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Preparation and Evaluation of Medical-Grade Plutonium-238 Fuels, July 1, 1967 - June 30, 1971

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Preparation and Development of Medical-Grade Plutonium-238 Fuels, July 1, 1967 - June 30, 1971*

Compiled by

L. J. Mullins

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*Work funded by the US AEC Division of Applied Technology, Isotopes Development.

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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.

N-2 --- calculated neutron multiplication factors and gamma dose rates for a variety of specified conditions. These calculations were useful in interpreting experimental measurements.

The work was done under the direction of R. D. Baker, CMB Division Leader. Principal investigators were J. A. Leary (1967 - 1970), C. F. Metz (1967 - 1970), L. J. Mullins (1970 - 1971), and G. R. Waterbury (1970 - 1971). Individual contributions to the program are indicated in the list of publications at the end of this report.

HIGHLIGHTS

1. Preparation of potential fuel forms in 1968 and 1969 led to development of four fuel forms with minimal radiation properties. These are ^{238}Pu metal, ^{238}Pu - 3 at.% Ga, $^{238}PuN^{15}$, and $^{238}PuO_2^{16}$. All were prepared from electrorefined metal of high chemical purity. Theoretical and experimental evaluations showed that $^{238}PuO_2^{16}$ is the best form for heat sources for artificial-heart devices.

2. Procedures have been developed for routine fabrication of $^{238}PuO_2^{16}$ sources having 1- to 50-W thermal outputs. Their radiation properties compare favorably to ^{238}Pu metal of high chemical purity.

3. The exchange of ^{18}O and ^{17}O with sintered pellets of $^{238}PuO_2^{16}$ has been studied in atmospheres of argon plus O_2, dry air, and air saturated with water vapor. Cylinders of $^{238}PuO_2^{16}$ as large as 50 W can be handled in air-filled gloveboxes without measurable oxygen exchange, in marked contrast to $^{238}PuO_2^{16}$ powders that must be handled in inert gloveboxes to avoid oxygen isotopic exchange.

4. Two flowsheets for preparation of $^{238}PuO_2^{16}$ powder are still under study. In one, the oxide is prepared by converting electrorefined metal to $^{238}PuO_2^{16}$. In the other, $PuO_2{}^{nat}$ is converted to $^{238}PuO_2^{16}$ by isotopic exchange. The electrorefined metal route is superior, and has been used for the preparation of all 30- and 50-W sources.

5. One 30-W ^{238}Pu - 3 at.% Ga source was fabricated and delivered to Battelle, Pacific Northwest Laboratory (PNL), in FY 1969, for in-phantom dosimetry studies.

6. One 30-W $^{238}PuO_2^{16}$ source (86.96 at.% ^{238}Pu, 0.25 ppm ^{236}Pu, 0.00 ppm ^{236}U) was fabricated and delivered to Battelle, PNL, in FY 1971 for in-phantom dosimetry studies.

7. Two 30-W $^{238}PuO_2^{16}$ sources were fabricated and delivered to Hittman Associates for heart heat-source subsystem studies in FY 1970.

8. Three 50-W $^{238}PuO_2^{16}$ sources were fabricated for delivery to the Atomic Energy Commission in FY 1971.

9. Approximately 63 g of medical-grade ^{238}Pu metal was delivered to Mound Laboratory in FY 1971 for fabrication of cardiac pacemaker fuel units.

10. Analytical methods were developed for determination of:

^{17}O and ^{18}O in H_2O^{16},
^{18}O in $^{238}PuO_2^{16}$,
^{228}Th in ^{238}Pu,
^{237}Np in ^{238}Pu by radiochemistry,
^{232}U and ^{234}U in ^{238}Pu,
Total plutonium in ^{238}Pu,
^{236}Pu in ^{238}Pu,
Neptunium in ^{238}Pu by spectrophotometry,
Trace impurities in ^{238}Pu metal and other fuel forms.

11. A water-bath calorimeter was fabricated and is now operational for 1- to 150-W sources.

12. Thermoluminescent-dosimetry techniques were developed for neutron and gamma dose-rate measurements.

13. The radiation characteristics of ^{238}Pu were evaluated theoretically from the literature, early in the program. Results were published in Los Alamos Scientific Laboratory report LA-3696 (1967).

14. A total of 2647 g of ^{238}Pu metal was prepared from PuO_2 by the bomb-reduction process. Typical products were evaluated.

