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THE THERMAL DIFFUSIVITY
AND THERMAL CONDUCTIVITY
OF STOICHIOMETRIC $(U_{0.8}Pu_{0.2})O_2$

By

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THE THERMAL DIFFUSIVITY AND THERMAL
CONDUCTIVITY OF STOICHIOMETRIC $(U_{0.8}Pu_{0.2})O_2$

R. L. Gibby

INTRODUCTION

Mixed UO_2 -(20-25 wt%) PuO_2 has been selected as the reference driver fuel for the Fast Test Reactor (FTR). Thermo-physical property data are necessary to establish an optimum fuel design commensurate with the required operating conditions of the reactor.

Although the thermal properties of UO_2 have been extensively investigated, only a few thermal property measurements have been reported for mixed uranium-plutonium oxides. Thermal conductivity has been measured by Schmidt⁽¹⁾ to 800 °C, and by Hetzler, Lannin, Perry, and Zebroski⁽²⁾ at temperatures above 800 °C. In addition, Bailey and Craig^(3,4) and Bailey, Hines, and Zebroski⁽⁵⁾ have reported thermal conductivity data deduced from in-reactor experiments.

The data of Schmidt⁽¹⁾ and Hetzler et al.⁽²⁾ appear to be in agreement. However, because their measurements were made in different temperature intervals, a direct comparison is not possible. In general, the agreement is poor between the results from in-reactor experiments and laboratory steady-state measurements.

The oxygen stoichiometry of the mixed oxide can be varied rather extensively by heat treatment in either reducing or oxidizing atmospheres. Thus, the presence of oxygen vacancies or excess oxygen atoms in the mixed-oxide lattice may result in a substantial reduction in thermal diffusivity and thermal conductivity when compared with the stoichiometric mixed oxide because such imperfections can act as phonon scattering centers. Schmidt⁽¹⁾ has shown that this is indeed the case and that only

slight deviations from stoichiometry ($O/M = 2.00$) result in a marked reduction of the already-low thermal conductivity of $(U, Pu)O_2$.

The purpose of this investigation was to measure the thermal diffusivity of sintered and pneumatically impacted stoichiometric mixed-oxide samples, and to calculate thermal conductivity from the results for comparison with reported steady-state results. Measurements of nonstoichiometric samples ($O/M \neq 2.00$) are currently in progress and will be reported later.

Thermal diffusivity measurements were made from 100 °C to 1600 °C using the "flash" method developed by Parker, Jenkins, Butler, and Abbott.⁽⁶⁾ Thermal conductivity was calculated from the measured diffusivities and estimated heat capacities.

Utilization of the "flash" method to determine thermal conductivity indirectly by measuring thermal diffusivity offers several major advantages over the more common steady-state methods. First, the "flash" method is rapid, allowing measurements at several temperatures to be obtained in a brief period of time. Second, the simplicity of the experimental method eliminates many of the experimental problems encountered in steady-state methods. Third, since only small specimens are required for measurement, many of the problems encountered in specimen preparation are also eliminated. Finally, because of the small sample size, cracking problems resulting from temperature gradients in the material are not encountered. The obvious disadvantage is that heat capacity must be estimated or measured independently to calculate thermal conductivity. However, individual assessment of the contributions of thermal diffusivity and heat capacity to thermal conductivity may permit a more basic understanding of the conduction process.

SUMMARY AND CONCLUSIONS

SUMMARY

The thermal diffusivities of sintered (96% TD) and pneumatically-impacted, annealed (95% TD) $(U_{0.80}Pu_{0.20})O_{2.00}$ were measured by a laser heat-pulse apparatus. The thermal diffusivity (α) for temperatures from 100 to 1200 °C obeys the relationship

$$\alpha = \frac{1}{0.110 T - 8.5} \text{ (cm}^2\text{/sec)}$$

where $T = ^\circ K$. Above 1200 °C, the thermal diffusivity data fall above this reciprocal temperature curve. This effect is attributed to an approach of the phonon mean-free path to a minimum value corresponding to the lattice dimensions of the primitive metal atom sublattice.

The thermal conductivity was calculated as a function of temperature from the thermal diffusivity data. The heat capacity data required in the calculations were estimated from the heat capacities reported for UO_2 and PuO_2 . The calculated thermal conductivities are in good agreement with reported steady-state measurements on UO_2 -20 wt% PuO_2 . Below approximately 500 °C, the calculated thermal conductivities are slightly higher than for both UO_2 and UO_2 -20 wt% PuO_2 , while above 500 °C, the calculated values are slightly less. The thermal conductivity (K) between 200 and 1400 °C is given by the equation

$$K = \frac{1}{0.0307 T - 0.45} \text{ (W/cm } ^\circ K\text{)}$$

where $T = ^\circ K$. Below 200 °C, the thermal conductivity approaches a maximum value, while above 1400 °C, it is constant.

CONCLUSIONS

The laser heat-pulse apparatus yielded reproducible thermal diffusivity results on stoichiometric sintered and annealed pneumatically-impacted $(U_{0.80}Pu_{0.20})O_2$ specimens. Agreement between results for the two samples was, from 100 to 1200 °C, better than $\pm 5\%$. The accuracy of the results was estimated as $\pm 8\%$ from measurements made on Armco iron and Pyroceram 9606 thermal conductivity standards.

