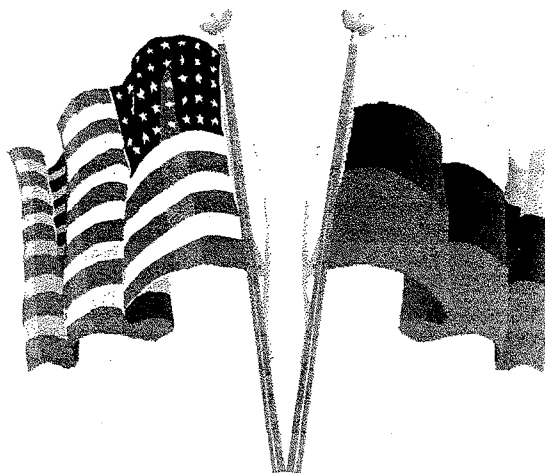


Thermophysical Properties of MOX and UO₂ Fuels Including the Effects of Irradiation

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Fissile Materials Disposition Program

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Engineering Technology Division

**THERMOPHYSICAL PROPERTIES OF MOX AND UO₂ FUELS
INCLUDING THE EFFECTS OF IRRADIATION**

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LIST OF NOMENCLATURE

A = constant in Eq. (6.3) (mk/W)
B = burnup (MWd/kgU or at. %)
C, C_i = constants (different units)
 C_p = heat capacity at constant pressure (J/kg·K)
 E_a = electron activation energy divided by the Boltzman constant [Eqs. (4.1) and (4.2)] (K)
FD = factor for effect of dissolved fission products
FM = factor for fuel porosity
FP = factor for effect of precipitated fission products
FR = factor for effect of radiation damage
H = enthalpy (J/kg)
L = length for expansion (m)
O/M = oxygen-to-metal (ratio)
 p = porosity $(\rho_{TD} - \rho)/\rho_{TD}$
TLTEC = true linear thermal expansion coefficient (1/K)
 $t = T/1000$ (K)
T = temperature (K)
 x = deviation from stoichiometry ($2 - O/M$)
 y = mole fraction of PuO_2 in MOX
 $\alpha(T)$ = coefficient of expansion (1/K)
 θ = Einstein temperature [Eqs. (4.1) and (4.2)] (K)
 ω = variable used in Eq. (6.4)
 ρ = density (kg/m³)
 Δ = increment (different units)
 λ = thermal conductivity

Subscripts

a = activation
L = liquidus
 ℓ = liquid
m = melting
P = pressure
o = unirradiated, fully dense fuel
s = solid
S = solidus
TD = theoretical density (for fully dense fuel with zero porosity)
v = volume

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ABSTRACT

Available open literature on thermophysical properties of both MOX and UO₂ fuels has been reviewed, and the best set of thermal properties has been selected. The properties reviewed are solidus and liquidus temperatures of the uranium-plutonium dioxide system (melting temperature), thermal expansion, density, heat of fusion, enthalpy, specific heat, and thermal conductivity. Only fuel properties are studied in this report. The selected properties will be used in thermal-hydraulic codes to study design basis accidents. The majority of the properties presented are for solid fuel.

1. INTRODUCTION

The excess weapons-grade plutonium available in the Russian Federation (R.F.) and the United States will be mainly disposed of by burning it as mixed-oxide (MOX) fuel in existing reactors. These reactors will be primarily VVER-1000s in the Russian Federation and pressurized-water reactors (PWRs) in the United States.

Safety analyses are required by the national regulatory bodies to prove that MOX fuel can be burned safely in these reactors. These safety analyses require calculations with safety codes that need the appropriate thermophysical properties of the MOX fuel.

The MOX fuel contains between 3 and 5% PuO₂ blended with natural or depleted uranium (in the proportion of 95–97%). The PuO₂ replaces the enriched fraction of ²³⁵U oxide in the regular UO₂ fuel. The uranium and plutonium dioxides are isostructural because their atomic masses are very close (238 and 239), and they form solid solutions.

A considerable body of data and correlations in the open literature covers measurements of thermal properties of UO₂, PuO₂, and MOX fuels. The data on MOX fuel are not so extensive as those on UO₂ fuel; but nevertheless they are sufficient to select adequate properties for MOX fuel.

In this report, the available open literature on fuel properties has been searched and reviewed, and the best set of thermal properties for MOX and UO₂ fuels has been selected.

The properties reviewed are solidus and liquidus temperatures of the uranium-plutonium dioxide system (melting temperature), enthalpy (or heat) of fusion, thermal expansion and density, enthalpy and specific heat, and thermal conductivity. Only fuel properties are studied in this report. The selected properties will be used in thermal-hydraulic codes that study only design basis accidents. Thus, no fuel melting will take place, and the majority of the properties presented are only for solid fuel. Properties needed to study severe accidents will be covered in a separate report.

For each property, the variables that influence the property are described, followed by a review of the available data and correlations. Variables considered are fuel composition, temperature (T), porosity (p) or fraction of the theoretical density (TD), burnup (B), and

oxygen-to-metal (O/M) ratio or deviation from stoichiometry ($x = O/M - 2$). It is expected that the use of these variables can accommodate fuel property variations due to different fuel manufacturing processes.

After the review, recommended values or correlations for both MOX and UO_2 fuels are presented, and the properties are given as graphs or tables of the property vs temperature with or without burnup (fresh fuel). These tables can be used readily as input for the properties required in thermal-hydraulic codes.

As expected, the properties of UO_2 and MOX fuels are very similar, but some differences were clearly identified. MOX fuel has a lower melting temperature and a lower thermal conductivity than UO_2 fuel, and burnup and/or deviation from stoichiometry reduce the thermal conductivity of both UO_2 and MOX fuels significantly.

This report was completed as a collaborative effort between the Kurchatov Institute (R.F.) and Oak Ridge National Laboratory (ORNL).

2. SOLIDUS AND LIQUIDUS TEMPERATURES OF URANIUM- PLUTONIUM DIOXIDE FUELS

2.1 INTRODUCTION

The melting temperature of fuel is a function of the composition (PuO_2 and UO_2 fraction), O/M ratio, and burnup (B). This temperature decreases with increasing PuO_2 mass or mole fraction (y), with increasing burnup, and with decreasing O/M ratio.

2.2 AVAILABLE DATA

The Adamson¹ equations provide the best fit to the data. The MATPRO² expressions are based on Lyon and Bailly³ data and yield the lowest values of the temperature, with the largest temperature reductions due to burnup. The Komatsu⁴ formula yields the highest temperatures; it has O/M ratio and burnup dependency. Tables 2.1 and 2.2 summarize the available data on melting temperatures.

2.3 RECOMMENDATIONS

Adamson¹ reported melting temperatures and equations that are the best fit to the experimental data. They are, therefore, recommended. This recommendation agrees with recent values reported by Fink⁸ for the International Nuclear Safety Center.

Table 2.1. Available data and formulas on melting temperatures of UO_2 and PuO_2

Reference	Pure UO_2	Pure PuO_2	Comments
Lyon and Bailly ³	$(3113 \pm 20) \text{ K}$	$(2663 \pm 20) \text{ K}$	No burnup effect
Aitken, Evans ⁵	$(3120 \pm 30) \text{ K}$	$(2718 \pm 35) \text{ K}$	No burnup effect
Latta, Fryxell ⁶	$(3138 \pm 15) \text{ K}$		No burnup effect
Adamson, Aitken ¹	$(3120 \pm 30) \text{ K}$	$(2701 \pm 35) \text{ K}$	Burnup effect: -0.4 K/MWd/kg
MATPRO ²	3113.15 K	2647 K	Burnup effect: -3.2 K/MWd/kg
Komatsu ⁴	$(3138 \pm 15) \text{ K}$	2718 K	Burnup effect: -0.7 K/MWd/kg

**Table 2.2. Available data and formulas on solidus
and liquidus temperatures of the system $\text{UO}_2\text{-PuO}_2$**

Reference	Experiment	Table	Correlation	Arguments
Lyon and Bailly ³	Yes	Yes	No	y
Aitken and Evans ⁵	Yes	Yes	No	y
Epstein ⁷	No	Yes	No	y
Adamson et al. ¹	No	No	Yes	y, B
Komatsu et al. ⁴	No	No	Yes	y, O/M, B
MATPRO, 1993 ²	No	No	Yes	y, B
Fink, 1999 ⁸	No	Yes	Yes	y

The recommended values for the melting temperatures of $\text{UO}_{2.00}$ and $\text{PuO}_{2.00}$ are

$$T_m(\text{UO}_{2.00}) = 3120 \pm 30 \text{ K} ,$$

$$T_m(\text{PuO}_{2.00}) = 2701 \pm 35 \text{ K} .$$

The recommended solidus and liquidus curves for stoichiometric fresh fuel solutions are based on detailed analyses of experimental data^{3,5} reported in Ref. 1. The liquidus [$T_L(y)$] and solidus [$T_S(y)$] temperatures (in K) are given by:¹

$$T_L(y) = 3120.0 - 388.1y - 30.4y^2 ,$$

$$T_S(y) = 3120.0 - 655.3y + 336.4y^2 - 99.9y^3 ,$$

where y is the mole fraction of PuO_2 . The solid-liquid phase diagram for the UO_2 - PuO_2 system is shown in Fig. 2.1. The enlarged portion of this figure for y less than 0.1 is shown in Fig. 2.2. The solidus and liquidus temperatures are tabulated in Table 2.3 as a function of the PuO_2 mole fraction.

For mole fractions of PuO_2 in fuel from 0 to 0.6, the two-standard-deviation estimated uncertainties are $\pm 35 \text{ K}$ for the solidus temperature and $\pm 55 \text{ K}$ for the liquidus temperature. For mole fractions of PuO_2 above 0.6, the two-standard-deviation uncertainties increase to $\pm 50 \text{ K}$ for the solidus and to $\pm 75 \text{ K}$ for the liquidus temperatures.

The effect of burnup on the solidus (melting) temperatures of MOX fuel and UO_2 was investigated in Refs. 1, 4, 9, and 10 (Fig. 2.3). We recommend a correction for burnup by decreasing the solidus temperature by 0.5 K/MWd/kgU for both MOX and UO_2 fuels.