15. Electrorefining processes were developed for preparing ^{238}Pu metal of high chemical purity on both the 15- and 100-g scales. A total of 70 g of metal was prepared on the 15-g scale, and 1814 g on the 100-g scale. This metal has been thoroughly evaluated and characterized. It is the purest ^{238}Pu ever produced and has minimal radiation levels.

16. Procedures were developed for preparation of $^{238}PuO_2^{16}$ powder from ^{238}Pu metal. Approximately 1700 g of $^{238}PuO_2^{16}$ was prepared and characterized.

17. Procedures were developed for preparation of sintered cylinders of $^{238}PuO_2^{16}$. Approximately a hundred and fifty 1-W, six 30-W, and four 50-W, cylinders were prepared and evaluated.

18. Procedures for preparation of $^{238}PuN^{15}$ were developed. Approximately 37 g of PuN^{15} was prepared and evaluated.

19. The growth of ^{236}Pu daughters in ^{238}Pu was measured.

20. The absolute intensities of ^{238}Pu gamma rays were measured.

21. Gamma-ray spectra were established for electrorefined ^{238}Pu metal, ^{238}Pu-Ga alloy, $^{238}PuO_2^{16}$, $^{238}PuO_2{}^{nat}$, $^{238}PuN^{15}$, and $^{238}PuN^{nat}$.

PREPARATION AND EVALUATION OF MEDICAL-GRADE PLUTONIUM-238 FUELS

ABSTRACT

The ^{238}Pu requirements, 55 to 90 g, for an artificial heart or circulatory-assist device demand a fuel having minimal radiation properties. Evaluation of potential ^{238}Pu fuel forms has led to development of four satisfactory fuel compositions, electrorefined metal, ^{238}Pu - 3 at.% Ga, $^{238}PuN^{15}$, and $^{238}PuO_2^{16}$. Theoretical and experimental studies of these fuels demonstrate that $^{238}PuO_2^{16}$ is the preferred composition for the artificial-heart program. This fuel is prepared as a pressed and sintered oxide. Procedures have been developed for preparing and characterizing cylindrical oxide sources varying in size from 1 to 50 W.

The chief sources of undesirable radiation in conventional fuels are low-Z impurity elements and radioactive daughters of ^{236}Pu. The low-Z impurity elements cause an increase in the neutron emission rate over that created by spontaneous fission of ^{238}Pu. This problem is eliminated by using electrorefined plutonium metal whose neutron emission rate is only 1 to 2% higher than that expected from spontaneous fission alone. The problems caused by ^{236}Pu are minimized by using plutonium that contains less than 0.3 ppm ^{236}Pu. Useful techniques based on high-resolution gamma spectrometry have been developed for identification of both low-Z elements and ^{236}Pu daughters in ^{238}Pu fuel materials.

I. INTRODUCTION

Successful utilization of ^{238}Pu in a radioistopic heat source for medical use depends on demonstrating that an economical fuel can be produced reliably in pure form, and that the fuel capsule can be used safely. The primary requirements are minimal external radiation and reliable containment of radioactive material during any credible accident. To achieve these objectives, the USAEC Division of Isotopes Development initiated a ^{238}Pu Fuel Development Program at the Los Alamos Scientific Laboratory. Emphasis has been placed on developing chemically pure fuel forms to minimize emission of neutrons and photons and fuel-container compatibility problems.

The radiation properties of ^{238}Pu fuel materials were calculated and reported early in this program.[1] Penetrating radiations from ^{238}Pu fuels having the isotopic composition shown in Table I derive primarily from the following sources:

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. In 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 (±0.6) x 10^{10} yr.[3] These values give a calculated value of 2586 (±400) n/sec-g ^{238}Pu. The present study gives 2785 (±55) n/sec-g ^{238}Pu for neutron emission from spontaneous fission of ^{238}Pu. Using this value and the

TABLE I

ISOTOPIC COMPOSITION OF CONVENTIONAL ^{238}Pu FUEL (TYPICAL LOT)

Pu Isotope	Abundance (wt.%)
236	1×10^{-4}
238	80.0
239	16.3
240	3.0
241	0.6
242	0.1

emission rate of 1.02×10^3 n/sec g ^{240}Pu for the spontaneous fission of ^{240}Pu, we calculate 2260 n/sec-g Pu as the neutron emission rate due to spontaneous fission alone for a metal containing 80 wt.% ^{238}Pu and 3.0 wt.% ^{240}Pu.