The thermal diffusivity of the unannealed pneumatically-impacted sample was approximately 16% lower than the annealed sample. The difference apparently is related to nonhomogeneity of the unannealed mixed oxide, although changes in grain size may have been partially responsible. The reciprocal thermal diffusivity of $(U_{0.80}Pu_{0.20})O_2$, linear from 100 to 1200 °C, indicates that thermal resistance arises from Umklapp processes. Thermal diffusivity in this temperature range is expressed by the relationship

$$\alpha = \frac{1}{0.110 T - 8.5} T(^{\circ}K)$$

Thermal diffusivity above 1200 °C is slightly higher than that calculated from the preceding expression. The minimum possible value for thermal diffusivity, assuming only lattice conduction with a phonon mean-free path equivalent to the unit cell dimensions of the primitive metal sublattice, is $5 \times 10^{-3} \pm 1 \times 10^{-3}$ cm²/sec, in good agreement with the observed minimum α of 5×10^{-3} cm/sec.

The thermal conductivity of $(U_{0.80}Pu_{0.20})O_{2.00}$ calculated from the thermal diffusivity data is in good agreement with steady-state measurements of thermal conductivity reported by other investigators. Within experimental error, the thermal conductivity of $(U_{0.80}Pu_{0.20})O_{2.00}$ is identical, between 100 °C

and 1300 °C, to the thermal conductivity of UO_2 . The thermal conductivity of the mixed oxide between 200 to 1400 °C is given by the relationship

$$K = \frac{1}{0.0307 T - 0.45} T(^{\circ}\text{K})$$

Below 200 °C, the thermal conductivity shows a negative deviation from the above relationship. This deviation is related to the rapid decrease in heat capacity. Above 1400 °C, the thermal conductivity of the mixed oxide reaches a minimum value consistent with a minimum in the phonon mean-free-path length.

EXPERIMENTAL PROCEDURE

EXPERIMENTAL METHOD

The "flash" method of measuring thermal diffusivity consists of instantaneously heating one surface of a thin sample with a heat pulse from an electron beam, a flash lamp or, as suggested by Deem and Wood,⁽⁷⁾ a laser and subsequently monitoring the transient temperature response of the rear surface of the sample with a thermocouple or an optical temperature monitor. When the energy pulse is absorbed uniformly over the front surface of the sample, when heat loss from the sample is negligible, when the pulse time of the heat source is negligible compared with the thermal pulse transit time through the sample, and when heat flow is normal to the heated surface, then thermal diffusivity can be determined from the shape of the temperature response curve using the simple relationship

$$\alpha = \frac{0.139 L^2}{t_{1/2}} \text{ (cm}^2\text{/sec)} \quad (1)$$

where L is the sample thickness (cm) and $t_{1/2}$ is the time (sec) to one-half the maximum temperature increase measured at the rear surface of the sample. By simply measuring L and $t_{1/2}$, α is readily determined from Equation (1).

When heat loss is not negligible, which is the case at high temperatures where radiation losses become appreciable, or when the pulse time of the heat source τ is only slightly less than the characteristic rise time t_c , the boundary conditions used in deriving Equation (1) are not satisfied and, consequently, thermal diffusivities determined from Equation (1) are not accurate.

The effects of radiation heat loss on measured thermal diffusivity using the "flash" method have been considered by Parker and Jenkins⁽⁸⁾ for heat losses from the pulse-heated surface of the sample. Cowan,⁽⁹⁾ however, has considered the case of general heat loss from both surfaces, and Cape and Lehman⁽¹⁰⁾ have studied radiation heat losses from a cylindrical specimen in both axial and radial directions. Cowan's method of correcting for general heat losses is particularly simple and is briefly described below. The method consists of determining from the temperature response curves either the ratio:

$$\Delta T(5 t_{1/2})/\Delta T(t_{1/2}); \text{ or } \Delta T(10 t_{1/2})/\Delta T(t_{1/2})$$

where ΔT is the incremental temperature rise above ambient temperature measured at the noted time intervals (multiples of the measured half-time). By using either of the above ratios, a quantity A is found from theoretical curves, where

$$A = \frac{\alpha t_{1/2}}{L^2} \quad (2)$$

When the value of either ratio is 2.00 (no heat loss condition), $A = 0.139$ and Equation (2) is identical to Equation (1). As the values of the ratios decrease with increasing heat losses, A also decreases. Consequently, thermal diffusivity calculated from Equation (1) will be high. Determination of A from the theoretical graphs requires a rough knowledge of the ratio of heat losses, r , from the front and rear surface of the sample. However, for radiation heat losses only, r can be estimated with sufficient accuracy from

$$r = \left(\frac{T_L}{T_O} \right)^3 \quad (3)$$

where T_L = temperature at front surface and T_O = temperature at rear surface. Normally, the temperature rise produced by the heat pulse at the front surface of the sample is small compared to ambient temperature; therefore, $r \approx 1$.

Cape and Lehman⁽¹⁰⁾ have noted that when the pulse time τ is comparable to the heat-pulse transit time through the sample, the rate of temperature rise on the rear surface of the sample is retarded. This has been referred to as the "finite-pulse-time" effect. They note that, while radiation heat losses are lessened by reducing sample thickness, finite-pulse-time effects are increased. Consequently, for a given material and a given τ , there is an optimum sample size which will result in minimum error when calculating α from Equation (1). However, it is not always possible or practical to use a sample of optimum size and, therefore, Taylor and Cape,⁽¹¹⁾ in their analysis of the finite-pulse-effect, constructed a graph of τ/t_c versus $t_{1/2}/t_c$, where t_c is a characteristic of time defined by the relationship

$$t_c \equiv \frac{L^2}{\pi \alpha} \quad (4)$$

and from which t_c can be determined from measurements of $t_{1/2}$ and hence α can be determined from Equation (4) for a square-wave pulse (which approximates a laser pulse) or a saw-tooth pulse (which approximates a flash lamp pulse). The equation of their linear graph for a square-wave pulse, rearranged to useful form, is

$$t_c = \frac{t_{1/2} - 0.55 \tau}{1.37} \quad (5)$$

When $t_c > \sim 50 \tau$, Equation (1) is accurate in calculating α to $\pm 1\%$. When $t_c < 50 \tau$, Equation (5) can be used in conjunction with Equation (4) to calculate α .

