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by

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HEAT TRANSFER IN $^{238}\text{PuO}_2$ SPHERES

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R. A. Kent and R. W. Zoehner

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ABSTRACT

The effective thermal conductivity for spherical $^{238}\text{PuO}_2$ specimens (80% ^{238}Pu) has been calculated from steady-state measurements of the surface-to-core thermal gradients. The specimens had an average density of 80% TD and thermal inventories ranging from 50 to 100 J/s. The experiments were performed in both argon-water and helium atmospheres over the range 300 to 1500°C. The results indicate that helium, produced by the alpha decay of ^{238}Pu , significantly affects the heat transfer, especially at low temperatures.

I. INTRODUCTION

The ^{238}Pu -isotope of plutonium has a relatively short half-life,¹ 87.8 yr, and produces thermal energy at the rate of approximately 0.5 J/s per gram. Because of this, isotopic heat sources, fueled with $^{238}\text{PuO}_2$ (80% ^{238}Pu), have been used in a number of space missions to supply the electrical energy required for onboard instruments. The thermal conductivity of the fuel must be known if heat fluxes and temperature gradients within advanced heat sources are to be accurately predicted. While the thermal conductivity of $^{238}\text{PuO}_2$ has been determined over the range 250–1200°C,² that for $^{238}\text{PuO}_2$ has not been measured. In these experiments, $^{238}\text{PuO}_2$ spheres were heated over the range 300–1400°C and the surface-to-core thermal gradients were measured.

II. EXPERIMENTAL

Three $^{238}\text{PuO}_2$ spheres (80% ^{238}Pu) were fabricated by hot pressing < 125 μm PuO_2 granules that had been thermally seasoned for 3 h at 1100°C. The graphite dies were designed so that each sphere was fabricated with a

0.155-cm-diam well extending from the surface to the core. Two spheres, SPP-59 and SPP-61, had thermal inventories of 100 J/s, the third sphere, SPP-60, had a thermal inventory of 50 J/s. Fabrication parameters for these spheres, together with as-pressed and sintered dimensions, are listed in Tables I, II, and III.

Normally, all $^{238}\text{PuO}_2$ specimens hot pressed in Los Alamos Scientific Laboratory (LASL) are postpress sintered in flowing $\text{Ar-H}_2^{16}\text{O}$. There are several reasons for this heat treatment. First, all plutonia specimens hot pressed in graphite dies have an as-pressed O/Pu ratio of about 1.89. Sintering in the $\text{Ar-H}_2^{16}\text{O}$ atmosphere causes a controlled reoxidation to $\text{PuO}_{2.0}$. The use of H_2^{16}O limits the (α , n) reactions that occur with ^{17}O and ^{18}O ; and thus, reduces the neutron emission rate. Finally, the grain growth that occurs during sintering provides a mechanism to anneal and homogenize the microstructure.

Thermal gradients within both as-pressed and sintered spheres were measured in the following manner. After the hot-pressing operation, the sphere was placed in a horizontal tube furnace and alumina-shielded Pt-Pt/Rh

TABLE I
FABRICATION PARAMETERS AND DIMENSIONS
FOR
SPHERE SPP-59

Feed Material	<125 μ m PuO ₂ granules seasoned 3 h at 1100°C
Hot-Pressing Conditions	3000 psi for 40 min at 1480°C
Post Press Sintering	Two sintering runs to 1440°C in Ar-H ₂ ¹⁶ O
Power	100.5 J/s (24.03 cal/s)

Condition	Dimensions ^a			
	Diam (cm)	Weight (g)	Stoichiometry (O/Pu)	Density (% TD)
As Pressed	3.703	251.2	1.89	83.0
Sintered ^b	3.698	252.85	2.00	83.3

a. Dimensions corrected to 25°C.

b. Dimensions after first sintering run. No change occurred during second sintering run.

thermocouples were positioned to measure the core and surface temperatures. Thermocouple outputs were read on a digital voltmeter attached to a two-pen recorder. The sphere was heated in flowing Ar-H₂¹⁶O to approximately 1400°C, held at that temperature for 16 h, and then cooled to ambient. The sphere was heated and cooled in stepwise fashion and both surface and core temperatures were determined under conditions of thermal equilibrium.

TABLE II
FABRICATION PARAMETERS AND DIMENSIONS
FOR
SPHERE SPP-61

Feed Material	<125 μ m PuO ₂ granules seasoned 3 h at 1100°C
Hot-Pressing Conditions	3500 psi for 30 min at 1480°C
Post Press Sintering	To 1500°C in Ar-H ₂ ¹⁶ O
Power	100.6 J/s (24.03 cal/s)

Condition	Dimensions ^a			
	Diam (cm)	Weight (g)	Stoichiometry (O/Pu)	Density (% TD)
As Pressed	3.746	251.3	1.89	80.2
Sintered	3.731	252.9	2.00	81.2

a. Dimensions corrected to 25°C.