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 (α,n) contribution in conventional-grade ^{238}Pu metal can vary from 9900 to 28,000 n/sec-g ^{238}Pu.[1,4] These neutrons are due to light-element impurities other than oxygen in the metal. The (α,n) contribution in present production-grade $^{238}PuO_2^{nat}$ from Savannah River varies from 12,575 to 17,475 n/sec-g ^{238}Pu. Most of these neutrons, 11,765 n/sec-g ^{238}Pu, are due to ^{17}O and ^{18}O. Clearly, these conventional fuels are unsuitable for medical applications.

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 multiplication of the neutron rate. The magnitude of this multiplication depends on the mass, density, and geometry, of the plutonium, as well as on the neutron flux. Gamma rays accompany the alpha decay of the plutonium isotopes and account for most of the gamma radiation 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 bombardments of impurity nuclides. This is particularly true if the impurities are nitrogen or fluorine.

For medical applications, a fuel's radiation must be understood quantitatively, and be kept to a minimum. Therefore, research was initiated to accomplish the following tasks during FY 1968:

1. to calculate the radiation properties of ^{238}Pu fuel materials[1]
2. to prepare, characterize, and evaluate high-purity ^{238}Pu metal
3. to measure the physical and radiation properties of ^{238}Pu metal
4. to provide pure materials for synthesizing ^{238}Pu compounds and alloys with minimal penetrating radiation
5. to provide a standard to which other fuel forms could be compared
6. to improve the accuracy of the spontaneous-fission-rate measurement of ^{238}Pu.

To achieve these objectives, we developed a small-scale (15 g of plutonium product) electrorefining process to prepare ^{238}Pu metal of high chemical purity.

During FY-1969, emphasis was placed on preparing and evaluating fuel forms other than the metal. We prepared and evaluated ^{238}Pu-3 at.% Ga, $^{238}PuO_2^{16}$, and $^{238}PuN^{15}$. To facilitate preparation of 30-W sources, we developed a 100-g scale process for electrorefining ^{238}Pu. Methods for preparing 30-W ^{238}Pu-3 at.% Ga sources were developed.

The preparation and evaluation of 30-W $^{238}PuO_2^{16}$ sources were of primary concern during FY-1970. We developed fabrication and diagnostic procedures that permitted preparation of five 30-W sources. Attention in FY-1971 was focused on the preparation, evaluation, and properties of 50-W $^{238}PuO_2^{16}$ sources.

II. POTENTIAL MEDICAL-GRADE FUEL COMPOSITIONS

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. However, the physical properties of this metal are complex because it exists in six allotropic modifications between room temperature and its melting point, 640 °C. Therefore the linear-expansion functions are discontinuous. For example, the linear-expansion coefficients vary from 58×10^{-6}/°C for the alpha phase to -118×10^{-6}/°C for the delta-prime phase. Large expansions and contractions also accompany some of the solid-state transitions. These transitions are summarized in Table III.[5] However, the metal can be stabilized in the delta phase by adding alloying agents such as aluminum, cerium, gallium, and scandium. For example, a Pu-3 at.% Ga alloy is stable in the delta phase from room temperature to 550 °C. The expansion of a polycrystalline rod of this alloy and

TABLE II

POTENTIAL MEDICAL-GRADE ^{238}Pu FUEL COMPOSITIONS

Composition	Melting Temp. (°C)	Power Density (W/cm^3)^a
Pu	640	7.2
Pu-3 Ga	670	7.1
Pu-10 Sc	750	6.6
Pu_4 Zr	800	5.9
($PuFe_2$ + Fe)	1165	3.9
$PuPt_2$	1475	3.2
PuC[12]	1650	5.4
PuN[15]	2200	5.6
PuO_2[16]	2400	4.2

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

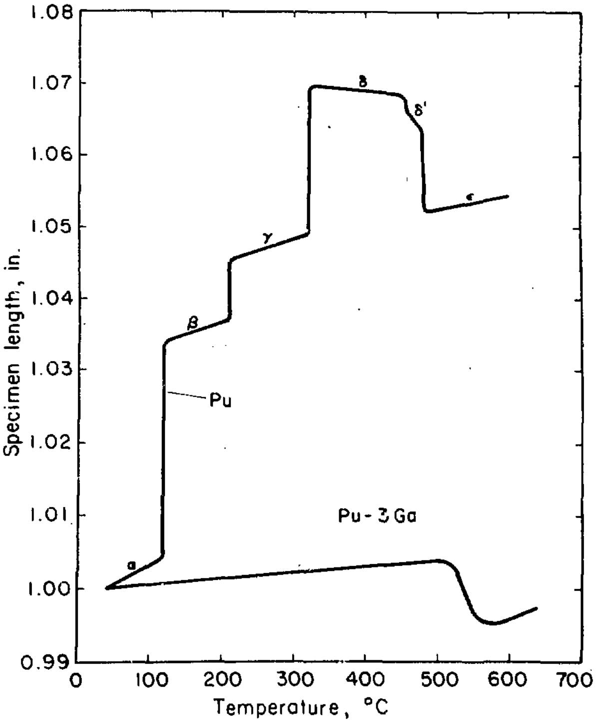


Fig. 1.
Thermal expansion of plutonium and Pu-3 at.% Ga.

