LA-5506-PR
Progress Report

UC-48
Issued: May 1974

Preparation and Evaluation of Medical-Grade Plutonium-238 Fuels

July 1, 1971 - June 30, 1973

Compiled by

L. J. Mullins

los alamos
scientific laboratory
of the University of California
LOS ALAMOS, NEW MEXICO 87544

UNITED STATES
ATOMIC ENERGY COMMISSION
CONTRACT W-7405-ENG. 36

Work funded by the US AEC Division of Applied Technology. The program was assigned to the Division of Biology and Environmental Research in July 1973.

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy $4.00 Microfiche $1.45

CONTENTS

ACKNOWLEDGMENTS

This work was performed principally by CMB-1 and CMB-11 personnel. However, the assistance of several other LASL groups is gratefully acknowledged. The following groups, in particular, rendered valuable contributions.

CNC-4 —	made and provided all the H_2O^{16} used in this project.
CMB-6 —	fabricated all of the ceramic ware and much of the specialized equipment; designed and tested the dies and punches for the fuel form for the vented heat source.

The work was done under the direction of R. D. Baker, CMB Division Leader. Principal investigators were L. J. Mullins and G. R. Waterbury. The following individuals contributed to the work reported here: A. J. Beaumont, R. G. Bryan, J. Bubernak, M. R. Conner, B. A. Dye, C. L. Foxx, N. L. Koski, G. M. Matlack, T. K. Marshall, L. J. Mullins, A. K. Murdock, M. E. Smith, A. N. Morgan, and R. L. Nance.

PREPARATION AND EVALUATION OF MEDICAL-GRADE PLUTONIUM-238 FUELS
JULY 1, 1971 - JUNE 30, 1973

Compiled by

L. J. Mullins

ABSTRACT

Work has continued on the preparation and properties of medical-grade $^{238}PuO_2{}^{16}$ fuel forms for the artificial heart program. Procedures were developed for fabricating the 33-W fuel cylinder for the artificial heart vented capsule. Three sources were fabricated and delivered to Mound Laboratory (MRC) for encapsulation in vented capsules. One 50-W and five 29-W nonvented heat sources were fabricated for animal implantation studies.

Studies of helium and radon release rates from medical-grade fuel forms have been initiated and are in progress.

The characterization of ^{238}Pu fuel forms by nondestructive techniques and chemical analysis has continued.

Most of the investigations discussed are of the continuing type. Results and conclusions described may therefore be changed or augmented as the work continues. Published references to results cited in the report should not be made without obtaining explicit permission to do so from the persons in charge of the work.

I. INTRODUCTION

The principal objectives of the medical-grade fuel program are (1) to prepare and evaluate medical-grade ^{238}Pu fuels for an artificial heart device, (2) to fabricate and characterize heat-source capsules (up to 50 W) for dosimetry, systems studies, and implantation studies, (3) to develop diagnostic procedures for evaluating ^{238}Pu fuel forms, (4) to establish the compatibility of medical-grade fuel forms with selected container materials, and (5) to measure properties of medical-grade fuel forms important to the design and safety of the heat source.

The ^{238}Pu requirements ($\sim$69 g) for an artificial heart or circulatory-assist device demand a fuel having minimal radiation properties. Preparing and evaluating potential ^{238}Pu fuel forms have led to the development of four fuel compositions having radiation properties approximately equal to the irreducible minimum values for ^{238}Pu. The fuel forms are electrorefined metal, ^{238}Pu-3 at.% Ga, $^{238}PuN^{15}$, and $^{238}PuO_2{}^{16}$. Theoretical and experimental studies of these fuels led to the conclusion that $^{238}PuO_2{}^{16}$ is the preferred composition for high-temperature ($\sim$870°C) application in the artificial heart program. This fuel is prepared as a pressed and sintered ceramic cylinder. Procedures have been developed for preparing and characterizing sources varying in size from 1 to 50 W.

The work done from the start of this program in July 1967 to June 30, 1971, was reported in detail in LA-4940.[1] LA-5506-PR covers the period July 1, 1971, to June 30, 1973.

II. REVIEW OF ^{238}Pu FUEL PROPERTIES

Penetrating radiations from ^{238}Pu fuels having the isotopic compositions shown in Table I are derived primarily from

1. spontaneous fission neutrons,
2. neutrons produced by (α,n) reactions in impurities of low atomic number,
3. fast fission neutrons,
4. photons from the decay of ^{238}Pu,
5. photons from other plutonium isotopes and their daughters, and
6. photons that result from alpha particle reactions with light element impurities.

An irreducible neutron emission rate is associated with the spontaneous fission of ^{238}Pu and ^{240}Pu. For the spontaneous fission of ^{238}Pu, the average number of neutrons per fission is 2.33(±0.08)[2] and the half-life is $5.0(\pm 0.6) \times 10^{10}$ yr.[3] These values give a calculated value of 2586(±400) n/s-g ^{238}Pu. Results obtained in the Los Alamos Scientific Laboratory (LASL) program give a value of 2785(±55) n/s-g ^{238}Pu for neutron emission resulting from the spontaneous fission of ^{238}Pu.[1,4]

The neutrons resulting from (α,n) reactions obviously depend on the particular elements present and on their concentrations. The important elements in this respect are lithium, beryllium, boron, carbon, oxygen, fluorine, sodium, magnesium, aluminum, and silicon. The neutrons from spontaneous fission and (α,n) reactions cause additional fission of the plutonium isotopes, which, in turn, produce excess neutrons. Thus, there is a self-multiplication of the neutron rate. The magnitude of this multiplication will depend on the mass, density, and geometry of the plutonium as well as on the neutron flux.[1] Gamma rays accompany the alpha decay of the plutonium isotopes and account for most of the gamma radiation observed from chemically pure ^{238}Pu. As the material ages, more gamma rays appear from daughter products of ^{236}Pu and ^{241}Pu. Gamma rays also arise from (α,n) or (α,p) reactions induced by alpha-particle bombardment of impurity nuclides. This is particularly evident if the impurities are nitrogen or fluorine.

Medical-grade ^{238}Pu fuels are fuels having minimal radiation properties, high chemical purity, and high specific power. The specifications for medical-grade ^{238}Pu metal and ^{238}Pu oxide are given in Ref. 1, Tables XLVIII and XLIX. Minimal radiation properties are those resulting from radiation sources 1, 3, 4, and 5, above. To achieve high specific power, the ^{238}Pu isotopic composition should be as high as possible. Presently available technology sets this value at 90 at.%. To minimize the gamma dose rate increase with time,[1] the ^{236}Pu concentration should be as low as possible. For the present, this value is set at 0.3 ppm or less. High chemical purity is important to both minimal radiation properties and fuel/container compatibility. (Note that conventional ^{238}Pu of 80 at.% composition (Table I) was used for most of the development work done in the present program.)

The ^{238}Pu fuel compositions considered for medical applications are listed in Table II. To achieve the minimum weight and volume, unalloyed ^{238}Pu metal would be the preferred form. The physical properties of the metal, however, are complex because it exists in six allotropic modifications between room temperature and its 640°C. melting point. However, the metal can be stabilized in the delta phase by adding alloying agents such as aluminum, cerium, gallium, and scandium. Of the alloys listed in Table II, only the ^{238}Pu-3 at.% Ga alloy has been characterized in the LASL program. Carbide, nitride, and oxide are the ceramic forms considered. To achieve minimal radiation levels, special isotopic enrichments of carbon, nitrogen, and oxygen are required. Although PuO_2 has a lower

TABLE I

PLUTONIUM ISOTOPIC COMPOSITION OF CONVENTIONAL AND MEDICAL-GRADE ^{238}Pu FUELS
(Typical Lots)

	Abundance, wt%	
Pu Isotope	Conventional	Medical-Grade
236	1×10^{-4}	$<3 \times 10^{-5}$[a]
238	80.0	90.4
239	16.3	9.0
240	3.0	0.6
241	0.6	0.03
242	0.1	<0.01

---

[a] That is, <0.3 ppm.

TABLE II

POTENTIAL MEDICAL-GRADE ^{238}Pu FUEL COMPOSITIONS

Composition	Melting Temp (C)	Power Density (megawatts per cubic meter)[a,b]
Pu	640	7.2
Pu - 3 Ga	670	7.1
Pu - 10 Sc	750	6.6
Pu_4 Zr	800	5.9
(Pu Fe_2 + Fe)	1165	3.9
Pu Pt_2	1475	3.2
Pu C^{12}	1650	5.4
Pu N^{15}	2200	5.6
Pu $O_2{}^{16}$	2400	4.2

---

[a] Calculations based on a ^{238}Pu isotopic composition of 80 at.% and a fuel density of 90% of theoretical.

[b] For the most part, SI units are used in this report as given in the Metric Practice Guide, E380-72, American Society for Testing and Materials. These units are compared to the units of Ref. 1 in the Appendix, Table A-I. Exceptions to SI usage are noted when used.

TABLE III

COMPARISON OF THE DOSE RATES OF ^{238}Pu FUEL FORMS[a,b,c]

(95.2-mm by 95.2-mm Cylinders)

Composition	Gamma R/h-kg ^{238}Pu	Neutron rem/h-kg ^{238}Pu
Pu	0.25	0.31
Pu - 3 at.% Ga	0.25	0.31
Pu N^{nat}	0.54	0.31
Pu N^{15}	0.28	0.31
$PuO_2{}^{nat}$	0.36	1.50
$PuO_2{}^{16}$	0.29	0.34

---

[a] Pu isotopic composition, 80 wt% ^{238}Pu.

[b] Dose rates in air through 0.76 mm Ta, 10 cm from the center of the source.

[c] All fuels prepared from electro refined metal.

power density than the nitride or carbide, its excellent chemical stability, high melting point, and ease of synthesis and fabrication make it the prime fuel candidate for most medical applications. $^{239}PuO_2$ has been studied for several years as a fast-reactor fuel and much information has been generated on its chemical and physical properties.

Fuel forms that have been prepared, characterized, and evaluated are

Elemental ^{238}Pu,
^{238}Pu - 3 at.% Ga,
$^{238}PuO_2{}^{nat}$,
$^{238}PuO_2{}^{16}$,
$^{238}PuN^{nat}$, and
$^{238}PuN^{15}$.

The gamma and neutron dose rates of these fuels are compared in Table III. The gamma dose rate of the metal 100 mm from the center of a 9.52- by 9.52-mm cylinder contained in a 0.76-mm-thick tantalum container is 0.25 R/h-kg ^{238}Pu. The neutron dose rate is 0.31 rem/h-kg ^{238}Pu. Dose rates for the Pu-3 at.% Ga alloy are identical to the metal. The neutron dose rate of PuN^{nat} is the same as the metal; however, the gamma dose rate is approximately doubled as the result of an (α,p) reaction on ^{14}N yielding an excited state of ^{17}O. Replacement of ^{14}N by ^{15}N gives a compound having a gamma dose rate only 10% higher than the metal. (The slightly higher gamma dose rate of the nitride is due primarily to its lower density.) Both the neutron and gamma dose rates of $^{238}PuO_2{}^{nat}$ are significantly higher than the metal. However, $^{238}PuO_2{}^{16}$ has dose rates comparable to the metal. Thus, the fuel forms having low neutron and gamma dose rates are ^{238}Pu, ^{238}Pu-3 at.% Ga, $^{238}PuN^{15}$, and $^{238}PuO_2{}^{16}$. Insofar as radiation is concerned, all of these materials would be suitable for implantable heat sources required to power pacemakers and artificial heart devices.

III. PLUTONIUM-238 FUEL DEVELOPMENT FACILITY

The medical-grade ^{238}Pu fuel development facility is shown in Fig. 1. This facility consists of three lines of glove boxes. The line of hydrogenous-shielded boxes on the right is used for aqueous and dry chemistry operations which can be performed in air-filled glove boxes. In addition to the air boxes, this line also has a small inert atmosphere glove box, argon-flush type, in which ^{238}Pu metal can be handled. The center line consists of two large inert glove boxes, recirculating-argon type with gas purification units. One of these boxes is used for preparing and casting electrorefined metal and alloys and for preparing $^{238}PuO_2{}^{16}$ powders. The other box is used to fabricate ceramics, and the milling, pressing, and sintering operations are done in

Fig. 1.
^{238}Pu fuel development facility.

this box. The line of boxes on the left is presently used for the temporary encapsulation of heat sources.

The fuel development facility is used to prepare medical-grade fuel forms, to recycle process residues, and for process development studies. (Flow sheets for the medical-grade fuel form preparation were presented in Ref. 1.)

IV. PROCESS DEVELOPMENT STUDIES

A. Fabrication of Artificial Heart Fuel Cylinder for Vented Heat Source

A drawing of the $^{238}PuO_2{}^{16}$ cylinder for the TRW*-designed 33.1-W (thermal) artificial heart heat source is given in Fig. 2. Fuel cylinders prior to this design were all right-circular cylinders of simple geometry and had a height-to-diameter ratio of $\sim$1.0.[1] The cylinder shown in Fig. 2 has a height-to-diameter ratio of 2.1 and has a cavity to accept a pressure relief device (PRD); all edges are chamfered. To use existing equipment and minimize development costs, ceramic fabrication procedures utilizing drypressing facilities were developed.

The following approach was used in developing the ceramic fabrication procedures.

1. Dies and punches were designed and made for dry-pressing a "green" plutonia cylinder in one piece. The green body contained all the geometrical features of the designed cylinder except for the top chamfer in the PRD cavity. The dies were designed assuming a minimum shrinkage of 5% during the sintering process. The green cylinder and die configuration are shown in Fig. 3. The top edge of the PRD cavity is chamfered after the cylinder is sintered. This operation is easily accomplished by hand with a conical diamond countersink.

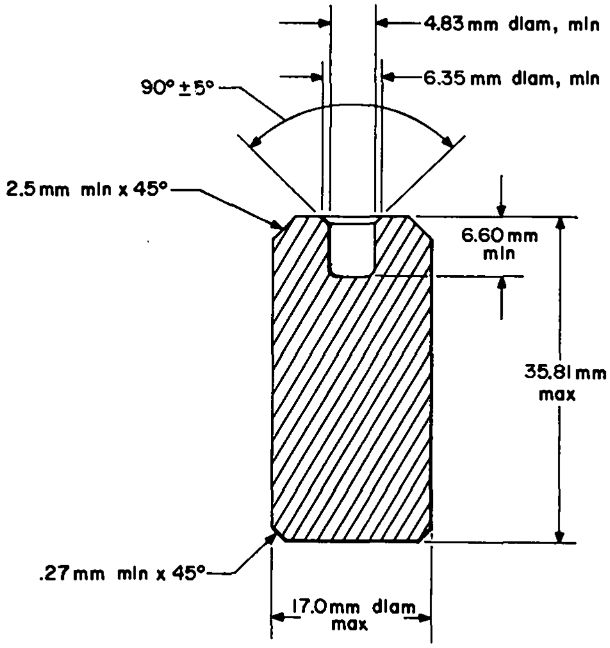


Fig. 2.
Artificial heart fuel cylinder, TRW design.

2. Thoria cylinders were made using these dies to demonstrate the practicability of the fabrication design.

3. Three $^{238}PuO_2{}^{16}$ cylinders, 80 at.% enrichment, were made to establish fabrication procedures before fabricating three medical-grade 33-W cylinders.

When the above work was concluded, ceramic fabrication problems still remained. The foremost problems were obtaining integral "green" bodies and "crack-free" sintered cylinders. A series of experiments was therefore initiated to examine these problems more fully and to establish the reproducibility of both product and process. Ten cylinders were prepared in this study, and all cylinders were made from nominal 80 at.% ^{238}Pu enrichment. The same weight of oxide powder was used in these preparations as would be used in the preparation of a 33.1-W medical-grade cylinder. The first five cylinders, 38 through 42, were made from the same batch of $^{238}PuO_2{}^{16}$ powder. This batch of

---

*TRW Systems Group, Redondo Beach, CA.

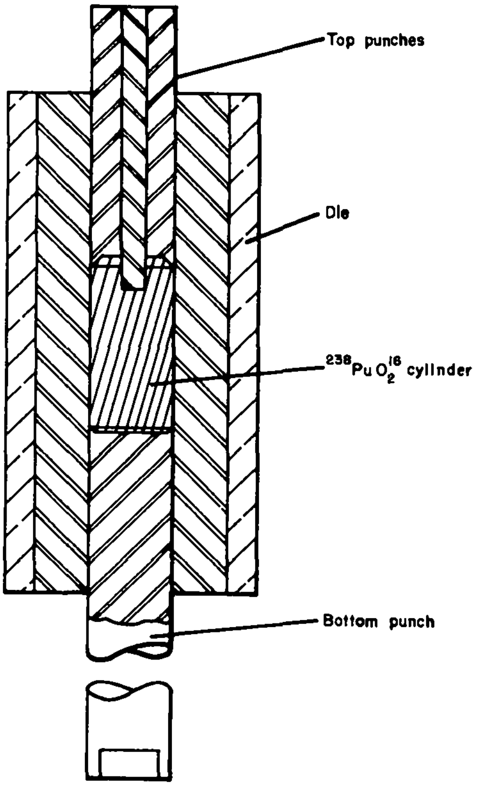


Fig. 3.
Cylinder and die configuration for pressing artificial heart fuel cylinder.

powder was prepared from old 50-W sources made for earlier program commitments. The procedure was as follows.