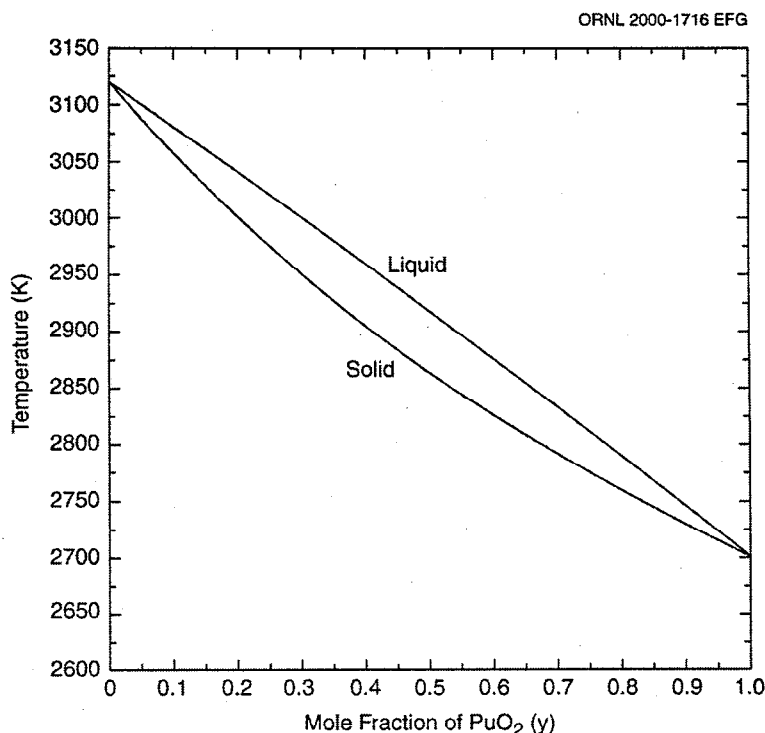


Fig. 2.1. Phase diagram of the UO_2 - PuO_2 system.¹

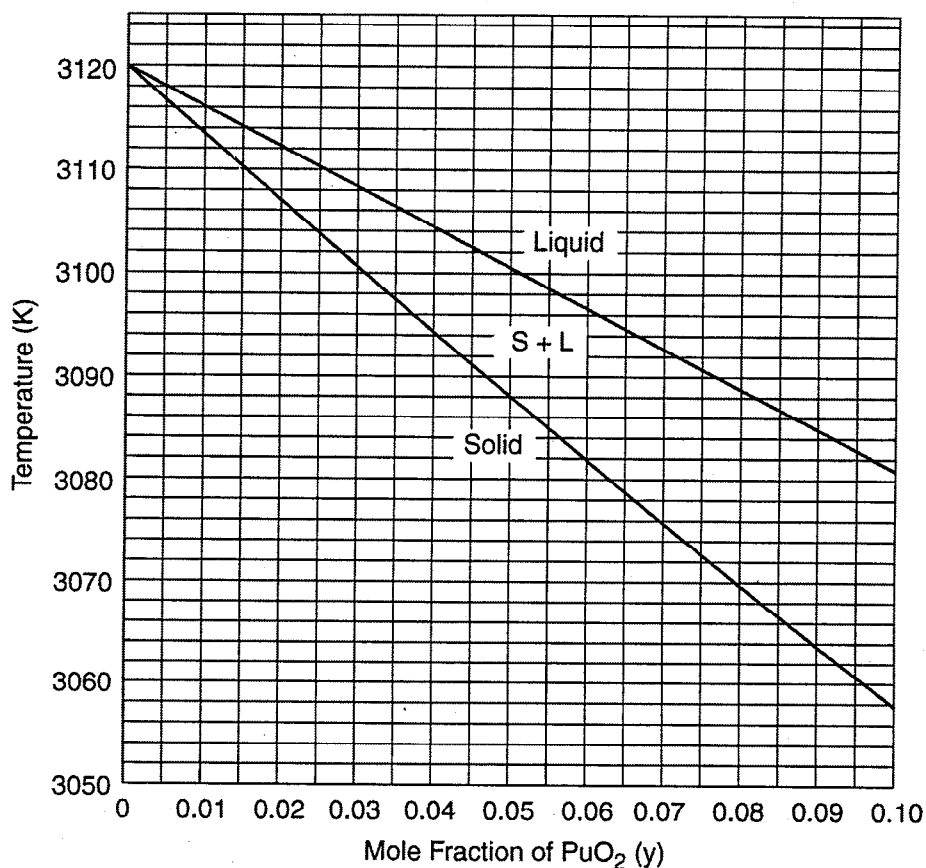


Fig. 2.2. Phase diagram of the UO_2 - PuO_2 system for PuO_2 mole fraction under 0.1.

Table 2.3. The solidus and liquidus temperatures of the UO_2 - PuO_2 system

Mole fraction of PuO_2	Solidus temperature (K)	Liquidus temperature (K)	Mole fraction of PuO_2	Solidus temperature (K)	Liquidus temperature (K)
0.00	3120.00	3120.00	0.10	3057.73	3080.89
0.01	3113.48	3116.12	0.11	3051.85	3076.94
0.02	3107.03	3112.23	0.12	3046.04	3072.99
0.03	3100.64	3108.33	0.13	3040.28	3069.03
0.04	3094.32	3104.43	0.14	3034.58	3065.07
0.05	3088.06	3100.52	0.15	3028.94	3061.10
0.06	3081.87	3096.60	0.16	3023.35	3057.13
0.07	3075.74	3092.68	0.17	3017.83	3053.14
0.08	3069.68	3088.76	0.18	3012.36	3049.16
0.09	3063.68	3084.82	0.19	3006.95	3045.16

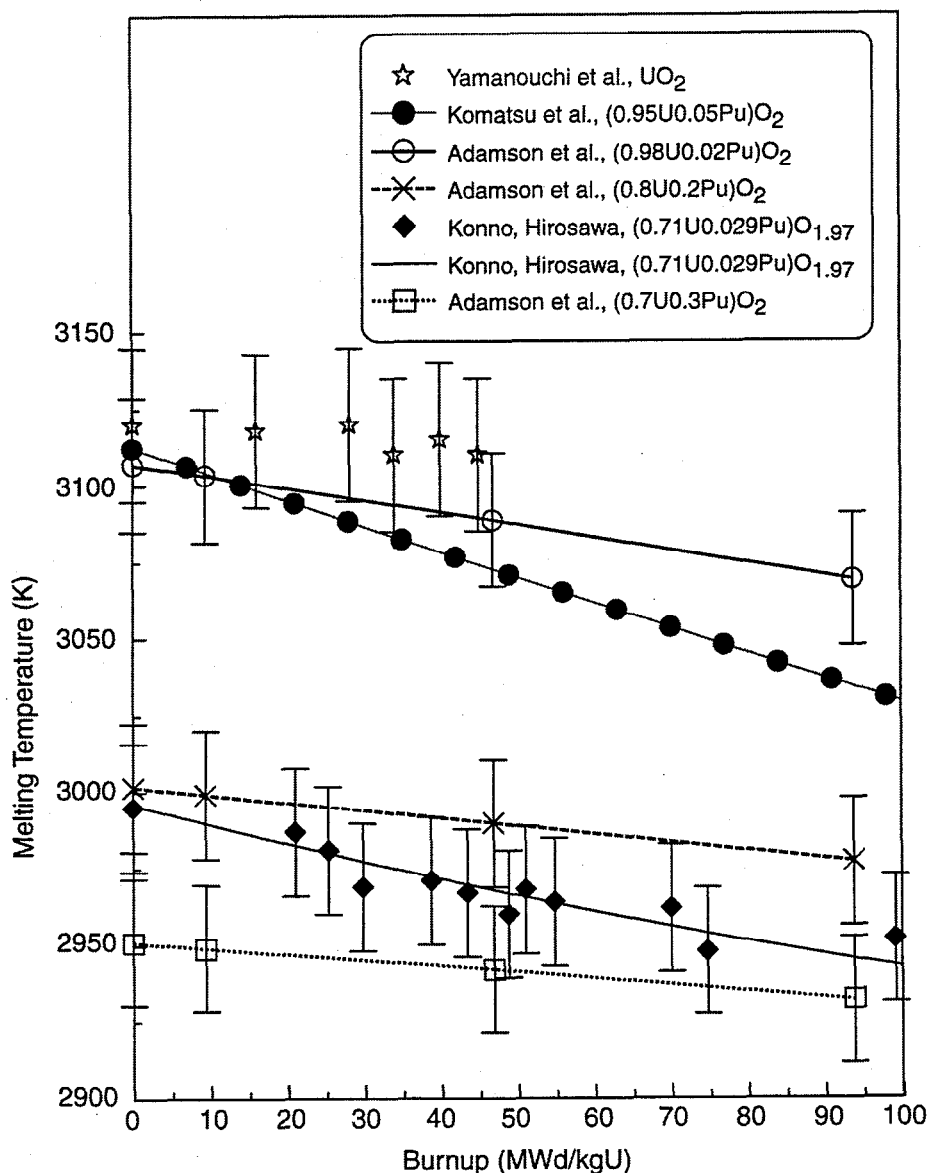


Fig. 2.3. The influence of burnup on the melting temperature of $\text{UO}_2\text{-PuO}_2$ fuel for various contents of PuO_2 .

2.4 REFERENCES

1. M. G. Adamson, E. A. Aitken, and R. W. Caputi, *J. Nucl. Mater.*, **130**, 245 (1985).
2. D. T. Hargman, Ed., *SCDAP/RELAP5/MOD3.1 Code Manual, Vol. 4: MATPRO—A Library of Materials Properties for Light-Water Reactor Accident Analysis*, USNRC Report NUREG/CR-6150 (EGGG-2720), 1993.
3. W. L. Lyon and W. E. Baily, *J. Nucl. Mater.*, **22**(3), 332 (1967).
4. J. Komatsu et al., *J. Nucl. Mater.*, **154**, 38 (1988).
5. E. A. Aitken and S. K. Evans, General Electric Report GEAP-5672, May 1968.
6. R. E. Latta and R. E. Fryxell, *J. Nucl. Mater.*, **35**, 195 (1970).

7. L. F. Epstein, *J. Nucl. Mater.*, **22**(3), 340 (1967).
8. J. K. Fink, INSC Material Properties Database, <http://www.insc.anl.gov/matprop> (1999).
9. S. Yamanouchi et al., *J. Nucl. Sci. Technology*, **25**, 538 (1988).
10. K. Konno and T. Hirosawa, *J. Nucl. Sci. Technology*, **35**, 494 (1998).

3. THERMAL EXPANSION AND DENSITY OF FUEL

3.1 INTRODUCTION

The fuel density ρ is a function of the fuel composition, temperature, fraction of theoretical density (TD) or porosity p [$p = (\rho_{TD} - \rho)/\rho_{TD}$], O/M ratio, and burnup (B).

The density of PuO_2 (or MOX fuel) is larger than the density of UO_2 . At 273 K, the density of UO_2 is $10970 \pm 70 \text{ kg/m}^3$, and the density of PuO_2 is $11460 \pm 80 \text{ kg/m}^3$ (for 100% TD). Therefore, the density increases with increasing PuO_2 content and decreases with increasing temperature.

The effect of the other variables (O/M ratio and burnup) is less important and usually does not need to be considered. The deviation from stoichiometry (O/M ratio different than 2) decreases the density. The burnup has two different effects: at low burnups the density increases by fuel densification; at higher burnups, the density decreases by fuel swelling.

3.2 AVAILABLE DATA AND CORRELATIONS

Table 3.1 shows the relevant correlations and data for the thermal expansion and density of the fuel.

Table 3.1. Thermal expansion and density of UO_2 and MOX fuels

Reference	UO_2 thermal expansion	MOX/ PuO_2 thermal expansion	$\rho(\text{UO}_2)$ at 273 K	$\rho(\text{PuO}_2)$ at 273 K
Martin ¹	X	X		
Tokar ²		X		
MATPRO ³	X	X	$10980 \pm 20 \text{ kg/m}^3$	
Fink ⁴	X		$10960 \pm 70 \text{ kg/m}^3$	
Benedict ⁵			$10970 \pm 70 \text{ kg/m}^3$	$11460 \pm 80 \text{ kg/m}^3$

3.3 RECOMMENDATION

The UO_2 , PuO_2 , and MOX fuels have very similar thermal expansions.¹⁻² The expression of Martin¹ can be used for UO_2 and MOX, with the corresponding basic density (at 273 K). The relative thermal expansion $[(L(T) - L(273))/L(273) = \Delta L(T)/L(273)]$, the true linear thermal expansion coefficient (TLTEC) $[\alpha_s(T) = 1/L(273) * (dL/dT)_p]$, and density $[\rho(T)]$ of solid stoichiometric UO_2 or MOX fuel in the temperature region of 273 to 923 K are recommended to be calculated by these correlations:¹

$$\Delta L(T)/L(273) = -2.66 * 10^{-3} + 9.802 * 10^{-6} * T - 2.705 * 10^{-10} * T^2 + 4.391 * 10^{-13} * T^3 ;$$

$$L(T) = L(273) * (9.9734 * 10^{-1} + 9.802 * 10^{-6} * T - 2.705 * 10^{-10} * T^2 + 4.391 * 10^{-13} * T^3) ;$$

$$\alpha_s(T) = 9.828 * 10^{-6} - 6.39 * 10^{-1} * T + 1.33 * 10^{-12} * T^2 - 1.757 * 10^{-17} * T^3 , (1/K) ;$$

$$\rho_s(T) = \rho_s(273)$$

$$* (9.9734 * 10^{-1} + 9.802 * 10^{-6} * T - 2.705 * 10^{-10} * T^2 + 4.391 * 10^{-13} * T^3)^{-3} , (\text{kg/m}^3) ;$$

in the temperature region of 923 K up to the UO_2 or MOX fuel melting temperature $T_{\text{ml}}[(\text{U}_{1-y}\text{Pu}_y)\text{O}_2]$:

$$\Delta L(T)/L(273) = -3.28 \times 10^{-3} + 1.179 \times 10^{-5} * T - 2.429 \times 10^{-9} * T^2 + 1.219 \times 10^{-12} * T^3 ;$$

$$L(T) = L(273) * (9.9672 \times 10^{-1} + 1.179 \times 10^{-5} * T - 2.429 \times 10^{-9} * T^2 + 1.219 \times 10^{-12} * T^3) ;$$

$$\alpha_s(T) = 1.1833 \times 10^{-5} - 5.013 \times 10^{-9} * T + 3.756 \times 10^{-12} * T^2 - 6.125 \times 10^{-17} * T^3 , (1/K) ;$$

$$\rho_s(T) = \rho_s(273)$$

$$* (9.9672 \times 10^{-1} + 1.179 \times 10^{-5} * T - 2.429 \times 10^{-9} * T^2 + 1.219 \times 10^{-12} * T^3)^{-3} , [\text{kg/m}^3] .$$

Densities $\rho_s(273)$ at 273 K of uranium and plutonium dioxides are 10,970 kg/m^3 and 11,460 kg/m^3 , respectively. Depending on mole fraction of PuO_2 , density of the solid solution of uranium and plutonium dioxides $(\text{U}_{1-y}\text{Pu}_y)\text{O}_2$ changes according to the linear law:

$$\rho_s(273) = 10,970 + 490y \text{ (in kg/m}^3\text{)} , \text{ for } (0 \leq y \leq 1) .$$

Errors in the values of the relative expansion are $\pm 2.6 \times 10^{-4}$, $\pm 4.4 \times 10^{-4}$, and $\pm 7 \times 10^{-4}$ in the temperature regions of 293–1273 K, 1273–2273 K, and 2273–2929 K, respectively. The corresponding errors in the values of the TLTEC are $\pm 0.11 \times 10^{-6}$, $\pm 0.22 \times 10^{-6}$, and $\pm 1.1 \times 10^{-6}$ 1/K. The recommended uncertainty in the density of UO_2 and MOX fuels is 1% for the entire temperature range. Recommended values of thermal expansion of stoichiometric MOX fuel are given in Table 3.2 and Figs. 3.1 and 3.2.