plutonium metal are compared in Fig. 1. The unalloyed plutonium expands ~7% between room temperature and 318 °C and then contracts ~2%. The Pu-3 at.% Ga alloy expands ~0.4% from room temperature to 500 °C and then contracts ~0.9%.

The phase transitions for Pu-3 at.% Ga are compared to those for Pu-10 at.% Sc alloy in Table IV. Both alloys are stable in the delta phase from room temperature to 500 °C. The gallium alloy starts to melt at 670 °C; the scandium alloy, at 750 °C. The values for Pu-10 at.% Sc are based on recently published measurements by Ballagny, Boucher, and Carrard.[6] Earlier phase diagrams[7,8] conflicted with respect to the delta-phase stability range. One diagram[7] indicated that at least 12 at.% Sc is required to stabilize the delta phase, and the other[8] indicated that about 8 at.% Sc is required. The more recent study[6] shows the alloy to be stable at scandium concentrations as low as 3 at.%.

The delta-stabilized gallium alloy was chosen for experimental study in the LASL program because the physical and mechanical properties of the ^{239}Pu-3 at.% Ga alloy were well established. (The delta-stabilized scandium alloy is used in the French pacemaker device.[9,10])

Other alloys considered are Pu-20 at.% Zr, Pu-81 at.% Fe, and Pu-66.7 at.% Pt. The zirconium alloy offers no particular advantages because the small increase in

TABLE III

VOLUME CHANGES ACCOMPANYING UNALLOYED PLUTONIUM PHASE TRANSITIONS

Phase Transition	Transition Temp (°C)	Volume Change (%)
Alpha → Beta	110	9.62
Beta → Gamma	190	2.67
Gamma → Delta	310	6.90
Delta → Delta Prime	452	−0.36
Delta Prime → Epsilon	480	−2.16
Epsilon → Liquid	639	−0.82

TABLE IV

PHASE TRANSFORMATIONS OF PU-3 AT.% Ga AND PU-10 AT.% Sc

	Transformation Temp. (°C)	
Transformation	Pu-3 at.% Ga	Pu-10 at.% Sc
Delta → Epsilon + Delta	550	548
Epsilon + Delta → Epsilon	580	623
Epsilon → Liquid + Epsilon	670	750
Liquid + Epsilon → Liquid	675	810

melting temperature is accompanied by decreased power density. The iron and platinum alloys are of interest for applications in which a high-melting metallic form is desired and low power density can be tolerated. The platinum alloy has been studied as a fuel candidate in the US AEC space program.[11]

The ceramic forms considered are the carbide, nitride, and oxide. To achieve minimal radiation levels, special isotopic enrichments of carbon, nitrogen, and oxygen are required. (This point is discussed more thoroughly later.) Although PuO_2 has a lower power density than the nitride or carbide, its excellent chemical stability, high melting point, and ease of synthesis and fabrication make it a prime fuel candidate. For several years, $^{239}PuO_2$ has been studied as a fast reactor fuel, and a wealth of information on its chemical, physical, and mechanical properties has been generated.

The nitride and carbide also are possibilities. Both have higher power densities and higher thermal conductivities than the oxide. The thermal conductivities[12-15] of these ceramic forms are compared in Table V. The higher thermal conductivity of the carbide and nitride is a distinct advantage in a fast reactor fuel, permitting operation at high power levels, but it offers no real advantage for heat sources.

The fuel forms prepared and evaluated in the present program are elemental ^{238}Pu, ^{238}Pu-3 at.% Ga, $^{238}PuO_2{}^{nat}$, $^{238}PuO_2^{16}$, $^{238}PuN^{nat}$, and $^{238}PuN^{15}$.