TABLE III
FABRICATION PARAMETERS AND DIMENSIONS
FOR
SPHERE SPP-60

Feed Material	<125 μ m PuO ₂ granules, seasoned 3 h at 1100°C
Hot-Pressing Conditions	3000 psi for 30 min at 1490°C
Post Press Sintering	One sintering run to 1400°C in Ar-H ₂ ¹⁶ O and one run to 1100°C in He
Power	50.2 J/s (11.99 cal/s)

Condition	Dimensions ^a			
	Diam (cm)	Weight (g)	Stoichiometry (O/Pu)	Density (% TD)
As Pressed	3.014	125.4	1.89	76.8
Sintered ^b	2.998	126.2	2.00	78.1

a. Dimensions corrected to 25°C.

b. Dimensions after first sintering run. No change occurred during second sintering run.

III. RESULTS AND DISCUSSION

A. Sphere SPP-59

This sphere was heated to 1440°C in flowing Ar-H₂¹⁶O, held at 1440°C for 16 h, and then cooled. Data obtained during this initial heating cycle are listed in Table IV and plotted in Fig. 1. The sphere remained integral and free from surface cracks throughout this experiment. The results obtained were surprising in that the surface-to-core ΔT decreased as the surface temperature was increased. This is contrary to what one would expect for a ceramic material based on calculations involving thermal conductivity values. Note also, that at an ambient surface temperature of 400°C, the surface-to-core ΔT is about 120°C lower for the sintered sphere. This, in part, is due to the annealing and sintering that occurred. The diameter for SPP-59, corrected to 25°C, was 3.703 cm as pressed and 3.698 cm after sintering. For the calculations that follow, only the sintered sphere is considered and the surface-to-core ΔT , calculated from the data in Table IV, is given by

$$\Delta T (^{\circ}\text{C}) = 168 - 3.20 \times 10^5 / T_s + 2.94 \times 10^8 / T_s^2 - 6.32 \times 10^{10} / T_s^3 \quad (1)$$

where T_s is the surface temperature in degrees C.

In a second experiment, an alumina plug, 0.836 cm long, was placed in the thermocouple well. The thermo-

TABLE IV
INITIAL SINTERING OF SPHERE SPP-59
IN FLOWING ARGON-WATER

Temperature (°C)					
Heat Up (As-Pressed Sphere)			Cool Down (Sintered Sphere)		
Surface	Core	ΔT	Surface	Core	ΔT
378	711	333	1435	1503	68
380	715	335	1390	1457	67
468	761	293	1336	1404	68
512	783	271	1289	1357	68
564	812	248	1226	1295	69
616	842	226	1175	1245	73
673	879	206	1113	1186	73
747	931	184	1046	1121	75
823	989	166	977	1059	82
915	1065	150	915	1003	88
1023	1156	133	849	946	97
1090	1212	122	770	882	112
1166	1275	109	700	828	128
1241	1348	107	621	772	151
1301	1405	104	547	724	177
1371	1459	88	500	695	195
1439	1524	85	400	643	243

Held for 16 hours

Heat Up: $\Delta T(^{\circ}\text{C}) = -33.7 + 1.79 \times 10^5/T_s - 1.24 \times 10^7/T_s^2$.

Cool Down: $\Delta T(^{\circ}\text{C}) = 1.68 \times 10^2 - 3.20 \times 10^5/T_s + 2.94 \times 10^9/T_s^2$
 $-6.32 \times 10^{10}/T_s^3$.

Fig. 1. Initial sintering cycle for sphere SPP-59 in flowing Ar-H₂¹⁶O.

couples were positioned at the surface and in the plugged well and the sphere was again heated to about 1400°C in flowing Ar-H₂¹⁶O. Surface and midradius temperatures were measured under conditions of thermal equilibrium. The surface-to-midradius ΔT , calculated from the data listed in Table V, is given by

$$\Delta T(^{\circ}\text{C}) = 19.9 - 2.16 \times 10^4/T_s + 1.24 \times 10^7/T_s^2 + 2.96 \times 10^9/T_s^3 \quad (2)$$

where, again, T_s is the surface temperature in degrees C. One can calculate the core-to-midradius (surface for the "inner sphere") ΔT by combining Eq. (1) and Eq. (2).

Surface-to-core and surface-to-midradius ΔT values for sphere SPP-59 are compared in Fig. 2.

