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THERMAL CONDUCTIVITY AND THERMAL
DIFFUSIVITY OF SOLID UO_2

by

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ABSTRACT

New equations for the thermal conductivity of solid UO_2 were derived based upon a nonlinear least squares fit of the data available in the literature. In the development of these equations, consideration was given to their thermodynamic consistency with heat capacity and density and theoretical consistency with enthalpy and heat capacity. Consistent with our previous treatment of enthalpy and heat capacity, 2670 K was selected as the temperature of a phase transition. A nonlinear equation, whose terms represent contributions due to phonons and electrons, was selected for the temperature region below 2670 K. Above 2670 K, the data were fit by a linear equation.

I. INTRODUCTION

A large number of publications exist on the thermal conductivity and thermal diffusivity of UO_2 . The most recent is a review by Brandt and Neuer,¹ who recommend an equation for thermal conductivity (k) of the form:

$$k = \frac{1}{A + BT} + DTe^{E/kT} + Fe^{G/kT} \quad (1)$$

where the first term is attributed to the contribution from phonons, the second term is due to electrons, and the third term is a conduction decrease owing to dislocations. The third term was first introduced by Weilbacher,² who found such a term essential to fit his high-temperature data.

In these analyses, the thermal-diffusivity data is converted to thermal conductivity *via* the relation

$$\kappa = \frac{k}{C_p \rho} \quad (2)$$

where k = thermal conductivity

κ = thermal diffusivity

C_p = heat capacity at constant pressure

ρ = density

New equations have been proposed for the heat capacity of UO_2 ^{3,4} since the analyses of thermal conductivity by Brandt and Neuer. The use of new equations for the calculation of the heat capacity used in Eq. 2 may significantly affect the fit of the combined data expressed as thermal conductivity because a significant portion of the data, particularly at high temperatures, is from thermal-diffusivity measurements. Thus, we have refit the combined thermal-diffusivity and thermal-conductivity data of UO_2 , using our recommended heat-capacity equations⁴ in Eq. 2. In fitting the combined thermal-conductivity and thermal-diffusivity data, we strived to maintain theoretical as well as thermodynamic consistency with related thermophysical properties.

II. METHODOLOGY

A. Selection of Data Base

The data base used for our analysis is essentially the same as that used by Brandt and Neuer.¹ Thermal-conductivity data consist of measurements by Godfrey *et al.*⁵ and Stora *et al.*⁶ and the round robin measurements at General Electric Co. and the Centre D'Etudes Nucleaires de Grenoble that are reported by Conway and Feith.⁷ Thermal-diffusivity data consist of the following: round robin measurements at Battelle Memorial Institute, Los Alamos Scientific Laboratory and General Electric Co. reported by Conway and Feith;⁷ round robin measurements at Battelle Memorial Institute reported by Bates;⁸ and measurements by Weilbacher.²

The only data excluded in the analysis are the data of Bates from cycles 1 and 2 of sample 1 and the heating portion of cycle 1 of sample 3 as recommended by Bates. Bates believes that during these heating cycles the uranium dioxide was changing from hypostoichiometric to nearly stoichiometric composition and that the results of these cycles are not meaningful. The results of these cycles were not reproducible. Bates found that his data were reproducible only after the samples were heated above 1470 K.

B. Method

First, the temperatures for the thermal-conductivity and thermal-diffusivity data were converted to the 1968 International Practical Temperature Scale. Second, the thermal diffusivities were converted to thermal conductivity using Eq. 2. For this conversion, the density equation recommended by O. D. Slagle⁹ based on measurements of Christensen¹⁰ was used. This equation is

$$\rho = \rho_0 \left(1.0056 - 1.6324 \times 10^{-5} T - 8.3281 \times 10^{-9} T^2 + 2.0176 \times 10^{-13} T^3 \right) \quad (3)$$