The parameters of thermal expansion of hypostoichiometric, $(\text{U}_{1-y}\text{Pu}_y)\text{O}_{2-x}$, solid MOX fuel are recommended to be calculated by the corresponding parameters of the stoichiometric fuel, multiplying them by a factor of $[1 + 3.9(\pm 0.9) * x]$. Analysis of the experimental data¹ showed that this recommendation is valid up to 1800 K; however, in the absence of high-temperature data, it can be used up to the melting temperature.

The recommended correlations for the coefficient of volume expansion and density of liquid UO_2 and MOX fuels as functions of temperature in the 3120–4500 K region are given by:⁶

$$\alpha_{v,l}(T) = 0.9285/[8860 - 0.9285(T - 3120)] , (1/K) ;$$

$$\rho_l(T) = 8860 - 0.9285 (T - 3120) , (\text{kg/m}^3) .$$

The relative uncertainties of these values are 4%.

These density correlations are for fully dense fuel with zero porosity. Generally, commercial fuel density ranges from 94–96% of TD prior to irradiation. During the early phases of irradiation, commercial fuel will densify by approximately 1–2% with maximum densification occurring by 5–15 MWd/kg burnup.⁷ Fuel also swells, due to progressive buildups of fission products, at a rate of 0.7–1.0% $\Delta v/v$ per 10 MWd/kg (Ref. 8). Maximum fuel density occurs between 5–15 GWd/MT burnup; thereafter, the fuel density decreases proportionally with increasing burnup.

Table 3.2. Parameters of thermal expansion of stoichiometric MOX fuel and density of UO_{2.00} as a function of temperature

Temperature (K)	Relative thermal expansion $\Delta L(T)/L(273)$	Average linear thermal expansion coefficient (1/K)	TLTEC (1/K)	Density of UO _{2.00} (kg/m ³)
3.0000E+02	2.6811E-04	9.9300E-06	9.7555E-06	1.0961E+04
4.0000E+02	1.2456E-03	9.8081E-06	9.7841E-06	1.0929E+04
5.0000E+02	2.2283E-03	9.8161E-06	9.8388E-06	1.0897E+04
6.0000E+02	3.2187E-03	9.8430E-06	9.9196E-06	1.0865E+04
7.0000E+02	4.2195E-03	9.8817E-06	1.0026E-05	1.0832E+04
8.0000E+02	5.2333E-03	9.9304E-06	1.0159E-05	1.0800E+04
9.0000E+02	6.2628E-03	9.9885E-06	1.0317E-05	1.0766E+04
1.0000E+03	7.3001E-03	1.0041E-05	1.0515E-05	1.0733E+04
1.1000E+03	8.3725E-03	1.0124E-05	1.0782E-05	1.0699E+04
1.2000E+03	9.4768E-03	1.0223E-05	1.1120E-05	1.0664E+04
1.3000E+03	1.0620E-02	1.0341E-05	1.1529E-05	1.0628E+04
1.4000E+03	1.1810E-02	1.0479E-05	1.2008E-05	1.0590E+04
1.5000E+03	1.3054E-02	1.0639E-05	1.2558E-05	1.0551E+04
1.6000E+03	1.4359E-02	1.0821E-05	1.3177E-05	1.0511E+04
1.7000E+03	1.5733E-02	1.1025E-05	1.3865E-05	1.0468E+04
1.8000E+03	1.7182E-02	1.1252E-05	1.4622E-05	1.0423E+04
1.9000E+03	1.8714E-02	1.1502E-05	1.5447E-05	1.0376E+04
2.0000E+03	2.0337E-02	1.1776E-05	1.6341E-05	1.0327E+04
2.1000E+03	2.2057E-02	1.2073E-05	1.7302E-05	1.0275E+04
2.2000E+03	2.3883E-02	1.2394E-05	1.8331E-05	1.0220E+04
2.3000E+03	2.5820E-02	1.2738E-05	1.9427E-05	1.0162E+04
2.4000E+03	2.7878E-02	1.3107E-05	2.0590E-05	1.0101E+04
2.5000E+03	3.0062E-02	1.3499E-05	2.1818E-05	1.0037E+04
2.6000E+03	3.2381E-02	1.3915E-05	2.3113E-05	9.9698E+03
2.7000E+03	3.4841E-02	1.4356E-05	2.4474E-05	9.8989E+03
2.8000E+03	3.7450E-02	1.4820E-05	2.5899E-05	9.8244E+03
2.9000E+03	4.0216E-02	1.5309E-05	2.7389E-05	9.7462E+03
3.0000E+03	4.3145E-02	1.5821E-05	2.8944E-05	9.6644E+03
3.1000E+03	4.6245E-02	1.6358E-05	3.0563E-05	9.5787E+03

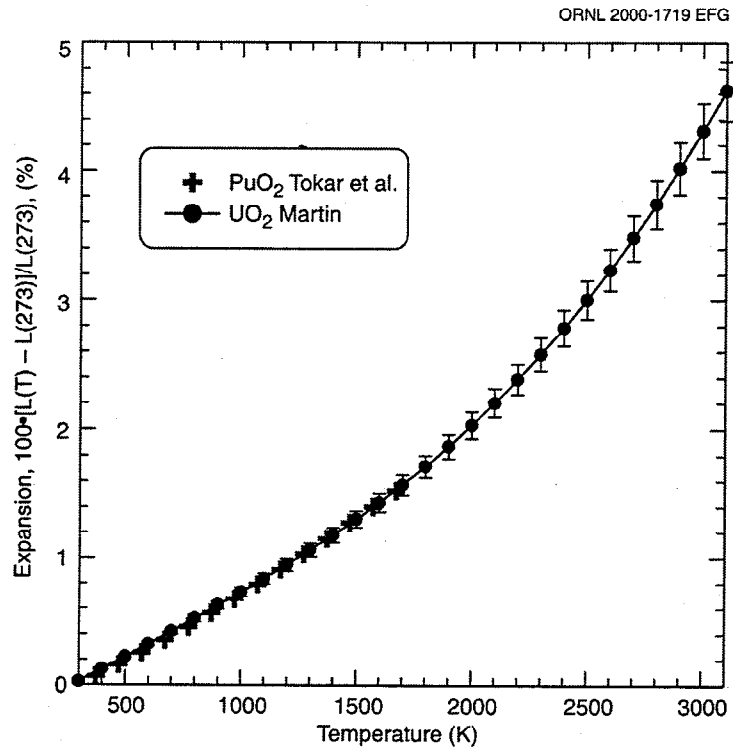


Fig. 3.1. Relative thermal expansion of UO_2 and PuO_2 fuels.

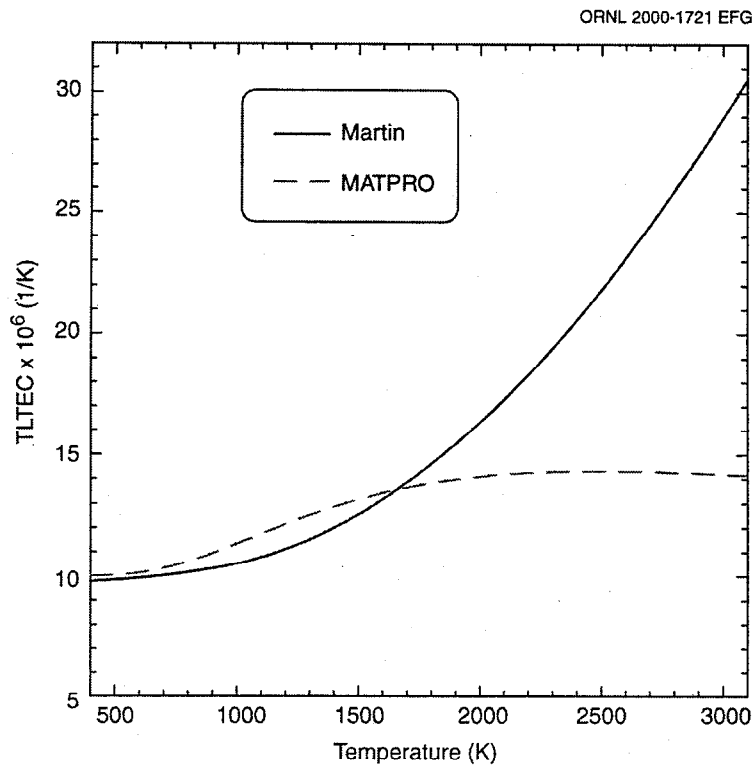


Fig. 3.2. Dependence of TLTEC of UO_2 on temperature.

3.4 REFERENCES

1. D. G. Martin, *J. Nucl. Mater.*, **152**, 94 (1988).
2. M. Tokar et al., *Nucl. Technol.*, **17**, 147 (1973).
3. *SCDAP/RELAP5/MOD2 Code Manual, Volume 4: MATPRO—A Library of Materials Properties for Light-Water-Reactor Accident Analysis*, 1993.
4. J. K. Fink, International Nuclear Safety Center Material Properties Data Base, <http://www.insc.anl.gov/matprop>, 1999.
5. M. Benedict, T. Pigford, and H. W. Levi, *Nuclear Chemical Engineering*, McGraw-Hill, 1981.
6. W. Breitung and K. O. Reil, *Nucl. Sci. Eng.*, **105**, 205 (1990).
7. S. A. Hodge, "Fission Gas Release and Pellet Swelling Within the Capsule Assembly During Phase IV of the Average Power Test," Section 6.2.2.1, ORNL Letter Report ORNL/MD/LTR-184, July 2000.
8. D. D. Lanning et al., *FRAPCON-3: Modifications to Fuel Rod Material Properties and Performance Models for High-Burnup Application*, USNRC Report NUREG/CR-6534 (PNNL-11513), Vol. 1, October 1997.

4. ENTHALPY AND SPECIFIC HEAT CAPACITY OF UO_2 , PuO_2 , AND MOX FUELS

4.1 INTRODUCTION

The fuel enthalpy and specific heat capacity are functions of fuel composition (UO_2 and PuO_2 fractions), temperature, burnup, and O/M ratio or deviation from stoichiometry. The effects of O/M ratio and burnup are very small and can be neglected. The specific heat capacity of MOX (or PuO_2) fuel is slightly larger than the values of UO_2 fuel up to a temperature of ~ 1800 K. Above this temperature, the reverse is true: UO_2 fuel has a larger heat capacity than MOX fuel.

4.2 AVAILABLE DATA AND CORRELATIONS

The available data on enthalpy and specific heat capacity are given in Table 4.1.