APPARATUS

The laser heat-pulse apparatus is shown schematically in Figure 1. Its essential components are (1) a ruby laser, (2) a sample furnace, and (3) a photodiode temperature sensor.

Laser

The sample was transiently heated with a five-joule pulse from a ruby laser ($\lambda = 6943 \text{ \AA}$). The laser head assembly consisted of a 1.3 cm diam x 15.2 cm ruby rod enclosed in an Electro Powerpac Model 4120 elliptical laser cavity. The laser cavity was continuously purged with nitrogen during operation to prevent oxidation discoloration of the elliptical mirrors from the heat generated by the flash lamp.

The laser pulses obtained were noted to approximate a square wave as shown in the oscilloscope trace in Figure 2. This trace was obtained using an indium antimonide photodiode infrared detector. Neutral density filters were used between the laser head and the detector to sufficiently attenuate the laser pulse for measurement. The output from the detector was recorded on an oscilloscope and photographed with a polaroid

FIGURE 1. Schematic of Thermal Diffusivity Apparatus

camera attachment. The laser pulses occurred approximately 1.2×10^{-3} sec after the flash lamp was triggered and lasted approximately 1.5×10^{-3} sec.

FIGURE 2. *Oscilloscope Trace of Ruby Laser Pulse
(sweep time = 0.5 msec/cm)*

Furnace

The sample was heated in a molybdenum-rhenium wound Marshall tube furnace. Power requirements for the maximum temperature (1700 °C) were 3 kW at 110 volts. Power input was hand regulated with a rheostat.

The furnace was located inside a glove box for measuring plutonium-bearing or plutonium-contaminated materials. Site ports on opposite sides of the glove box, were located coincident with the axis of the furnace tube to provide direct light paths between the sample and the laser, and between the sample and the photodiode detector. The ends of the furnace tube, enclosed for atmosphere control, also contained windows. Quartz windows were provided on the laser side of the sample, and sapphire, with better infrared transmission characteristics, was used on the photodiode side.

Ambient sample temperature was measured with a Pt - Pt/10% Rh thermocouple located immediately adjacent to the sample. The thermocouple output was measured with a Hewlett Packard Model 3440-A digital voltmeter equipped with a Hewlett Packard Model 3443-A high gain/auto range plug-in unit.

Transient Temperature Monitoring System

The temperature rise on the rear surface of the sample resulting from the instantaneous heating of the front surface with the laser pulse was measured with an indium antimonide (InSb) infrared detector manufactured by the Davers Corporation. The detector was liquid-nitrogen cooled for maximum sensitivity and optimum signal-to-noise ratio.

The thermal radiation emitted from the rear surface of the samples was focused on the detector with a silicon lense. Silicon is transparent only at wavelengths greater than 1μ . Thus, the lense also protected the detector from accidental irradiation by the direct laser beam ($\lambda = 0.6943 \mu$). A sight tube was also placed between the sample and detector to allow only the central area on the rear surface to be viewed by the detector, thus minimizing radial heat flow effects in the measurements.

The output signal from the detector was amplified with a Tektronic Type E plug-in unit, displayed on the cathode ray tube of a Tektronic 535A oscilloscope, and photographed with a polaroid camera attachment. A typical temperature response curve is shown in Figure 3. The minimum ambient sample temperature on which a measurable temperature rise was detectable with the system was approximately $100^\circ\text{C} \pm 50^\circ\text{C}$, depending on thickness and heat capacity of the material being measured.

The output from the delay sweep circuit of the oscilloscope was amplified to trigger the laser a short time after starting the trace. Discharge of the capacitors in the power

supply across the flash tube caused a small disturbance on the trace, which was conveniently used as the zero time in measuring $t_{1/2}$ from the transient response curve.

FIGURE 3. *Oscilloscope Trace of Temperature Transient on Rear Surface of $(U_{0.80}Pu_{0.20})O_{2.00}$ Pellet (sample temp. = 304 °C. sweep time = 100 msec/cm)*

Accuracy and Precision

The accuracy of the method was estimated by comparing thermal diffusivity measurements on Armco iron and Pyrocera Code 9606 with the results of other investigators. The comparatively large difference between the thermal diffusivities of Armco iron and Pyrocera allowed accuracy of the method to be evaluated for a broad range of thermal diffusivities. The sizes of the three Armco iron samples studied were 0.63 cm diam x 0.0704 cm, 0.63 cm diam x 0.0711 cm, and 0.63 cm diam x 0.0544 cm. Both surfaces of the Pyrocera sample were coated with a colloidal graphite suspension to prevent transmission of the laser beam through the sample. All calculations were corrected for finite-pulse-time effects using Equation (4),

and for heat loss using Cowan's method. Although the derivation of the finite-pulse-time effect assumes negligible heat losses, and the heat loss correction assumes negligible finite-pulse-time effects, utilization of both corrections for the Armco iron data resulted in good agreement of the data with reported values. The thermal diffusivity of Armco iron is compared in Figure 4 with the data of Fulkerson, Moore, and McElroy;⁽¹³⁾ Cody, Abeles, and Beers;⁽¹⁴⁾ Laubitz,⁽¹⁵⁾ and Shanks, Klein, and Danielson.⁽¹⁶⁾

The thermal diffusivity of Pyroceram 9606 is compared in Figure 5 with the data of Flieger;⁽¹⁷⁾ Plummer, Campbell, and Cowstock;⁽¹⁸⁾ and Rudkin.⁽¹⁹⁾

The thermal diffusivities determined for Armco iron agree to within $\pm 4\%$ with data reported in the literature. Pyroceram data were within $\pm 2\%$ of Rudkin's data, also determined by the "flash" method, and within 8% of the values determined by Plummer, Campbell, and Comstock. In general, the thermal diffusivities of Armco iron and Pyroceram 9606 determined in this investigation were slightly less than most reported values. This result may be related to the simultaneous application of both heat loss and finite-pulse-time corrections.