TABLE V
SECOND SINTERING OF SPHERE SPP-59
IN FLOWING ARGON-WATER

Temperature (°C)		
Surface	Midpoint	ΔT^a
447	514	67
531	579	48
610	641	31
791	810	19
872	888	16
1004	1017	13
1080	1093	13
1148	1160	12
1205	1216	11
1265	1276	11
1326	1339	13
1329	1341	12
1394	1407	13
1416	1430	14

$$\Delta T(^{\circ}\text{C}) = 19.9 - 2.16 \times 10^4/T_s + 1.24 \times 10^7/T_s^2 + 2.96 \times 10^9/T_s^3$$

a. The data were obtained with thermocouples located at surface and midway to the core.

Fig. 2. Temperature profile for sintered sphere SPP-59 heated in flowing Ar-H₂¹⁶O.

The surface-to-core ΔT values for self-heating spheres may be calculated from the equation

$$\Delta T = Qr^2/6K \quad (3)$$

where Q is the heat generated per unit volume per unit time, r is the radius, and K is the thermal conductivity. This relationship is derived in the appendix as Eq. (A-9). With the heat expressed in cal/s (100 J/s = 23.9 cal/s), sphere dimensions in cm, and K in cal/cm s °C, the value of ΔT is given in degrees C.

In order to employ Eq. (3) over the experimental temperature range, the thermal expansion and the thermal conductivity of PuO₂ must be known as functions of temperature.

The thermal expansion of ²³⁸PuO₂, over the range 25-1600°C, was measured at LASL³ and is given by the equation

$$\Delta L/L_0 = 2.249 \times 10^{-4} + 9.020 \times 10^{-6}T - 1.130 \times 10^{-9}T^2 + 1.525 \times 10^{-12}T^3 \quad (4)$$

where T is in degrees C.

Lagedrost et al.² measured the thermal diffusivity of two ²³⁸PuO₂ specimens, 81.9% TD and 96.5% TD, over the range 250-1200°C. From their data, we calculated the following equation for the thermal conductivity of PuO₂, corrected to 100% TD.

$$K(\text{cal/cm s } ^\circ\text{C}) = 8.14 \times 10^{-3} - 7.45/T + 6.41 \times 10^3/T^2 - 1.11 \times 10^6/T^3, \quad (5)$$

where, again, T is in degrees C. These authors observed reduction of the PuO₂ specimens at temperatures greater than 1000°C. For this work, Eq. (5) was assumed to hold to 1400°C because the experiments were performed in an oxidizing atmosphere.

The calculations for the full sphere, SPP-59, are summarized in Tables VI and VII. Values for the surface temperature, in 100° increments, are listed in the first column of Table VI. Values for the corresponding midradius and core temperatures, calculated from Eqs. (1) and (2), are listed in the second and third columns. The values from these three columns were used to calculate the average temperature values, T_{av} , listed in column four. The values of $\Delta L/L_0$ listed in column five were calculated at T_{av} from Eq. (4). Likewise, the values of K_{PuO_2} (100% TD) listed in column six were calculated at T_{av} from Eq. (5).

Sphere SPP-59 weighed 252.85 g, corresponding to 100.5 J/s or 24.03 cal/s. The sintered diameter (at 25°C) was 3.698 cm. This value, combined with the $\Delta L/L_0$ values listed in Table VI, was used to calculate the diameters listed in Table VII. The corrected diameters were used to calculate the sphere volume and hence, density over the experimental temperature range. The assumption was made that the value of K_{PuO_2} at some density, e.g., 80% TD, was equal to 80% of the value of K_{PuO_2} at 100% TD. Calculated and observed surface-to-core ΔT values are listed in Table VII in columns four and five. Finally, values for the effective conductivity of the sphere, K_{eff} , are listed in the last column.

The agreement between calculated and observed ΔT values is good near the middle of the experimental temperature range. The agreement at higher temperatures is reasonable considering the extrapolation of the K_{PuO_2} values to temperatures above 1000°C, and the fact that the effective thermal conductivity varies directly with

TABLE VI
DATA FOR FULL SPHERE SPP-59

Surface	Temperature (°C)			Calculated at Average T K _{PuO₂} (100% TD)	
	Midradius	Core	Average	$\Delta L/L_0$ (x10 ³)	(cal/cm s °C) (x10 ³)
400	490	621	504	4.23	9.96
500	550	700	583	4.95	8.65
600	632	760	664	5.71	7.69
700	723	828	750	6.55	6.99
800	818	905	841	7.47	6.49
900	915	990	935	8.47	6.16
1000	1014	1080	1031	9.54	5.94
1100	1113	1173	1129	10.71	(5.81) ^a
1200	1212	1270	1227	11.96	(5.73)
1300	1312	1368	1327	13.32	(5.70)
1400	1412	1467	1426	14.76	(5.69)

a. Values in parentheses are extrapolated.