1. The 50-W cylinder was broken up by milling for 30 min in a SPEX Mill.
2. The resulting powder was heated to 760°C in an atmosphere of (Ar + H_2O^{16}) and held at this temperature for approximately 100 h. The powder was then heated to 1540°C, held at this temperature for 8 h, and then cooled to ambient temperature.
3. The powder was ball milled for 18 h in a uranium-molybdenum ball mill.
4. The powders (395-g total weight) were mixed in a large, shallow, water-cooled pan.
5. The cylinders were prepared by the procedure given in Ref. 1, p. 59.

The composition of the blended powder is given in Table IV. The powder was made from cylinders and control pellets from the preparation of cylinders 10, 11, 13, and 24. (Note: Only a small amount of 10 was available for use because most of this fuel was used to prepare the TRW developmental cylinder 34.)

TABLE IV

COMPOSITION OF POWDER FOR 29-W CYLINDERS

Serial Numbers
38 through 42

Total Weight of Powder 395.12 g

Composition 50-W Cylinder Serial No.	Weight Fraction
10	0.0605
11	0.3147
13	0.3096
24	0.3154

---

[a] These cylinders were formerly identified as 50-3, 50-4, 50-5, and 50-6. The nomenclature system in present use is defined in the Appendix.

Three cylinders (38, 39, and 40) and the control pellets were pressed during one work period and were then sintered by the procedure given in Ref. 1, footnote (b), p. 60. Cylinders 41 and 42 were pressed during a later work period. As was the case with the earlier 33-W TRW cylinders, some difficulty was encountered in obtaining integral green bodies during the pressing operation. (For preparations in which the pressing was unsuccessful, the carbowax binder was allowed to vaporize, the cylinder was rescreened, and the pressing operation was repeated.) Two pressings were required for 38 and three for 40. The first pressing was successful for 39, 41, and 42. (Note: The fuel ejection equipment and procedures were modified after fabricating 40. These modifications resulted in obtaining integral green pellets on the first pressing attempt.)

The physical dimensions of the five sintered cylinders are given in Table V. The same weight of oxide feed was used in each pressing. The difference in weights of the sintered bodies is due to varying process losses. The average weight was 74.19 (±0.05) g. Assuming a power density of 0.3914 kW/kg fuel, the average power of the five cylinders should be 29.04 (±0.02) W. Thus the preparation of sources of constant wattage should pose no problem if the specific power of the feed material is known accurately. The average length of the cylinders was 34.80 (±0.15) mm. A comparison of the maximum

TABLE V

PHYSICAL DIMENSIONS OF 29-W $^{238}PuO_2{}^{16}$ CYLINDERS

	Cylinder Serial Number					
	38	39	40	41	42[a]	47[b]
Wt, g fuel	74.20	74.11	74.16	74.18	74.30	73.83
Dimensions, mm (specification - length 35.81 max, diam 17.02 max)						
Length	34.67	34.80	34.59	35.05	34.92	34.52
Diam						
Max	16.71	16.69	16.69	16.76	16.69	16.66
Min	16.66	16.61	16.66	16.71	16.64	16.64
Cavity for PRD, mm (specification diam 4.83 min, depth 6.60 min)						
Diam	5.08	5.05	5.05	5.11	5.08	5.08
Depth	6.86	6.81	6.86	6.96	6.93	6.83

[a] A piece of the cylinder wall, weighing 1.9 g, broke out at the midpoint of 42 during the sintering cycle.

[b] Cylinder 47 prepared by recycling 42 through ceramic fabrication.

and minimum diameters of Table V shows that very little "hourglassing" occurs in the sintering of these cylinders. The average maximum diameter was 16.71 (±0.02) mm and the average minimum diameter was 16.6 (±0.02) mm. The average dimensions for the PRD cavity were 5.08 (±0.02) mm by 6.88 (±0.05) mm deep. The physical appearance of cylinders 38 through 41 was excellent. A piece of the cylinder wall of 42 weighing 1.9 g broke out at midpoint during the sintering process. Subsequent radiographic examination of the other cylinders revealed no defects in 38, 39, and 41. At least one transverse crack and one oblique crack were detected in 40. Cylinder 42 was reprocessed through ceramic fabrication (milling, screening, pressing, and sintering) to produce a ceramic of excellent physical appearance. Subsequent radiographic inspection, however, revealed a crack about midway along the length of the cylinder. This cylinder is designated as 47.

The second group of cylinders, 43 and 44, was prepared from Lot LAS 14 $^{238}PuO_2{}^{16}$ powder which was prepared 18 months before ceramic processing. After removal from storage in an inert atmosphere, the material was subjected to the following operations.

1. Oxygen isotopic exchange at 750°C,
2. 6-h heating at 1540°C, and
3. 18-h ball milling.

Cylinders 43 and 44 were pressed three months after operation 3, above. No difficulty was experienced with the pressing operation; integral pellets were obtained on the first attempt Sintering of 43 and 44, however, resulted in low-density cylinders that did not meet dimensional specifications. The diameters and lengths were too large. See Table VI. The density of the control pellets for these cylinders was only 81% of theoretical as compared to the usual 87 to 88%. Subsequent analysis of the particle size of the powder used for preparing 43 and 44 showed a significant difference from the usual ball-milled powder. The particle size distribution of the 43 and 44 powder is compared to the powder for preparations 38 through 42 and the powder for preparation 47, as shown in Fig. 4. (The latter two powders gave high-density cylinders and are typical of those measured on the usual ball-milled powder.[1]) The 43 and 44 powder contains much larger particles than either of the other two powders and its distribution curve is much flatter. The larger size of the 43 and 44 powder may be due to sintering of the powder during the 3-month storage of the ball-milled powder.

TABLE VI

PHYSICAL DIMENSIONS OF 29-W $^{238}PuO_2{}^{16}$ CYLINDERS

Serial Numbers
43 through 46

	43	44	45	46
Wt (g)	74.08	74.07	74.33	74.17
Dimensions, mm (specification - length 35.81 max, diam 17.02)				
Length	36.32	36.07	34.52	35.23
Diam				
Max	17.35	17.32	16.64	16.51
Min	17.32	17.27	16.66	16.51
Cavity for PRD, mm (specification - diam 4.83 min, depth 6.60 min)				
Diam	5.28	5.31	5.08	5.11
Depth	7.11	7.16	6.83	6.91

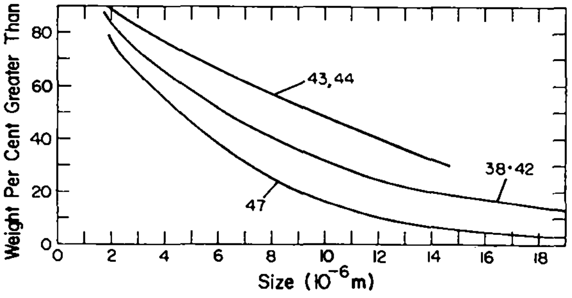


Fig. 4.
Particle size distribution of milled powders, preparations 38 through 44 and 47.

The control pellets for 43 and 44 were broken up in a SPEX mill and reprocessed through operations (2) and (3), above. The purpose of these operations was to resize the feed powder and to release stored helium gas, which might also affect pellet densities. This powder was then pressed into two small pellets and sintered. The resultant densities were 88% of theoretical. The particle size distribution for this feed powder is shown in Fig. 5. Cylinders 43 and 44 were then broken up and reprocessed through steps (1) through (3). The resulting powder, see Fig. 5 for particle size distribution, was pressed into cylinders 45 and 46. No difficulty was encountered in obtaining integral green units on the first attempt, as shown in Table VII which summarizes the pressing of cylinders 38 through 47. Sintering of the pellets resulted in cylinders well within the dimensional specifications shown in Table VI.

The reproducibility of product weights is important to meeting the wattage specifications. The weight of product feed is adjusted to the desired amount before adding carbowax (Ref. 1, p. 59). Small losses are encountered in the subsequent operations which include loading the die, pressing and extracting the fuel, sintering the cylinder, and, finally, chamfering the top edge of the PRD cavity. The material balances for fabricating cylinders 38 through 47 are given in Table VIII. The average weight of the final sintered and chamfered pellets was 74.14 (±0.10) g.

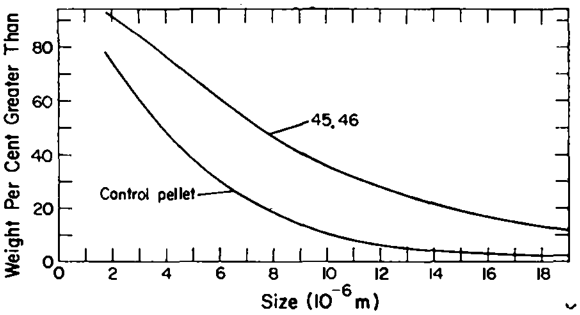


Fig. 5.
Particle size distribution of resized control pellet powder and resized powder for preparations 45 and 46.

TABLE VII

NUMBER OF PRESSINGS REQUIRED TO PRODUCE INTEGRAL "GREEN" CYLINDER

Cylinder Serial Number	No. of Pressings
38	2
39	1
40	3
41	1
42	1
43	1
44	1
45	1
46	1
47	1

The following conclusions can be drawn from these experiments.

a. Artificial heart heat sources of the design shown in Fig. 2 can be fabricated routinely by dry-pressing and sintering techniques.

b. The product weight can be controlled to better than 0.2 g. Thus the power output of a 33-W source could be controlled to 0.1 W.

c. Powders should be pressed as soon as possible after preparation.

The remaining problem areas in fabricating cylinders of the TRW design are

1. to fabricate "crack-free" cylinders on a routine basis. (Approximately half of the pellets have contained cracks detectable by radiography.)
2. to eliminate trichloroethylene as a solvent for carbowax, which is used as a pressing and sintering aid. (Trichloroethylene is decomposed by the heat and radiation from ^{238}Pu and releases a corrosive gas or vapor. Since this gas is generated in an inert glove box, its residence time in the box is relatively long and corrosion of equipment and work areas takes place. Thus, replacement of trichloroethylene by an inocuous solvent is desirable.)

TABLE VIII

MATERIAL BALANCES FOR FABRICATION OF CYLINDERS

Serial Numbers
38 through 47

Serial No.	Wt Oxide Feed (g)	Wt Sintered Pellet (g)	Wt Chamfered Pellet (g)	Processing Loss (g)
38	74.5312	74.2157	74.1970	0.334
39	74.5185	74.1255	74.1065	0.412
40	74.5169	74.1802	74.1588	0.358
41	74.5032	74.2050	74.1758	0.327
42	74.5392	74.3470	74.2975	0.242
43	74.5058	74.1125	74.0904	0.393
44	74.5030	74.0412	74.0252	0.195
45	74.5265	74.3590	74.3317	0.195
46	74.5338	74.1907	74.1717	0.362
47	74.502	73.8595	73.8314	0.671

B. Preparation of $^{238}PuO_2{}^{16}$ by Oxygen-Isotopic Exchange

Preparing $^{238}PuO_2{}^{16}$ by the reaction

$$^{238}PuO_2{}^{nat} + H_2O^{16} \longrightarrow {}^{238}PuO_2{}^{16} + H_2O^{nat}$$

was first reported by Porter and Thompson[5] who prepared powder having neutron emission rates as low as 5500 n/s-g ^{238}Pu. Several lots of $^{238}PuO_2{}^{16}$ powders have been prepared in the present program[1,6] by passing argon gas saturated with H_2O^{16} over plutonia at 700 to 750°C. The neutron emission rates of these powders were 300 to 1500 n/s-g ^{238}Pu higher than the measured rate for oxides prepared from electrorefined metal. Oxygen isotopic exchange was complete in all cases and the higher rates were due to chemical impurities in the oxide feed.

Only a limited effort was devoted to the exchange process during the present reporting period. This effort was devoted to

1. fabricating and evaluating a vertical column that allows H_2O^{16} and a carrier gas (helium) to flow down through a bed of $^{238}PuO_2$. The pressure in the apparatus can be lowered to 1 x 10^3 Pa to increase H_2O^{16} transport rate and thus increase the exchange rate.
2. improving the chemical purity of the oxide feed material. The present effort was limited to the volatilization of impurities from Savannah River oxide feed at 900°C.

The equipment for the isotopic exchange experiments is shown in Fig. 6. The reaction tube is platinum-lined nickel with a platinum frit close to the bottom to support the bed of $^{238}PuO_2$ powder. The small tube used has a capacity of ~40 g of powder. A quartz bubbler contains the H_2O^{16}. All H_2O in the off-gas is trapped in two series-connected liquid N_2-cooled Pyrex traps. A Welch Model 1397 vacuum pump was adequate to handle the flows used. All inner metallic surfaces between the bubbler and the platinum in the reaction tube are gold plated to eliminate any possible reaction with H_2O^{16}.

Two experiments were done in the above equipment. In the first one, as-received Savannah River Oxide (SRO), $^{238}PuO_2{}^{nat}$, Lot HBO 907545, was used as feed material. The chemical analysis of this material has already been reported.[1] In the second run, the same lot of oxide was heated in air at 900°C for 10 h before introducing it into the isotopic-exchange equipment.

The neutron emission rate of the oxide was monitored in situ during the experiments. Although these measurements were semiquantitative, they did indicate when exchange was complete as evidenced by a leveling-off of the neutron count. The operating pressure above the bed of 10 g of oxide was 4 x 10^3 – 9 x 10^3 Pa depending on helium flow through the H_2O^{16}. The H_2O^{16} flow rate through the oxide was

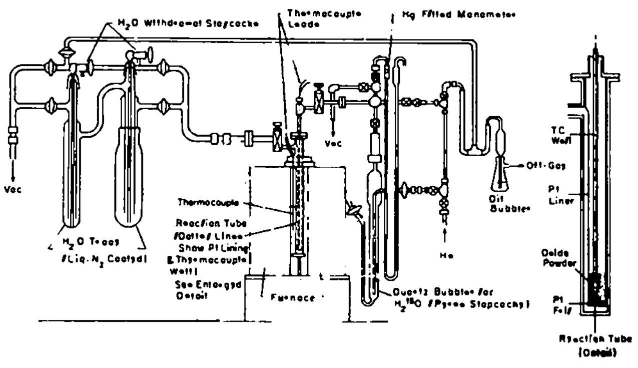


Fig. 6.
Oxygen isotopic exchange apparatus.

~1.6 g/h. The results of the neutron monitoring are shown in Fig. 7 where the relative neutron count rate is plotted as a function of time. (Relative neutron count rate = 100 x [counts/min at time(t)/counts/min at time 0]. In both experiments, approximately 5 h were required to reach complete exchange as evidenced by a "constant" neutron count rate. The experiments were continued for a total time of 12 h at 800°C. When the experiments were completed, samples of the powder were encapsulated and evaluated. Table IX gives the neutron emission rates for the oxide feed and the $^{238}PuO_2{}^{16}$ products. The neutron emission rates of the oxide feed powders were 18 600 and 16 500 n/s-g ^{238}Pu. The neutron emission rate of the products were 3850 and 3740 n/s-g ^{238}Pu. The lower emission rate of the feed for Experiment 2 was primarily due to volatilization of fluorine by the 900°C ignition in air. However, as shown in Table X, this pretreatment of powder did not remove fluorine completely from the natural oxide. Future volatilizations of impurities will be done in high vacuum at higher temperatures. Table X also shows a definite pickup of nitrogen which was subsequently largely removed during isotopic exchange. An apparent increase of sodium is indicated by these results, but the precision of the measurements must be improved before definite conclusions can be drawn. (The results given in Table X were obtained by gamma spectrometry, which, at present, is used to estimate relative amounts of impurities.) The oxygen isotopic analyses of the

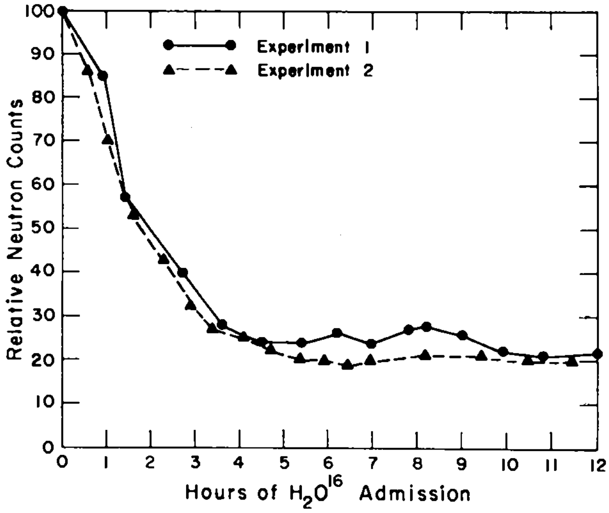


Fig. 7.
Relative neutron count rate as a function of time, oxygen-isotopic exchange experiments.

TABLE IX

NEUTRON EMISSION RATES OF OXIDE FEEDS AND PRODUCTS, ISOTOPIC EXCHANGE EXPERIMENTS

	(n/s-kg ^{238}Pu) x 10^{-3}	
	Expt 1	Expt 2
$^{238}PuO_2{}^{nat}$ feed	18 600	16 500
$^{238}PuO_2{}^{16}$ product	3 850	3 740

TABLE X

ANALYSIS OF LIGHT ELEMENT IMPURITIES IN IGNITED $^{238}PuO_2{}^{nat}$ and $^{238}PuO_2{}^{16}$

	Ratio, g Impurity in Oxide/g Impurity in S. R. Oxide			
Impurity	S. R. Oxide	Ignited S. R. Oxide	$^{238}PuO_2{}^{16}$ Expt 1	$^{238}PuO_2{}^{16}$ Expt 2
N	1.00	1.6	0.1	0.1
F	1.00	0.3	0.3	0.3
Na	1.00	1.1	1.1	1.1
Cl	1.00	0	0.2	0

H_2O^{16} water and the $^{238}PuO_2{}^{16}$ products are given in Table XI. These results show that oxygen-isotopic exchange was complete. The neutron and gamma dose rates of the $^{238}PuO_2{}^{16}$ powders are given in Table XII. The neutron dose rates were 0.39 and 0.38 R/h-kg ^{238}Pu and the gamma dose rates were 0.91 and 0.84 R/h-kg ^{238}Pu at 100 mm.