The maximum error in the thermal diffusivity determinations was estimated to be $\pm 9\%$, while the most probable error, assuming random independent errors, was estimated to be $\pm 5\%$. Major sources of error were: oscilloscope sweep time $\pm 3\%$, sample thickness $\pm 1\%$, and heat loss and finite pulse corrections (estimate) $\pm 4\%$. Experimental results on the Pyroceram and the mixed oxide indicated the precision to be $\pm 5\%$ for measurements made above approximately 300 °C, and $\pm 8\%$ at lower temperatures where the infrared detector was less sensitive to small temperature transients. A much greater scatter was noted in results for the Armco iron samples at all temperatures due to smaller temperature transients produced in the iron by the laser heat pulse.

FIGURE 4. Thermal Diffusivity of Armco Iron

FIGURE 5. Thermal Diffusivity of Pyroceram 9606

SAMPLE PREPARATION AND CHARACTERIZATION

A high-purity ($\text{U}_{0.80}\text{Pu}_{0.20}$) O_2 powder was prepared from a mixed plutonium-uranyl nitrate solution using a co-precipitation process. The co-precipitate was calcined for approximately one hour at 1000 °C in a nitrogen-6 vol% H_2 atmosphere to obtain an oxide powder.

Analysis of the powder by X-ray diffraction showed only a single phase. However, the back-reflection diffraction lines were broad and poorly resolved, indicating either (1) a non-homogeneous solid solution, or (2) fine particle size.

The plutonium and uranium contents of the powder, determined by isotopic dilution analysis, were found to be 0.20 mole fraction plutonium and 0.80 mole fraction uranium.

The dense specimens required for thermal diffusivity measurements were prepared from the powder by two methods: (1) cold pressing and sintering, and (2) pneumatic impaction.

Dense sintered bodies were prepared as follows: The powder was pressed into pellets of 0.954 cm diam x 1.5 cm, at 3000 dyne/cm². Approximately two drops of water per gram of powder was required to form strong, lamination-free pellets. After drying, the pellets were heated over a 24-hr period to a sintering temperature of 1650 °C, allowed to soak at this temperature for 16 hr, and slowly cooled to room temperature. The entire sintering process was performed in a protective atmosphere of flowing argon-8 vol% H_2 . The density of the sintered pellets was 10.60 g/cm³ or 96% TD (theoretical density).

The pneumatic impaction process consisted of placing the powder in a small stainless-steel can, loading into a mild-steel die assembly, and then enclosing the entire assembly in a larger stainless-steel can. Void areas between the die and the outer can were filled with alumina powder. After sealing, the

complete assembly was evacuated through a small tube in the outer can and simultaneously heated to 1200 °C. The hot assembly was then impacted at 20,000 atmospheres. The densified material, upon removal from the inner container by machining, fractured into many pieces. The larger pieces were salvaged for specimens. The density of the impacted material, essentially the same as the sintered material, was 10.57 g/cm³ (95.5% TD). Properties of the densified material are summarized in Tables I and II.

TABLE I. *Properties of Pneumatically Impacted Material*
(U_{0.80}Pu_{0.20})O₂

Lattice Parameter, O/M = 2.00	5.4553 ± 0.0003 Å
Weight Percent Plutonium (Isotopic Dilution)	17.6%
Density (Mercury Displacement at 10 μ)	10.57 g/cm ³
Percent of Theoretical Density (11.08 g/cm ³)	96%
Grain Size	Unresolvable at 500X
Chemical Impurities (Spectrochemical)	
Ag 1 ppm	Mg <5 ppm
Al 50	Mn 5
As <20	Mo 20
B 10	Na 5
Bi <1	Ni 23
Ca 2	P <5
Cd <1	Pb <1
Cr 45	Si 320
Cu 10	Sm 14
Fe 68	Ti <1
Ge <10	V <10
K 20	Zn 2
Li <5	

TABLE II. *Properties of Sintered Material*
 $(U_{0.80}Pu_{0.20})O_{2.00}$

Lattice Parameter, O/M = 2.00	5.4553 ± 0.0003 Å		
Weight Percent Plutonium (Isotopic Dilution)	17.6%		
Density (Mercury Displacement at 10 μ)	10.60 g/cm ³		
Percent of Theoretical Density (11.08 g/cm ³)	96%		
Grain Size	30 μ		
Chemical Impurities (Spectrochemical)			
Ag	<1 ppm	Mg	10 ppm
Al	10	Mn	2
As	<20	Mo	<5
B	<1	Na	5
Bi	<1	Ni	50
Ca	20	P	<100
Cd	<1	Pb	2
Cr	50	Si	5
Cu	2	Sm	20
Fe	<25	Tl	<1
Ge	<10	V	<10
K	<5	Zn	50
Li	<5		

Pieces of sintered and pneumatically impacted material were polished and etched to observe the microstructures. A well-defined microstructure was produced by etching for 60-90 min with a solution of 80 ml H₂O, 20 ml HNO₃, 3 drops HF, and 0.1 g Ce (NO₃)₃ · 0.6H₂O. Photomicrographs of the sintered and pneumatically impacted specimens are shown in Figures 6 and 7, respectively. The average grain size of the sintered material was approximately 30 μ, while that of the impacted material was not resolvable at 500X. Porosity in the sintered material was located both in the grain boundaries and in the grains.