TABLE V

THERMAL CONDUCTIVITIES OF PuO_2, PuN, AND PuC[a]

Temp. (°C)	PuO_2	Pu C		PuN
	Ref. 12	Ref. 13	Ref. 14	Ref. 15
100	----	----	0.043	----
200	----	0.082	0.048	----
300	0.050	0.096	0.053	0.12
400	0.042	0.111	0.064	0.11
500	0.036	----	0.067	0.10
600	0.032	----	0.074	0.10
700	0.029	----	0.081	0.10
800	0.026	----	0.084	0.11
900	0.025	----	----	0.11
1000	0.023	----	----	0.11

---

[a] Fuel forms are 85% of theoretical density.

III. DOSE RATES OF VARIOUS ^{238}Pu FUEL FORMS

The gamma and neutron dose rates of the fuel forms prepared and evaluated in this program are compared in Table VI. The gamma dose rate of the metal 10 cm from the center of a 3/8-in.-diam source contained in a 0.030-in.-thick tantalum container is 0.25 mR/h-g ^{238}Pu. The neutron dose rate is 0.31 mRem/h-g ^{238}Pu. Dose rates for the Pu-3 at.% Ga alloy are identical to those for the metal. The neutron dose rate for $^{238}PuN^{nat}$ is also identical to that for the metal. However, the gamma dose rate is approximately doubled by an (α,p) reaction on ^{14}N that yields an excited state of ^{17}O. Replacement of ^{14}N by ^{15}N gives a compound whose gamma dose rate is only 10% higher than that of 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 those of the metal. However, $^{238}PuO_2^{16}$ has dose rates comparable to the metal. Thus, the fuel forms with low neutron and gamma dose rates are ^{238}Pu, ^{238}Pu-3 at.% Ga, $^{238}PuN^{15}$, and $^{238}PuO_2^{16}$. With respect to radiation, all of these materials would therefore be suitable for implantable heat sources required to power pacemakers and artificial-heart devices.

IV. PREPARATION OF ^{238}PU FUEL FORMS

A. Production of ^{238}Pu.

Plutonium-238 is produced by neutron irradiation of ^{237}Np. The nuclear reactions involved, starting with ^{235}U, are

$$^{235}U \xrightarrow[7.13 \times 10^{8} y]{(n,\gamma)} {}^{236}U \xrightarrow[2.39 \times 10^{7} y]{(n,\gamma)}$$

$$^{237}U \xrightarrow[6.8 d]{\beta} {}^{237}Np \quad , \qquad (1)$$

TABLE VI

DOSE RATES OF ^{238}Pu FUEL FORMS[a,b]
(3/8- by 3/8-in. cylinders)

Composition	Dose Rates Gamma (mR/h - g ^{238}Pu)	Dose Rates Neutron (mRem/h - g ^{238}Pu)
Pu	0.25	0.31
Pu-3 at.% Ga	0.25	0.31
PuN^{nat}	0.54	0.31
PuN^{15}	0.28	0.31
PuO_2^{nat}	0.36	1.50
PuO_2^{16}	0.29	0.34

---

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

[b] Dose rates in air through 0.030-in.-thick tantalum, 10 cm from center of source.

and

$$^{237}\mathrm{Np} \xrightarrow[2.20 \times 10^6\,\mathrm{y}]{(n,\gamma)} {}^{238}\mathrm{Np} \xrightarrow[2.1\,\mathrm{d}]{\beta} {}^{238}\mathrm{Pu}\ (t_{1/2} = 87.4\mathrm{y}) \quad . \qquad (2)$$

Kilogram quantities of ^{238}Pu are produced by recycling the ^{236}U and separating and recycling the ^{237}Np. Heavier plutonium isotopes are produced in amounts that depend on the reactor operating conditions. These isotopes are produced through multiple neutron capture by ^{238}Pu and subsequent nuclides. Small amounts of ^{236}Pu also are formed by

$$^{237}\mathrm{Np} \xrightarrow[2.20 \times 10^6\,\mathrm{y}]{\gamma,n} {}^{236}\mathrm{Np} \xrightarrow[22\,\mathrm{h}]{\beta} {}^{236}\mathrm{Pu}\ (t_{1/2} = 2.85\ \mathrm{y}) \quad , \qquad (3)$$

and

$$^{237}\mathrm{Np} \xrightarrow[2.20 \times 10^6\,\mathrm{y}]{n,\,2n} {}^{236}\mathrm{Np} \xrightarrow[22\,\mathrm{h}]{\beta} {}^{236}\mathrm{Pu} \quad . \qquad (4)$$

The (γ,n) route, shown in series (3), is the principal source of ^{236}Pu present in production grade ^{238}Pu. Although the threshold for this reaction has the high value of 6.5 MeV, there are enough γ rays of this energy in production reactors to yield ^{236}Pu concentrations of $>$ 1 ppm unless special precautions are taken to shield the ^{237}Np. The importance of minimizing both ^{236}Pu and ^{232}U (a daughter of ^{236}Pu) in medical-grade fuel forms is discussed below.