The results are compared with those obtained on $^{238}PuO_2{}^{16}$ powders prepared from electrorefined metal and show that the product from Isotopic-Exchange Experiment 2 has a neutron emission rate 360 n/s-g ^{238}Pu higher than the average rate measured on nine lots of powder.[1,4] The gamma dose rate of the Experiment 2 product is approximately twice that of the electrorefined-metal oxide powders. The high gamma dose rates are due primarily to ^{236}Pu daughters.

V. PREPARATION AND PROPERTIES OF PROCESS INTERMEDIATES

A. Electrorefined Metal and $^{238}PuO_2{}^{16}$ Powder

The two principal intermediates in the fabrication of medical-grade products are electrorefined metal and $^{238}PuO_2{}^{16}$ powder. Procedures for their preparation were given earlier[1] with results for electrorefining runs through 100-29 and for powder preparations through LAS 13. An additional nine electrorefining runs were made during the present reporting period to produce a total of 606 g of product. Approximately 420 g of this material had a ^{238}Pu isotopic content of 90 at.% and the balance was 80 at.% material. The chemical purity of the metal was comparable to the product from earlier runs;[1] however, to conserve ^{238}Pu, samples were not taken in every case. The neutron emission rates of product metal from several runs are given in Table XIII. The average emission rate for the nine 1- to 2-g samples was 3080 (±30) n/s-g ^{238}Pu. This rate is comparable to that reported earlier,[1] 3095 (±46) for 18 samples containing 1- to 2-g plutonium. The chemical purity of a typical lot of electrorefined metal is given in Table XIV.

Few, if any, impurities are introduced into the product in preparing $^{238}PuO_2{}^{16}$ powder. The chemical purity of a typical oxide is given in Table XV. A further indication of the purity of the oxide powders is given by the gamma spectrum which can detect trace amounts of fluorine, sodium, chlorine, nitrogen, oxygen, phosphorus, and aluminum. The impurity elements detected in the gamma spectrum of powders are given in Table XVI. Most of the lots showed no detectable impurities. The only element detected that would produce (α,n) neutrons is sodium, which was found in lot LAS 17. Spectrochemical analysis of this powder gave a value of 50 ppm for the sodium concentration, which shows that sodium can be detected at very low concentrations by gamma-ray spectrometry. This method can also be used for estimating ^{17}O and ^{18}O in $^{238}PuO_2{}^{16}$. Oxygen isotopic analyses for several lots of powder by gamma-ray spectrometry is given in Table XVII. The ^{18}O values are consistent with the ^{18}O concentration measured on H_2O^{16} used in the preparations. The ^{17}O values are higher than the ^{17}O measured on H_2O^{16}. Most of the lots of H_2O^{16} used in the preparations contained ~75 ppm ^{17}O (Ref. 4).

Work is continuing on the use of gamma-ray spectrometry for the nondestructive quantitative determination of ^{17}O, ^{18}O, and light element impurities (see Sec. X). The neutron emission rates of the eight lots of powder prepared during the present reporting period are given in Table XVIII. The average neutron emission rate of the eight lots was 3375 (±45) n/s-g ^{238}Pu. This compares to an earlier value of 3378

TABLE XI

OXYGEN ISOTOPIC ANALYSIS OF H_2O^{16} AND $^{238}PuO_2{}^{16}$

Description	^{17}O, ppm[a]	^{18}O, ppm[a]
H_2O^{16}	80[b]	15[b]
$^{238}PuO_2{}^{16}$, Expt 1	80[c]	10[c]
$^{238}PuO_2{}^{16}$, Expt 2	70[c]	10[c]

[a] g oxygen isotope/1000-kg oxygen.
[b] Measured by mass spectrometry.
[c] Measured by gamma spectrometry.

TABLE XII

DOSE RATES OF $^{238}PuO_2{}^{16}$ PRODUCTS, ISOTOPIC EXCHANGE EXPERIMENTS

Dose Rates at 100 mm[a]	Expt 1	Expt 2
Neutron, rem/h-kg ^{238}Pu	0.39	0.38
Gamma, R/h-kg ^{238}Pu	0.91	0.84

[a] Measured in air through 7.6-mm Ta on 2-g samples.

TABLE XIII

NEUTRON EMISSION RATES OF ELECTROREFINED METAL

Run No.	Sample No.	Pu (g)	Isotopic Comp wt% ^{238}Pu	Neutron Emission Rate, (n/sec-kg ^{238}Pu) x 10^{-3}
100-30	382	1.61[a]	89.87	3115
	385	5.09[b]	89.87	3125
100-31	408	1.52[a]	89.92	3120
	409	1.66[b]	89.92	3105
100-35	461	1.94[b]	90.42	3040
	467	14.08	90.42	3124
100-36	463	1.84[b]	90.42	3050
	464	2.07[b]	90.42	3030
	469	17.21	90.42	3110
100-42	524	1.60	79.81	3105
100-45	528	1.68	79.81	3080
	529	1.65	79.81	3090

---

[a] Sample from top of casting.

[b] Sample from bottom of casting.

TABLE XIV

CHEMICAL PURITY OF A TYPICAL LOT OF ELECTROREFINED ^{238}Pu METAL, RUN 100 - 45

Element	ppm[a]	Element	ppm[a]	Element	ppm[a]	Element	ppm[a]
Li	<2	V	<2	Y	<2	Ta	<100
Be	<1	Cr	5	Zr	<2	W	50
B	<1	Mn	1	Nb	<10	Re	<2
Na	<50	Fe	50	Mo	<2	Pb	<2
Mg	8	Co	<2	Cd	<2	Bi	<1
Al	<10	Ni	<2	Sn	<2	^{237}Np	360
Si	5	Cu	2	Cs	<10	^{241}Am	0
K	4	Zn	5	Ba	<2	^{234}U	0
Ca	20	Rb	<2	La	<2		
Ti	<2	Sr	<2	Hf	<2	Pu, wt% by titration	99.8

---

[a] g element/1000-kg sample.

TABLE XV

CHEMICAL PURITY OF A TYPICAL LOT OF $^{238}PuO_2{}^{16}$ POWDER, LOT LAS 16

Element	ppm[a]	Element	ppm[a]
Li	<1	Y	<2
Be	<1	Zr	<2
B	<1	Nb	<10
Na	<50	Mo	<2
Mg	2	Cd	<2
Al	15	Sn	2
Si	10	Cs	<10
K	5	Ba	<2
Ca	10	La	<2
Ti	30	Hf	<2
V	<2	Ta	<100
Cr	6	W	<50
Mn	<1	Re	<2
Fe	20	Pb	<5
Co	<2	Bi	<1
Na	<5	^{237}Np	410
Cu	3		
Zn	<10		
Rb	<2		
Sr	<2		

[a] g element/1000-kg sample.

TABLE XVI

IMPURITIES DETECTED IN THE GAMMA SPECTRUM, $^{238}PuO_2{}^{16}$ POWDERS

Lot	Element Detected
LAS 14	N
LAS 15	None
LAS 16	None
LAS 17	Na, Al
LAS 18	None
LAS 19	None
LAS 20	None

TABLE XVII

ESTIMATE OF ^{17}O, ^{18}O CONCENTRATIONS IN $^{238}PuO_2{}^{16}$ BY GAMMA-RAY SPECTROMETRY

	Isotopic Comp (ppm)[a]	
Lot	^{17}O	^{18}O
LAS 16	168	14
	169	21
LAS 17	172	18
	175	25
LAS 18	180	20
LAS 19	170	13
LAS 20	110	16
	100	25
LAS 21	120	20

[a] g oxygen isotope/1000-kg oxygen.

TABLE XVIII

NEUTRON EMISSION RATES OF $^{238}PuO_2{}^{16}$ POWDERS

Lot No.	Sample No.	Sample (g)	Isotopic Comp, wt% ^{238}Pu	Neutron Emission Rate, (n/s-kg ^{238}Pu) x 10^{-3}
LAS 14	344	1.06	79.73	3425
LAS 15	345	1.12	86.20	3375
LAS 16	386	1.84	89.93	3370
	387	4.43	89.93	3235
	391	1.97	89.93	3447
LAS 17	413	2.03	89.93	3465
	414	5.03	89.93	3390
LAS 18	444	2.12	90.0	3465
LAS 19	450	2.02	90.0	3310
LAS 20	517	2.03	90.42	3345
	518	2.06	90.42	3370
LAS 21	527	4.42	79.8	3290
	530	1.89	79.8	3320

(±63) n/s-g ^{238}Pu reported for nine lots of powder.[1] Thus the average increase in converting electrorefined metal to oxide was 295 n/s-g ^{238}Pu. This compares to an earlier value of 275 n/s-g ^{238}Pu.[1] As discussed below, most of this increase is due to the presence of small amounts of ^{17}O and ^{18}O in the oxide.

B. (α,n) Contribution from ^{17}O and ^{18}O in $^{238}PuO_2{}^{16}$

Most of the previous[1,5,7] estimates of the (α,n) contribution from ^{18}O and ^{17}O in $^{238}PuO_2{}^{nat}$ and $^{238}PuO_2{}^{16}$ were based on measurements made by Russian workers of the neutron yield from polonium nitrate solutions.[8] The conclusion drawn from this study was that the (α,n) cross section for ^{17}O was approximately 10% of that for ^{18}O. More recent experiments by workers at Lawrence Livermore Laboratory (LLL)[9] give data on the cross sections of the (α,n) reaction on ^{17}O and ^{18}O for alpha particle energies from 5 to 12.5 MeV. The ratio of ^{17}O to ^{18}O cross section at 4.6 MeV is 0.41 showing that more neutrons can be expected from (α,n) on ^{17}O than indicated by the measurements made on polonium. Thus for $^{238}PuO_2{}^{16}$ containing 10 ppm ^{18}O and 100 ppm ^{17}O, the contribution from ^{18}O and ^{17}O would be ~250 n/s-g ^{238}Pu* using the LLL cross sections and ~100 n/s-g ^{238}Pu* using the earlier Russian cross sections. As shown above, the average increase in neutron emission rate in converting electrorefined metal to $^{238}PuO_2{}^{16}$ powder is ~275 n/s-g ^{238}Pu. Our results appear to be in good agreement with the LLL values.

VI. PREPARATION AND PROPERTIES OF $^{238}PuO_2{}^{16}$ PRODUCTS

A. One-Watt Pellets

The major emphasis in fuel preparations has been on the fabrication and evaluation of 29-, 30-, 33-, and 50-W sources for dosimetry experiments, animal implantations, and systems studies. However, small sources, ~1-W, were prepared for other laboratories for studies not connected with the heart program. The properties of the pellets prepared for Monsanto Research Corporation and Lawrence Livermore Laboratory are given in Table XIX. These pellets were made from $^{238}PuO_2{}^{16}$ powder having an isotopic composition of 86.90 wt% ^{238}Pu and 0.25 ppm ^{236}Pu. The dimensions were 6.9 mm diam by ~8.5 mm tall and the power was 1.3 W. The average neutron emission rate of the pellets was 3250 n/s-g ^{238}Pu, which is somewhat lower than the rate usually observed on pellets of this size. The same lot of $^{238}PuO_2{}^{16}$ powder was used to fabricate 0.65-W pellets for Mound Laboratory. The dimensions of these pellets were 4.8 mm diam by ~4.9 mm tall. The properties of the pellets are given in Table XX. The average neutron emission rate was 3310 n/s-g ^{238}Pu. The chemical purity of the pellets described in Table XIX and XX is given in Table XXI.

One-watt $^{238}PuO_2{}^{16}$ pellets, having an isotopic composition of 89.95 wt% ^{238}Pu and 0.18-ppm ^{236}Pu, were made for Donald W. Douglas Laboratory. The properties of these pellets are given in Table XXII. The average neutron emission rate was 3350 n/s-g ^{238}Pu, which compares favorably to a value of 3390 n/s-g ^{238}Pu for the oxide powder (Lot LAS 17) used in the preparation.

B. Fifty-Watt Cylinders

The preparation of 50-W sources 8, 9, 10, 11, and 13 was reported in Ref. 1. Two additional 50-W sources were prepared during this reporting period. The properties of these cylinders are given in Table XXIII. The neutron emission rates of the cylinders were 3550 and 3591 n/s-g ^{238}Pu. It should be noted that the neutron multiplication factor in these relatively large sources is approximately 6%. The measured rates on the 1-W control pellets were 3320 n/s-g ^{238}Pu for cylinder 24 and 3410 n/s-g ^{238}Pu for cylinder 25. Both cylinders were prepared from oxide powder made from electrorefined metal. The chemical purities of the metal, powder, milled powder, and sintered cylinder are compared in Table XXIV. The analyses are fairly typical of the process used for the 50-W cylinders. The electrorefined metal is the purest material of the four items listed. The $^{238}PuO_2{}^{16}$ powder compares favorably to the metal. The ball milling and screening operations introduce small amounts of impurities, some of which are removed in the subsequent sintering operation. Plutonium isotopic analyses for cylinders 24 and 25 are given in Table XXV and the O/M ratios are given in Table XXVI. The oxides are stoichiometric $PuO_{2.00}$ within the limits of the chemical analyses used. (The O/M ratios were based on chemical analyses for oxygen and plutonium. A more precise method is now available; see Sec. X below.)

---

*In making these calculations a value of 1.18 n/s-g ^{238}Pu was used for the (α,n) contribution of natural oxide,[1] which contains oxygen having an isotopic content of 99.759 at.% ^{16}O.

TABLE XIX

PROPERTIES OF 1.3-W $^{238}PuO_2{}^{16}$ PELLETS FOR MONSANTO RESEARCH CORPORATION AND LAWRENCE LIVERMORE LABORATORY

	Pellet Serial Number				
	319[a,b]	18[a]	19[a]	22[c]	23[c]
Wt of fuel pellet (g)	3.123	3.082	3.152	3.074	3.075
Wt of ^{238}Pu (g)[d]	2.364	2.329	2.382	2.343	2.348
Pu isotopic composition					
Wt% ^{238}Pu isotope	86.90	86.90	86.90	86.30	86.30
ppm ^{236}Pu isotope	0.25	0.25	0.25	0.21	0.21
Dimensions					
Diam, mm	6.91	6.91	6.91	6.91	6.91
Height, mm	8.69	8.66	8.89	8.33	8.36
Density (Mg fuel/m^3)	9.63	9.49	9.45	9.83	9.84
Power (W)	1.34	1.32	1.35	1.33	1.33
Neutron emission rate					
n/s-capsule	7525	7650	7790	7620	7670
(n/s-kg ^{238}Pu) x 10^{-3}	3185	3285	3270	3255	3265
Dose rates at 10 cm					
Neutron rem/h-kg ^{238}Pu	0.32	0.33	0.33	0.33	0.33
Gamma R/h-kg ^{238}Pu[e]	—	—	—	0.25	0.25

---

[a] Pellets for Monsanto Research Corporation.

[b] A serial number was not assigned to this pellet. The sample number is therefore used for identification.

[c] Pellets for Lawrence Livermore Laboratory.

[d] Value based on calorimetry and Pu isotopic analysis.

[e] Measured in air through 0.076-cm Ta.

C. Thirty-Three-Watt Cylinders for Vented Heat Sources

Three 33-W fuel cylinders were made for encapsulation by Mound Laboratory in the artificial heart vented fuel capsule. The dimensions and geometry of this 33-W cylinder are given in Sec. II.A and in Fig. 2. Because of the tight time schedule for delivery of the cylinders to Mound Laboratory, the cylinders, designated as 35, 36, and 37, were fabricated before completing the ceramic fabrication development program (see Sec. IV.A, above). Three experiments were done with $^{238}PuO_2{}^{16}$, 80 wt% ^{238}Pu, before fabricating the 33-W cylinders, 90 wt% ^{238}Pu. These experiments demonstrated the feasibility of fabricating the cylinder as one unit, since integral, but cracked, cylinders were obtained in two of the three experiments. The procedures developed in these experiments were used to fabricate the 33-W cylinders.

The fabrication of 35 proceeded smoothly. An integral green pellet was readily obtained and was sintered to meet dimensional specifications. Considerable difficulty was experienced in obtaining integral green bodies for 36 and 37. Three attempts to press 36 powder resulted in laminated cylinders with the break occurring at the bottom of the cylinder. Similar difficulties were encountered with 37, but

TABLE XX

PROPERTIES OF 0.65-W $^{238}PuO_2$ [16] PELLETS FOR MOUND LABORATORY

	Pellet Serial Number							
	14	15	16	311[a]	312[a]	313[a]	17	315[a]
Wt of fuel pellet (g)	1.551	1.519	1.503	1.327	1.434	1.521	1.433	1.383
Wt of ^{238}Pu (g)[b]	1.171	1.147	1.134	1.002	1.083	1.148	1.081	1.044
Pu isotopic composition								
Wt% ^{238}Pu isotope	86.90	86.90	86.90	86.90	86.90	86.90	86.90	86.90
ppm ^{236}Pu isotope	0.25	0.25	0.25	0.25	0.25	0.25	0.25	0.25
Dimensions								
Diam (mm)	4.83	4.88	4.88	4.88	4.85	4.88	4.88	4.83
Height (mm)	8.84	8.64	8.64	7.92	8.64	8.92	8.61	8.39
Density (Mg PuO_2/m^3)	9.55	9.41	9.33	9.04	9.00	9.14	8.94	9.03
Power (W)	9.664	0.650	0.643	0.568	0.614	0.651	0.613	0.592
Neutron emission rate								
n/s-capsule	3920	3815	3785	3305	3450	3815	3640	3445
(n/s-kg ^{238}Pu) x 10^{-3}	3345	3325	3335	3300	3185	3325	3370	3300

---

[a] A serial number was not assigned to this pellet. The sample number is therefore used for identification.