where $\rho_0 (= 10970 \text{ kg/m}^3)$ is the density of UO_2 at 273 K¹¹ and ρ is in kg/m^3 and T is in kelvins. The following equations were used to calculate the heat capacity of UO_2 to be used in Eq. 2. For $298 \leq T \leq 2670 \text{ K}$,

$$C_P = \frac{C_1 \theta^2 e^{\theta/T}}{T^2 (e^{\theta/T} - 1)^2} + 2 C_2 T + C_3 k e^{-E_a/kT} \left[1 + \frac{(T - T^*) E_a}{kT^2} \right] \quad (4)$$

where $C_1 = 78.212 \text{ J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$

$C_2 = 3.8616 \times 10^{-3} \text{ J}\cdot\text{mol}^{-1}\cdot\text{K}^{-2}$

$C_3 = 3.3993 \times 10^8 \text{ J}\cdot\text{mol}^{-1}\cdot\text{eV}^{-1}$

$E_a = 1.8815 \text{ eV}$

$\theta = 516.11 \text{ K}$

$T^* = 298.15 \text{ K}$

$k = 8.6144 \times 10^{-5} \text{ eV}\cdot\text{K}^{-1}$ (the Boltzmann constant)

In this equation, C_p is in $\text{J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1}$ and T is in kelvins. For $2670 \leq T \leq 3120 \text{ K}$,

$$C_p = 167.0 \text{ J}\cdot\text{mol}^{-1}\cdot\text{K}^{-1} \quad (5)$$

Third, nonlinear least-squares fits to the combined data for thermal conductivity and thermal diffusivity expressed as thermal conductivity were performed using the method described by Bevington.¹²

The general form of thermal-conductivity equation used is

$$k = \frac{1}{A + BT + CT^2} + DTe^{E/kT} + Fe^{G/kT} \quad (6)$$

with the three terms representing contributions from phonons, electrons, and dislocations, respectively. Brandt and Neuer chose a form with $C = 0$ and G fixed at -3.29 to fit the thermal-conductivity data. The various forms of conductivity expressions investigated in our study include (1) an equation with phonon and electronic terms, (2) an equation with phonon, electronic, and dislocation terms, and (3) the first form from 298 to 2670 K and the following linear equation above 2670 K:

$$k = L_1 + L_2 \cdot T \quad (7)$$

In these three forms, the phonon contribution was represented by either

$$\frac{1}{A + BT} \quad (8)$$

or

$$\frac{1}{A + BT + CT^2} \quad (9)$$

Equation 8 contains contributions due to phonon-impurity or grain-boundary collisions (the constant A) and contributions from phonon-phonon collisions also called umklapp three-phonon processes (the term BT). In addition to including contributions from these processes, Eq. 9 incorporates contributions

from phonon-phonon collisions by four-phonon processes. This contribution is in the CT^2 term. The temperature dependence in the second representation of the phonon contribution was suggested by Cabannes¹³ and Pomeranchuk.¹⁴

For convenience, the following terminology will be used to describe the various forms of Eq. 6 investigated:

$$\text{Form 1: } \frac{1}{A + BT} + DTe^{E/kT}$$

$$\text{Form 2: } \frac{1}{A + BT + CT^2} + DTe^{E/kT}$$

$$\text{Form 3: } \frac{1}{A + BT} + DTe^{E/kT} + Fe^{G/kT}$$

$$\text{Form 4: } \frac{1}{A + BT} + DTe^{E/kT} + Fe^{-3.29/kT}$$

$$\text{Form 5: } \frac{1}{A + BT + CT^2} + DTe^{E/kT} + Fe^{G/kT}$$

$$\text{Form 6: } \begin{cases} \frac{1}{A + BT + CT^2} + DTe^{E/kT} & \text{below 2670 K} \\ L_1 + L_2 \cdot T & \text{above 2670 K} \end{cases}$$

III. RESULTS AND DISCUSSION

Table 1 gives the best values for the constants in Eq. 6 for each form considered, the constants recommended by Brandt and Neuer, and the constants L_1 and L_2 for the linear equation. Table 2 gives the standard deviation of each form from (1) the combined data for thermal conductivity and thermal diffusivity expressed as thermal conductivity and (2) the thermal-diffusivity data from diffusivity values calculated using Eq. 2. Figures 1 through 6 show the fits of each functional form to the combined data expressed as thermal conductivity.