After chemical separation of plutonium from neptunium and impurity elements, the plutonium is converted to PuO_2. This production-grade oxide served as the starting material for the studies described here.

B. Methods Available for Preparing High-Purity ^{239}Pu Metal.

Two general methods of preparing pure plutonium metal are available, bomb reduction and electrorefining. Meticulous quality control of the bomb-reduction process can produce ^{239}Pu of 99.8 to 99.96% chemical purity. To obtain metal of this quality requires chemically pure PuF_4, calcium, and I_2 because bomb reduction provides little or no purification. The electrorefined product, on the other hand, is relatively insensitive to purity of the feed metal. Plutonium-239 of >99.98% purity is obtained routinely by electrorefining impure ^{239}Pu (~99.8% Pu) and plutonium alloys (Pu-2.5 wt% Fe and Pu-1 wt% Ga). Electrorefining procedures have been developed to prepare this high-purity metal on 0.5- and 3-kg scales.[16,17] A plant with a plutonium capacity of 10 kg/week has been operated at the Los Alamos Scientific Laboratory for the past 7 yr. Some of this high-purity metal is distributed by the National Bureau of Standards as a primary chemical plutonium standard. Because of the superior chemical purity of the product and the assured quality control, we adapted these electrorefining procedures to the present ^{238}Pu work.

C. Preparation of High-Purity ^{238}Pu Metal.

The feed material for electrorefining is bomb-reduced ^{238}Pu metal. $^{238}PuO_2{}^{nat}$ is converted to metal using the flowsheet shown in Fig. 2. This is the conventional procedure for preparation of ^{239}Pu metal; necessary modifications are described elsewhere.[18] Briefly, the process consists of dissolving the oxide in HNO_3-HF, precipitating the peroxide, converting the peroxide to fluoride, and reducing the fluoride to metal using calcium. The flowsheet is for the preparation of 50 g of plutonium, the present scale of operation. Earlier in the program, this process was done on a 25-g scale. Detailed operating procedures for conversion of oxide to metal are given in Appendix A. A typical material balance for conversion of oxide to metal is given in Table VII. The processing loss, which is recoverable, was 4%. Plutonium material balances for all of the oxide-to-metal conversions are given in Table BI, Appendix B. A total of 1900 g of ^{238}Pu metal was made from $^{238}PuO_2{}^{nat}$ lots received from Mound Laboratory, Savannah River, and Atlantic Richfield, Hanford Co. An additional 747 g of metal was recovered from LASL process-waste streams and R & D-generated wastes.

The present flowsheet for preparation of electrorefined metal is shown in Fig. 3. Bomb-reduced metal is cast into 5/8-in.-diam rods containing ~50 g of plutonium each. Two rods are then introduced into the electrorefining cell and converted to high-purity metal which is then cast into rods and encapsulated.

Figure 4 shows the equipment and feed materials for the 15- to 20-g scale electrorefining process used earlier in the program. The bomb-reduced metal is placed in the anode leg (the tapered leg) of the MgO-Y_2O_3 crucible. A cylinder of equimolar NaCl-KCl also is placed in the

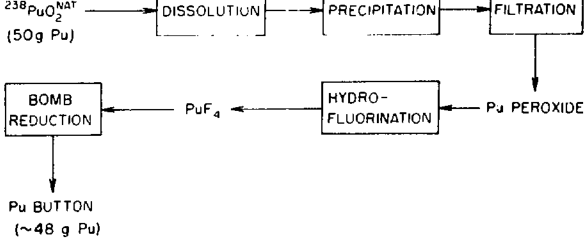


Fig. 2.

Flowsheet for conversion of $^{238}PuO_2{}^{nat}$ to ^{238}Pu metal.