[b] Value based on calorimetry and Pu isotopic analysis.

only one pressing was attempted. The powders for 36 and 37 were then sintered at 1530°C using the standard sintering cycle[1] and were remilled for 18 h. These powders were successfully pressed into green cylinders using a new set of die punches. The chemical purity of the three cylinders* is given in Table XXVII. The higher-than-usual values for aluminum, silicon, chromium, and iron are due to the excessive handling and processing of the powders during ceramic fabrication. Virtually all of the ^{234}U present in the fuel is due to decay of ^{238}Pu after fuel purification.

The isotopic composition of the three 33-W cylinders is given in Table XXVIII. The ^{238}Pu isotopic composition was greater than 90 wt% and the ^{236}Pu concentration was less than 0.20 ppm. The specifications and properties of the 33-W cylinders are given in Table XXIX. The average neutron emission rate of the three cylinders was 3460 n/s-g ^{238}Pu.

To ensure geometrical specifications, a fuel acceptance gauge designed and fabricated by TRW was used (TRW Drawing G411268, "Fuel Gauge Assy., AHHS). Each of the three cylinders fit readily into this gauge. The physical appearance of the cylinders was good; however, subsequent radiographic examination revealed one prominent transverse crack at the midlength of 35 and a crack at the upper chamfer on 36. Radiographic examination also indicated either a low-density area or a possible crack about 20 mm from the upper end of 36. There were no radiographically detectable defects in 37.

The cylinders were packaged for shipment as shown in drawing CMB-11-9868, September 15, 1972. An earlier shipment of one of the 80 at.% development cylinders, 34, showed that this method of shipping did not result in damage to the fuel cylinder. Shipment of the three cylinders to Mound Laboratory was accompanied by a LASL escort. Subsequent radiographic inspection of the sources at Mound Laboratory showed no apparent shipping damage. Calorimetric measurements at Mound Laboratory gave values of 33.042, 32.881, and 33.085 W, for 35, 36, and 37. The TRW design specification is 33.1

---

*Chemical and isotopic analyses are done on the control pellets that accompany all source fabrications. See Ref. 1, p. 59, for procedures.

TABLE XXI

CHEMICAL PURITY OF 1-W PELLET

Element	ppm[a]	Element	ppm[a]
Li	<0.2	Rb	<1
Be	<0.2	Sr	<0.2
B	<1	Y	<0.2
Na	40	Mo	18
Mg	8	Cd	<1
Al	40	Sn	<5
Si	15	Cs	<4
K	2	Ba	1
Ca	8	La	<2
Ti	<0.4	Hf	<1
V	<1	Re	<1
Cr	60	Pb	<1
Mn	4	Bi	<1
Fe	150		
Co	<1		
Ni	8	^{237}Np	269
Cu	10		
Zn	<10		

[a] g element/1000-kg sample.

(±0.2) W. All cylinders met both the fuel gauge dimensional specifications and the power specifications.

The welded tantalum shipping containers were opened at Mound Laboratory and the fuel cylinders were encapsulated in the TRW-designed vented fuel capsule. The three fuel cylinders were integral units and no apparent difficulties were encountered in the fueling operation. Mound Laboratory shipped two of the heat sources to Westinghouse Astro Nuclear Laboratory for engine studies and one source to LASL for storage and safety studies.

D. Twenty-Nine-Watt Heat Sources for Implantation Studies

Five 29-W $^{238}PuO_2{}^{16}$ heat sources, 80 wt% ^{238}Pu, were fabricated for implantation studies at Battelle PNL and Cornell University. The encapsulation of these cylinders is described below in Sec. VII. Fabrication of the fuel cylinders was done as part of the ceramic fabrication program described in Sec. IV.A. The cylinders were made from $^{238}PuO_2{}^{16}$ sources made for earlier program commitments. The composition of the feed powder is given above in Table IV, and the dimensions of the fuel cylinders 38, 39, 40, 41, and 47 are given in Table V. The fabrication procedure is given in Sec. IV.A.

The chemical purity of the fuel for cylinders 38, 39, 40, and 41 is given in Table XXX. The chemical analysis was made on one of the control pellets prepared for these cylinders. The principal impurity is ^{234}U resulting from the decay of ^{238}Pu. Crushing and grinding the 50-W cylinders introduced small amounts of silicon, chromium, iron, and nickel. The properties of the control pellets for the 29-W cylinders are given in Table XXXI. The average density of the four control pellets is 87.3% of theoretical and the average neutron emission rate is 3465 n/s-g ^{238}Pu. As shown in Table XXXII, the O/M ratio of the control pellets is 1.998 (±0.004) and the water concentration is less than 2 ppm. The properties of the encapsulated 29-W sources are given in Table XXXIII. Measurements were made on the encapsulated heat source, which consisted of a 0.50-mm-thick tantalum liner, a 1.52-mm-thick Ta-10W strength member, and a 0.50-mm-thick Pt-20Rh outer jacket. Fuel cylinder 47 was prepared by recycling 42 through ceramic fabrication as described in Sec. II.A; 38, 39, 40, 41, and 42 were prepared from the same lot of $^{238}PuO_2{}^{16}$ powder. The average neutron emission rate of 29LPX3038, 29LPX3039, 29LPX3040, and 29LLX3041 is 3525 n/s-g ^{238}Pu. This rate is comparable to that measured on the 33-W cylinders, above, where an average rate of 3560 n/s-g ^{238}Pu was obtained. The plutonium isotopic analysis of the 29-W sources is given in Table XXXIV. A typical gamma spectrum is given in Fig. 8 and a typical gamma dose rate plot is given in Fig. 9. Sodium was the only light element impurity detectable in the gamma spectrum; the peak at 1806 keV is due to $^{23}Na(\alpha,p)\,^{26}Mg$.

VII. ENCAPSULATION OF NONVENTED HEAT SOURCES

The nonvented ^{238}Pu sources fabricated in the medical-grade fuel program for dosimetry, implantation, and system studies have been made at LASL. In past years, these sources have included

One 30-W Pu-3 at.% Ga source for dosimetry studies at Battelle, PNL,

Two 30-W $^{238}PuO_2{}^{16}$ sources for systems studies at Hittman Associates, and

One 32-W $^{238}PuO_2{}^{16}$ source for implantation studies at Battelle, PNL.

TABLE XXII

PROPERTIES OF 1-W $^{238}\mathrm{PuO}_2{}^{16}$ PELLETS FOR DONALD W. DOUGLAS LABORATORY

	Capsule Number									
	26	27	28	400[a]	29	402[a]	403[a]	404[a]	30	31
Wt of fuel pellet (g)	2.263	2.269	2.280	2.257	2.277	2.282	2.244	2.250	2.288	2.264
Wt of ^{238}Pu (g)[b]	1.784	1.784	1.789	1.775	1.791	1.795	1.765	1.770	1.752	1.782
Pu isotopic composition										
Wt% ^{238}Pu isotope	89.95	89.95	89.95	89.95	89.95	89.95	89.95	89.95	89.95	89.95
ppm ^{236}Pu isotope	0.18	0.18	0.18	0.18	0.18	0.18	0.18	0.18	0.18	0.18
Dimensions										
Diam (mm)	7.98	7.98	7.98	7.95	7.98	7.98	7.98	7.95	7.98	7.98
Height (mm)	4.50	4.47	4.52	4.62	4.47	4.47	4.52	4.50	4.39	4.55
Density (Mg fuel/m^3)	10.1	10.2	10.1	--	10.2	10.2	—	10.1	10.1	9.98
Power (W)	1.011	1.011	1.014	1.006	1.015	1.017	1.000	1.003	0.993	1.010
Neutron emission rate										
n/s - capsule	5845	5930	5950	6005	6025	6035	5940	5935	5995	5920
(n/s-kg ^{238}Pu) x 10^{-3}	3275	3325	3325	3380	3365	3365	3365	3355	3420	3320

---

[a] A serial number was not assigned to this pellet. The sample number is therefore used for identification.

[b] Value based on calorimetry and Pu isotopic analysis.

The following sources were fabricated and delivered during the current reporting period.

One 50-W $^{238}\mathrm{PuO}_2{}^{16}$ source to Battelle, PNL for implantation studies,

Four 29-W $^{238}\mathrm{PuO}_2{}^{16}$ sources to Battelle, PNL for implantation studies, and

One 29-W $^{238}\mathrm{PuO}_2{}^{16}$ source to Cornell University for implantation studies.

A. Quality Control Program

A quality control program was initiated in FY 1972 for fabricating heat sources in the present program at LASL. This program is defined in document 0820-QCP-1 "LASL Quality Control Program for Division of Applied Technology Products (Program 0820, Preparation and Evaluation of Pu-238 Radioisotopic Heat Sources), March 31, 1972." Fabrication of the 50-W heat sources is defined in Experimental Plan 0820-EP-2. The experimental plans cover the fabrication of the fuel, material procurement and acceptance, fabrication, and preparation of capsule components, assembly, and fabrication of the heat source, product index sheet, and packaging and shipping.

B. Encapsulation

1. Fifty-Watt Sources. Three 50-W $^{238}\mathrm{PuO}_2{}^{16}$ cylinders, 13, 24, and 25, were encapsulated as shown in LASL Drawing 26Y-74527 (7-24-72) and Experimental Plan 0820-EP-1, Revision O. This capsule consists of three containers, an inner tantalum liner (0.51-mm wall thickness), a Ta-10W strength member (1.52-mm wall thickness), and a Pt-20Rh outer container (0.51-mm wall thickness). The closure welds in the tantalum liner and Pt-20Rh capsules are cap welds and the closure weld in the Ta-10W capsule is a circumferential weld. The capsule is shown in Fig. 10.

Considerable difficulty was encountered in the circumferential closure weld of the Ta-10W strength member. Pressurization of the capsule during TIG welding resulted in radical thinning of the wall thickness in two of the three capsules welded. The thinning was detected visually in 50LEX1024 and the

TABLE XXIII

PROPERTIES OF 50-W $^{238}\mathrm{PuO}_2$[16] CYLINDERS

Serial Numbers
24 and 25

	24	25
Wt of fuel pellet (g Pu)	126.51	125.01
Wt of ^{238}Pu (g)	89.21[a]	88.31[a]
Oxygen/Pu Ratio	1.99	2.03
Pu isotopic composition		
Wt% ^{238}Pu isotope	79.65	80.14
ppm ^{236}Pu isotope	0.64	0.64
Dimensions		
Diam (mm)	25.50	25.22
Height (mm)	25.48	25.15
Density (Mg fuel/m^3)	9.73	9.95
Power (W)	50.57	50.07
Power Density (W/cm^3)	3.89	3.99
Neutron emission rate		
(n/s-kg Pu) x 10^{-3}	2827	2878
(n/s·kg ^{238}Pu) x 10^{-3}	3550	3591
Dose rates at 10 cm		
Neutron, rem/h·kg ^{238}Pu	0.36	0.36
Gamma, R/h-kg ^{238}Pu	0.20[b]	0.19[b]

---

[a] Calculated from calorimetry and Pu mass spectrometry.

[b] Measured through 0.051-cm Ta and 0.051-cm stainless steel.

attempted repair resulted in a "blow-out." The thinning in 50LEX1013 was detected by radiography. The welding of 50LPX1025 was satisfactory; the minimum weld thickness of the Ta-10W capsule in this case was ~83% of the wall thickness. No difficulty was encountered with any of the tantalum liners or Pt-20Rh capsule weldings; all welds passed inspection. Heat sources 50LEX1013 and 50LEX1024 were rejected because of the Ta-10W welds. Heat source 50LPX1025 was accepted and was shipped to Battelle, PNL, for implantation studies.

2. Twenty-Nine-Watt Sources. The capsule design for the 50-W sources was used for encapsulating 29-W sources for implantation studies. However, the poor results obtained in welding the Ta-10W capsules, above, pointed out the obvious need for a weld development program before proceeding to the 29-W sources. An experimental series of Ta-10W closure welds was therefore initiated to determine adequate welding parameters.

a. Ta-10W Weld Development Program. Cost considerations led to using test weld capsules fabricated from two pieces of 25.4-mm-diam Ta-10W tubing as shown in Fig. 11. An inner tantalum liner (not shown) was used to approximate the clearances and void volumes of the heat-source capsule shown in Fig. 10.

Approximately 24 experiments were made using various procedures and welding parameters. The frequency of success was encouraging with a vented double overlap scheme. This method uses a 0.25-mm tungsten heat shield between the Ta-10W and tantalum in the weld area. A tantalum shim is also used to ensure centering the liner. The procedure developed is given below. Prior to its use with the 29-W sources, it was tried successfully on a full-scale encapsulation of a $\mathrm{ThO_2}$ cylinder and a full-scale encapsulation of a 30-W cylinder (H/D = 1).

b. Encapsulation of 29-W Cylinders. Five 29-W cylinders, 38, 39, 40, 41, and 47, have been encapsulated as per Quality Control Program 0820-QCP-1, Revision O, and Experimental Plan 0820-EP-2. The configuration of the capsule is given in Fig. 12. All encapsulations were successful; all heat sources were accepted. The procedure for welding the Ta-10W capsule is given below.

Procedures for Ta-10W Encapsulations of 29-W Sources.

All part numbers refer to LASL Drawing 26Y-74997.

Procedures preceding welding are as follows.

1. Vent the capsule, part 4, at 90° intervals.
2. Lap parts 3 and 4 to gain intimate contact and then clean as per Quality Control Procedures PP-C-1, Rev. 1, paragraphs 4-6.
3. The source in its decontaminated and leak-checked tantalum liner is placed in Ta-10W, part 4, along with tantalum felt, tantalum shim, and tungsten strip (parts 14, 5, and 15, respectively).
4. Place the Ta-10W lid, part 3, on top and clamp the assembly in the welding fixture chuck. Adjust to eliminate run-out of more than 0.05 mm for part 4 and then align the lid using a fixture.

Welding Procedures for Ta-10W.

1. After a 50% Ar-50% He atmosphere has been established, tack weld the lid on at 120° intervals using 120 A.
2. Remove the alignment fixture, re-establish a 50% Ar-50% He atmosphere and perform

TABLE XXIV

CHEMICAL PURITY OF 50-W CYLINDERS 24, 25, AND PROCESS INTERMEDIATES IMPURITY CONCENTRATION, ppm

	Serial Number 24				Serial Number 25			
Element	Metal	Powder	Milled Powder	Cylinder	Metal	Powder	Milled Powder	Cylinder
Li	<0.1	<0.2	<0.2	1	<0.1	<0.2	0.3	0.3
Be	<0.1	<0.2	<0.2	<1	<0.1	<0.2	<1	<1
B	<1	1	4	<1	<1	<1	4	2
C	45	150	35	110	60	55	120	30
Na	<1	20	50	<50	5	20	<50	<50
Mg	<1	10	8	5	5	5	120	120
Al	15	3	10	25	7	1	10	15
Si	<25	20	100	60	<1	20	40	25
K	<0.5	<1	250	45	<3	<1	20	38
Ca	<3	<6	100	<10	<3	<6	25	5
Ti	<0.2	12	35	35	1	20	10	--
V	<0.5	<1	<1	<2	<0.5	<1	<2	<2
Cr	6	6	50	100	1	8	50	25
Mn	1	3	4	5	1	2	5	<2
Fe	20	85	250	150	55	80	200	30
Co	<0.5	<1	<5	<2	<0.5	<1	<2	<5
Ni	5	10	10	15	3	10	15	<5
Cu	2	4	75	<1	2	5	50	<2
Zn	<5	<10	50	<10	<5	<10	50	<10
Rb	<0.5	<1	<1	<2	<0.5	<1	<2	
Sr	<0.1	<0.2	<0.2	<2	<0.1	<0.2	<2	<10
Y	<0.1	<0.2	<0.2	<2	<0.1	<0.2	<2	<2
Zr	<0.1	<0.2	<0.2	8	<0.1	<0.2	<2	<1
Mo	<0.5	1	2	<2	<0.5	<1	3	12
Cd	<0.5	<1	<1	<2	<0.5	<1	<2	<20
Sn	<0.5	2	50	<5	<0.5	1	25	5
Cs	<2	<4	<4	<2	<2	<4	4	<10
Ba	4	10	<0.2	4	5	2	4	1
La	<1	<2	<2	<2	<1	<2	<2	<2
Hf	<0.5	<1	<1	<2	<0.5	<1	<2	<2
	<0.5	<1	<1	<2	<0.5	<1	<2	<2
Pb	1	2	5	<5	1	2	5	<5
Bi	<1	<1	<1	<1	<1	<1	<1	<1
Np	470	--	—	516	664	--	--	654

TABLE XXV

PLUTONIUM ISOTOPIC COMPOSITION
Cylinders 24 and 25

Pu Mass No.	Wt%[a] 24	Wt%[a] 25
236	0.64×10^{-4}	0.64×10^{-4}
238	79.65	80.14
239	16.29	15.91
240	2.77	2.70
241	0.94	0.90
242	0.35	0.35

---

[a] g of isotope/0.100·kg Pu.