TABLE VII
CALCULATIONS FOR FULL SPHERE SPP-59

Surface Temp (°C)	Diam (cm)	Density (% TD)	Surface-to-Core ΔT (°C)		Calculated K _{eff} (cal/cm s °C) (x10 ³)
			Calculated ^a	Observed	
400	3.713	82.3	63	221	2.3
500	3.716	82.1	72	200	2.6
600	3.719	81.9	82	160	3.2
700	3.722	81.7	90	128	4.0
800	3.725	81.5	97	105	4.9
900	3.729	81.3	103	90	5.7
1000	3.733	81.0	106	80	6.4
1100	3.737	80.7	109	73	7.0
1200	3.742	80.4	111	70	7.3
1300	3.747	80.1	112	68	7.5
1400	3.753	79.7	112	67	7.6

a. Values were calculated based on K_{PuO₂}, corrected for density.

the density which was assumed to be the same throughout the sphere.

At lower temperatures, however, the calculated ΔT values are much lower than those observed. It was thought that this difference could be due to helium within the internal cracks and pores, especially near the core, which would probably be the lowest density region of the sphere, despite the assumptions made above. For this reason, the second experiment was performed with the "inner" sphere.

The surface of the inner sphere is taken to be at the midradius of the full sphere. Thus, the surface-to-core ΔT values for the inner sphere are obtained by combining Eqs. (1) and (2). Values for the average temperature of the inner sphere, as a function of the surface temperature of the full sphere, are listed in Table VIII. Values for K_{PuO_2} (100% T.D.) and K_{He} ,⁴ calculated for the average temperature of the inner sphere, are also listed in Table VIII.

The calculations for the inner sphere are summarized in Table IX. Two assumptions were made,

1. At 20°C, the full sphere diameter was 3.698 cm and the length of the Al_2O_3 plug (radius of the inner sphere) was 0.836 cm. It was assumed that the values of K_{PuO_2} and $K_{\text{Al}_2\text{O}_3}$ were such that the radius of the inner sphere was 45.2% that of the full sphere over the range 25–1400°C.

2. It was assumed that, at each temperature, the density of the inner sphere was that of the full sphere. Thus, the density values listed in Table VII were used to calculate the corrected values for K_{PuO_2} . This assumption results in the inner sphere weighing 23.427 g corresponding to 2.226 cal/s.

Values for the inner sphere surface-to-core ΔT were calculated from Eq. (3) based on K_{PuO_2} (corrected for density) and K_{He} . These values are compared to the observed ΔT values in Table IX. Values for the effective conductivity, K_{eff} , are listed in the last column of Table IX.

The data listed in Table IX clearly indicate that the observed ΔT values, near the core at low temperatures,

are due to helium within the internal cracks and pores. It is also evident that, even at 1400°C, a substantial amount of helium remains within the sphere, especially near the core.

B. Sphere SPP-61

Sphere SPP-61, like sphere SPP-59, was fabricated with a nominal thermal inventory of 100 J/s. After the hot-pressing operation, the sphere was heated to 1440°C in flowing $\text{Ar-H}_2^{16}\text{O}$, held at 1440°C for 16 h, and then cooled. Data obtained during this heating cycle are listed in Table X. Again, the calculations that follow are for the sintered sphere only, and the surface-to-core ΔT , calculated from the data in Table X is given by

$$\Delta T(^{\circ}\text{C}) = -2.15 + 5.10 \times 10^4/T_s + 4.01 \times 10^7/T_s^2 - 1.09 \times 10^{10}/T_s^3 \quad (6)$$

where T_s is the surface temperature in degrees C.

The calculations for sphere SPP-61 are summarized in Tables XI and XII. Values for the surface temperature, in 100° increments, are listed in the first column of Table XI. Values for the corresponding core and average temperatures are listed in the second and third columns. Values for $\Delta L/L_0$ and K_{PuO_2} (100% T.D.), calculated at T_{av} , are listed in the last two columns of Table XI.

Sphere SPP-61 weighed 252.9 g, corresponding to 24.03 cal/s. The sintered diameter (at 25°C) was 3.731 cm. This value, together with the values of $\Delta L/L_0$, listed in Table XI, was used to calculate the diameters and sphere volumes listed in Table XII. Calculated and observed surface-to-core ΔT values are listed in column four and five of Table XII. Values for the effective thermal conductivity of this sphere are listed in the last column in Table XII.

The values listed in Table XII for sphere SPP-61 are in reasonably good agreement with those listed in Table VII for sphere SPP-59.

C. Sphere SPP-60

Sphere SPP-60 was fabricated with a nominal thermal inventory of only 50 J/s. After the hot-pressing operation, the sphere was heated to about 1300°C, in flowing $\text{Ar-H}_2^{16}\text{O}$, held at that temperature for 16 h, and then cooled.