FIGURE 6. Photomicrograph (250X) of Sintered $(U_{0.80}Pu_{0.20})O_{2.00}$

FIGURE 7. Photomicrograph (500X) of Pneumatically Impacted $(U_{0.80}Pu_{0.20})O_{2.00}$

The oxygen-to-metal ratio (O/M) of the material was determined from weight-gain measurements following a six-hour heat treatment at 800 °C in an atmosphere of argon-8 vol% H₂ passed over water at 0 °C. The O/M of the sintered material was 1.99, while that of the impacted material was 2.00. The samples, during thermal diffusivity experiments, were contained in a flowing atmosphere of argon-8 vol% H₂ passed over water at 0 °C to maintain stoichiometric composition at elevated temperatures. In this atmosphere, an O/M = 2.00 would be maintained to approximately 1350 °C.⁽¹²⁾

A single-phase fluorite structure was observed in both sintered and pneumatically impacted material from Debye-Scherrer X-ray diffraction patterns. The lattice parameter was $a_0 = 5.4553 \pm 0.0005 \text{ \AA}$, corresponding to the value expected from a Vegard's law relationship. The resolution of the back-reflection lines of the pneumatically impacted material was equally as poor as those of the starting powder. An alpha-autoradiograph, however, indicated plutonium distribution to be uniform within the resolving power of the microscope (0.5 μ).

Both materials were sliced with a diamond saw into thin sample wafers. Measurements reported in this investigation were made on one sample of pneumatically impacted material (GCP) 0.63 cm diam x 0.0610 cm, and one sample of sintered material (GCPB) 0.63 cm diam x 0.0734 cm.

RESULTS AND DISCUSSION

THERMAL DIFFUSIVITY

The thermal diffusivity of (U_{0.80}Pu_{0.20})O_{2.00} is shown in Figure 8 as a function of temperature. The experimental data are tabulated in Table III for the sintered material, and Tables IV and V for the pneumatically impacted material. All data were corrected for heat-loss effects using Cowan's method, and for finite-pulse-time effects using Equations (4) and (5) and a laser pulse time, τ , of 0.0015 seconds.

FIGURE 8. Thermal Diffusivity of $(U_{0.80}Pu_{0.20})O_{2.00}$

TABLE III. Thermal Diffusivity ($U_{0.80}Pu_{0.20}$) $O_{2.00}$ Sintered
(GCPB $L = 0.0734$ cm)

Temp. °C	$t_{1/2}$ sec	α cm ² /sec	Temp. °C	$t_{1/2}$ sec	α cm ² /sec
105	0.0281	0.0241	1498	0.0991	0.00576
154	0.0290	0.0232	1544	0.1018	0.00550
172	0.0289	0.0237	1597	0.0981	0.00577
189	0.0296	0.0234	1550	0.0956	0.00603
216	0.0315	0.0218	1494	0.0975	0.00590
243	0.0331	0.0206	1452	0.1021	0.00541
306	0.0373	0.0182	1402	0.0964	0.00610
369	0.0414	0.0164	1351	0.0954	0.00611
400	0.0436	0.0154	1306	0.0924	0.00642
478	0.0493	0.0135	1220	0.0911	0.00660
520	0.0516	0.0129	1175	0.0893	0.00681
566	0.0542	0.0122	1142	0.0873	0.00695
608	0.0577	0.0116	1100	0.0860	0.00695
673	0.0620	0.0105	1050	0.0857	0.00698
708	0.0634	0.0103	1014	0.0842	0.00720
751	0.0647	0.0101	943	0.0791	0.00784
801	0.0696	0.00915	880	0.0771	0.00803
849	0.0728	0.00885	847	0.0727	0.00867
928	0.0755	0.00831	795	0.0711	0.00901
956	0.0784	0.00792	754	0.0677	0.00951
1020	0.0799	0.00769	701	0.0649	0.00997
1080	0.0830	0.00728	637	0.0603	0.0109
1119	0.0857	0.00699	593	0.0573	0.0115
1151	0.0872	0.00683	546	0.0537	0.0125
1200	0.0897	0.00672	240	0.0329	0.0208
1252	0.0918	0.00640	231	0.0320	0.0214
1299	0.0929	0.00626	172	0.0293	0.0234
1352	0.0949	0.00614	144	0.0267	0.0256
1415	0.0963	0.00605	108	0.0257	0.0271
1449	0.0948	0.00609	90	0.0247	0.0274

TABLE IV. *Thermal Diffusivity ($U_{0.80}Pu_{0.20}$) $O_{2.00}$ Pneumatically Impacted (GCP $L = 0.0610$ cm)*

Temp. °C	$t_{1/2}$ sec	cm^2/sec	Temp. °C	$t_{1/2}$ sec	cm^2/sec
140	0.0195	0.0232	1141	0.0651	0.00656
211	0.0229	0.0212	1248	0.0717	0.00560
254	0.0275	0.0173	1318	0.0727	0.00563
285	0.0278	0.0172	1371	0.0696	0.00598
347	0.0311	0.0142	1419	0.0741	0.00536
389	0.0339	0.0138	1497	0.0801	0.00484
729	0.0528	0.00847	1507	0.0710	0.00549
829	0.0529	0.00880	1454	0.0756	0.00529
862	0.0545	0.00840	1386	0.0746	0.00525
895	0.0566	0.00790	1327	0.0738	0.00538
932	0.0576	0.00775	1238	0.0623	0.00651
974	0.0596	0.00762	1151	0.0626	0.00640
1028	0.0666	0.00633	984	0.0616	0.00706
1037	0.0594	0.00691	761	0.0527	0.00862
1114	0.0681	0.00628			

At temperatures below 800 °C, the thermal diffusivity of the unannealed pneumatically impacted material was approximately 16% lower than that of the sintered material. However, after heating the impacted material to 1600 °C, the thermal diffusivity at the lower temperatures increased and agreed to better than $\pm 5\%$ with that of the sintered material. It is impossible to conclude from the available data what structural change or changes in the impacted mixed oxide were responsible for the slight improvement in thermal diffusivity after annealing. However, three possible candidates were determined to be (1) grain growth, (2) improvement in solid solution homogeneity, and/or (3) relaxation of residual strain imparted during the impaction process.