Examination of only the standard deviations of each form from the combined data expressed as thermal conductivity leads to the conclusion that form 5 is the best. However, this conclusion is not obvious from examination of the figures. Thus, further study is needed to determine whether or not the dislocation term is needed and which phonon term should be used. Comparison of forms 3 and 5, which have phonon, electronic, and dislocation terms, is inconclusive regarding the need for CT^2 in the phonon term (see Fig. 7). However, comparison of the standard deviations for forms 1 and 2, which have only phonon and electronic terms, shows that inclusion of the contribution from collisions due to four-phonon processes does improve the fit. Examination of Fig. 8 indicates that improvement is in the temperature region of 1300 to 2100 K.

Table 1. Parameters for Each Form of Thermal-Conductivity Equation Investigated

Form	Phonon Term			Electronic Term		Dislocation Term		Linear Equation	
	A $\times 10^2$	B $\times 10^4$	C $\times 10^8$	D $\times 10^2$	E	F	G	L ₁	L ₂ $\times 10^4$
1	5.3037	2.1669	--	4.7211	-1.0295	--	--	--	--
2	7.4229	1.4466	5.0182	1.3660	-0.68877	--	--	--	--
3	5.0526	1.5530	--	1.4538	-0.69807	-0.87495	2.3325×10^{-2}	--	--
4	5.3805	2.1504	--	0.92006	-1.5973	-1.2567×10^6	-3.29 ^a	--	--
5	7.1551	1.5493	4.1546	3.0292	-0.85907	-2.857×10^{-5}	-5.6543	--	--
6	6.8337	1.6693	3.1886	12.783	-1.1608	--	--	4.1486	-2.2673
BN ^b	4.39	2.16	--	11.2	-1.18	-4.18×10^5	-3.29 ^a	--	--

^aG is fixed at -3.29.^bThe Brandt-Neuer Equation

Table 2. Standard Deviations of Data from Each Function Form

Form	Standard Deviation, %	
	Thermal Conductivity	Thermal Diffusivity
1	6.53	7.51
2	6.15	6.88
3	6.15	6.88
4	6.26	7.07
5	6.05	6.68
6	6.23	6.58
BN	7.34	7.75

Fig. 1.

Fit of Combined Data Expressed
as Thermal Conductivity to
 $\frac{1}{A + BT} + DTe^{E/kT}$ (Form 1)

Fig. 2.

Fit of Combined Data Expressed
as Thermal Conductivity to
 $\frac{1}{A + BT + CT^2} + DTe^{E/kT}$ (Form 2)

Fig. 3.

Fit of Combined Data Expressed
as Thermal Conductivity to

$$\frac{1}{A + BT} + DTe^{E/kT} + Fe^{G/kT}$$

(Form 3)

Fig. 4.

Fit of Combined Data Expressed
as Thermal Conductivity to

$$\frac{1}{A + BT} + DTe^{E/kT} + Fe^{-3.29/kT}$$

(Form 4)

Fig. 5.

Fit of Combined Data Expressed
as Thermal Conductivity to

$$\frac{1}{A + BT + CT^2} + DTe^{E/kT} + Fe^{G/kT}$$

(Form 5)

Fig. 6.

Fit of Combined Data Expressed as
Thermal Conductivity to

$$\frac{1}{A + BT + CT^2} + DTe^{E/kT}$$

below 2670 K and to $L_1 + L_2 \cdot T$
above 2670 K (Form 6)

Fig. 7.