Data obtained during the heating cycle in Ar-H₂¹⁶O are listed in Table XIII and plotted in Fig. 3. Again, the calculations that follow are for the sintered sphere only, where the surface-to-core ΔT , calculated from the data in Table XIII, is given by

$$\Delta T(^{\circ}\text{C}) = 69.0 - 6.31 \times 10^4/T_s + 6.88 \times 10^7/T_s^2 - 1.15 \times 10^{10}/T_s^3 \quad (7)$$

where, again, T_s is the surface temperature in degrees C.

The calculations for sphere SPP-60, heated in Ar-H₂¹⁶O, are summarized in Tables XIV and XV. Values for the surface, core and average temperatures, together with values for $\Delta L/L_0$ and K_{PuO_2} (100% TD), calculated at T_{av} , are listed in Table XIV. Sphere SPP-61 weighed 126.2 g, corresponding to 11.99 cal/s. The diameter for the sintered sphere (at 25^oC) was 2.998 cm. This value, together with the $\Delta L/L_0$ values listed in Table XIV, was used to calculate the diameters and densities listed in Table XV. Also listed in Table XV are the observed and calculated surface-to-core ΔT values, and the values for the effective conductivity of this sphere. Note that the values for K_{eff} calculated for the 50 J/s sphere, SPP-60,

in Ar-H₂¹⁶O, are in agreement with those obtained in Ar-H₂¹⁸O, for the 100 J/s spheres SPP-59 and SPP-61.

The data obtained for the three spheres in Ar-H₂¹⁶O, especially the data for the inner sphere SPP-59, indicate that, in part, the heat flow is dependent on the amount of helium trapped within cracks and pores. This is especially true near the sphere core and at low surface temperatures. Thus, one would expect that if a sphere were to be heated in helium instead of Ar-H₂¹⁶O, the surface-to-core ΔT values would be lower at low surface temperatures and higher at the higher surface temperatures.

Sphere SPP-60 was, therefore, reheated in flowing helium to 1100^oC and then cooled. Data collected during this cycle are listed in Table XVI and plotted in Fig. 4. The value for the surface-to-core ΔT , in helium, calculated from the data in Table XVI, is given by

$$\Delta T(^{\circ}\text{C}) = 88.6 - 2.50 \times 10^4/T_s + 2.77 \times 10^7/T_s^2 - 4.58 \times 10^9/T_s^3 \quad (8)$$

where T_s is the surface temperature in degrees C. The data for sphere SPP-60, heated in flowing helium, are summarized in Tables XVII and XVIII. Note that, at low

Fig. 3. Initial heating cycle for sphere SPP-60 in flowing Ar-H₂¹⁶O.

Fig. 4. Heating cycle for sintered sphere SPP-60 in flowing helium.

surface temperatures, the ΔT values are lower (greater K_{eff}) when helium is substituted for the $\text{Ar-H}_2^{16}\text{O}$. At higher surface temperatures, the surface-to-core ΔT values are higher (smaller K_{eff}) in helium than in $\text{Ar-H}_2^{16}\text{O}$. These data, again, indicate the dependency of the heat flow on the amount of helium trapped within the cracks and pores in the specimen.

At the conclusion of the experiments, metallographic samples were taken from both the surface and the core of each of the three spheres. The micrographs for sphere SPP-61 are shown in Fig. 5 and indicate that the sphere microstructure was homogeneous with respect to grain size ($A_v = 20 \mu\text{m}$) and density.

Fig. 5. Micrograph (etched, X500) for sphere SPP-61. One inch equals $50 \mu\text{m}$.

IV. SUMMARY

In the experiments described above, the $^{239}\text{PuO}_2$ thermal conductivity data of Lagedrost² were used to calculate the surface-to-core ΔT values for $^{239}\text{PuO}_2$ spheres. While the agreement between calculated and observed values was fair in the range 700-900°C, it was somewhat poorer at higher temperatures and very poor at lower temperatures. The differences between the observed and calculated values are primarily due to helium gas present in the $^{239}\text{PuO}_2$ microstructure.

The alpha decay of ^{239}Pu generates approximately 0.5 cm^3 (at STP) of helium per yr per g of ^{239}Pu . Thus, a 100 J/s $^{239}\text{PuO}_2$ sphere will produce between 90 and 100 cm^3 of helium in a year. The results of these experiments indicate that sufficient helium remains trapped within the microstructure to significantly affect heat flow within these specimens. As would be expected, the effect of helium is greater at low temperatures because relatively more helium remains within the microstructure. However, even at 1400°C, not all of the helium is removed from the microstructure. In addition, some of the disagreement at higher temperatures is probably due to the extrapolation of the Lagedrost data beyond 1000°C.