TABLE V. Thermal Diffusivity ($U_{0.80}Pu_{0.20}O_{2.00}$) Pneumatically-Impacted-Annealed (GCP-1 $L = 0.0610$ cm)

Temp. °C	$t_{1/2}$ sec	α cm ² /sec	Temp. °C	$t_{1/2}$ sec	α cm ² /sec
87	0.0179	0.0283	835	0.0496	0.00915
116	0.0200	0.0244	870	0.0518	0.00876
129	0.0193	0.0246	929	0.0534	0.00857
158	0.0183	0.0260	1005	0.0566	0.00787
186	0.0201	0.0234	1069	0.0573	0.00782
228	0.0219	0.0218	1096	0.0598	0.00741
242	0.0225	0.0211	1163	0.0601	0.00724
254	0.0232	0.0200	1239	0.0619	0.00693
288	0.0250	0.0186	1258	0.0634	0.00722
322	0.0264	0.0180	1006	0.0585	0.00750
360	0.0283	0.0165	916	0.0531	0.00846
422	0.0316	0.0150	831	0.0496	0.00915
431	0.0327	0.0141	716	0.0453	0.0102
459	0.0331	0.0142	600	0.0381	0.0123
491	0.0352	0.0133	468	0.0342	0.0136
535	0.0369	0.0127	442	0.0325	0.0144
561	0.0375	0.0125	338	0.0259	0.0181
652	0.0424	0.0110	304	0.0237	0.0192
696	0.0437	0.0105	268	0.0237	0.0194
724	0.0441	0.0104	195	0.0203	0.0232
764	0.0472	0.00980	167	0.0226	0.0208

As shown in the photomicrograph (Figure 9), the grain size of the impacted material was extremely fine ($<0.5 \mu$). However, according to Charvat and Kingery,⁽²⁰⁾ the grain size of a poor conductor such as the mixed-oxide must be much smaller than 0.5μ before grain boundaries would measurably affect thermal conductivity (or diffusivity). Since the microstructure of the impacted material could not be resolved with an optical microscope, the actual grain size of the mixed oxide was not

a. As-Impacted

b. After Annealing at 1600 °C

FIGURE 9. *Photomicrographs (500X) Showing Microstructure of Pneumatically Impacted Material*

determined and, consequently, estimates of grain-size effects on thermal diffusivity could not be determined.

The evidence in support of solid-solution heterogeneity as the cause of lower thermal diffusivity in the as-impacted materials is found in the X-ray diffraction patterns of the calcined co-precipitated powder and the pneumatically impacted material, both of which showed poorly resolved high angle diffraction peaks. Although such effects might also be associated with fine particle size, mixed oxide powders made from the co-precipitation process and displaying sharp diffraction lines have indicated a calcination temperature of 1000 °C as sufficiently high to eliminate particle-size line broadening. Moreover, since the impacted material was heated to 1200 °C prior to impaction, it is even less likely that grain size was responsible for the poor resolution of diffraction lines. Since the increase in thermal diffusivity appeared to be associated with improved resolution of high angle X-ray lines, solid-solution homogeneity appears, lacking more conclusive evidence, to be responsible for the differences in thermal diffusivity of the impacted and annealed material.

The effect of residual strain from pneumatic impaction on thermal diffusivity is probably very slight. Experimental evidence for pneumatically impacted UO_2 shows the stored energy from the impaction process to be less than 2 cal/g. Measurements on pneumatically impacted PuO_2 also indicated residual strain to be negligible. It is of interest to note, however, that residual strain in impacted UO_2 was sufficient to result in recrystallization at 1400 °C. The heterogeneous grain structure of the annealed mixed oxide sample shown in Figure 9 may, in fact, be related to a recrystallization process.

The thermal resistance of a dielectric solid above the Debye temperature results from the phonon interactions called Umklapp processes. The phonon mean-free path varies inversely

with absolute temperature, and thermal diffusivity can be related to the phonon mean-free path by the classical relationship

$$\alpha = \left(\frac{1}{3}\right) \ell \bar{V} \quad (6)$$

where ℓ is the mean-free path and $\bar{V}$ is an average phonon velocity. According to this relationship, and assuming the average phonon velocity to be independent of temperature, α will vary inversely with absolute temperature. The reciprocal thermal diffusivity for sintered and pneumatically impacted material $(\text{U}_{0.80}\text{Pu}_{0.20})\text{O}_{2.00}$ is shown in Figures 10 and 11, respectively. All data fall on a line satisfying the relationship

$$1/\alpha = AT + B \quad (7)$$

The values under A and B in Table VI are for the three curves in Figures 10 and 11. The experimental data for the sintered sample is plotted in Figure 8 along with the empirical relationship [Equation (7)].