Table 4.1. Available data on enthalpy and heat capacity of UO_2 , PuO_2 , and MOX fuels

Reference	UO_2	PuO_2	MOX	Comments
Gibby et al., ¹ 1974			Experiment, formula	T
Fink, ² 1982	Formula, table	Formula, table	Formula	T
Cordfunke, ³ 1990	Formula, table	Formula, table		T
MATPRO, ⁴ 1993	Formula	Formula	Formula	T, y, O/M
Ronchi et al., ⁵ 1993	Experiment, table			T, liquid
Lucuta et al., ⁶⁻⁹ 1992-1997	Experiment, table, formula			T, O/M, B (SIMFUEL)
Ronchi et al., ¹⁰ 1999	Experiment, table			T, solid
Fink, ¹¹ 1999	Formula, table			T

Recently, Ronchi et al.¹⁰ measured the heat capacity of UO_2 from 2000 to 2900 K. However, these measurements lacked the sensitivity required to detect the phase transition peak at 2670 K, which has been confirmed by high-temperature neutron diffraction and scattering experiments reported by Hutchings et al.¹² and by thermal analysis of UO_{2+x} cooling curves from 2300 to 3000 K by Hiernaut et al.¹³ It was shown that above the λ -phase transition, the heat capacity has a temperature dependence that is similar to that prior to the phase transition. The heat capacity equation of MATPRO⁴ does not fit these recent high-temperature data perfectly although the equation gives heat capacity values that increase with temperature (Figs. 4.1 and 4.2). Previously recommended equations developed by Fink,² which give a constant heat capacity above 2670 K, are not consistent with the heat capacity data of Ronchi et al.¹⁰ above 2670 K. More recently, Fink¹¹ reviewed all available heat capacity and enthalpy data for solid UO_2 and made a combined analysis of enthalpy and heat capacity data to obtain equations for the enthalpy increments and heat capacities that are consistent with each other and with the experimental data.

Lucuta et al.⁶⁻⁹ measured the specific heats of simulated high-burnup UO_2 -based fuel (SIMFUEL) with an equivalent burnup from 3 to 8 at. % between room temperature and 1673 K. The results indicate that the burnup effect caused by fission products is rather small. The specific heat measurements of SIMFUEL have shown: (1) the specific heat increases slightly with burnup, as predict by the Kopp-Neymann rule; (2) higher oxygen contents increase the specific heat, but only slightly; (3) the dependence on burnup and on deviation from stoichiometry can be

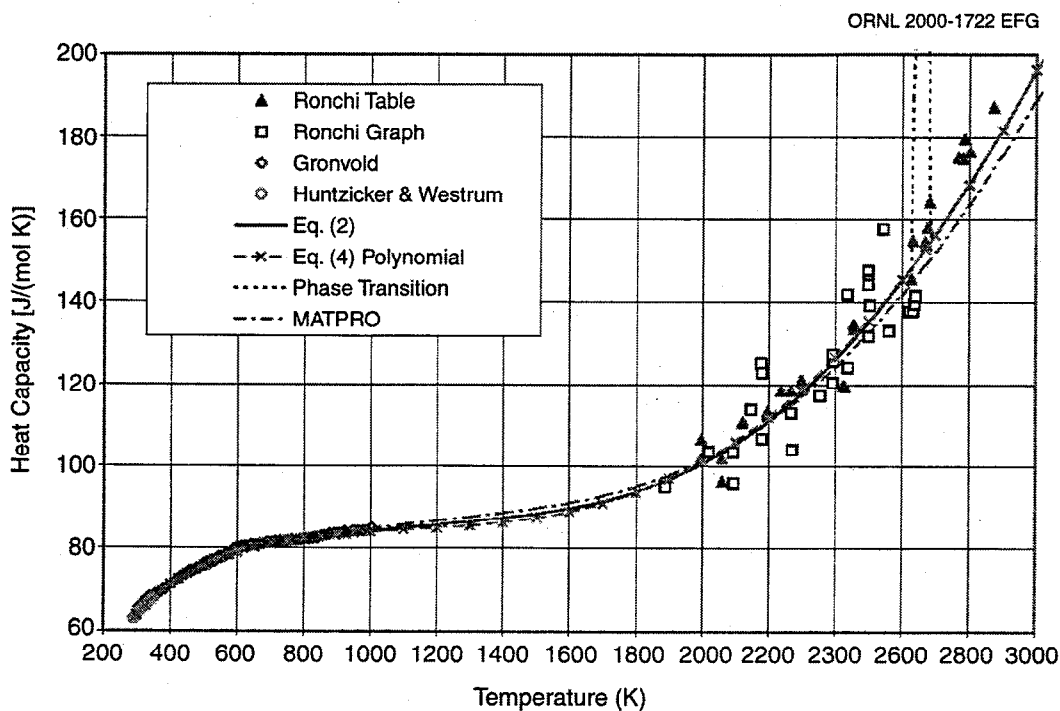


Fig. 4.1. Comparison of the MATPRO calculated heat capacity for UO_2 with the Fink Eqs. (2) and (4) of Ref. 11 and with experimental data. Source: Reproduced from Ref. 11.

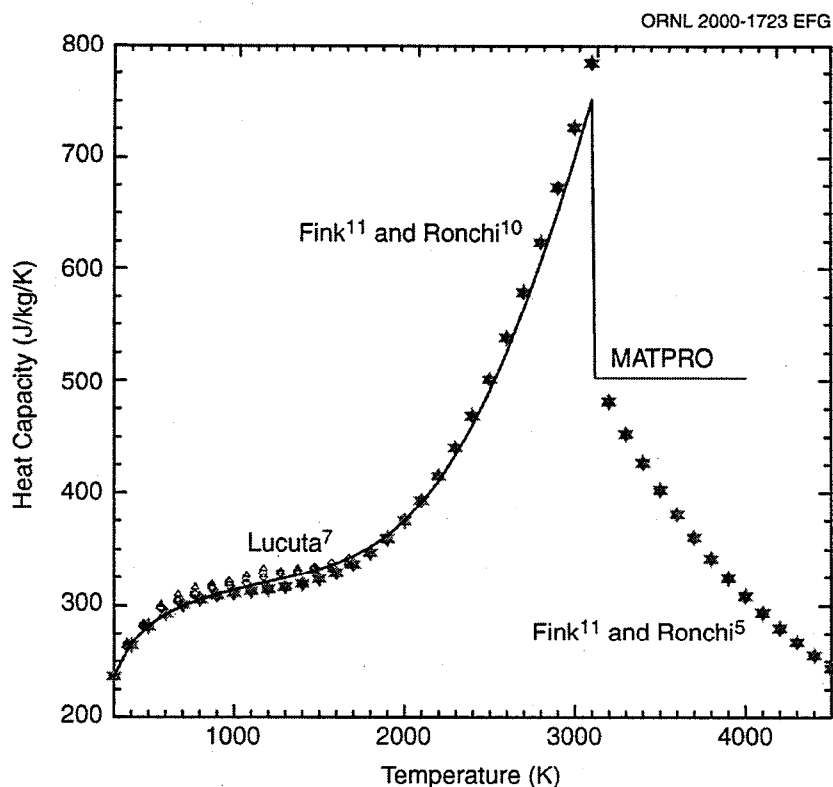


Fig. 4.2. Heat capacity of UO_2 .

expressed by slightly modifying the coefficients used for the analytical expression recommended for UO_2 [Eq. (4.2)].

Equations for the enthalpy and heat capacity of liquid UO_2 were obtained by Fink.¹¹ They are a combined fit of the liquid UO_2 heat capacity data of Ronchi et al.⁵ and the enthalpy data of Leibowitz et al.¹⁵ and Hein et al.¹⁴

Data sources for pure PuO_2 are scarce. Enthalpy has been measured by Kruger and Savage¹⁶ from 298 to 1404 K, Ogard¹⁷ from 1500 to 2715 K, and Oetting¹⁸ from 353 to 1610 K. In MATPRO, the constants for PuO_2 are determined by fitting only the data of Kruger.¹⁶ All measurements of Refs. 16–18 have been examined by Fink,² who fitted the enthalpy measurements to an expression representing two heat capacity contributions—a lattice contribution (taken to be a classical Einstein harmonic oscillator) and a term proportional to T , representing the contribution from lattice expansion. Fink's values have been refitted to the more usual polynomial form by Cordfunke and Konings.³ Figure 4.3 compares the MATPRO⁴ results with the Fink²–Cordfunke and Konings³ formula.

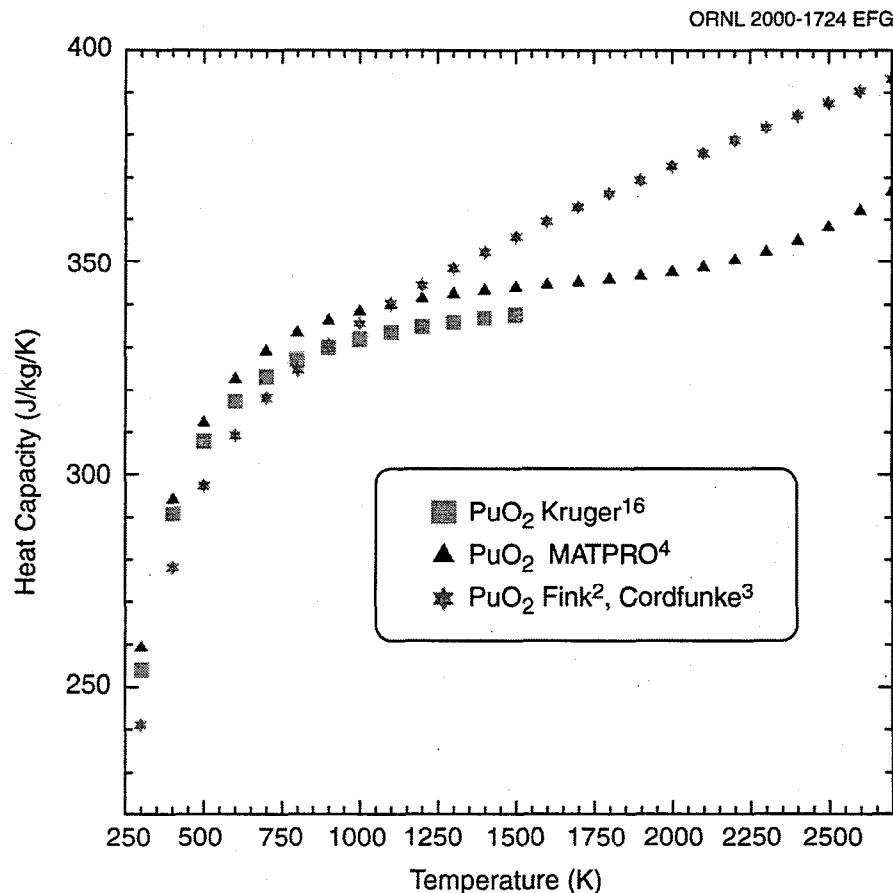


Fig. 4.3. Heat capacity of PuO_2 .

4.3 RECOMMENDATION

Recommended equations for the enthalpy and heat capacity of solid UO_2 are based on the combined analysis made by Fink.¹¹ The best fit to the data was obtained with the equations:

For $298.15 \text{ K} \leq T \leq 3120 \text{ K}$,

$$H_s(T) - H_s(298) = C_1 * \theta \{ [\exp(\theta/T) - 1]^{-1} - [\exp(\theta/298) - 1]^{-1} \} + C_2(T^2 - 298^2) + C_3 \exp(-E_a/T) , \quad (\text{J/kg}) ; \quad (4.1)$$

$$C_p(T) = C_1 * (\theta/T)^2 * \exp(\theta/T) / [\exp(\theta/T) - 1]^2 + 2C_2T + C_3 E_a \exp(-E_a/T) / T^2 , \quad [\text{J}/(\text{kg} * \text{K})] ; \quad (4.2)$$

where

- $H_s(T) - H_s(298)$ = the enthalpy increment (J/kg);
- C_p = specific heat capacity (J/kg/K);
- T = temperature (K);
- θ = Einstein temperature (K);
- E_a = electron activation energy divided by the Boltzman constant (K);

and the value of the constants used in Eqs. (4.1) and (4.2) are given in Table 4.2.

Table 4.2. Constants used in the enthalpy and heat capacity correlations

Constant	UO ₂	PuO ₂	Units
C ₁	302.27	322.49	J/kg/K
C ₂	8.463×10^{-3}	1.4679×10^{-2}	J/kg/K ²
C ₃	8.741×10^7	0	J/kg
θ	548.68	587.41	K
E _a	18,531.7		K

The same Eqs. (4.1) and (4.2) are recommended for the enthalpy and heat capacity of solid PuO₂ using the constants for PuO₂ from Table 4.2; these constants are from Ref. 2. These equations are applicable from 298.15 K to 2701 K, the melting point of PuO₂.

The uncertainty in the recommended enthalpy increments is $\pm 2\%$ from 298 to 1800 K and $\pm 3\%$ from 1800 K to the melting point. The heat capacity uncertainty is $\pm 2\%$ from 298 to 1800 K and $\pm 13\%$ from 1800 K to the melting point.