TABLE VII

TYPICAL MATERIAL BALANCE FOR CONVERSION OF $^{238}PuO_2^{nat}$ TO PLUTONIUM METAL, 50-g SCALE

Lot No.	HB0 907547
Pu in oxide feed (g)	204.4
Pu in samples (g)	3.6
Pu-to-metal conversion (g)	200.8

	Fraction			
	A	B	C	D
Pu in oxide feed (g)	50.0	50.7	51.0	49.1
Pu losses (g)				
Peroxide filtrate	0.5	0.6	0.6	0.4
Slag and crucible	1.0	1.3	1.2	2.7
Pu metal regulus (g Pu)	48.5	48.8	49.2	46.0

crucible. The entire assembly is loaded into a commercial resistance-heated furnace, shown in Fig. 5, and brought to an operating temperature of 740 °C. The slotted tantalum stirrer, which also serves as the anode lead, and the cathode rod are lowered into position, and direct current at 1 A is passed through the cell to generate $PuCl_3$ and sodium metal. During this phase of the operation, the cell reaction is

$$\underset{\text{(anode)}}{Pu\,(\ell)} + \underset{\text{(molten salt)}}{3NaCl\,(\ell)} \rightarrow \underset{\text{(molten salt)}}{PuCl_3\,(\ell)} + \underset{\text{(cathode)}}{3\,Na\,(\ell)} \; .$$

The back emf of the cell is monitored periodically to ensure that this reaction is taking place. When $PuCl_3$ generation is complete, the stirrer is activated and a brief pre-electrolysis at 0.5 A is conducted. The cathode rod is then raised, and a second tungsten cathode rod is lowered into the cathode leg of the crucible. The normal electrorefining process is then conducted at 0.8 A. The cell reaction during electrorefining is

$$\underset{\text{(anode)}}{\text{Impure Pu}\,(\ell)} \rightarrow \underset{\text{(molten salt)}}{Pu^{+3}} \rightarrow \underset{\text{(cathode)}}{\text{Pure Pu}\,(\ell)} \; .$$

The process is monitored by back-emf measurements, and is discontinued when these measurements indicate serious anode polarization or when ~90% of the plutonium has been dissolved anodically. The cell is then heated to 850 °C, vibrated briefly to improve metal collection, and cooled to room temperature. Figure 6 shows the cell components after removal from the furnace.

The material balance for a typical small-scale electrorefining run is given in Table VIII. Although we have not tried to optimize the process, unintentional losses are low, 1.05 g or 3.7% of the plutonium feed, and are recoverable.

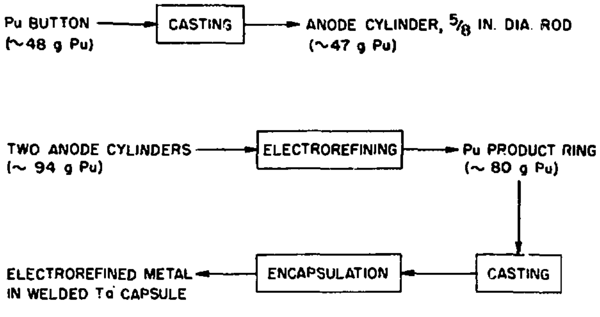


Fig. 3.
Flowsheet for preparation of electrorefined ^{238}Pu metal.

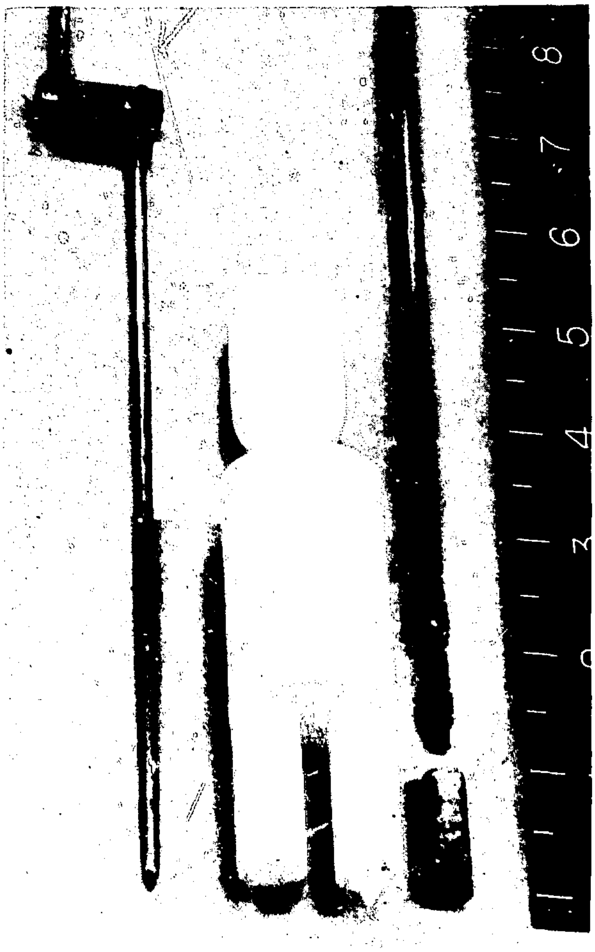


Fig. 4.
Small-scale (15- to 20-g ^{238}Pu) electrorefining components (scale in inches).