TABLE VI. Values of A and B for the Mixed Oxide Receiving Different Treatment

<u>Material</u>	<u>A</u>	<u>B</u>
Pneumatically-Impacted	0.110	-1.5
Pneumatically-Impacted Annealed	0.104	-3.6
Sintered	0.110	-8.5

The negative deviation of reciprocal thermal diffusivity above 1450 °K suggests thermal resistance approaching a maximum value. A similar result has been observed for UO_2 .⁽²⁴⁾ Here, the deviation from a reciprocal temperature relationship at higher temperatures was attributed to a small electronic

FIGURE 10. Reciprocal of Thermal Diffusivity of Sintered $(U_{0.80}Pu_{0.20})O_{2.00}$ as a Function of Absolute Temperature

FIGURE 11. Reciprocal of Thermal Diffusivity of Pneumatically Impacted $(U_{0.80}Pu_{0.20})O_{2.00}$ as a Function of Absolute Temperature

contribution to thermal conductivity. The thermal resistance, however, might also be expected to approach a maximum value as the phonon-mean-free path length approaches a minimum value, assuming, of course, that phonon velocity is independent of temperature. Consequently, a minimum thermal diffusivity based on a minimum phonon scattering distance can be estimated using Equation (5).

If one assumes that the minimum phonon mean-free path corresponds to the lattice parameter, a_0 , of the mixed-oxide fluorite unit cell, and that the phonon velocity is equal to the sound velocity in the solid which, for UO_2 , is estimated as approximately 4×10^5 cm/sec, then the minimum thermal diffusivity expected would be

$$\alpha = \frac{(5.45 \times 10^{-8})(4 \times 10^5)}{3} = 7 \times 10^{-3} \text{ cm}^2/\text{sec}$$

The lowest determined value for α in this experiment was 5×10^{-3} cm²/sec, somewhat lower than the calculated value, but in fair agreement considering the rough assumptions. However, a lower minimum value of α results if one assumes that the oxygen or the metal atom sublattices constitute the minimum scattering distance in the fluorite structure. Thus, for the simple cubic oxygen sublattice, $\ell_o = 2.73 \times 10^{-8}$ cm, and for the primitive face-centered cubic (rhombohedral symmetry) metal sublattice, $\ell_m = 3.90 \times 10^{-8}$ cm. Consequently,

$$\alpha_o = \frac{(2.73 \times 10^{-8})(4 \times 10^5)}{3} = 4 \times 10^{-3} \text{ cm}^2/\text{sec}$$

$$\alpha_m = \frac{(3.90 \times 10^{-8})(4 \times 10^5)}{3} = 5 \times 10^{-3} \text{ cm}^2/\text{sec}$$

Based on these assumptions, thermal diffusivities as low as $5 \times 10^{-3} \pm 1 \times 10^{-3}$ cm²/sec are predicted.

THERMAL CONDUCTIVITY

The calculated thermal conductivity of $(U_{0.80}Pu_{0.20})O_{2.00}$ is shown in Figure 12 as a function of temperature. Thermal conductivity was calculated from the thermal diffusivity data using the relationship

$$K = 4.183 \alpha \rho C_p \text{ (W/cm } ^\circ\text{K)}$$

where ρ is the density (g/cm^3), and C_p is the heat capacity ($\text{cal/g } ^\circ\text{K}$). The thermal conductivity data are compiled in Tables VII and VIII. The variation of density with temperature was estimated from the X-ray lattice expansion measurements on $(U_{0.80}Pu_{0.20})O_{2.00}$.⁽²¹⁾ Heat capacity, reported in the literature, was estimated from UO_2 ⁽²²⁾ and PuO_2 ⁽²³⁾ heat capacity data by assuming the mixed oxide heat capacity to be a linear function of the mole fraction of UO_2 and PuO_2 in the solid solution (a Kopp's law relationship).

The thermal conductivities calculated from the thermal diffusivity measurements are in reasonable agreement ($\pm 20\%$) with results reported from steady-state measurements⁽²⁻³⁾ (Figure 13). A more meaningful comparison could be made if measured heat capacity data were available. The results of this investigation indicate, as do the experiments of Schmidt⁽²⁾ and Hetlzer et al.,⁽³⁾ that the thermal conductivity of the mixed oxide is essentially the same as that for pure UO_2 .⁽²⁴⁾ This result is not unexpected inasmuch as it confirms Klemens⁽²⁵⁾ postulation that introduction of a substitutional atom decreases thermal conductivity only if there is (1) a mass difference, (2) an atomic size difference, and/or (3) a bonding energy difference. Since the mass and size of the uranium⁺⁴ and plutonium⁺⁴ ions are very similar, one would not expect an appreciable change in thermal conductivity when substituting plutonium⁺⁴ for uranium⁺⁴. At very high

FIGURE 12. Calculated Thermal Conductivity of $(U_{0.80}Pu_{0.20})O_{2.00}$

TABLE VII. Calculated Thermal Conductivity ($U_{0.80}Pu_{0.20}O_{2.00}$ Sintered (GCPB))