The effect of simulated burnup on specific heat can also be incorporated in the analytical expression of the heat capacity by modifying the constant C₂ accordingly. Lucuta et al.⁹ obtained:

$$C_2^* = C_2 (1 + 0.011 * B) , \quad (4.3)$$

where C₂ is the constant in Eq. (4.2) (see Table 4.2), and B is the numerical value of burnup (at. %).

The enthalpy of liquid UO₂ from 3120 to 4500 K is:¹¹

$$H_l(T) - H_s(298.15\text{K}) = 2.977 \times 10^6 + 0.931T - 4.9215 \times 10^9/T , \quad (\text{J/kg}) . \quad (4.4)$$

The uncertainty in the recommended enthalpy increments is $\pm 2\%$ from 3120 to 3500 K and $\pm 10\%$ from 3500 K to 4500 K.

The specific heat capacity of UO_2 in the liquid state from 3120 to 4500 K is given by¹¹

$$C_{P,l}(T) = 0.931 + 4.9215 \times 10^9/T^2, \quad [\text{J}/(\text{kg K})] \quad (4.5)$$

The heat capacity uncertainty is $\pm 10\%$ from T_m to 3400 K and increases linearly from $\pm 10\%$ at 3400 K to 25% at 4500 K. For PuO_2 or MOX the values for UO_2 can be used.

The solid solutions, formed in the system $\text{UO}_2\text{--PuO}_2$, are almost ideal. The Kopp–Neumann rule is true for an ideal system. We recommend the calculation of specific heat of solid MOX fuel $[(\text{U}_{1-y}\text{Pu}_y)\text{O}_2]$ by the Kopp–Neumann rule: for a mixture of UO_2 and PuO_2 , the specific heat capacity of the solid is determined by combining the contribution from each constituent in proportion to its mole fraction.

$$C_P(T) = (1 - y) C_P(T, \text{UO}_{2.00}) + y C_P(T, \text{PuO}_{2.00}) \quad (4.6)$$

When the material is partially molten, the heat capacity is determined similarly with a weighed sum of the solid and liquid portions.

Table 4.3 shows calculated values of the density (calculated as recommended in Sect. 3), heat capacity, and the product “density” times “heat capacity” for UO_2 and MOX fuel (assuming 5% PuO_2 with 95% TD). The values of this table can be used as input for thermal-hydraulic codes.

Table 4.3. Density and heat capacity of UO_2 and MOX fuels
(densities are for 100% TD fuel; products of $\rho \times C_P$ are for 95% TD fuel)

Fuel T (K)	UO_2			MOX (5% PuO_2)		
	$\rho \times 10^{-4}$ (kg/m^3)	$C_P \times 10^{-2}$ ($\text{J}/\text{kg}/\text{K}$)	$0.95 \times \rho \times C_P \times 10^{-6}$ ($\text{J}/\text{m}^3/\text{K}$)	$\rho \times 10^{-4}$ (kg/m^3)	$C_P \times 10^{-2}$ ($\text{J}/\text{kg}/\text{K}$)	$0.95 \times \rho \times C_P \times 10^{-6}$ ($\text{J}/\text{m}^3/\text{K}$)
300	1.0961	2.3658	2.4635	1.0986	2.3698	2.4732
400	1.0929	2.6432	2.7443	1.0954	2.6521	2.7598
500	1.0897	2.8153	2.9144	1.0921	2.8253	2.9314
600	1.0865	2.9299	3.0241	1.0889	2.9403	3.0416
700	1.0832	3.0071	3.0945	1.0856	3.0181	3.1127
800	1.0800	3.0584	3.1378	1.0824	3.0702	3.1570
900	1.0766	3.0918	3.1624	1.0790	3.1050	3.1829
1000	1.0733	3.1140	3.1752	1.0757	3.1286	3.1972
1100	1.0699	3.1306	3.1819	1.0723	3.1466	3.2054
1200	1.0664	3.1465	3.1877	1.0688	3.1640	3.2125
1300	1.0628	3.1666	3.1971	1.0652	3.1850	3.2229
1400	1.0590	3.1950	3.2144	1.0614	3.2139	3.2407
1500	1.0551	3.2357	3.2434	1.0575	3.2545	3.2695
1600	1.0511	3.2925	3.2876	1.0534	3.3102	3.3127
1700	1.0468	3.3688	3.3502	1.0491	3.3845	3.3733
1800	1.0423	3.4679	3.4340	1.0447	3.4803	3.4540
1900	1.0376	3.5926	3.5415	1.0400	3.6005	3.5571
2000	1.0327	3.7457	3.6748	1.0350	3.7475	3.6848
2100	1.0275	3.9297	3.8359	1.0298	3.9239	3.8387
2200	1.0220	4.1468	4.0261	1.0243	4.1316	4.0204
2300	1.0162	4.3989	4.2468	1.0185	4.3726	4.2308

Table 4.3. (continued)

Fuel T (K)	UO ₂			MOX (5% PuO ₂)		
	$\rho \times 10^{-4}$ (kg/m ³)	$C_P \times 10^{-2}$ (J/kg/K)	$0.95 \times \rho \times C_P \times 10^{-6}$ (J/m ³ /K)	$\rho \times 10^{-4}$ (kg/m ³)	$C_P \times 10^{-2}$ (J/kg/K)	$0.95 \times \rho \times C_P \times 10^{-6}$ (J/m ³ /K)
2400	1.0101	4.6878	4.4986	1.0124	4.6486	4.4709
2500	1.0037	5.0151	4.7821	1.0060	4.9610	4.7411
2600	0.9969	5.3820	5.0974	0.9992	5.3109	5.0414
2700	0.9898	5.7896	5.4445	0.9921	5.6995	5.3718
2800	0.9824	6.2386	5.8226	0.9846	6.1275	5.7317
2900	0.9746	6.7297	6.2310	0.9768	6.5954	6.1203
3000	0.9664	7.2632	6.6685	0.9685	7.1035	6.5364
3100	0.9578	7.8392	7.1335	0.9600	7.6520	6.9787

4.4 REFERENCES

1. R. L. Gibby et al., *J. Nucl. Mater.*, **50**, 155 (1974).
2. J. K. Fink, *Intl. J. Thermophys.*, **3**, 165 (1982).
3. E. H. P. Cordfunke and R. J. M. Konings, *Thermochemical Data for Reactor Materials and Fission Products*, North-Holland, Amsterdam, 1990.
4. D. T. Hagman, Ed., *SCDAP/RELAP5/MOD3.1 Code Manual, Vol. 4: MATPRO—A Library of Materials Properties for Light-Water Reactor Accident Analysis*, USNRC Report NUREG/CR-6150 (EGGG-2720) (1993).
5. C. Ronchi et al., *Nucl. Sci. Eng.*, **113**, 1 (1993).
6. P. G. Lucuta et al., *J. Nucl. Mater.*, **188**, 198 (1992).
7. P. G. Lucuta et al., *J. Nucl. Mater.*, **223**, 51 (1995).
8. P. G. Lucuta et al., *J. Nucl. Mater.*, **228**, 251 (1996).
9. P. G. Lucuta et al., *J. Nucl. Mater.*, **247**, 121 (1997).
10. C. Ronchi et al., *J. Applied Phys.*, **85**, 776 (1999).
11. J. K. Fink, INSC Material Properties Database, <http://www.insc.anl.gov/matprop> (1999).
12. M. T. Hutchings, *J. Chem. Soc. Faraday Trans. II*, **83**, 1083 (1987).
13. J. P. Hiernaut et al., *Int. J. Thermophys.*, **14**, 259 (1993).
14. R. A. Hein, P. M. Flagella, and J. B. Conway, *J. Amer. Ceram. Soc.*, **51**(5), 291 (1968).
15. L. Leibowitz et al., *J. Nucl. Mater.*, **39**, 115 (1971).
16. O. L. Kruger and H. J. Savage, *J. Chem. Phys.*, **49**, 4540 (1968).
17. A. E. Ogard, *Proc. 4th Int. Conf. Plutonium and Other Actinides, Sante Fe, New Mexico* (1970), Vol. 1, p. 78.
18. F. L. Oetting, *J. Nucl. Mater.*, **105**, 257 (1982).

5. HEAT OF FUSION OF UO_2 , PuO_2 , AND MOX FUELS

5.1 INTRODUCTION

The phase transition from solid to liquid (melting) represents the loss of long-range order. For stoichiometric oxide compounds, this change is the phase transition of the first kind and occurs uniquely at one temperature. If the compound is nonstoichiometric or involves added components in solid solution, then melting may involve a liquid phase coexisting with a solid phase at different compositions. Composition changes are required to maintain the lowest free energy for the mixture at a given temperature. This transition is the phase transition of the second kind and occurs over a temperature range. The free energy content of the liquid exceeds the free energy content of the solid by an amount approximately equal to the heat of fusion, which is a measure of the loss of long-range order.

The experimental data about heat of fusion of pure UO_2 are not numerous; and for pure PuO_2 they are absent in the open literature. The enthalpy or heat of fusion of pure UO_2 appears to be larger than the values for pure PuO_2 or MOX fuel. There is significant scatter in the reported values, with some values for PuO_2 larger than for UO_2 . Table 5.1 summarizes the most relevant values.

Table 5.1. Available data on the heat of fusion (kJ/mol) in the system UO_2 – PuO_2

Reference	UO_2	MOX	PuO_2	Comments
Epstein, ¹ 1967	77.8 ± 5.4		66.5 ± 5.4	Calculation from phase diagram
	88.7 ± 5.4		70.3 ± 5.4	Evaluation
Hein, Flagella, ² 1968	76.1 ± 2.0			Drop calorimetry, experiment
Leibowitz et al., ³ 1971	74 ± 3			Drop calorimetry, experiment
Leibowitz et al., ⁴ 1975		67 ± 3 ($\text{U}_{0.8}\text{Pu}_{0.2}\text{O}_2$)		Drop calorimetry, experiment
Rand et al., ⁵ 1978; Fink et al., ⁶ 1981	74.8 ± 3			Combined analysis of the available enthalpy data to 1978
Fink, ⁷ 1999	70 ± 4			Combined analysis of the available enthalpy and heat capacity data up to 1999

5.2 RECOMMENDATIONS

The recommended value for the heat of fusion of $\text{UO}_{2.00}$ was calculated from Eqs. (4.1) and (4.4) recommended in Sect. 4 for the enthalpy of solid and liquid UO_2 at the melting point of 3120 K.

$$\Delta H_m(\text{UO}_{2.00}) = 70 \pm 4 \text{ kJ/mol} = 259.3 \pm 14.8 \text{ kJ/kg}$$

This value was chosen as the best estimate value based on the recent analysis completed by Fink⁷ for the International Nuclear Safety Center.

The recommended value for the heat fusion of $\text{PuO}_{2.00}$ is

$$\Delta H_m(\text{PuO}_{2.00}) = 66.5 \pm 5.4 \text{ kJ/mol} = 245.4 \pm 19.9 \text{ kJ/kg}$$

This value is based on evaluation by Epstein¹ from the liquid and solid data of the UO₂-PuO₂ system. This PuO₂ fusion heat is the lowest reported value and, therefore, the most conservative value.

For MOX fuel, a value of 67 ± 3 kJ/mol (248 ± 12 kJ/kg) can be used, or the fusion heat of stoichiometric MOX fuel with the given content of the mole fraction, y , of PuO₂ in (U_{1-y}Pu_y)O₂ can be calculated by the weighted average of the MOX components:

$$\Delta H_m((U_{1-y}Pu_y)O_2) = (1 - y) \Delta H_m(UO_{2.00}) + y \Delta H_m(PuO_{2.00}) ,$$

where $\Delta H_m(UO_{2.00})$ and $\Delta H_m(PuO_{2.00})$ are the heats of fusion for UO₂ and PuO₂, respectively. The estimated error for this value is ~15 kJ/kg.

Values of the heat of fusion, $\Delta H_m((U_{1-y}Pu_y)O_2)$, and the melting temperature of MOX fuel are tabulated in Table 5.2 as a function of the mole fraction of PuO₂.