Weld No. 1 as shown in Fig. 13 at about 6 x 10^4 Pa.

3. After 1/2 h cooling, perform Weld No. 2 as shown in Fig. 13.
4. Cool 1/2 h before breaking the inert atmosphere and then leak check.

The actual TIG welding parameters used for the five 29-W Ta-10W capsules varied slightly, but are typified in Fig. 13.

VIII. HELIUM RELEASE FROM $^{238}PuO_2{}^{16}$ FUEL CYLINDER

A. Introduction

The alpha decay of ^{238}Pu continuously generates helium within the ceramic fuel body. The rate and manner of helium release from the oxide pellet influence the internal pressures occurring within a heat-source capsule and dictate the helium flow rate

TABLE XXVI

OXYGEN TO METAL RATIO

Cylinder Serial Number	Oxygen/Metal
24	1.98
25	2.03

TABLE XXVII

CHEMICAL PURITY OF 33-W CYLINDERS
Serial Numbers 35, 36, and 37

Element	Impurity (ppm) 35	Impurity (ppm) 36	Impurity (ppm) 37
Li	<2	<2	<2
Be	<1	<1	<1
B	<1	<1	<1
Na	<50	<50	<50
Mg	30	40	25
Al	20	60	85
Si	110	200	280
K	2	6	4
Ca	10	25	15
Ti	<25	<25	25
V	<2	10	<2
Cr	7	65	380
Mn	2	4	15
Fe	140	360	600
Co	<2	<2	20
Ni	6	<2	15
Cu	<1	<1	2
Zn	<10	<10	<10
Rb	<2	<2	<2
Sr	<2	<2	<2
Y	<2	<2	<2
Zr	2	<2	<2
Mo	<2	<2	<2
Cd	<2	<2	<2
Sn	<2	<2	<2
Cs	<10	<10	<10
Ba	6	4	6
La	<2	<2	<2
Hf	<2	<2	<2
Re	<2	<2	<2
Pb	<2	<2	<2
Bi	<1	<1	<1
^{241}Am	6	4	9
^{237}Np	1570	541	1150
^{232}U	0.015	0.008	0.020
^{234}U	1590[a]	1040[b]	2610[c]
^{228}Th	0.00012	0.00005	0.00011

---

[a] Date of fuel purification (LAS 18) by electrorefining was 9/11/72; date of analyses for U and Th was 11/14/72.

[b] Date of fuel purification (LAS 19) by electrorefining was 9/15/72; date of analyses for U and Th was 11/14/72.

[c] Fuel composed of powders from Lots LAS 16, 17, 18, and 19. Average purification date was 6/22/72; date of analyses for U and Th was 11/15/72.

TABLE XXVIII

PLUTONIUM ISOTOPIC COMPOSITION OF 33-W CYLINDERS

Pu Isotope	Weight Percent (g Pu isotope/0.100-kg Pu) 35[a]	36[a]	37[a]
236	0.14×10^{-4}	0.17×10^{-4}	0.18×10^{-4}
238	90.14	90.24	90.04
239	9.22	9.13	9.30
240	0.595	0.589	0.614
241	0.030	0.031	0.031
242	0.009	0.008	0.008

[a] Date of Isotopic Analysis was 12/21/72.

of a practical capsule vent. Thus the release characteristics of the fuel cylinder are important considerations for the safe containment of the fuel material.

Investigation of helium release from actinide fuels employed in heat sources has produced a general description of the sort of release behavior that might be expected from each fuel form. A study of release from $^{244}Cm_2O_3$ pellets[10] showed that release was quantitative as the helium was formed at a temperature of 1280°C. Release from $^{238}PuO_2$ microspheres was found to be complete within minutes at 1300°C, or above, at Oak Ridge National Laboratory (ONRL).[11] This was supported by bakeout studies on SNAP-27 fuel at Mound Laboratory[12] and by microsphere studies at LASL.[13,14] The last work also examined the behavior of several other $^{238}PuO_2$ fuel forms including cold-pressed and sintered cylinders (6.4 by 6.4 mm), solid solution of plutonia with thoria and zirconia, solid solution cermets (SSC), plutonia-molybdenum cermets (PMC), and 51-mm-diam by 6.4-mm-thick PMC disks. The cermets acted much like the

TABLE XXIX

SPECIFICATIONS AND PROPERTIES OF 33-W $^{238}PuO_2$[16] CYLINDERS

	Specifications	Cylinder Serial Number 35	36	37
Wt of fuel (g)	—	73.545	73.506	74.071
Wt of ^{238}Pu (g)	—	58.32[a]	58.03[a]	58.39[a]
Power (W)	33.1 ± 0.2	33.042[b]	32.881[b]	33.085[b]
Dimensions				
Length (mm)	35.81 max	34.71	34.25	34.59
Diam (mm)	17.02 max	16.71 max 16.61 min	16.66 max 16.54 min	16.64 max 16.56 min
Cavity for PRD				
Diam (mm)	4.83 min	5.11	>4.83	>4.83
Depth (mm)	6.60 min	6.96	>6.60	>6.60
Neutron emission rate				
(n/s-kg fuel) $\times 10^{-3}$	--	2695	2795	2715
(n/s-kg ^{238}Pu) $\times 10^{-3}$	--	3400	3540	3445

[a] Value based on calorimetry.

[b] Wattage measured by Mound Laboratory (MRC).

TABLE XXX

CHEMICAL PURITY OF 29-W CYLINDERS
Serial Numbers 38 through 41

Element	ppm[a]	Element	ppm[a]
Li	<2	Rb	<2
Be	<1	Sr	<2
B	<1	Y	<2
Na	<50	Zr	4
Mg	10	Mo	<2
Al	55	Cd	<2
Si	200	Sn	4
K	6	Sc	<10
Ca	55	Ba	4
Ti	30	La	<2
V	<2	Hf	2
Cr	130	Re	<2
Mn	9	Pb	5
Fe	430	Bi	<1
Co	4	^{241}Am	970[b]
Ni	45	^{237}Np	470
Cu	2	^{232}U	0.300[b]
Zn	5	^{234}U	13600[b]
		^{228}Th	0.0026[b]

---

[a] g element/1000-kg fuel.

[b] Values as of 5/1/73.

microspheres by generally producing larger gas release rates at a given temperature than did the solid solution and pure PuO$_2$ 6.4-mm pellets. Despite the differences between microspheres and pellets, a different release mechanism was detected for both forms below 1000°C from that which prevailed at higher temperatures.

Neither these investigations nor an analysis of the microsphere data at Sandia Laboratories[15] have provided mathematical descriptions which permit reliable long-term predictions of release behavior, given the characteristics of a fuel material, except in instances of total release at elevated temperatures, 1300 to 1900°C. Thus experimental observation continues to be the most certain indicator of what behavior may be expected from a fuel form.

The objectives of the present investigation include (1) determining the rate of releasing helium from the fuel form at operating temperature and the variation of rate with time, (2) determining the amount of helium retained within the fuel material at any given time, and (3) determining the rate and extent of releasing helium from the fuel under accident conditions.

B. Experimental

1. Apparatus

a. Steady-State Testing. The vacuum system and equipment employed in measuring helium release and sampling radon release from the fuel pellet under steady-state conditions is illustrated in Fig. 14. The oxide cylinder was contained in a Ta-10W inner container and a nickel outer container both of which were connected to a vacuum system manifold. The outer container shown in Fig. 15 served as a furnace tube. Both containers normally operated under a vacuum of 10^{-3} Pa but could be backfilled with argon or helium. The vacuum manifold contained a solenoid valve that isolated the fuel from the remainder of the system during a power failure.

A Lindberg Hevi-Duty Model M-3012 resistance furnace with covered nickel chrome heating elements was used to heat the sample. The furnace and its associated vacuum manifold are shown in Fig. 16. Power was supplied by a Minneapolis Honeywell Pyrovane Controller as regulated by the input from a Chromel-Alumel thermocouple. A redundant controller system provided excess temperature protection. The fuel temperature was determined by a Chromel-Alumel thermocouple placed directly on top of the cylinder.

The helium was continually evacuated through the main vacuum system which was in turn vented into the glove box exhaust system. A measurement of the helium released from the fuel body was achieved by valving off the main vacuum system and evacuating through the calibrated Consolidated Electrodynamics Model 24-120B helium leak detector shown in Fig. 17. The mass spectrometer leak detector had a detection range of 2×10^{-10} to 2×10^{-4} std cm^3 He/s. It was calibrated with a Veeco type SC-4 sensitivity calibrator.

The furnace, encapsulation, and vacuum manifold were contained inside an air-atmosphere glove box, whereas the leak detector, which was vented back into the glove box, was attached to the system outside the enclosure. No filters were placed in the lines leading to the leak detector. Rather, the integrity of the ^{238}PuO$_2$[16] pellet and a minimum of oxide dust particles were relied upon to keep the external system free of significant radioactive contamination.

TABLE XXXI

PROPERTIES OF CONTROL PELLETS FOR 29-W CYLINDERS
Serial Numbers 38 through 41

	Sample Number			
	487	488	489	490
Wt of pellet (g)	2.0045	2.0013	2.0103	5.4597
Wt of ^{238}Pu (g)	1.376	1.374	1.380	3.749
Dimensions				
Diam (mm)	0.808	0.810	0.808	0.803
Height (mm)	0.394	0.388	0.395	1.060
Density (Mg fuel/m^3)[a]	9.94	10.00	9.91	10.18
Percent of theoretical density[b]	86.7	87.3	86.5	88.8
Neutron emission rate				
(n/s-kg fuel) x 10^{-3}	2405	2370	2390	2360
(n/s-kg ^{238}Pu x 10^{-3}	3500	3450	3480	3435

---

[a] Calculated from measured geometry and weight.

[b] Based on a theoretical density of 11.46 Mg/m^3.

TABLE XXXII

OXYGEN-TO-METAL RATIO AND H_2O CONCENTRATION OF 29-W PELLETS

Oxygen/Metal	1.998 (±0.004)
H_2O Concentration (ppm)	
g H_2O/1000-kg fuel)	<2

This approach avoided the potential problems of holdup of the helium flow and outgassing from the filter material. No contamination was found in the leak detector by standard alpha-monitoring procedures.

b. Temperature-Excursion Testing. The test material is contained in a Ta-10W container vented by a tantalum tube to the vacuum manifold. The container sits on a tantalum stand and both are located inside quartz vacuum-envelope connected to the main vacuum system. The container and the envelope normally operate under a vacuum of 10^{-3} Pa, although either or both can be back-filled with argon or helium.

The sample is heated by an induction coil wound about the exterior of the quartz envelope. A 15-kW Tocco motor generator set and capacitor bank, model 1MG-152-1, supply power to the coil. A manual variac provides control of the power level and, in turn, the temperature of the sample. The latter is monitored with platinum, Pt-10Rh thermocouple in contact with the bottom of the capsule and with a Leads and Northrup optical pyrometer, model 8622-C.

Several safeguards are built into the system. If the cooling water flow to the motor generator set, the induction coil, or envelope lid fails, the motor generator set automatically shuts down. If cooling water to the diffusion pump fails, it shuts off. If a pressure of more than 6.7 Pa occurs within the vacuum system, power is removed from the mechanical pump, diffusion pump, and motor generator set. None of these units can resume functioning until a reset switch is manually engaged.

The helium release during temperature excursion is determined by the helium leak detector described above with certain modifications. The latter are designed to decrease the sensitivity of the leak detector because the higher release rates expected at higher temperatures could readily overwhelm the instrument. The alterations consist of a less sensitive

TABLE XXXIII

PROPERTIES OF 29-W $^{238}PuO_2{}^{16}$ HEAT SOURCES[a]

	29LPX3038	29LPX3039	29LPX3040	29LLX3041	29LPX3047
Wt of heat source (g)	400	399	399	399	398
Wt of fuel (g)	74.18	74.09	74.15	74.14	73.83
Wt of ^{238}Pu (g)[b]	50.98	50.84	50.91	50.91	50.45
Dimensions of heat source					
o.d. (mm)					
max	32.51	32.51	32.51	32.51	32.51
min	32.38	32.21	32.23	32.23	32.21
Length (mm)					
max	50.93	50.83	50.77	50.77	50.85
min	50.72	50.77	50.65	50.65	50.70
Power (W)[c]	28.91	28.83	28.87	28.87	28.61
Specific power (kW/kg fuel)	0.3897	0.3891	0.3893	0.3893	0.3875
Neutron emission rate (n/s-kg ^{238}Pu) x 10^{-3}	3540	3470	3570	3510	3660
Dose rates in air at 10 cm					
Gamma, R/h-kg ^{238}Pu[d]	0.23	0.23	0.22	0.25	0.25
Neutron, rem/h-kg ^{238}Pu	0.35	0.35	0.36	0.35	0.37

---

[a] All properties were measured on the fueled capsule as shown on LASL drawing 26Y-74997.

[b] ^{238}Pu values are based on calorimetry and Pu isotopic analysis.

[c] Power measured calorimetrically.

[d] Measured in air through 0.050-cm Ta, 0.152-cm Ta-10W, and 0.050-cm Pt-20Rh.

direct current amplifier within the detection unit and admission of the gas sample into the leak detector directly above the diffusion pump. The sensitivity now extends over a range of about 2 x 10^{-8} to 2 x 10^{-2} std cm^3 He/s-g $^{238}PuO_2{}^{16}$.

Still, the leak detector can be saturated with helium. In that event the release is estimated by measuring the pressure within a known volume of the vacuum system with a Wallace Tiernan differential pressure gauge, Model FA 141. This pressure is corrected for heating effects within the sample container. The Wallace Tiernan gauge is calibrated against a mercury manometer.

2. Fuel Cylinder. The characteristics of the fuel cylinder used to date are shown in Table XXXV. Cylinder 5, shown in Fig. 18, was representative of the 30-W artificial heart fuel form[1] at the time the helium release studies were initiated. The microstructure of this fuel form is given in Fig. 19. The fuel is of open porosity, which should permit relatively free release of helium. Cylinder 5 was in shelf storage for 857 days before initiating the steady-state helium release experiment.

3. Procedure

a. Steady-State Testing. The fuel container was connected to the vacuum system, with both containers having a helium atmosphere. The furnace was open with water flowing through the cooling coils and with a blower cooling the surface of the outer container. Under these conditions the cylinder temperature was ~350°C. The experiment was initiated by pulling a vacuum on the inner container and allowing the fuel to self-heat to 552°C. The blower was then stopped, the furnace was closed, and the cylinder was gradually heated to the 870°C operating temperature. This temperature was maintained throughout the experiment except when equipment failure or maintenance work necessitated a shutdown. During the initial period of rapidly changing release rate, the flow was monitored closely, but as the rate varied less and less, it was measured twice a week.

TABLE XXXIV

ISOTOPIC ANALYSIS OF 29-W CYLINDERS
Serial Numbers 38, 39, 40, 41, and 47

Pu Isotope	Weight Percent (g Pu isotope/0.100-kg Pu)
236	0.40×10^{-4}
238	79.63
239	16.39
240	2.81
241	0.83
242	0.34

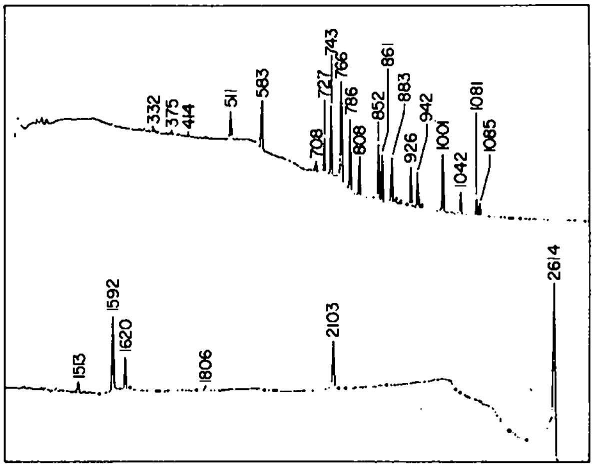


Fig. 8.
Gamma spectrum of PNL 29-4 (energy in keV).

b. Temperature-Excursion Testing. The temperature excursion apparatus has just been assembled and actual experimentation has not yet begun. The planned procedure involves bringing the pellet used in steady-state testing to 870°C and holding it there until the helium release is stable. The temperature excursion will be to 1370°C over a 45-min interval.

C. Results

The steady-state helium release of the $^{238}PuO_2{}^{16}$ cylinder 5 is shown in Fig. 20. The duration of the experiment was 5325 h, but downtime reduced the time at temperature to 5041 h. Each interruption of heating was characterized by a very low release rate.