Fits of Combined Data Expressed
as Thermal Conductivity to
Form 3 (dashed curve) and
Form 5 (solid curve)

Fig. 8.

Fits of Combined Data Expressed
as Thermal Conductivity to
Form 1 (dashed curve) and
Form 2 (solid curve)

Comparison of Figs. 2 and 3 indicates that, graphically, forms 2 and 3 are equivalent; examination of the standard deviations of these forms given in Table 2 confirms this. Thus, in a form without CT^2 in the phonon term, the dislocation term will compensate for this missing phonon contribution. The dislocation term contributes to the fit of the high-temperature data only when Eq. 9 is used to represent the phonon contribution as in form 5 and when the exponent in the dislocation term is fixed at a negative value such as -3.29 , as was done by Brandt and Neuer and in form 4. Consequently, inclusion of the contribution due to four-phonon collisions is more statistically important than inclusion of a dislocation term. Examination of the standard deviations for forms 2 and 5 and of Fig. 9, in which the fits of the forms are compared, did not indicate whether inclusion of the dislocation term is statistically justified. This term appears to mainly contribute to the fit of eight high-temperature data points. Comparison of the deviations of individual data points from the fits of each of the forms and F-tests indicated that statistically the dislocation term should not be included.

Fig. 9.

Fits of Combined Data Expressed as Thermal Conductivity to Form 2 (solid curve) and Form 5 (dashed curve)

The dislocation term was introduced by Weilbacher,² who did not use a nonlinear least-squares method to fit his data for the entire temperature range of 773–3023 K. Instead, he divided the data into temperature-dependent regions and separately fit each temperature region by reducing the data to a form that could be fit with a linear equation. He assumed that, from 773 to 1800 K, the phonon term dominated and fit his data in this temperature region to Eq. 8. The phonon contribution was then subtracted

from the remaining data, which were fit to $DTe^{E/kT}$ by assuming that the electronic term dominated between 1800 and 2700 K. Since the phonon and electronic terms were not sufficient to fit his data above 2700 K, the contributions from these terms were subtracted from the data above 2700 K, and the remainder was fit to $Fe^{G/kT}$, which Weilbacher attributed to a conduction decrease due to dislocations.

In fitting these data, we considered consistency with other thermo-physical properties as well as closeness of fit. For theoretical consistency with the enthalpy and heat capacity of UO_2 , there should be evidence of a phase transition at 2670 K and only phonon and electronic contributions.^{4,15} Calculation of thermal diffusivity from the fits of thermal conductivity and comparison with experimental thermal-diffusivity data indicated that a linear equation for the thermal conductivity is needed above the transition temperature of 2670 K. Figure 10 is a plot of thermal diffusivities calculated from Eq. 2 with form 5 and experimental thermal-diffusivity data. A discontinuity occurs near 2670 K because at the transition temperature the heat capacity becomes constant. Similar discontinuities occur in plots of thermal diffusivity calculated with forms 1 through 4 and with the Brandt and Neuer equation. Consequently, the thermal conductivity data were fit to a non-linear equation below 2670 K and to a linear equation above 2670 K. This is form 6, which is shown graphically in Fig. 6. To prevent any discontinuities in the calculated values of the thermal diffusivity of UO_2 , the thermal-conductivity equation used to fit the combined data below 2670 K was constrained to agree with the linear equation at 2670 K. The use of a linear equation above 2670 K is consistent with the mathematical treatment of enthalpy and heat capacity.⁴

Fig. 10.