Obviously, thermal conductivity measurements for PuO_2 should be extended beyond 1000°C. However, baseline measurements must be made on $^{239}\text{PuO}_2$ specimens since some helium will always be present in $^{239}\text{PuO}_2$ specimens. Finally, if quantitative heat flow calculations are to be made for future isotopic generators fueled with $^{239}\text{PuO}_2$, additional data must be obtained for $^{239}\text{PuO}_2$ specimens containing helium. Additional experiments will be performed at LASL to determine the effects of specimen density on the amount of helium retained and subsequently on the heat flow within the specimen.

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TABLE VIII
DATA FOR INNER SPHERE SPP-59

Temperature ($^{\circ}\text{C}$)		Values at Average Temperature ^a	
Surface (Full Sphere)	Average (Inner Sphere)	K (cal/cm s $^{\circ}\text{C}$)	
		PuO ₂ (100% TD) ($\times 10^3$)	He ($\times 10^3$)
400	556	9.05	0.75
500	625	8.11	0.80
600	696	7.40	0.85
700	776	6.82	0.90
800	862	6.40	0.95
900	953	6.11	1.01
1000	1047	5.91	1.07
1100	1143	(5.79)	1.13
1200	1241	(5.73)	1.19
1300	1340	(5.70)	1.25
1400	1439	(5.69)	1.31

a. Values in parentheses are extrapolated.

TABLE IX
CALCULATIONS FOR INNER SPHERE SPP-59

Surface Temp (Full Sphere) (°C)	Inner Sphere Diam (cm)	Inner Sphere Surface-to-Core ΔT (°C)			Calculated K _{eff} (cal/cm s °C) (x10 ³)
		Calculated		Observed	
		^a	^b		
		K PuO ₂	K He		
400	1.680	14	141	131	0.8
500	1.681	16	132	150	0.7
600	1.683	17	124	128	0.8
700	1.684	19	117	105	1.0
800	1.686	20	111	87	1.2
900	1.688	21	104	75	1.4
1000	1.690	22	98	66	1.6
1100	1.692	22	93	60	1.7
1200	1.694	23	88	58	1.8
1300	1.695	23	84	56	1.9
1400	1.698	23	80	55	1.9

a. ΔT values were calculated based on K_{PuO_2} , corrected for density.

b. ΔT values were calculated based on K_{He} .

TABLE X
SINTERING CYCLE FOR SPHERE SPP-61
IN FLOWING ARGON-WATER

Temperature (°C)					
Heat Up (As-Pressed Sphere)			Cool Down (Sintered Sphere)		
Surface	Core	ΔT	Surface	Core	ΔT
469	690	231	1342	1398	56
467	669	208	1374	1426	52
509	717	206	1482	1529	47
565	754	189	1292	1349	57
598	777	179	1152	1215	63
638	806	168	1055	1125	70
694	852	158	983	1061	78
761	909	146	917	1004	87
824	963	139	819	920	101
897	1028	131	816	918	102
958	1064	126	627	769	142
1052	1168	116	532	698	166
1117	1232	115	440	633	193
1186	1302	116	399	605	206
1253	1365	112	Stop Ar-H ₂ ¹⁶ O flow, Introduce He		
1355	1453	98	359	525	166
1440	1532	92			

Held for 16 hours

Heat Up: $\Delta T(^{\circ}\text{C}) = -63.1 + 3.41 \times 10^5/T_s - 2.05 \times 10^6/T_s^2 + 5.15 \times 10^{10}/T_s^3$.

Cool Down: $\Delta T(^{\circ}\text{C}) = -2.15 + 5.10 \times 10^4/T_s + 4.01 \times 10^7/T_s^2 - 1.09 \times 10^{10}/T_s^3$.

TABLE XI
DATA FOR SPHERE SPP-61

Temperature (°C)			Calculated at Average T	
Surface	Core	Average	$\Delta L/L_0$ ($\times 10^3$)	K_{PuO_2} (100% TD) (cal/cm s °C) ($\times 10^3$)
400	606	503	4.22	9.98
500	673	587	4.99	8.59
600	744	672	5.79	7.61
700	821	760	6.65	6.92
800	903	851	7.57	6.45
900	989	945	8.58	6.13
1000	1078	1039	9.64	5.93
1100	1169	1135	10.79	(5.80) ^a
1200	1262	1231	12.01	(5.73)
1300	1356	1328	13.33	(5.70)
1400	1451	1425	14.75	(5.70)
1500	1546	1523	16.28	(5.70)

a. Values in parentheses are extrapolated.