Table 5.2. Dependence of the melting temperature and the heat of fusion on mole fraction of PuO₂ in MOX fuel (U_{1-y}Pu_y)O₂

Mole fraction of PuO ₂ , y	Melting temperature (K)	Heat of fusion (kJ/kg)	Mole fraction of PuO ₂ , y	Melting temperature (K)	Heat of fusion (kJ/kg)
0.00	3120	259.3	0.10	3057.7	258.0
0.01	3113.5	259.2	0.11	3051.8	257.8
0.02	3107	259.1	0.12	3046.0	257.7
0.03	3100.6	258.9	0.13	3040.3	257.6
0.04	3094.3	258.8	0.14	3034.6	257.5
0.05	3088.1	258.6	0.15	3028.9	257.3
0.06	3081.9	258.5	0.16	3023.4	257.2
0.07	3075.7	258.4	0.17	3017.8	257.0
0.08	3069.7	258.2	0.18	3012.4	256.9
0.09	3063.7	258.1	0.19	3007.0	256.8

5.3 REFERENCES

1. L. F. Epstein, *J. Nucl. Mater.*, **22**(3), 340 (1967).
2. R. A. Hein, P. M. Flagella, and J. B. Conway, *J. Amer. Ceram. Soc.*, **51**(5), 291 (1968).
3. L. Leibowitz et al., *J. Nucl. Mater.*, **39**, 115 (1971).
4. L. Leibowitz et al., ANL-8082, February 1975.
5. M. N. Rand et al., *Rev. Hautes Temp. Refract.*, **15**, 355-365 (1978).
6. J. K. Fink, M. G. Chasanov, and L. Leibovitz, *J. Nucl. Mater.*, **102**, 17 (1981).
7. J. K. Fink, INSC Material Properties Database, <http://www.insc.anl.gov/matprop> (1999).

6. THERMAL CONDUCTIVITY OF FUEL

6.1 INTRODUCTION

The thermal conductivity of both UO_2 and MOX fuels is a function of temperature, fuel composition, fraction of the fuel theoretical density (TD) or porosity p [$p = (\rho_{\text{TD}} - \rho)/\rho_{\text{TD}}$], O/M ratio or deviation from stoichiometry (x), and burnup (B). The thermal conductivity decreases with temperature up to ~ 2000 K and then increases with temperature. The addition of PuO_2 to the fuel slightly decreases the thermal conductivity. Deviation of the O/M ratio from 2 significantly decreases the thermal conductivity and so does the burnup. The burnup slightly changes the O/M ratio, so both effects are interrelated.

6.2 AVAILABLE DATA AND CORRELATIONS

There is a very large body of data and correlations for the thermal conductivity of fuel. The data are not so extensive for MOX fuel as they are for UO_2 fuel. Table 6.1 shows the most relevant data published after 1989. Burnup is considered by four references in this table, and only one of them considers burnup for MOX fuel.

Table 6.1. Some data on thermal conductivity of fuel (after 1989)

Reference	UO_2	MOX	Parameters
Harding, Martin, ¹ 1989	Formula		T
MATPRO, ² 1993	Formula, table, graphs	Formula, table, graphs	T, U/Pu, O/M, p
Lucuta et al., ³⁻⁶ 1992–1996	Measurement, formulas, tables, graphs (SIMFUEL)		T, p, O/M, B
Philipponneau, ⁷ 1992		Formula	T, p, O/M, B
Wiesenak, ⁸ 1997	Measurement, formula		T, 0.95 TD, B
Ohira, Itagaki, ⁹ 1997	Measurement, formula		T, p, B
Ronchi et al., ¹⁰ 1999	Measurement, table		T, p
Fink, ¹¹ 1999	Formula		T, p
Duriez, Philipponneau, et al., ¹² 2000		Measurement, formula	T, p, O/M

Since 1981 theoretical research and new measurements have led to improvements in equations for the thermal conductivity of unirradiated UO_2 . The physically based equation of Harding and Martin¹ included lattice (phonon-defect and phonon-phonon scattering processes) and small-polaron ambipolar contributions. Since the publication of this equation, Casado, Harding, and Hyland¹³ have shown that the temperature dependence for small-polaron ambipolar contribution is incorrect. They report the correct temperature dependence, which was used Ronchi et al.¹⁰ for fitting the thermal conductivity obtained from their thermal diffusivity measurements. The 2000–2900 K thermal diffusivity data of Ronchi et al.¹⁰ indicate that the high-temperature thermal diffusivity values reported by Weilbacher,¹⁴ which were the main high-temperature data available prior to 1999, are too high. All data for the thermal diffusivity and thermal conductivity of solid unirradiated UO_2 , including the latest Ronchi et al.¹⁰ data, have been reassessed by Fink.¹¹ The Fink¹¹ equation for the thermal conductivity of 95% dense solid unirradiated UO_2 consists of a lattice term and a term suggested by Ronchi et al.¹⁰ to represent the small-polaron ambipolar contribution to the thermal conductivity.

Wiesenack⁸ proposed a modification of the correlation for irradiated urania thermal conductivity based on data from the Halden Ultra-High-Burn Experiment. The data indicated a steady degradation in urania fuel thermal conductivity (averaged over the temperature range from 750 to 1200 K) of 5 to 7% per 10 MWd/kgU for burnups up to 88 MWd/kgU. The uncertainty on the burnup degradation rate must reflect the uncertainty on temperature measurement (~2%), heat generation rate (10%), and on the analysis (~10%); it is therefore about 15% relative standard deviation. The thermal conductivity values have the same uncertainty. The data evaluation accounts for concurrent effects (e.g., gap closure and fission gas release), which also have an influence on temperature. Otherwise, the resulting correlation for urania thermal conductivity reflected the "total" effect of solid and volatile fission products entrained in the fuel matrix, irradiation damage, microcracking, and others.

The degradation rate of urania fuel thermal conductivity reported by Wiesenack⁸ is qualitatively consistent with the results of laser-flash diffusivity measurements on unirradiated SIMFUEL performed by Lucuta et al.³⁻⁶ SIMFUEL replicates the composition and the microstructure of irradiated fuel by introducing 11 stable additives into the UO₂, both those that are soluble in the fluorite-type lattice of the fuel and others that precipitate as second phases. The thermal conductivity was determined for different simulated burnups (1.5, 3, 6, and 8 at. % burnup). The accuracy of diffusivity measurements (better than 5% relative standard deviation) combined with the extremely accurate specific heat measurements indicates conductivity measurement accuracy of better than 8% relative standard deviation. It is better (less) than the estimated uncertainty (15%) in Wiesenack⁸ measurements. The use of SIMFUEL permitted the assessment and analysis of "single" effects on thermal conductivity. The effect of fission products and the effect of deviation from stoichiometry were quantified from the results in the form of factors applied to thermal conductivity of unirradiated UO₂. The expression for each factor is derived from the available experimental data obtained by studying the single effect. Lucuta et al.⁶ provide an analytical expression for the thermal conductivity of irradiated UO₂ fuel as a function of the temperature; it accounts for all the changes taking place during irradiation. There are solid fission-product buildup (dissolved and precipitated), pores and fission-gas-bubble formation, radiation damage, and changes in the oxygen-to-uranium ratio (O/U). The expression for the parametric dependence of irradiated UO₂ thermal conductivity, λ , has the form of a product with contributing factors for each individual effect.

Ohira and Itagaki⁹ measured thermal conductivity of high-burnup BWR pellets using the "laser flash" method. Their correlation is similar to the Wiesenack⁸ correlation and fits their data well.

The "laser flash" method was used to measure the thermal diffusivity of fresh MOX fuel with plutonium contents ranging from 3 to 15 wt % and an O/M ratio ranging from 2.00 to 1.95 by Duriez et al.¹² The temperature range extends from 700 to 2300 K. Duriez et al. have shown that the MOX fuel thermal conductivity seems not to depend on the plutonium concentration. However, experimental data¹² are lower in the investigated temperature range than the Harding and Martin¹ UO₂ recommendation; this indicates that even a small amount of plutonium leads to a decrease in the thermal conductivity. Duriez et al. observed an effect of the deviation from stoichiometry on the thermal conductivity smaller than what is recommended for high plutonium content (~20% Pu) fast breeder reactors fuels by Philipponneau.⁷ Duriez et al.¹² proposed an equation for the thermal conductivity of 95% dense solid unirradiated MOX fuel as a function of the deviation from stoichiometry [$x = 2 - (O/M)$].

6.3 RECOMMENDATION

Analysis of available data shows that all changes taking place during irradiation [solid fission-product buildup (dissolved and precipitated), pores and fission-gas-bubble formation, and

radiation damage] in UO₂ and MOX fuels are equivalent. We recommended use of the physically based Lucuta model⁶ for calculation of the irradiated UO₂ and MOX thermal conductivity:

$$\lambda(T, B, p, x) = \lambda_0(T, x) * FD * FP * FM * FR, \quad (W * m^{-1} * K^{-1}), \quad (6.1)$$

where $\lambda_0(T, x)$ is the expression for thermal conductivity of unirradiated, fully dense UO₂ or MOX fuel (in $W * m^{-1} * K^{-1}$); the FD factor (unitless) describes the effect of the dissolved solid fission products in the UO₂ or MOX fuel matrix; the FP factor (unitless) describes the effect of the precipitated solid fission products in the UO₂ or MOX fuel matrix; the FM factor (unitless) describes the effect of fuel porosity; the FR factor (unitless) describes the effect of radiation damage; T is temperature (in K); B is burnup [in at. % (1 at. % = 9.375 MWd/kgU)]; p is porosity, $p = (\rho_{TD} - \rho)/\rho_{TD}$; ρ is fuel density; ρ_{TD} is theoretical density (TD) for fully dense UO₂ or MOX fuel; and $x = 2 - (O/M)$ is deviation from stoichiometry.

The recommended equation for the thermal conductivity of 100% dense solid UO₂ is

$$\lambda_0(T) = 115.8/(7.5408 + 17.692t + 3.6142t^2) + 7410.5 t^{-5/2} \exp(-16.35/t), \quad (W * m^{-1} * K^{-1}), \quad (6.2)$$

with $t = T(K)/1000$.

This correlation was chosen as the best equation based on the latest analysis completed by Fink¹¹ for the International Nuclear Safety Center. The recommended equation consists of a lattice term and a term suggested by Ronchi et al.¹⁰ to represent the small-polaron ambipolar contribution to the thermal conductivity. Uncertainties were determined from the scatter in the available data and the deviations of the data from the recommended equation. From 298 to 2000 K the uncertainty is 10%. From 2000 to 3120 K, the uncertainty has been increased to 20% because of the large discrepancies between measurements by different investigators.

Our recommended equation for the thermal conductivity of 100% dense solid MOX fuel consists of a lattice term from Duriez et al.¹² and the term suggested by Ronchi et al.¹⁰ to represent the small-polaron ambipolar contribution to the thermal conductivity:

$$\lambda_0(T, x) = 1.1579/(A + CT) + 2.3434 * 10^{11} * T^{-5/2} \exp(-16350/T), \quad (W * m^{-1} * K^{-1}), \quad (6.3)$$

where

$$\begin{aligned} A &= A(x) = 2.85x + 0.035 \text{ (mK/W)}, \\ C &= C(x) = (-7.15x + 2.86) * 10^{-4} \text{ (m/W)}, \end{aligned}$$

in the range of $700 \leq T \leq 3100$ K; $3\% \leq Pu/(Pu + U) \leq 15\%$. This equation was selected because the lattice contribution is based on the best experimental data by Duriez et al.;¹² and it is in good agreement with an earlier equation of Philipponneau⁷ for stoichiometric MOX fuel. At high temperatures ($T > 2000$ K), heat is conducted predominantly by small polarons, but the contribution from phonons is by no means negligible. The physically based term for the small-polaron ambipolar contribution is given by Ronchi et al.¹⁰ Figure 6.1 shows the total thermal conductivity, the lattice contribution, and the ambipolar contribution as a function of temperature for UO₂ and MOX fuels. Below 1300 K the ambipolar term is insignificant, and the total conductivity equals the lattice contribution. The UO₂ lattice contribution is larger than that of MOX. Although the ambipolar term begins to have a significant contribution to the total thermal conductivity above 1300 K, it is not larger than the lattice contribution until 2800 K. At 3120 K the lattice

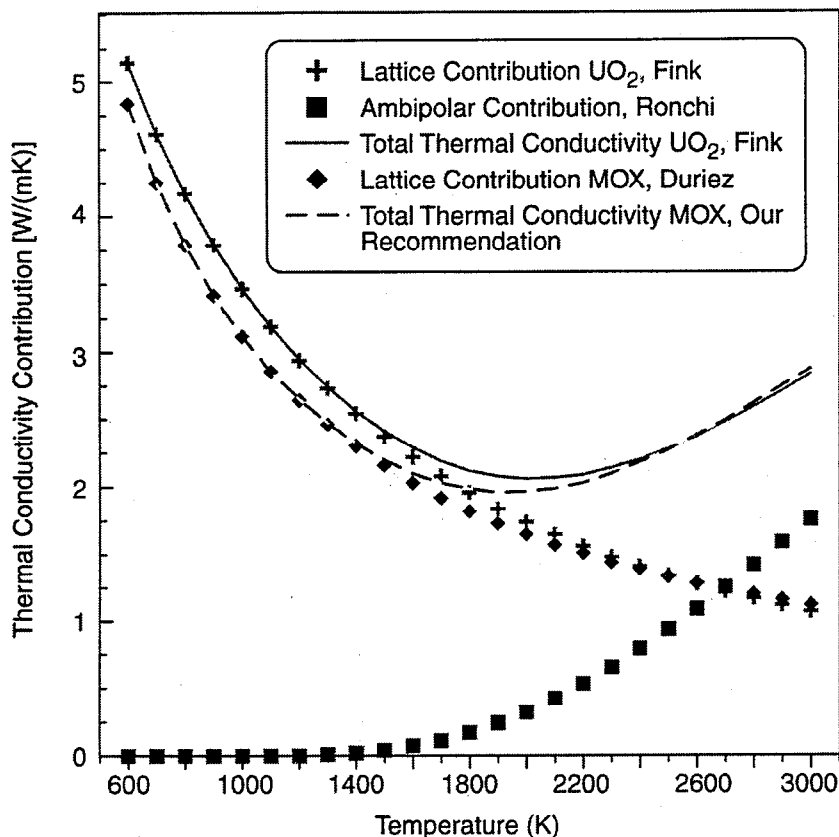


Fig. 6.1. Thermal conductivity contributions and total thermal conductivity of 95% dense unirradiated $\text{UO}_{2.00}$ and MOX fuels.

contribution is still significant. We estimated the uncertainties in thermal conductivity calculated with the equation as 7% from 700 to 1800 K, increasing to 20% at 3100 K.