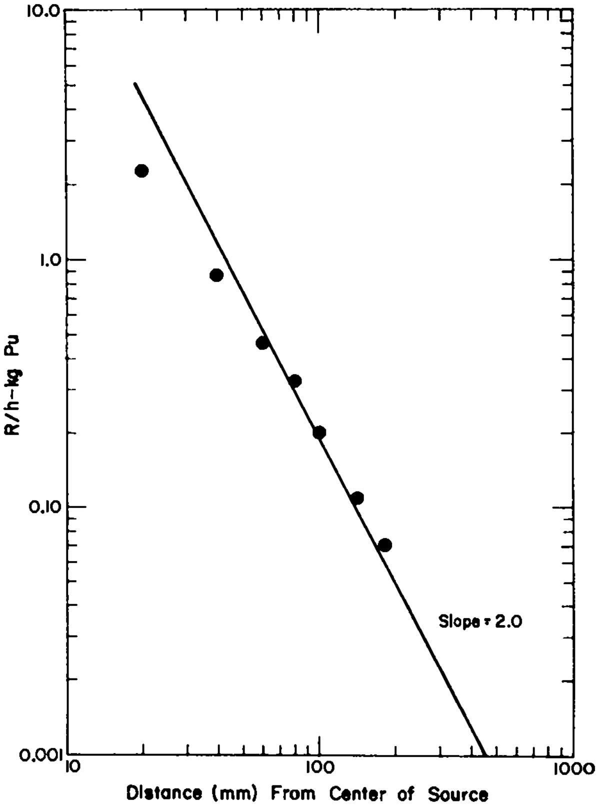


Fig. 9.
TLD measurements on 29LLX3041.

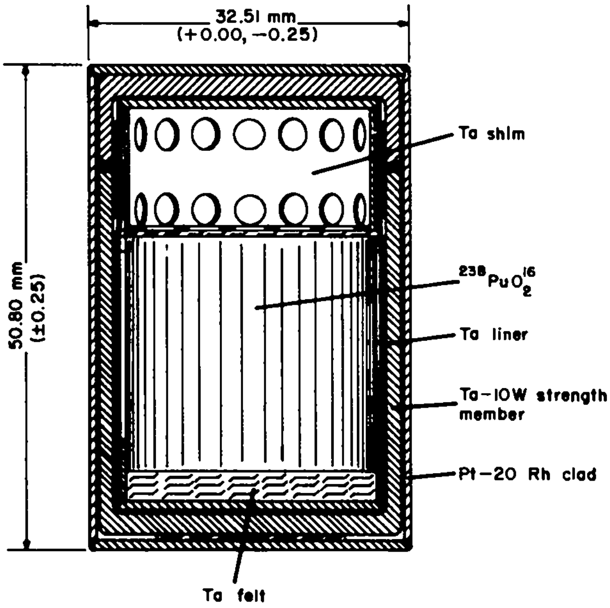


Fig. 10.
Sealed 50-W heat source.

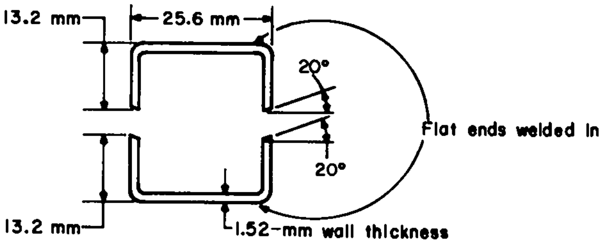


Fig. 11.
Ta-10W weld development capsule.

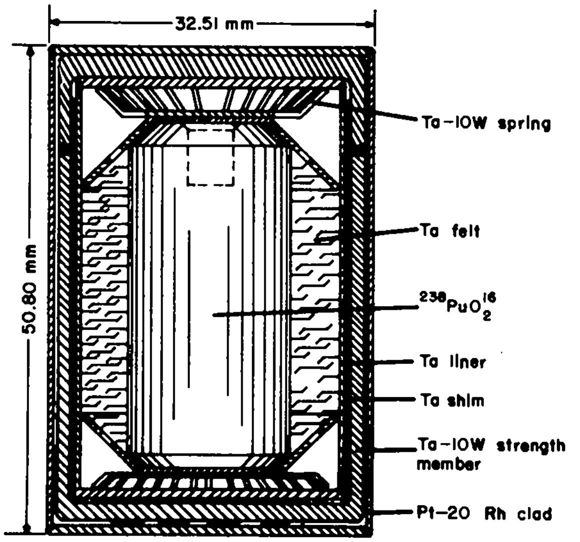
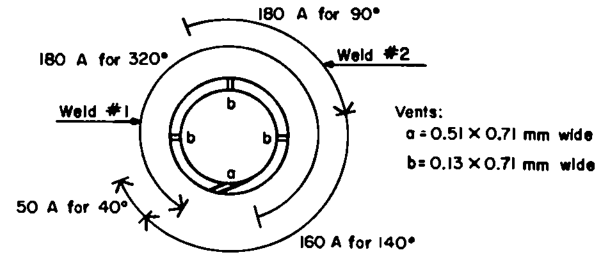


Fig. 12.
Sealed 29-W heat source.



Weld directions are shown looking down on capsule

Capsule rotational speed = 127 mm/min

Fig. 13.
Ta-10W 29-W welding diagram.

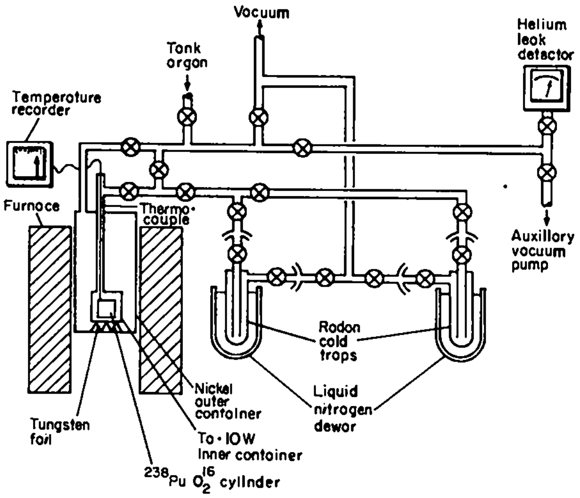


Fig. 14.
Vacuum system and equipment for He and Rn release measurements.

Resumption of heating produced somewhat more helium after which the release rate returned to near the previously prevailing level within a few hours.

As can be seen, the fuel was brought to temperature over three working days. The large initial helium inventory recorded in Table XXXV produced a large initial release rate that would have overwhelmed the leak detector if the heatup had been any faster. After the fuel cylinder reached 870°C, the release rate first decreased rapidly for about 15 days, and then decreased more slowly. The release rate approached, but remained greater than, the helium generation rate for the fuel material throughout the observation period. It was still 150% of the generation rate after ~5000 h. This means that the helium stored within the oxide cylinder continually decreased during the experiment from an initial quantity of ≤ 89.9 cm^3 to a final amount ≤ 39.0 cm^3. The total measured helium release during the experiment, as found by graphical integration, was 73.9 cm^3 or 65.4% of that generated between sintering the pellet and terminating the run.

Radiography of the encapsulated pellet following termination of the experiment revealed no cracks or breaks in the pellet. The results thus pertain to an integral 30-W cylinder.

The limits of helium release behavior are complete storage of all the gas generated, and immediate release of the gas as it is formed. The behavior extremes observed for $^{238}PuO_2$ fuel forms at about 870°C have included release of most stored helium from microspheres after relatively short intervals[14,16]

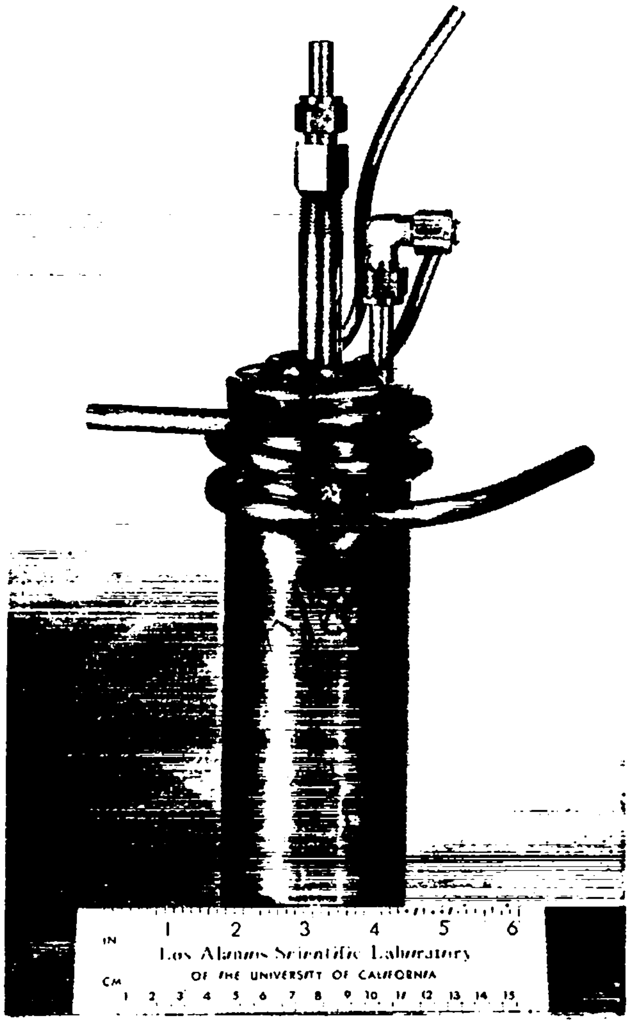


Fig. 15.
Welded nickel container for gas release experiments.

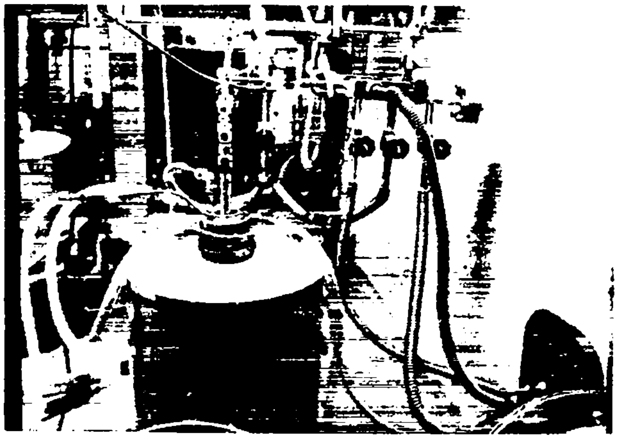


Fig. 16.
Furnace and vacuum manifold assembly (inside glove box) for gas release experiments.

and the accumulation of additional helium with time in a plutonia cylinder and SSC and PMC fuel disks following an initial "burst" of gas.[17,18] Cylinder 5 apparently exhibited an intermediate type of release despite being a bulk fuel form like the cylinder and disks mentioned above. This could be the result of the open porosity of cylinder 5 (see Fig. 19). The pores may have given rise to intermediate length diffusion paths by which helium escaped from the fuel. The large helium inventory of cylinder 5 may have been an important factor in determining the release behavior of this fuel form. The comparison with microspheres is uncertain because most data for that fuel form were derived from short experiments (usually not exceeding 48 h). A comparison requires extrapolation of microsphere release to longer periods.

IX. RADON RELEASE FROM ^{238}PuO$_2$[16] FUEL CYLINDERS

A. Introduction

Nominal ^{238}Pu embraces a number of plutonium isotopes as shown in Table I. The decay chains of several give rise to isotopes of radon. Thus ^{236}Pu and ^{240}Pu eventually decay to ^{220}Rn, ^{238}Pu and ^{242}Pu to ^{222}Rn, and ^{239}Pu to ^{219}Rn. However, only the decay of ^{236}Pu and ^{238}Pu contribute significant radon over the useful life span (~10 yr) of an artificial heart device.[19]

At first consideration the ^{222}Rn activity appears to be insignificant when compared to that of ^{220}Rn. The latter derived from ^{236}Pu grows in rather rapidly. Ten years after purification, a fuel which originally contained 1 ppm ^{236}Pu; will have a ^{220}Rn activity of

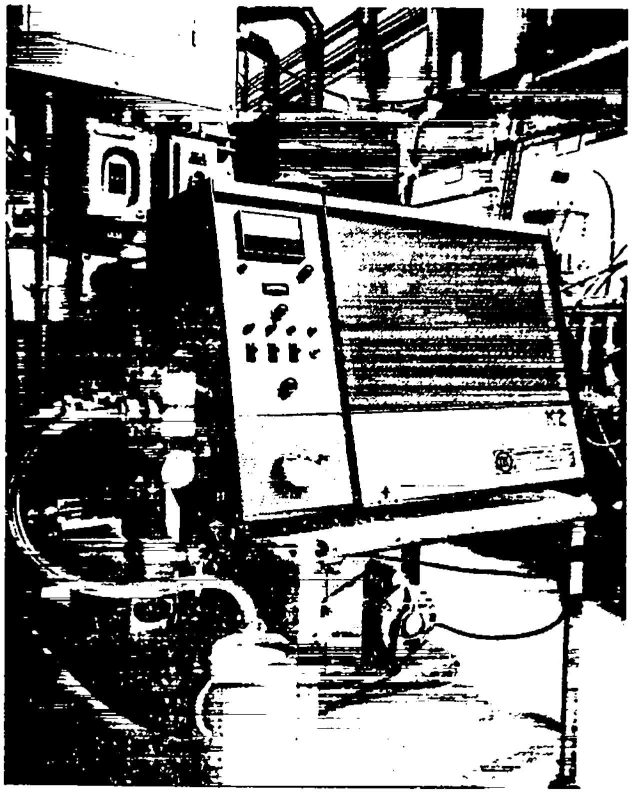


Fig. 17.
Helium leak detection equipment.

5.76 x 10^5 dis/s/g ^{238}Pu. In contrast, ^{222}Rn generated by the decay chain from ^{238}Pu grows in very slowly due to the 248 000 half-life of ^{234}U. The same 10-yr-old fuel will possess a ^{222}Rn activity of 1 dis/s/g ^{238}Pu. In a 30-W heat source this is equivalent to 60 dis/s.

But the amounts of the radon isotopes actually released by the fuel may be influenced by their half-lives. ^{220}Rn has a short half-life of 51.5 s. Much of it may decay before diffusing out of the fuel. The longer 3.8 day half-life for ^{222}Rn presents the possibility for a much higher escape probability. Thus the ^{222}Rn activity becomes important in an aged plutonium fuel and both radon isotopes must be considered in the design of a vented heat source capsule.

TABLE XXXV

CHARACTERISTICS OF $^{238}PuO_2{}^{16}$ CYLINDER
Serial Number 5

History

Date of Chemical Purification (date of electrorefining)	9/28/69
Date of Cylinder Sintering	11/25/69

Plutonium Isotopic Composition

Isotope	Wt%	Date of Analysis
236	0.70 x 10^{-4}	12/8/69
238	79.82	12/29/69
239	16.21	12/29/69
240	2.92	12/29/69
241	0.80	12/29/69
242	0.25	12/29/69

Physical Properties (11/25/69)

Diam (mm)	21.54
Height (mm)	20.40
Weight (g)	73.66
Weight of Pu (g)	64.92
Weight of ^{238}Pu (g)	51.82
Density (Mg fuel/m^3)	10.11
% of theoretical density	88.2
Power (W)	29.8
Power density MW/m^3	4.03

Helium Content

857 days after sintering, cm^3	89.94

There is no existing experimental data pertinent to the release of radon from actinide heat source fuels. However, some attention has been focused on the problem. Parsont[20] has made calculations for the SNAP-19 heat source relating to the internal pressures attributable to radon and the biological risks due to radon released from $^{238}PuO_2{}^{16}$. He concluded that the pressures would be negligible and that the potential biological effect would be small. However, the latter conclusion was based on several assumptions that are not appropriate for an artificial heart heat source. The objective of the present work, therefore, is to determine experimentally the portions of the radon isotopes that are released from a $^{238}PuO_2{}^{16}$ cylinder.

Fig. 18.
$^{238}\mathrm{PuO}_2{}^{16}$ cylinder, serial number 5 (scale in inches).

B. Experimental

1. Apparatus. The vacuum system and equipment used in sampling radon release from the ceramic fuel body at the operating temperature is presented in Fig. 14 and has been discussed in Sec. VIII.B.1.(a). The liquid nitrogen-cooled radon traps shown in Fig. 21 were located outside the glove box, although the vacuum lines returned into the box. Again fuel integrity, rather than a filter in the vacuum line, was relied upon to prevent severe ^{238}Pu contamination of the cold traps. A filter could have delayed the migration of the radon gas to the trap long enough to allow complete decay before entering the trap.

The cold traps were placed in parallel but at different distances from the oxide pellet. Collecting samples at two locations would make it possible to calculate the amount of decay occurring between the pellet and the traps. With a correction for decay it would then be possible to determine the amount of radon that escaped through the surface of an oxide cylinder.

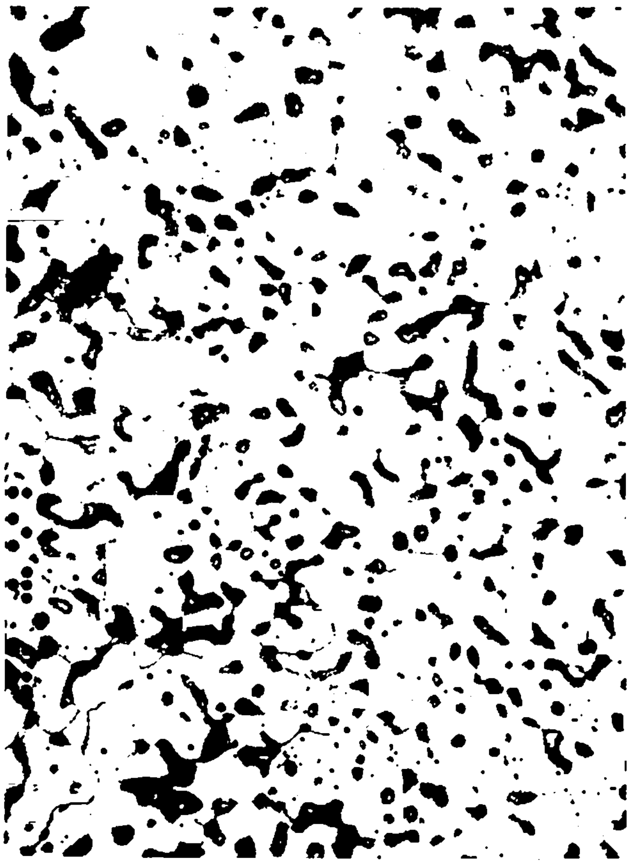


Fig. 19.
Typical microstructure of $^{238}\mathrm{PuO}_2{}^{16}$ cylinder (500 X magnification).