TABLE XII
CALCULATIONS FOR SPHERE SPP-61

Surface Temp (°C)	Diam (cm)	Density (% TD)	Surface-to-Core ΔT (°C)		Calculated K_{eff} (cal/cm s °C) ($\times 10^3$)
			Calculated ^a	Observed	
400	3.746	80.2	64	206	2.5
500	3.749	80.0	74	173	2.9
600	3.752	79.8	84	144	3.5
700	3.755	79.6	92	121	4.2
800	3.759	79.4	99	103	4.9
900	3.763	79.1	105	89	5.7
1000	3.767	78.9	109	78	6.5
1100	3.771	78.6	111	69	7.4
1200	3.775	78.3	113	62	8.2
1300	3.780	78.0	114	56	9.0
1400	3.786	77.7	114	51	9.9
1500	3.791	77.3	114	46	11.0

a. Values were calculated based on K_{PuO_2} , corrected for density.

TABLE XIII

INITIAL SINTERING OF SPHERE SPP-60
IN FLOWING ARGON-WATER

Temperature (°C)					
Heat Up (As-Pressed Sphere)			Cool Down (Sintered Sphere)		
Surface	Core	ΔT	Surface	Core	ΔT
299	507	208	1379	1434	55
406	575	169	1305	1360	55
481	628	147	1214	1274	60
558	685	127	1025	1091	66
667	772	105	947	1015	68
739	834	95	854	926	72
808	898	90	790	866	76
862	947	85	712	796	84
914	997	83	655	746	91
973	1051	78	544	658	114
975	1049	74	427	578	151
1003	1080	77	325	515	190
1004	1076	72	322	514	192
1040	1115	75			
1059	1131	72			
1083	1164	81			
1106	1182	76			
1140	1207	67			
1249	1324	75			
1306	1377	71			

Held for 16 hours

Heat Up: $\Delta T(^{\circ}\text{C}) = 93.6 - 8.51 \times 10^4/T_s + 8.36 \times 10^7/T_s^2 - 1.43 \times 10^{10}/T_s^3$.

Cool Down: $\Delta T(^{\circ}\text{C}) = 69.0 - 6.31 \times 10^4/T_s + 6.88 \times 10^7/T_s^2 - 1.15 \times 10^{10}/T_s^3$.

TABLE XIV

DATA FOR SPHERE SPP-60
IN ARGON-WATER

Temperature (°C)			Calculated at Average T	
Surface	Core	Average	$\Delta L/L_0$ ($\times 10^3$)	K_{PuO_2} (100% TD) (cal/cm s °C) ($\times 10^3$)
300	497	399	3.29	12.33
400	562	481	4.02	10.43
500	626	563	4.77	8.94
600	702	651	5.59	7.82
700	786	743	6.48	7.04
800	875	838	7.87	6.51
900	968	934	8.46	6.16
1000	1063	1032	9.56	5.94
1100	1160	1130	10.73	5.81
1200	1258	1229	11.98	5.73
1300	1356	1328	13.33	(5.70) ^a
1400	1455	1427	14.78	(5.69)

a. Values in parentheses are extrapolated.

TABLE XV

CALCULATIONS FOR SPHERE SPP-60 IN ARGON-WATER

Surface Temp (°C)	Diam (cm)	Density (% TD)	Surface-to-Core ΔT (°C)		Calculated K_{eff} (cal/cm s °C) ($\times 10^3$)
			Calculated ^a	Observed	
300	3.008	77.3	33	197	1.6
400	3.010	77.1	39	162	2.0
500	3.012	76.9	46	126	2.5
600	3.015	76.8	53	102	3.1
700	3.017	76.6	59	86	3.7
800	3.022	76.2	64	75	4.2
900	3.023	76.1	67	68	4.6
1000	3.027	75.9	70	63	5.0
1100	3.030	75.6	72	60	5.2
1200	3.034	75.3	73	58	5.4
1300	3.038	75.0	74	56	5.6
1400	3.042	74.7	74	55	5.7

a. Values were calculated based on K_{PuO_2} , corrected for density.

TABLE XVI
SINTERING OF SPHERE SPP-60
IN FLOWING HELIUM

Temperature ($^{\circ}\text{C}$)		
Surface	Core	ΔT
228	389	161
329	473	144
455	581	126
575	686	111
659	762	103
757	854	97
834	926	92
911	1001	90
977	1066	89
1068	1154	86
1071	1159	88
981	1065	84
944	1030	86
830	918	88
746	839	93
648	747	99
555	662	107
377	507	130
225	378	153

TABLE XVII
DATA FOR SPHERE SPP-60 IN HELIUM

Temperature ($^{\circ}\text{C}$)			Calculated at Average T	
Surface	Core	Average	$\Delta L/L_0$ ($\times 10^3$)	K_{PuO_2} (100% TD) (cal/cm s $^{\circ}\text{C}$) ($\times 10^3$)
300	444	372	3.05	12.95
400	528	464	3.87	10.79
500	613	556	4.70	9.05
600	703	651	5.59	7.82
700	796	748	6.53	7.00
800	892	846	7.52	6.47
900	989	944	8.57	6.13
1000	1087	1043	9.68	5.92
1100	1185	1143	10.89	5.79
1200	1284	1242	12.16	5.72

Sintered Sphere; $\Delta T (^{\circ}\text{C}) = 88.6 - 2.50 \times 10^4/T_g + 2.77$
 $\times 10^7/T_g^2 - 4.58 \times 10^3/T_g^3$.