The effect of the “dissolved” fission products is reflected by a burnup and temperature-dependent factor FD (Lucuta⁶):

$$\text{FD} = \omega * \arctan(1/\omega) , \quad (6.4)$$

where $\omega = 1.09/B^{3.265} + 0.0643(T/B)^{1/2}$.

The effect of the “precipitated” fission products is reflected by a burnup and temperature-dependent factor FP (Lucuta⁶):

$$\text{FP} = 1 + 0.019B/(3 - 0.019B) * \{1 + \exp[-(T - 1200)/100]\}^{-1} . \quad (6.5)$$

The effect of “porosity” is accounted for by the well-known Maxwell-Eucken factor, FM:

$$\text{FM} = (1 - p)/(1 + 2p) . \quad (6.6)$$

The “radiation effect” is given by the factor FR (Lucuta⁶):

$$\text{FR} = 1 - 0.2/\{1 + \exp[(T - 900)/80]\} . \quad (6.7)$$

This factor has a significant effect at temperatures below 900 K, sharply decreasing as temperatures increase above 900 K. This factor is not dependent on the burnup.

Values for the thermal conductivity for irradiated 95% dense UO_2 calculated from Eqs. (6.1, 6.2, 6.4–6.7) and from the Wiesenack⁸ equation are given in Table 6.2 and are shown in Fig. 6.2 (our recommendation is labeled as Lucuta and Fink). The deviations for the equation of Wiesenack⁸ from our recommendations are greater than 20% above 2800 K.

Values for the thermal conductivity for irradiated 95% dense stoichiometric and hypo-stoichiometric ($x = 0.02; 0.05$) MOX fuel calculated from Eqs. (6.1, 6.3–6.7) are given in Tables 6.3–6.5 and are shown in Figs. 6.3–6.5.

In Fig. 6.6 the recommended values for the thermal conductivity for irradiated 95% dense stoichiometric UO_2 and MOX fuels are compared. It is shown that values of the thermal conductivity of MOX fuel at equivalent burnup are less than for UO_2 from 600 to 2600 K; at temperatures above 2600 K, the thermal conductivity MOX fuel is slightly larger than for UO_2 .

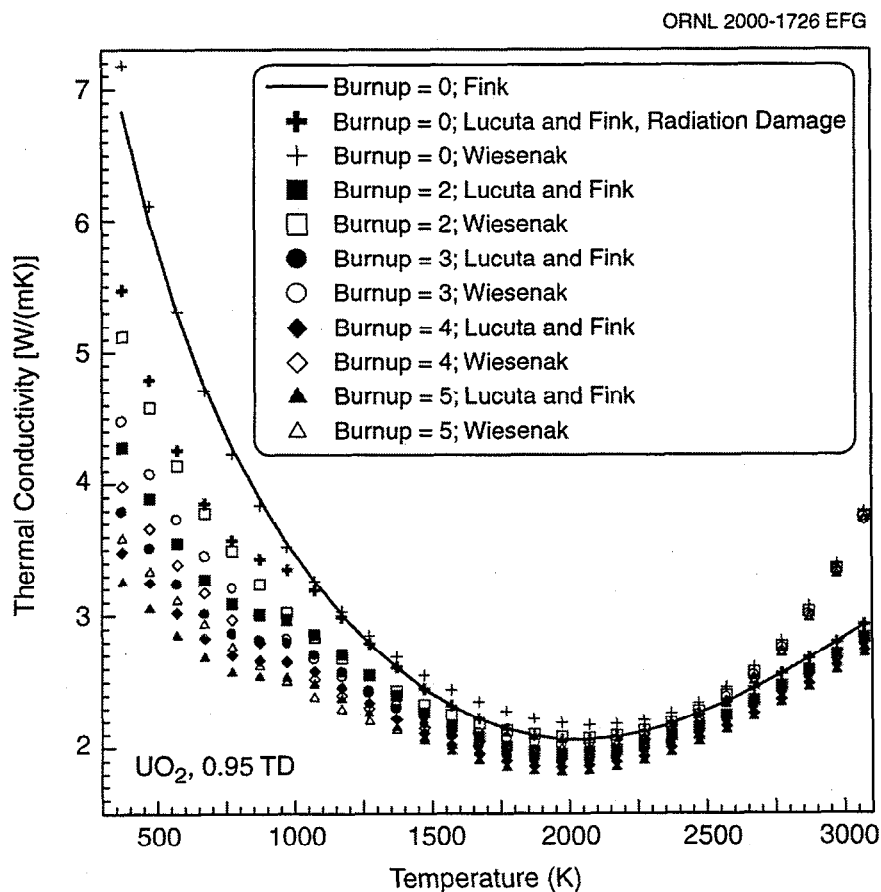


Fig. 6.2. Comparison of thermal conductivity equations for 95% dense $\text{UO}_{2.00}$ with different burnups.

Table 6.2. Thermal conductivity, λ [W/(mK)], of stoichiometric $\text{UO}_{2.00}$ fuel with 0.95 TD for burnup of 0, 2, 3, 4, and 5 (at. %)

T (K)	Burnup (at. %)											
	Recommended		Ref. 8	Recommended		Ref. 8	Recommended		Ref. 8	Recommended		Ref. 8
	0	0 ^a	0	2	2	3	3	4	4	5	5	
673	4.74	3.85	4.71	3.27	3.78	3.01	3.45	2.83	3.17	2.68	2.93	
773	4.28	3.57	4.23	3.09	3.49	2.86	3.21	2.70	2.97	2.57	2.76	
873	3.89	3.43	3.84	3.01	3.24	2.81	3.00	2.66	2.80	2.54	2.62	
973	3.55	3.35	3.52	2.97	3.02	2.79	2.82	2.65	2.65	2.53	2.50	
1073	3.26	3.19	3.26	2.86	2.84	2.70	2.67	2.58	2.52	2.48	2.38	
1173	3.01	2.99	3.03	2.71	2.68	2.57	2.54	2.46	2.40	2.37	2.29	
1273	2.79	2.79	2.85	2.55	2.55	2.43	2.42	2.34	2.30	2.26	2.20	
1373	2.61	2.61	2.69	2.40	2.43	2.30	2.32	2.22	2.22	2.15	2.13	
1473	2.45	2.45	2.55	2.27	2.33	2.18	2.24	2.11	2.15	2.05	2.07	
1573	2.32	2.32	2.44	2.16	2.25	2.09	2.17	2.02	2.09	1.97	2.01	
1673	2.22	2.22	2.35	2.08	2.18	2.01	2.11	1.95	2.04	1.90	1.97	
1773	2.14	2.14	2.28	2.01	2.13	1.95	2.07	1.90	2.00	1.85	1.95	
1873	2.09	2.09	2.22	1.97	2.10	1.91	2.04	1.86	1.98	1.82	1.93	
1973	2.06	2.06	2.19	1.95	2.08	1.90	2.02	1.85	1.98	1.81	1.93	
2073	2.06	2.06	2.17	1.96	2.07	1.90	2.03	1.86	1.98	1.82	1.94	
2173	2.08	2.08	2.18	1.98	2.09	1.93	2.05	1.89	2.01	1.85	1.97	
2273	2.12	2.12	2.21	2.03	2.13	1.98	2.09	1.93	2.06	1.90	2.02	
2373	2.18	2.18	2.26	2.09	2.19	2.04	2.16	2.00	2.13	1.96	2.10	
2473	2.26	2.26	2.34	2.16	2.28	2.11	2.25	2.07	2.22	2.04	2.20	
2573	2.35	2.35	2.46	2.25	2.40	2.20	2.38	2.16	2.35	2.13	2.33	
2673	2.45	2.45	2.61	2.36	2.57	2.31	2.54	2.26	2.52	2.23	2.50	
2773	2.56	2.56	2.81	2.47	2.77	2.42	2.75	2.37	2.73	2.34	2.71	
2873	2.68	2.68	3.07	2.59	3.03	2.54	3.01	2.49	3.00	2.46	2.98	
2973	2.80	2.80	3.39	2.71	3.36	2.66	3.34	2.62	3.33	2.58	3.31	
3073	2.93	2.93	3.79	2.84	3.76	2.79	3.75	2.74	3.73	2.71	3.72	

^aRadiation effect (factor FR) with no burnup.

Table 6.3. Thermal conductivity, λ [W/(mK)], of stoichiometric ($x = 0.00$) MOX fuel with 0.95 TD for burnup of 0, 2, 3, and 5 (at. %)

T (K)	Burnup (at. %)				
	0	0 ^a	2	3	5
673	4.40	3.57	3.04	2.79	2.48
773	3.91	3.26	2.82	2.61	2.34
873	3.51	3.10	2.72	2.54	2.29
973	3.19	3.01	2.67	2.51	2.28
1073	2.93	2.87	2.57	2.43	2.22
1173	2.70	2.69	2.43	2.31	2.13
1273	2.52	2.51	2.30	2.19	2.04
1373	2.36	2.36	2.17	2.08	1.95
1473	2.23	2.23	2.07	1.99	1.87
1573	2.13	2.13	1.98	1.91	1.80
1673	2.05	2.05	1.92	1.85	1.76
1773	2.00	2.00	1.88	1.82	1.73
1873	1.97	1.97	1.86	1.80	1.71
1973	1.96	1.96	1.86	1.81	1.72
2073	1.98	1.98	1.88	1.83	1.75
2173	2.02	2.02	1.92	1.87	1.79
2273	2.08	2.08	1.98	1.93	1.86
2373	2.15	2.15	2.06	2.01	1.93
2473	2.24	2.24	2.15	2.10	2.02
2573	2.35	2.35	2.25	2.20	2.12
2673	2.46	2.46	2.37	2.32	2.24
2773	2.58	2.58	2.49	2.44	2.36
2873	2.71	2.71	2.62	2.57	2.49
2973	2.85	2.85	2.75	2.70	2.62
3073	2.98	2.98	2.89	2.84	2.75

^aRadiation effect (factor FR) with no burnup.