Thermal-Diffusivity Data along
with Fit to Thermal-Conductivity
Data Obtained from Form 5

IV. CONCLUDING REMARKS

The equations that we recommend for calculation of the thermal conductivity of UO_2 are given below. For $298.15 \leq T \leq 2670$ K,

$$k = \frac{1}{A + BT + CT^2} + DTe^{E/kT} \quad (10)$$

where $A = 6.8337 \times 10^{-2} \text{ m}\cdot\text{K}\cdot\text{W}^{-1}$

$B = 1.6693 \times 10^{-4} \text{ m}\cdot\text{W}^{-1}$

$C = 3.1886 \times 10^{-8} \text{ m}\cdot\text{W}^{-1}\cdot\text{K}^{-1}$

$D = 1.2783 \times 10^{-1} \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-2}$

$E = -1.1608 \text{ eV}$

$k = 8.6144 \times 10^{-5} \text{ eV}\cdot\text{K}^{-1}$ is the Boltzmann constant

For $2670 \leq T \leq 3120$ K,

$$k = 4.1486 - 2.2673 \times 10^{-4} T \quad (11)$$

In Eqs. 10-11, T is in kelvins and thermal conductivity is in $\text{W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$.

The parameters A through E in Eq. 10 were determined by the best nonlinear least square fit to the experimental thermal conductivity and thermal diffusivity data. The parameter E represents the electron activation energy which has been determined to be -1.0 eV from solid state theory. The value -1.1608 eV determined from the fit to the data is in reasonable agreement with this theoretical value. Unfortunately, the other parameters in Eq. 10 have not been independently determined from theory so no evaluation of their consistency with theory may be made.

Since the T^2 dependence in the phonon term improves the fit in the temperature region of 1300 to 2100 K, it was included in the nonlinear function. Figure 11 compares our fit of the data with that recommended by Brandt and Neuer.¹ While the equation given by Brandt and Neuer fits the low-temperature data better, overall our recommended equations are a better fit. The relative standard deviation for our fit is 6.23%, whereas that for the equation given by Brandt and Neuer is 7.34%.

For each set of data analyzed, Table 3 gives the number of experimental data points used, the temperature range of the data, and the relative standard deviation of the recommended equations from the experimental data. The data is grouped according to property measured. There appears to be no bias in the fit for thermal-conductivity or thermal-diffusivity measurements. Figure 6, which designates which data points are from thermal-conductivity measurements and which are from thermal-diffusivity measurements, confirms this. As shown, the majority of high-temperature measurements are thermal-diffusivity ones.

Fig. 11.

Fit of Experiment Data to Brandt-Neuer Equation (dashed curve) and Eqs. 10 and 11 (solid curve)

Table 3. Standard Deviations of Data Expressed as Conductivity from the Recommended Equations

Experimenter	Temperature Range, K	No. of Points	Standard Deviation, %	Property Measured	Reference	Year
Godfrey	323-1573	105	3.51	k	5	1964
Stora	473-2777	19	5.92	k	6	1964
General Electric Co. Nuclear Systems Programs	1229-2661	71	7.45	k	7	1969
Centre D'Etudes Nucleaires de Grenoble France	373-2577	14	7.12	k	7	1969
Battelle Memorial Institute, Ohio	457-2271	55	5.80	κ	7	1969
Los Alamos Scientific Lab.	299-2083	34	8.79	κ	7	1969
General Electric Co. Nuclear Systems Programs	575-1668	36	6.22	κ	7	1969
Bates, Sample 1	539-2388	46	4.08	κ	8	1970
Bates, Sample 2	361-1762	45	5.73	κ	8	1970
Bates, Sample 3	289-2777	97	7.88	κ	8	1970
Weilbacher Run 3	773-2973	16	3.91	κ	2	1972
Weilbacher Run 4	773-3023	16	5.67	κ	2	1972

Thermal-diffusivity values may be calculated from Eq. 2, using Eq. 3 for the density, Eqs. 4 and 5 for the heat capacity, and Eqs. 10 and 11 for the thermal conductivity. Figure 12 shows the fit of these calculated thermal diffusivities to the measured thermal diffusivities. The relative standard deviation of the calculated values from the measured data is 6.58%.

Fig. 12.

Thermal-Diffusivity Data and the Fit Obtained from Eqs. 10 and 11

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