TABLE XVIII
CALCULATIONS FOR SPHERE SPP-60 IN HELIUM

Surface Temp ($^{\circ}\text{C}$)	Diam. (cm)	Density (% TD)	Surface-to-Core ΔT ($^{\circ}\text{C}$)		Calculated K_{eff} (cal/cm s $^{\circ}\text{C}$) ($\times 10^3$)
			Calculated ^a	Observed	
300	3.007	77.3	32	144	2.2
400	3.010	77.2	38	128	2.5
500	3.012	77.0	45	113	2.8
600	3.015	76.8	53	103	3.1
700	3.018	76.5	59	96	3.3
800	3.021	76.3	64	92	3.4
900	3.024	76.1	68	89	3.5
1000	3.027	75.8	70	87	3.6
1100	3.031	75.6	72	85	3.7
1200	3.034	75.3	73	84	3.7

a. Values were calculated based on K_{PuO_2} , corrected for density.

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APPENDIX A

CONDUCTION OF HEAT IN A SPHERE

The rate at which heat crosses from the inside to the outside of an isothermal surface per unit area per unit time at a point is

$$-K \frac{\partial T}{\partial r}, \quad (\text{A-1})$$

where K is the thermal conductivity and $\partial T / \partial r$ denotes differentiation along an outward-drawn normal to the surface.

For a homogeneous isotropic solid whose thermal conductivity is independent of temperature

$$\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} - \frac{1}{D} \frac{\partial T}{\partial t} = 0 \quad (\text{A-2})$$

where the diffusivity D is given by K/c_p where c is the specific heat and ρ is the density. If the temperature T is independent of time, t , there is a steady state and the Laplace equation applies.

$$\nabla^2 T = \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} = 0. \quad (\text{A-3})$$

If we let

$$\begin{aligned} x &= r \sin \theta \cos \phi \\ y &= r \sin \theta \sin \phi \\ z &= r \cos \theta \end{aligned}$$

and

$$ds^2 = dr^2 + r^2 d\theta^2 + r^2 \sin^2 \theta d\phi^2$$

then, in polar coordinates, Eq. (A-3) becomes

$$\frac{\partial T}{\partial t} = \frac{D}{r^2} \left[\frac{\partial}{\partial r} \left(r^2 \frac{\partial T}{\partial r} \right) + \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial T}{\partial \theta} \right) + \frac{1}{\sin^2 \theta} \frac{\partial^2 T}{\partial \phi^2} \right]. \quad (\text{A-4})$$

Under steady-state conditions, with radial heat flow, Eq. (A-4) reduces to

$$\frac{d}{dr} \left(r^2 \frac{dT}{dr} \right) = 0 \quad (\text{A-5})$$

with the solution

$$\Delta T = \frac{A}{r} + B, \quad (\text{A-6})$$

where A and B are constants determined by the boundary conditions.

If heat is produced in the solid so that at a point $P(x, y, z)$ heat is produced at a rate $Q(x, y, z, t)$ per unit time per unit volume, then Eq. (A-3) becomes

$$\nabla^2 T - \frac{1}{D} \frac{\partial T}{\partial t} = - \frac{Q(x, y, z, t)}{K}. \quad (\text{A-7})$$

Then for a solid sphere with $0 \leq r \leq a$, with heat produced in it at a constant rate Q per unit time per unit volume, we have, combining Eqs. (A-4) and (A-7),

$$\frac{1}{r^2} \frac{1}{dr} \left(r^2 \frac{dT}{dr} \right) = - \frac{Q}{K}, \quad (\text{A-8})$$

which requires a finite solution at $r = 0$, or

$$\Delta T = \frac{Qr^2}{6K}. \quad (\text{A-9})$$

Equation (A-9) applies to the work described here because the surface temperature of the sphere is known. If it is not known, and if there is radiation at $r = a$ into a medium, the solution to Eq. (A-8) becomes

$$\Delta T = \frac{Q}{6hK} \left[h(a^2 - r^2) + 2a \right], \quad (\text{A-10})$$

where h is a constant reflecting the radiant heat transfer.