Table 6.4. Thermal conductivity, λ [W/(mK)], of hypostoichiometric ($x = 0.02$) MOX fuel with 0.95 TD for burnup of 0, 2, 3, 4, and 5 (at. %)

T (K)	Burnup (at. %)					
	0	0 ^a	2	3	4	5
673	3.64	2.95	2.51	2.31	2.17	2.05
773	3.31	2.76	2.39	2.22	2.09	1.99
873	3.04	2.68	2.35	2.20	2.08	1.98
973	2.81	2.65	2.35	2.20	2.09	2.00
1073	2.61	2.55	2.29	2.16	2.06	1.98
1173	2.44	2.42	2.20	2.08	2.00	1.92
1273	2.29	2.29	2.09	2.00	1.92	1.86
1373	2.17	2.17	2.00	1.92	1.85	1.79
1473	2.07	2.07	1.92	1.84	1.79	1.73
1573	1.99	1.99	1.86	1.79	1.73	1.69
1673	1.93	1.93	1.81	1.75	1.70	1.65
1773	1.89	1.89	1.78	1.72	1.68	1.64
1873	1.88	1.88	1.77	1.72	1.68	1.64
1973	1.89	1.89	1.79	1.73	1.69	1.66
2073	1.91	1.91	1.82	1.77	1.73	1.69
2173	1.96	1.96	1.87	1.82	1.78	1.74
2273	2.03	2.03	1.93	1.89	1.85	1.81
2373	2.11	2.11	2.02	1.97	1.93	1.89
2473	2.20	2.20	2.11	2.06	2.02	1.99
2573	2.31	2.31	2.22	2.17	2.13	2.09
2673	2.43	2.43	2.34	2.29	2.25	2.21
2773	2.56	2.56	2.46	2.41	2.37	2.34
2873	2.69	2.69	2.60	2.55	2.50	2.47
2973	2.83	2.83	2.73	2.68	2.64	2.60
3073	2.97	2.97	2.87	2.82	2.78	2.74

^aRadiation effect (factor FR) with no burnup.

Table 6.5. Thermal conductivity, λ [W/(mK)], of hypostoichiometric ($x = 0.05$) MOX fuel with 0.95 TD for burnup of 0, 1, 2, 3, 4, and 5 (at. %)

T (K)	Burnup (at. %)						
	0	0 ^a	1	2	3	4	5
673	2.89	2.34	2.25	2.00	1.84	1.72	1.63
773	2.70	2.25	2.16	1.94	1.80	1.70	1.62
873	2.53	2.23	2.15	1.96	1.83	1.73	1.65
973	2.38	2.24	2.17	1.99	1.87	1.77	1.70
1073	2.24	2.20	2.13	1.97	1.86	1.77	1.70
1173	2.13	2.11	2.06	1.91	1.82	1.74	1.68
1273	2.03	2.02	1.97	1.85	1.76	1.70	1.64
1373	1.94	1.94	1.90	1.79	1.71	1.65	1.60
1473	1.87	1.87	1.83	1.73	1.67	1.61	1.57
1573	1.81	1.81	1.78	1.69	1.63	1.58	1.54
1673	1.78	1.78	1.75	1.67	1.61	1.56	1.52
1773	1.76	1.76	1.73	1.66	1.60	1.56	1.52
1873	1.76	1.76	1.74	1.66	1.61	1.57	1.54
1973	1.78	1.78	1.76	1.69	1.64	1.60	1.57
2073	1.82	1.82	1.80	1.73	1.68	1.65	1.61
2173	1.88	1.88	1.86	1.79	1.75	1.71	1.67
2273	1.96	1.96	1.93	1.87	1.82	1.78	1.75
2373	2.05	2.05	2.02	1.96	1.91	1.87	1.84
2473	2.15	2.15	2.13	2.06	2.01	1.97	1.94
2573	2.27	2.27	2.24	2.18	2.13	2.09	2.05
2673	2.39	2.39	2.37	2.30	2.25	2.21	2.17
2773	2.52	2.52	2.50	2.43	2.38	2.34	2.30
2873	2.66	2.66	2.63	2.57	2.52	2.48	2.44
2973	2.80	2.80	2.78	2.71	2.66	2.62	2.58
3073	2.95	2.95	2.92	2.85	2.80	2.76	2.72

^aRadiation effect (factor FR) with no burnup.

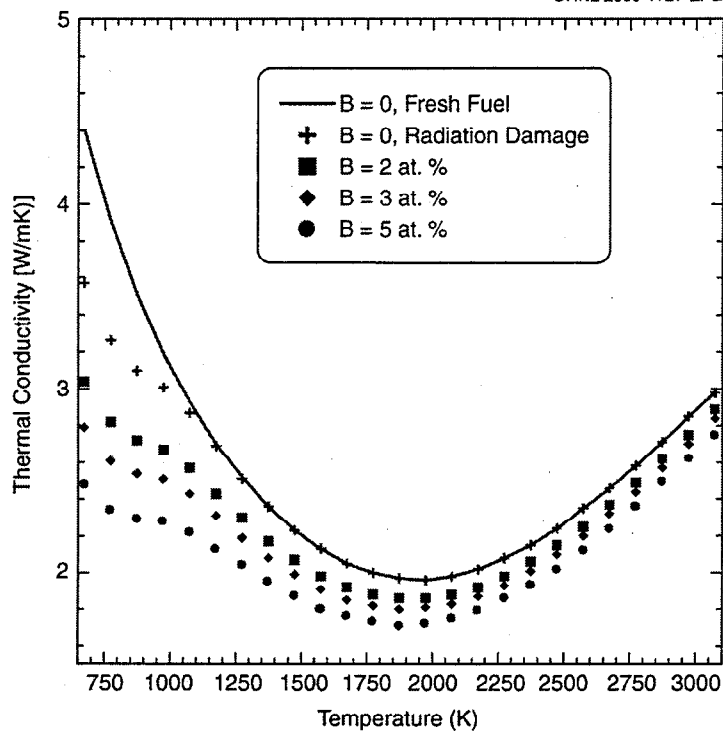


Fig. 6.3. Thermal conductivity of irradiated stoichiometric 95% dense MOX fuel.

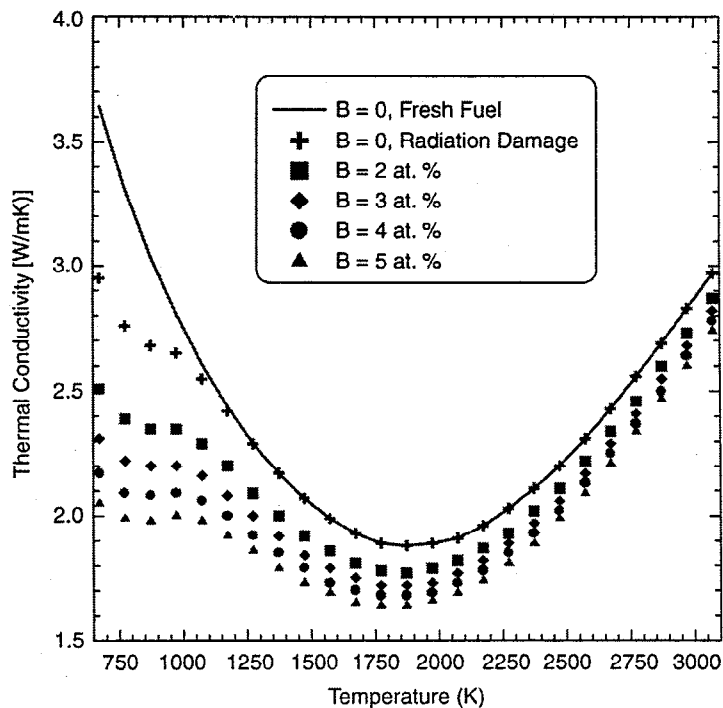


Fig. 6.4. Thermal conductivity of irradiated hypostoichiometric ($x = 0.02$) 95% dense MOX fuel.

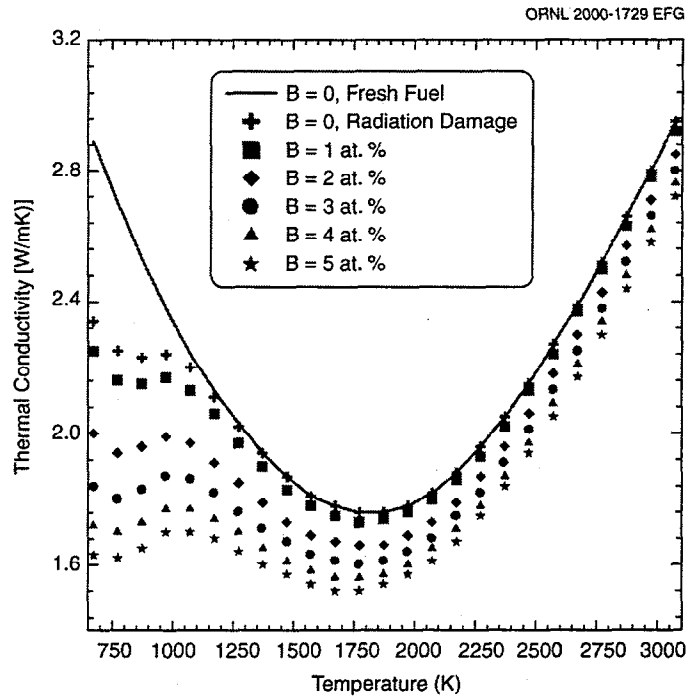


Fig. 6.5. Thermal conductivity of irradiated hypostoichiometric ($x = 0.05$) 95% dense MOX fuel.

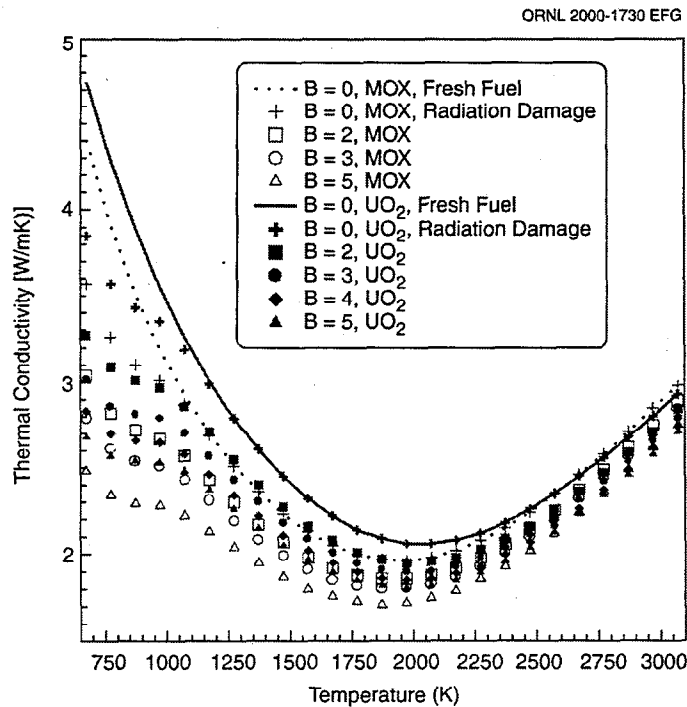


Fig. 6.6. Comparison of recommended equations for the thermal conductivity of irradiated stoichiometric 95% dense UO_2 and MOX fuels.

6.4 REFERENCES

1. J. H. Harding and D. G. Martin, *J. Nucl. Mater.*, **166**, 223–226 (1989).
2. D. T. Hagrman, Ed., *SCDAP/RELAP5/MOD3.1 Code Manual, Vol. 4: MATPRO—A Library of Materials Properties for Light-Water Reactor Accident Analysis*, USNRC Report NUREG/CR-6150 (EGGG-2720) (1993).
3. P. G. Lucuta, H. J. Matzke, R. A. Verrall, and H. A. Tasman, *J. Nucl. Mater.*, **188**, 198 (1992).
4. P. G. Lucuta, H. J. Matzke, and R. A. Verrall, *J. Nucl. Mater.*, **217**, 279 (1994).
5. P. G. Lucuta, H. J. Matzke, and R. A. Verrall, *J. Nucl. Mater.*, **223**, 51 (1995).
6. P. G. Lucuta, H. J. Matzke, and I. J. Hastings, *J. Nucl. Mater.*, **232**, 166–180 (1996).
7. Y. Philipponneau, *J. Nucl. Mater.*, **188**, 194–197 (1992).
8. W. Wiesenack, *Proc. ANS Inter. Topical Meeting on LWR Fuel Performance, Portland, Oregon*, pp. 507–511 (1997).
9. K. Ohira and N. Itagaki, *Proc. ANS International Topical Meeting on LWR Fuel Performance, Portland, Oregon*, pp. 541–549 (1997).
10. C. Ronchi et al., *J. Applied Phys.*, **85**, 776–789 (1999).
11. J. K. Fink, International Nuclear Safety Center Materials Property Data Base, <http://www.anl.insc.gov/matprop>, 1999.
12. C. Duriez, J-P. Alessandri, T. Gervais, and Y. Philipponneau, *J. Nucl. Mater.*, **277**, 143–158 (2000).
13. J. M. Casado, J. H. Harding, G. J. Hyland, *J. Phys. Condens. Matter.*, **6**, 4685 (1994).
14. J. C. Weilbacher, *High Temp.-High Pressures*, **4**, 431 (1972).

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