysical and transport property values for candidate advanced nitride fuels which may reasonably be used until additional experimental or theoretical

TABLE 15. Thermal Conductivity of Nitride Fuels (100% Theoretical Density)

Compound	K_s at T_m, cal/(sec)(cm)(°C)	K_ℓ at T_m, cal/(sec)(cm)(°C)
UN	0.067	0.058
PuN	0.043	0.037
$(U_{0.8}Pu_{0.2})N$	0.057[a]	0.049

[a] Alexander *et al.*[2] gave values for mixed nitride of only 93% of theoretical density, but suggested multiplication by 1.1 to estimate thermal conductivity for 100% theoretically dense samples. The value reported here for mixed nitride is obtained using this suggestion.

TABLE 16. Viscosity of Nitride Fuels

Compound	μ_m (centipoise) Calcd. from Eq. 34	μ_m (centipoise) Calcd. from Eq. 35	μ_m (centipoise) Recommended
UN	1.5	5.7	4
PuN	1.9	5.3	4
$(U_{0.8}Pu_{0.2})N$	1.4	5.7	4

data are available. For the liquid region, almost all the property values were estimated, and an attempt was made to use estimation techniques similar to those used for carbide fuels. Recommended data are given in tabular form and in terms of standard equations. No attempt was made to optimize the form of the equations. Throughout, efforts were made to preserve internal consistency, consistency with other properties, and consistency with low-temperature data (that is, data below the melting point). Due care should be taken when using these data.

As was found in work on carbide fuels, this work has revealed a significant gap in the information for nitride fuels in the liquid region. Experimental work and additional analysis are needed.

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