The identification and determination of radon released from the fuel is achieved by alpha counting and gamma spectroscopy.

2. Fuel Cylinder. The cylinder employed in the radon release experiment was described under material for helium release. This fuel had the advantages of being well characterized (see Table XXXV) and being a representative material with a known thermal history. It also had attained an age (more than 900 days since chemical purification) commensurate with an abundance of ^{220}Rn.[21] It was not sufficiently aged, however, to contain a detectable quantity of ^{222}Rn.[19]

3. Procedure. Sampling occurred, after the fuel had reached operating temperature, by evacuating the inner capsule through one of the cold traps shown in Fig. 22. An examination of the ^{236}Pu decay

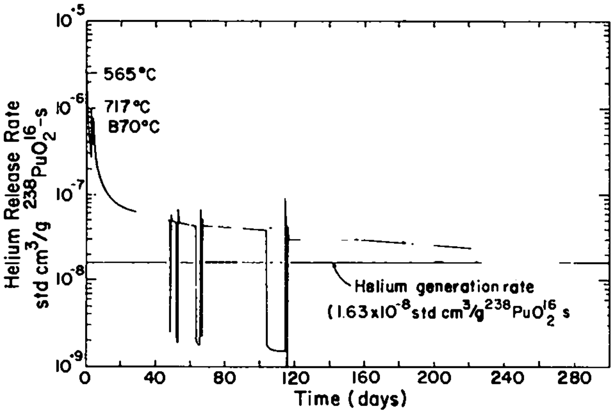


Fig. 20.
Helium release rate from $^{238}PuO_2{}^{16}$ cylinder, 5.

series shown in Eq. (1) indicated that the short half-lives of ^{220}Rn and ^{216}Po would cause them to decay before the sample could be counted, but that ^{212}Pb should be ideal for detection purposes. Equation (2) reflected the relationships prevailing in the system provided that the actual ^{220}Rn release rate was constant with time or at least showed no trend during sample collection. Since $R_{g^{220}\mathrm{Rn}}$ was constant over the collection period, the formation of ^{212}Pb represented an instance of approach to secular equilibrium. As a compromise between largest possible sample size and greatest possible accuracy in $R_{p^{212}\mathrm{Pb}}$ the sample was collected over approximately two half-lives of ^{212}Pb.

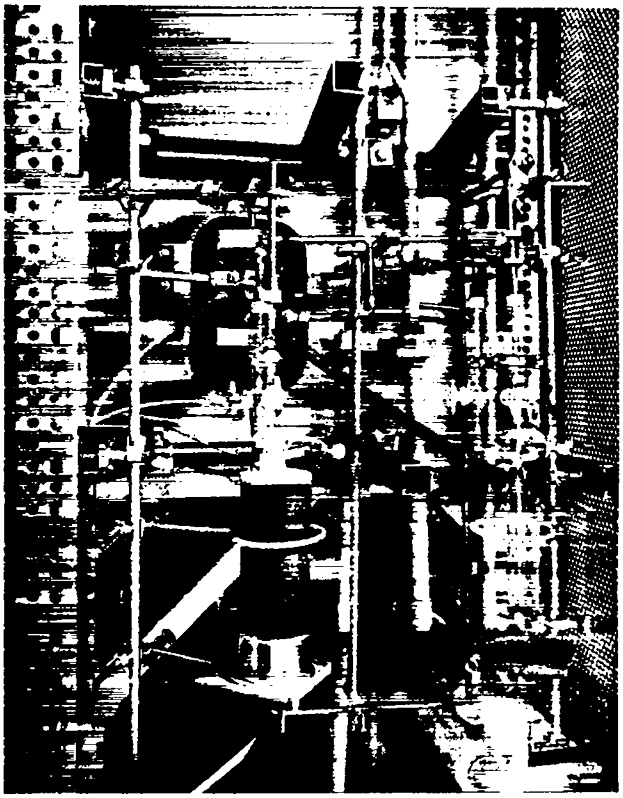


Fig. 21.
Radon traps and associated vacuum system external to glove box enclosure.

$$\begin{tikzcd}
{}^{236}\mathrm{Pu} \arrow[r, "2.85\,\mathrm{y}", "\alpha"'] & {}^{232}\mathrm{U} \arrow[r, "72\,\mathrm{y}", "\alpha"'] & {}^{228}\mathrm{Th} \arrow[r, "1.91\,\mathrm{y}", "\alpha"'] & {}^{224}\mathrm{Ra} \arrow[r, "3.64\,\mathrm{d}", "\alpha"'] & {}
\end{tikzcd}$$

$$\begin{tikzcd}
{}^{220}\mathrm{Rn} \arrow[r, "51.5\,\mathrm{s}", "\alpha"'] & {}^{216}\mathrm{Po} \arrow[r, "0.16\,\mathrm{s}", "\alpha"'] & {}^{212}\mathrm{Pb} \arrow[r, "10.64\,\mathrm{h}", "\beta"'] & {}^{212}\mathrm{Bi} \arrow[r, "60.6\,\mathrm{m}", "\alpha\,(34\%)"'] \arrow[d, "\beta\ (66\%)"', "60.6\,\mathrm{m}"] & {}^{208}\mathrm{Tl} \arrow[d, "\beta"', "3\,\mathrm{m}"] \\
& & & {}^{212}\mathrm{Po} \arrow[r, "0.3\,\mu\mathrm{s}", "\alpha"'] & {}^{208}\mathrm{Pb}\ (\mathrm{stable})
\end{tikzcd} \tag{1}$$

The sample was then isolated from the vacuum system while noting the duration of the sample collection period. After the ^{220}Rn had completely decayed, the majority of the contents was dissolved out of the trap with 5 ml of concentrated HNO_3 followed by 5 ml of agua regia. The solutions were combined and were subjected to radiochemical analysis.

The measured ^{212}Pb activity was corrected for the decay that occurred during the interval between trapping termination and activity determination. The activity was converted to ^{212}Pb abundance in the trap at termination. The $R_{p^{212}\mathrm{Pb}}$ in Eq. (2) was calculated from Eq. (3), which assumes that the abundance of ^{212}Pb in the trap at t = 0 was zero.

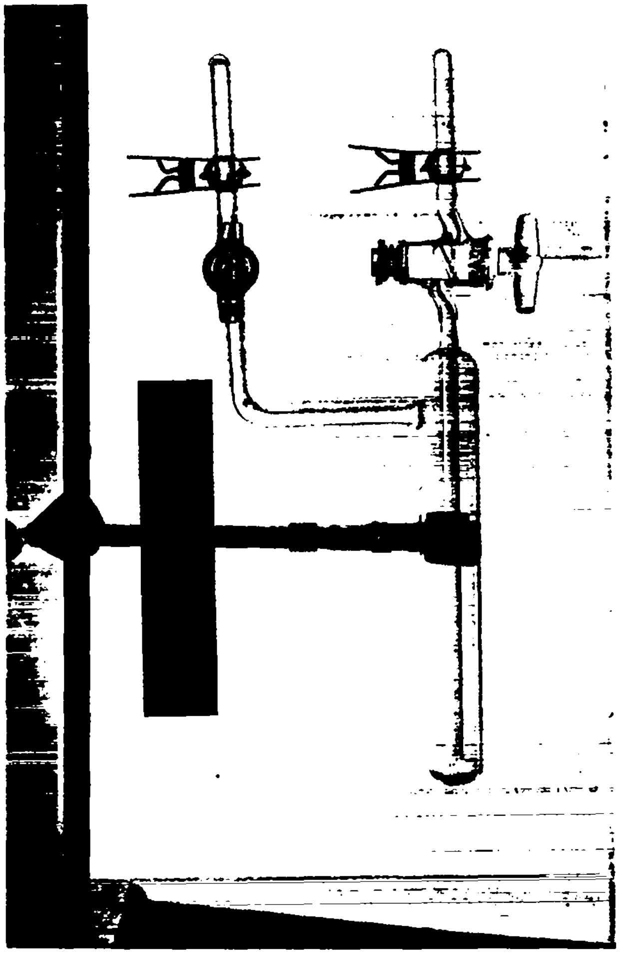


Fig. 22.
Glass reservoir for trapping Rn.

$$R_{p^{212}Pb} = R_{p^{216}Po} = R_{t^{220}Rn} \alpha R_{r^{220}Rn} \alpha R_{g^{220}Rn} , \quad (2)$$

where R_p = rate of production within the trap, R_t = rate of trapping, R_r = rate of release from the pellet, and R_g = rate of generation within the pellet.

$$R_{p^{212}Pb} = \lambda_2 N_2/(1 - e^{-\lambda_2 t}) , \quad (3)$$

where λ_2 = decay constant for ^{212}Pb, in s^{-1}, N_2 = number of atoms of ^{212}Pb at termination, and t = duration of production or trapping in seconds.

C. Results

The sample was collected over 20.5 h. The α and γ spectrums revealed the presence of ^{212}Pb, ^{212}Po, ^{212}Bi, and ^{208}Tl (all ^{220}Rn daughters) as well as ^{238}Pu. The present procedure did not permit detection of ^{222}Rn. The ^{212}Pb activity corresponded to $R_{t^{220}Rn}$ of ~86 atoms/s, which is about 1.5×10^{-3}% of the ^{220}Rn generated per second. $R_{r^{220}Rn}$ is proportional to $R_{t^{220}Rn}$,additional experiments are required to determine the proportionality factor.

The ^{238}Pu activity in the sample was 44 dis/s. The associated ^{236}Pu would yield a ^{212}Pb activity of only 2×10^{-7} of that actually measured. Thus the measured ^{212}Pb activity was the result of ^{220}Rn release and not the result of contamination of the sample by plutonia.

The radon release experiments are a continuing project. Future experiments are designed to determine release rates from the fuel cylinder.

X. FUEL CHARACTERIZATION

A. Nondestructive Analysis

Emphasis has been placed on developing nondestructive methods for characterizing medical-grade ^{238}Pu fuels. At present, the most valuable single measurement to ascertain fuel radiation quality is the neutron count. This measurement is supplemented by detailed gamma-ray spectroscopy, which is capable of detecting small amounts of light element impurities, such as fluorine, sodium, chlorine, nitrogen, oxygen, phosphorus, and aluminum. Gamma-ray spectroscopy is also used for routine measurements of ^{17}O and ^{18}O in $^{236}PuO_2{}^{16}$.

1. **Neutron Counting.** Neutron counting of small samples is done routinely inside a well surrounded by four detectors, but large samples must be counted on top of the well about 42 cm above the in-well position and 20 cm above the top of the moderator. Counting inside the well is much less sensitive to sample positioning, however, than counting outside the well. Quantitative measurements showed 0.1% variation of count rate for each millimeter difference in height inside the well as compared with a 2% variation above the well. These results point to the importance for accurate positioning of the sample and standard for above-well counting, and for the desirability of constructing a new well counter to handle large samples. Development work is also being continued on the "long counter" detection system. It consists of five matched ^3He detectors in a

polyethylene moderator arranged to achieve a similar response to a wide variety of initial neutron energies.

2. Gamma Spectrometry

a. Measurement of ^{17}O and ^{18}O in $^{238}PuO_2{}^{16}$. The reliability of gamma spectrometry for measuring ^{17}O and ^{18}O in ^{238}PuO$_2{}^{16}$ was estimated from computer-compiled spectra to be ±10% for ^{18}O contents down to 10 μg/g total oxygen and ±25% for ^{17}O contents down to 50 μg/g of total oxygen. This estimate was based on measurements made by compiling a series of gamma spectra with a PDP-11 computer from definite ratios of a ^{238}PuO$_2{}^{nat}$ and ^{238}Pu metal spectrum. This was done by accumulating gamma spectra of the ^{238}PuO$_2{}^{nat}$(2040 ppm ^{18}O and 370 ppm ^{17}O) and for ^{238}Pu metal. Successive additions of data from the latter spectrum to that of the former produced a series of new spectra with ^{18}O and ^{17}O abundances effectively reduced by half each time. Channel-by-channel addition of the spectral data was done on a PDP-11 minicomputer. From this series of spectra we concluded that gamma-ray analysis is dependable for determining ^{18}O abundances down to less than 10 ppm. The estimated accuracy in the 10- to 2040-ppm range is about 10%. Calculations of ^{17}O abundances are unreliable below the 50-ppm level. Poor accuracy is due mainly to low statistics in counting the peaks of interest. The accuracy is estimated to be 25%.

b. Determination of ^{228}Th. The measurement of ^{228}Th in ^{238}Pu sources by gamma spectrometry was investigated by comparing the intensity of the 861-keV peak, from the ^{208}Tl daughter of ^{228}Th, to that of the 851-keV peak of ^{238}Pu in aged (>28 days) sources. Results for five samples showed a high positive bias relative to those obtained with a chemical method for determining ^{228}Th content. A possible reason for the high values obtained by gamma spectrometry may be that ^{220}Rn (54.5-s half-life) diffuses toward the surface of the sample. The ^{208}Tl activity resulting from the diffused ^{220}Rn would be subject to less attenuation by the sample than the ^{238}Pu activity to which it is compared. If this mechanism proves valid, then gamma dose rates, an important specification for medical-grade ^{238}Pu, could be greater than calculated from original ^{236}Pu content. It seems likely that gamma spectrometry is not a reliable method for the nondestructive determination of ^{238}Th content.

c. Minicomputer Data Reduction of Gamma-Ray Spectra. A DPD-11 computer with 8 K of core memory has been interfaced with the 4096-channel analyzer for measuring gamma-ray spectra. Associated software provides a means of data reduction, including the calculation of gamma-ray energies and peak area. A first application may involve the determination of 345- and 351-keV peak areas for use in ^{18}O analysis in ^{238}Pu. Preliminary results indicate a linear relationship of calculated 351-/345-keV peak ratios with ^{18}O content, but the linearity does not hold true when the spectrum shifts slightly. We observed this effect in spectra prepared by combining a spectrum of PuO$_2{}^{nat}$ (2040 ppm ^{18}O) with that of plutonium metal (0 ppm ^{18}O) in suitable proportions by using the computer. Thus, equal proportions of each individual spectra produce an effective standard containing 1020 ppm O. Two distinct linear segments, separated by a break, were obtained for the relationship of 351-/345-keV peak area ratios with ^{18}O content, as calculated by the computer. Examination of the computer program disclosed the fact that shifting the peak channel, upon which the peak area is based, caused the break in the curve. Efforts are being made to modify the program so that there will be less dependence of peak area to peak channel.

3. Calorimetry. An evaluation of LASL's water bath calorimeter[1] was made using a 30-W ^{238}PuO$_2{}^{16}$ heat source, 30ULX0007, which had been standardized by Mound Laboratory(MRC). The standardization was done in a high-precision isothermal twin-bath calorimeter, calorimeter No. 87. MRC provided wattage tables (i.e., wattage as a function of time), which are accurate to a least ±0.04% at the 95% confidence level, for the standardized source.

Over a one month period, 15 determinations were made of the wattage. These results, together with the MRC values, are given in Table XXXVI. Eight LASL results were higher than the MRC values with an average difference of 0.024 W and seven were lower with an average difference of 0.022 W. The average difference between the two sets of values is ±0.023 W or ±0.08%. Thus the performance of the LASL water bath calorimeter is within its design criterion of ±0.1% at 30 W.

B. Chemical Analysis

1. Mass Spectrometric Analysis of ^{17}O and ^{18}O in ^{238}PuO$_2{}^{16}$. Since a low neutron emission rate is essential for medical-grade fuels, measuring traces of ^{18}O and ^{17}O in ^{238}PuO$_2{}^{16}$ is necessary because these impurities contribute significantly through

TABLE XXXVI

COMPARISON OF LASL AND MRC CALORIMETRIC MEASUREMENTS ON ${}^{238}PuO_2{}^{16}$ HEAT SOURCE 30ULX0007

Det No.	Date	LASL Value (W)	MRC Value[a] (W)	Difference (LASL-MRC) (W)
1	8- 5-71	28.917	28.932	−0.015
2	8- 6-71	28.935	28.932	+0.003
3	8-10-71	28.972	28.929	+0.043
4	8-11-71	28.901	28.929	−0.028
5	8-16-71	29.012	28.926	+0.086
6	8-18-71	28.943	28.925	+0.018
7	8-19-71	28.930	28.924	+0.006
8	8-23-71	28.926	28.922	+0.004
9	8-24-71	28.893	28.921	−0.028
10	8-25-71	28.910	28.921	−0.011
11	8-26-71	28.910	28.920	−0.010
12	8-27-71	28.931	28.920	+0.011
13	8-30-71	28.867	28.918	−0.051
14	8-31-71	28.904	28.917	−0.013
15	9- 1-71	28.937	28.916	+0.021

---

[a] Values from MRC's Calorimetric Standardization Calendar for 30ULX0007.