Temp. °C	C _p cal/gm °C	K W/cm °C	K ⁻¹ cm °C/W	Temp. °C	C _p cal/gm °C	K W/cm °C	K ⁻¹ cm °C/W
105	0.0634	0.0676	14.8	1498	0.0819	0.0198	50.5
154	0.0655	0.0677	14.8	1544	0.0831	0.0192	52.1
172	0.0656	0.0700	14.3	1597	0.0845	0.0205	48.9
189	0.0675	0.0700	14.3	1550	0.0830	0.0210	47.6
216	0.0687	0.0664	15.1	1494	0.0820	0.0203	49.3
243	0.0698	0.0635	15.7	1452	0.0809	0.0185	54.0
306	0.0717	0.0577	17.3	1402	0.0802	0.0206	48.5
369	0.0732	0.0534	18.7	1351	0.0791	0.0205	48.8
400	0.0739	0.0504	19.8	1306	0.0784	0.0213	47.0
478	0.0749	0.0449	22.3	1220	0.0778	0.0218	45.9
520	0.0755	0.0433	23.1	1175	0.0773	0.0224	44.6
566	0.0758	0.0410	24.4	1142	0.0769	0.0226	44.2
608	0.0760	0.0386	25.9	1100	0.0766	0.0227	44.0
673	0.0764	0.0351	28.5	1050	0.0766	0.0230	43.5
708	0.0764	0.0342	29.2	1014	0.0765	0.0238	42.0
751	0.0765	0.0337	29.7	943	0.0765	0.0258	38.8
801	0.0765	0.0304	32.9	880	0.0765	0.0267	37.5
849	0.0765	0.0294	34.0	847	0.0765	0.0286	35.0
928	0.0765	0.0274	36.5	795	0.0765	0.0300	33.3
956	0.0765	0.0261	38.3	754	0.0765	0.0318	31.5
1020	0.0765	0.0252	39.7	701	0.0764	0.0333	30.0
1080	0.0767	0.0240	41.6	637	0.0764	0.0365	27.4
1119	0.0769	0.0228	43.9	593	0.0759	0.0383	26.1
1151	0.0770	0.0224	44.6	546	0.0757	0.0420	23.8
1200	0.0774	0.0220	45.4	240	0.0698	0.0645	15.5
1252	0.0779	0.0211	47.4	231	0.0687	0.0650	15.4
1299	0.0783	0.0208	48.1	172	0.0660	0.0685	14.6
1352	0.0791	0.0206	48.6	144	0.0646	0.0734	13.6
1415	0.0803	0.0205	48.9	108	0.0631	0.0760	13.2
1449	0.0810	0.0208	48.1	90	0.0620	0.0755	13.3

TABLE VIII. *Calculated Thermal Conductivity ($U_{0.80}Pu_{0.20}O_{2.00}$)
Pneumatically-Impacted-Annealed (GCP-1)*

Temp. °C	C_p cal/gm °C	K W/cm °C	K^{-1} cm °C/W	Temp. °C	C_p cal/gm °C	K W/cm °C	K^{-1} cm °C/W
87	0.0621	0.0779	12.9	764	0.0765	0.0328	30.5
116	0.0639	0.0690	14.5	835	0.0765	0.0304	32.9
128	0.0646	0.0705	14.2	870	0.0765	0.0291	34.4
158	0.0660	0.0760	13.2	929	0.0765	0.0291	35.5
186	0.0673	0.0698	14.3	1005	0.0764	0.0259	38.6
228	0.0692	0.0669	14.9	1069	0.0764	0.0257	38.9
242	0.0699	0.0653	15.3	1096	0.0767	0.0245	40.8
254	0.0701	0.0622	16.1	1163	0.0773	0.0239	41.8
288	0.0711	0.0586	17.1	1239	0.0782	0.0230	43.5
322	0.0722	0.0577	17.3	1006	0.0764	0.0247	40.5
360	0.0731	0.0534	18.7	916	0.0765	0.0278	36.0
422	0.0745	0.0495	20.2	831	0.0765	0.0304	32.9
431	0.0745	0.0465	21.5	716	0.0764	0.0341	29.3
459	0.0749	0.0471	21.2	600	0.0759	0.0411	24.3
491	0.0752	0.0444	22.5	468	0.0750	0.0451	22.2
535	0.0757	0.0426	23.5	442	0.0745	0.0475	21.0
561	0.0760	0.0420	23.8	338	0.0728	0.0581	17.2
652	0.0764	0.0369	27.1	304	0.0716	0.0608	16.4
696	0.0764	0.0352	28.4	268	0.0705	0.0606	16.5
724	0.0765	0.0347	28.8	195	0.0676	0.0695	14.4

FIGURE 13. A Comparison of Calculated Thermal Conductivity for $(U_{0.80}Pu_{0.20})O_{2.00}$ with Steady-State Measurements on $(U_{0.80}Pu_{0.20})O_{2.00}$ and UO_2

temperatures, however, where electronic and radiation heat transfer processes as well as small changes in oxygen stoichiometry due to preferential losses of oxygen from the lattice become possible, plutonium substitutions may result in an appreciable change in thermal conductivity. Measurements at temperatures above 1600 °C are required to resolve this important question.

The thermal resistance of the mixed oxide between 200-1400 °C as determined in this investigation is given by the relationship

$$\frac{1}{K} = 0.0307 T - 0.45, T(^{\circ}\text{K})$$

Below 200 °C, the data show a positive deviation from this relationship (Figure 14). A maximum in the thermal conductivity of UO_2 at approximately room temperature has been observed by Béthoux, Thomas, and Weil,⁽²⁶⁾ and at approximately 50 °C by Godfrey et al.⁽²⁴⁾ The maximum was ascribed to a rapid decrease in heat capacity below approximately 200 °C. The results of this investigation are also explained in the same manner. The thermal diffusivity of the mixed oxide deviates only slightly from a reciprocal temperature relationship below 200 °C. Because of the poor sensitivity of the apparatus in this temperature region, however, the results are questionable. The thermal conductivity deviates much more noticeably from a reciprocal temperature relationship below 200 °C because of the rapid decrease of the heat capacity at the lower temperatures. The consequence, therefore, is an approach to a maximum value in thermal conductivity below 200 °C.

At approximately 1400 °C, the thermal conductivity of the mixed oxide reaches a minimum value. The thermal diffusivity also has reached a minimum at this temperature, and heat

capacity is increasing very slowly while density is decreasing slowly. If the heat capacity of the mixed oxide were to show the same increase reported for $\text{UO}_2^{(22)}$ at very high temperatures, the thermal conductivity might also increase.

FIGURE 14. Thermal Resistance of $(\text{U}_{0.80}\text{Pu}_{0.20})\text{O}_{2.00}$ as a Function of Absolute Temperature

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