(α,n) reactions to the neutron count. Absolute measurement of these isotopes by coupling inert gas fusion and mass spectrometry is being studied. The oxide sample is heated to 2000°C in a helium atmosphere with elemental ${}^{13}C$ to produce CO which is collected in a silica gel trap and then analyzed using a mass spectrometer. The ratio of the ${}^{13}C{}^{17}O$, 30, peak to the ${}^{13}C{}^{16}O$, 29, peak is used to determine the ${}^{17}O/{}^{16}O$ atom ratio and the ratio of ${}^{13}C{}^{18}O$, 31, peak to the ${}^{13}C{}^{16}O$, 29, peak is used to determine the ${}^{18}O/{}^{16}O$ atom ratio.

In initial tests, a tungsten crucible was used to contain the oxide sample while it was heated, but this was replaced by a tantalum crucible to avoid the volatilization of tungsten oxides. Attempts to pelletize a sample of ${}^{238}PuO_2$ mixed with ${}^{13}C$ before it was heated in the analysis apparatus to 2000°C was not practical because of the poor pelletizing characteristics of the ${}^{13}C$.

Analyses of powdered ${}^{238}PuO_2$-${}^{13}C$ mixtures showed the precision of the method to be unsatisfactory. Disassembly and inspection of the analysis apparatus indicated that significant amounts of powdered samples were blown out of the crucible and adhered to the walls of the fused silica furnace tube during analysis. To avoid sample loss, containment of powdered ${}^{238}PuO_2$ - ${}^{13}C$ mixtures in small tantalum envelopes was tried successfully. However, a high apparatus background developed upon subsequent analyses. The furnace tube, which was coated with carbon and sample on the inside, was replaced with a new fused-silica tube. Further determinations were planned to estimate the precision of the method.

2. Measurement of O/M Ratios in ${}^{238}PuO_2$. The reliability of the thermogravimetric measurement of oxygen-to-metal atom ratios (O/M) in ${}^{238}PuO_2$ fuels was investigated using oxides of known composition prepared from ${}^{238}Pu$ metal (80% enriched in ${}^{238}Pu$ isotope). In this method, originally developed for analyzing ${}^{239}PuO_2$ materials, the oxide is heated to 1000°C in air to form a hyperstoichiometric oxide which is then reduced in Ar-6% H_2 at 1000°C to form the stoichiometric dioxide.[22] The change in weight is used to calculate the original O/M. In these tests the ${}^{238}Pu$ metal was first carefully weighed in an inert atmosphere and then slowly converted to a hyperstoichiometric oxide by heating in air. Weighed portions of this oxide containing known amounts of ${}^{238}Pu$ were oxidized and then reduced as described above. Twelve determinations showed that the standard deviation in measuring O/M in ${}^{238}PuO_2$ was ±0.004. Studies of the effect of uranium impurity on the measurement of O/M are planned.

REFERENCES

1. "Preparation and Evaluation of Medical-Grade Plutonium-238 Fuels, July 1, 1967 - June 30, 1971," Los Alamos Scientific Laboratory report LA-4940 (1972).

2. D. A. Hicks, J. Ise, Jr., and R. V. Pyle, "Probabilities of Prompt-Neutron Emission from Spontaneous Fission," Phys. Rev. 101, 1016 (1956).

3. V. A. Druin, V. P. Perelygin, and G. I. Khlebnikou, "Spontaneous Fission Periods of ${}^{237}Np$, ${}^{238}Pu$, and ${}^{242}Pu$," Soviet Phys. JETP **13**, 5, 913 (1961).

4. L. J. Mullins, G. M. Matlack, J. Bubernak, and J. A. Leary, "Characterization and Properties of Medical-Grade ${}^{238}Pu$ Fuels," Second International Symposium on Power from Radioisotopes, Madrid, May 29 - June 1 , 1972, OCDE, AEN, 49-67.

5. J. A. Porter and M. C. Thompson, "Preparation of ${}^{238}PuO_2{}^{16}$," Savannah River Laboratory report DP-1153 (1968).

6. "Preparation and Evaluation of Plutonium-238 Radioisotopic Heat Sources, Annual Report for the Period July 1, 1968 - June 30, 1969," LASL internal document (1970).

7. M. Taherzadeh, "Neutron Yield from the (α,n) Reaction in the Isotope Oxygen-18," Nucl. Sci. Eng. **44**, 190-193 (1971).

8. I. A. Serdivkova, A. G. Khabakhpashev, and E. M. Tsenter, "Investigation of the (α,n) Reaction on Oxygen," Bulletin of the Academy of Sciences of the USSR, Physical Series No. 7, Vol. 21, (1957).

9. L. F. Hansen, J. D. Anderson, J. W. McClure, B. A. Pohl, M. F. Stelts, J. E. Wesolowski, and C. Wong, "The (α,n) Cross Sections on ^{17}O and ^{18}O between 5 and 12.5 MeV," Nucl. Phys. **A 98**, 25-32 (1967).

10. K. W. Haff and D. W. Ramey, "One-Kilowatt $^{244}Cm_2O_3$ Heat Source Test," Trans. Amer. Nucl. Soc. **15**, 2, 689-690 (1972).

11. Peter Angelini, R. E. McHenry, J. L. Scott, W. S. Ernst. Jr., and J. W. Prados, "Helium Release from $^{238}PuO_2$ Microspheres," Oak Ridge National Laboratory report ORNL-4507 (1970).

12. "Mound Laboratory Isotopic Power Fuels Program: January - March 1971," Mound Laboratory report MLM-1817, pp. 28-32 (June 15, 1971).

13. B. A. Mueller and R. N. R. Mulford. "Helium Migration and Release in $^{238}PuO_2$ Fuel Forms," Trans. Amer. Nucl. Soc. **15**, No. 2 , 686 (1972).

14. R. N. R. Mulford and B. A. Mueller, "Measurements of Helium Release from Materials Containing $^{238}PuO_2$," Los Alamos Scientific Laboratory report LA-5215 (July 1973).

15. Walter A. Stark, "Simple Diffusion Analysis of Helium Release Experiments," Sandia Laboratories report SC-DC-69-1954 (August 1969).

16. W. A. Stark, Jr., "Helium Release Data for PuO_2 Microspheres," Sandia Laboratories report SC-DC-69-1892B (November 1969).

17. "Quarterly Status Report on Plutonium-238 Space Electric Power Fuel Development Program, July 1 - Sept. 30, 1970," Los Alamos Scientific Laboratory report LA-4647-MS (March 1971).

18. "Quarterly Status Report on Plutonium-238 Space Electric Power Fuel Development Program, January 1 - March 31, 1971," Los Alamos Scientific Laboratory report LA-4819-MS (October 1971).

19. J. Bubernak, Los Alamos Scientific Laboratory, Private communication (May 1, 1971).

20. M. A. Parsont, "Radon Production in $^{238}PuO_2$ Fuels," Sandia Laboratories report SC-RR-68-832 (December 1968).

21. George M. Matlack and Charles F. Metz, "Radiation Characteristics of Plutonium-238," Los Alamos Scientific Laboratory report LA-3696 (1967).

22. J. W. Dahlby, T. K. Marshall, G. R. Waterbury, and G. C. Swanson, "Measurement of Oxygen-to-Metal Atom Ratios in Uranium and Plutonium Oxides," Los Alamos Scientific Laboratory report LA-5329 (1973).

APPENDIX

NOMENCLATURE

I. SI UNITS

SI units, as given in the Metric Practice Guide, E 380-72, American Society for Testing and Materials, are used in this report. Some of these units differ from those used in earlier reports. For the convenience of the reader, Table A-I gives the conversion factors for the older units.

II. STANDARDIZED NOMENCLATURE SYSTEM FOR HEAT-SOURCE IDENTIFICATION

LASL and the USAEC/DAT made a joint study to standardize the nomenclature system for encapsulated ^{238}Pu heat sources made at LASL. The following system was adopted and was put into effect in July 1973, and is used in this report.

Definition of encapsulated heat source. An encapsulated AEC heat-source product made by LASL destined for off-site shipment or for prolonged use at LASL. This latter category would include calorimetry and neutron standards as well as heat-sources earmarked for future use.

Nomenclature system. The encapsulated heat source is identified by a combination of six numbers and letters as follows.

Columns 1 and 2 give the wattage of the product at the time of fabrication to the nearest integer.

Column 3 gives the pellet encapsulator of the quality-assured capsule
- D for Donald Douglas
- L for LASL
- M for Mound Laboratory (MRC)
- U for unencapsulated source or for sources encapsulated in temporary containers.

Column 4 gives the present location
- B for Lawrence Livermore Laboratory (LLL)
- C for Cornell
- D, L, M as above
- E for destroyed or expended pellets
- O for Monsanto
- P for Battelle, PNL
- Q for unknown or questionable location
- W for Westinghouse

Column 5 gives the plutonium isotopic enrichment at time of fabrication:
- X for 80 wt% ^{238}Pu
- Y for 90 wt% ^{238}Pu
- Z for 99 +wt% ^{238}Pu
- F for other enrichments.

Column 6 gives the year of pellet encapsulation or fabrication.

Columns 7, 8, and 9 give the pellet serial number.

A list of all the encapsulated heat sources made at LASL for the present program is given in Table A-II. The products are given in two size groups, one for 29 W and larger, and the other for less than 29 W. The first column gives the old identification and the last column gives the new six-column identification. To facilitate cross-referencing, the products of Table A-II are listed by serial number in Table A-III.

TABLE A-I

FACTORS FOR CONVERTING FORMERLY USED UNITS TO SI UNITS

SI Unit	Former Unit	Conversion Factor[a]
Megawatts per cubic meter (MW/m^3)	watts per cm^3	1.000 000[b]
R/h-kg ^{238}Pu	mR/h-g ^{238}Pu	1.000 000[b]
rem/h·kg ^{238}Pu	mrem/h-g ^{238}Pu	1.000 000[b]
Megagram fuel per cubic meter (Mg/m^3)	g/cm^3	1.000 000[b]
Pascal (Pa)	torr	$1.333\ 224 \times 10^2$
n/s-kg ^{238}Pu	n/s-g ^{238}Pu[c]	$1.000\ 000 \times 10^3$[b]

---

[a] (Former unit) x (conversion factor) = SI unit.

[b] Conversion factor is exact and all subsequent digits are zero.

[c] This unit is used frequently in the text because of its present well-established usage.

TABLE A-II

LIST OF ENCAPSULATED ^{238}Pu HEAT SOURCES MADE AT LASL FOR USAEC

Fuel Cylinders (29 W and larger)

Old Designation	History	Present Location	New System Designation
LASL 1	30-W Pu-Ga source made for Battelle, PNL	LASL, in storage	30LLX8002
30-1	30-W $^{238}PuO_2{}^{16}$ source made for Hittman Assoc.	destroyed	30LEX9003
30-2	Same as above	destroyed	30UEX9004
30-3	30-W $^{238}PuO_2{}^{16}$ cylinder made for fuel property studies	LASL, in He-Rn release experiments	30ULX0005
30-4	Same as 30-1	destroyed	30LEX0006
30-5	Same as 30-3	LASL calorimetry standard	30ULX0007
30-6	32-W medical-grade $^{238}PuO_2{}^{16}$ made for PNL	LASL, for compatibility evaluation	30LLF0012
50-1	Process development $^{238}PuO_2{}^{16}$ cylinder	destroyed	50UEX0008
50-2	50-W $^{238}PuO_2{}^{16}$ cylinder made for NHLI	destroyed	50UEX0009
50-3	Same as 50-2	destroyed	50UEX0010
50-4	Same as 50-2	destroyed	50UEX0011
50-5	50-W $^{238}PuO_2{}^{16}$ cylinder made for PNL	destroyed	50LEX1013
50-6	Same as 50-5	destroyed	50LEX1024
50-7	Same as 50-5	PNL, implanted in a miniature swine	50LPX1025

TABLE A-II (cont)

Old Designation	History	Present Location	New System Designation
TRW-DP-1	29-W process development $^{238}PuO_2{}^{16}$ cylinder	destroyed	29UEX2032
TRW-DP-2 (CAP 437)	Same as TRW-DP-1	LASL, being stored for He-Rn release	29ULX2033
TRW-DP-3 (CAP 438)	Same as TRW-DP-1	LASL, calorimetry standard	29ULX2034
33-1 (Hf 1)	33-W medical-grade $^{238}PuO_2{}^{16}$ cylinder made for vented fuel capsule	LASL, will be used in He-Rn test at fuel operating temperature	33MLY2035
33-2 (HF 2)	Same as HF-1	Westinghouse	33MWY2036
33-3 (HF 3)	Same as HF-1	Westinghouse	33MWY2037
PNL 29-1	29-W $^{238}PuO_2{}^{16}$ cylinder for animal implantation at PNL or Cornell	Battelle, PNL	29LPX3038
PNL 29-2	Same as PNL 29-1	Battelle, PNL	29LPX3039
PNL 29-3	Same as PNL 29-1	Battelle, PNL, for shipment	29LPX3040
PNL 29-4	Same as PNL 29-1	LASL, ready for shipment	29LLX3041
29-5	Same as PNL 29-1	destroyed	29UEX3042
PNL 29-10	Same as PNL 29-1	Battelle, PNL	29LPX3047
29-6	29-W process development cylinder	destroyed	29UEX3043
29-7	Same as PNL 29-6	destroyed	29UEX3044
29-8	Same as PNL 29-6	LASL, CMB-1 neutron standard	29ULX3045
PNL 29-9	Same as PNL 29-6	LASL, fuel property studies	29ULX3046

TABLE A-II (cont)

Fuel Cylinders (less than 29 W)

Old Designation	History	Present Location	New System Designation
CAP 823-6	First electrorefined metal source made by LASL in 1967, 4.8 W	LASL, CMB-1, neutron standard	05ULX7001
CAP 320	Medical-grade $^{238}\mathrm{PuO}_2{}^{16}$ pellet shipped to Monsanto Research Corp, see monthly report CMB-1822, May 12, 1971, p.4, Table IV; 1.32 W	Monsanto[a]	01UOF1018[a]
CAP 321	Same as CAP 320, 1.35 W	Monsanto[a]	01UOF1019[a]
CAP 308	Medical-grade $^{238}\mathrm{PuO}_2{}^{16}$ pellet shipped to Mound Lab. see CMB-1822, Table VII, 0.66 W	Monsanto[a]	01UOF1014[a]
CAP 309	Same as CAP 308, 0.65 W	Monsanto[a]	01UOF1015[a]
CAP 310	Same as CAP 308, 0.64 W	Monsanto[a]	01UOF1016[a]
CAP 314	Same as CAP 308, 0.61 W	Monsanto[a]	01UOF1017[a]
CAP 330-1	32.2-g electrorefined metal medical-grade shipped to MRL for pacemaker fuel fabrication, see monthly report CMB-1835, July 14, 1971, Tables IV, V, VI; 15.4 W	MRL[a]	15UMF1020[a]
CAP 333	30.4g of electrorefined metal same history as CAP 330-1, 14.6 W	MRL[a]	15UMF1021[a]
CAP 351	Medical-grade $^{238}\mathrm{PuO}_2{}^{16}$, 1.3 watts, pellet shipped to LLL, see monthly report CMB-1870 Feb. 14, 1972, Table I	LLL[a]	02UBF1022[a]
CAP 352	Same as CAP 351	LLL[a]	02UBF1023[a]
CAP 397	Medical-grade $^{238}\mathrm{PuO}_2{}^{16}$ pellet 1.6 W, shipped Donald W. Douglas Lab., Richland, WA, see monthly report CMB, July 19, 1972, Table IV	DWDL[a]	01UDF2026[a]

TABLE A-II (cont)

Old Designation	History	Present Location	New System Designation
CAP 398	Same as CAP 397	DWDL[a]	01UDF2027[a]
CAP 399	Same as CAP 397	DWDL[a]	01UDF2028[a]
CAP 401	Same as CAP 397	DWDL[a]	01UDF2029[a]
CAP 405	Same as CAP 397	DWDL[a]	01UDF2030[a]
CAP 406	Same as CAP 397	DWDL[a]	01UDF2031[a]

---

[a] Location to which product was shipped is shown.

TABEL A-III

IDENTIFICATION OF SOURCES
IN ORDER OF SERIAL NUMBER

New System I.D.	Old Designation
05ULX7001	CAP 823-6
30LLX8002	LASL 1
30LEX9003	30-1
30UEX9004	30-2
30ULX0005	30-3
30LEX0006	30-4
30ULX0007	30-5
50UEX0008	50-1
50UEX0009	50-2
50UEX0010	50-3
50UEX0011	50-4
30LLF0012	30-6
50UEX1013	50-5
01UOF1014	CAP 308
01UOF1015	CAP 309
01UOF1016	CAP 310
01UOF1017	CAP 314
01UOF1018	CAP 320
01UOF1019	CAP 321
15UOF1020	CAP 330-1
15UMF1021	CAP 333
02UBF1022	CAP 351
02UBF1023	CAP 352
50UEX1024	50-6
50LPX1025	50-7
01UDF2026	CAP 397
01UDF2027	CAP 398
01UDF2028	CAP 399
01UDF2029	CAP 401
01UDF2030	CAP 405
01UDF2031	CAP 406
29UEX2032	TRW-DP 1
29ULX2033	TRW-DP 2 (CAP 437)
29ULX2034	TRW-DP 3 (CAP 438)
33MLY2035	33-1 (HF-1)
33MWY2036	33-2 (HF-2)
33MWY2037	33-3 (HF-3)
29LPX3038	PNL 29-1
29LPX3039	PNL 29-2
29LPX3040	PNL 29-3
29LLX3041	PNL 29-4
29UEX3042	29-5
29UEX3043	29-6
29UEX3044	29-7
29UEX3045	29-8
29ULX3046	29-9
29LPX3047	PNL 29-10