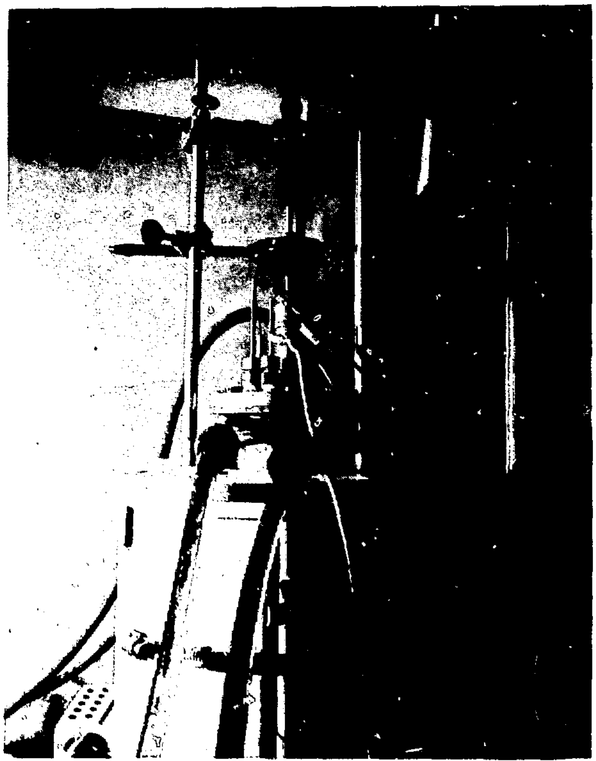


Fig. 5.
Electrorefining furnace.

The geometry of the small-scale MgO-Y_2O_3 refining cell was dictated principally by the small volume, ~1 cm^3, of liquid plutonium metal used in the process. Extrapolation of this cell design to a 100-g scale did not provide a good yield of ^{238}Pu metal. Although the 100-g cell, shown in Fig. 7, gave 96% collection yields with ^{239}Pu in process development studies, its use with ^{238}Pu gave yields of only 50%. We believed that excessively high temperatures at the metal-salt and metal-crucible interfaces in the cathode product zone caused the poor ^{238}Pu collection yields. To avoid these difficulties, we designed an annular cell, shown in Fig. 8, which consists of two MgO-2 wt% Y_2O_3 crucibles cemented together. The outer crucible is 1-5/8 in. o.d. by 4 in. long, the inner is 15/16 in. o.d. by 1-1/2 in. long. The impure bomb-reduced metal is placed in the inner cup and covered by a cylinder of NaCl-KCl. Operation of this cell is like that described above for the 20-g process. Detailed operating procedures for the 100-g electrorefining process are given in Appendix A. The essential equipment is shown in Fig. 9. Material balance for a well-executed run is given in Table IX. Unintentional losses, which are recoverable, were 5.9 g, or 6.0% of the metal dissolved at the anode. The production collection yield was therefore 94.0%.

Fig. 6.
Small-scale electrorefining components after removal from electrorefining furnace (scale in inches).

TABLE VIII

TYPICAL SMALL-SCALE ELECTROREFINING MATERIAL BALANCE

	Weight of Pu(g)
Pu feed to cell	28.32
Electrorefined ingot	20.97
Anode metal	4.45
$PuCl_3$ generated in salt	1.20
Pu adhered to cathode	0.65
Loss as slag and shot (by difference)	1.05

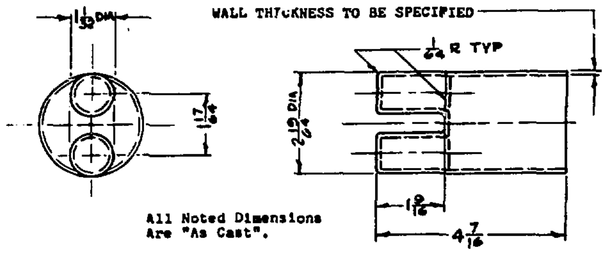


Fig. 7.
First design of 100-g ^{238}Pu electrorefining crucible.

Plutonium material balances for all 100-g scale electrorefining runs are given in Table BII. A total of 26 runs were made. One, 238-100-13, was aborted because of a broken stirrer. The average metal collection yield for the other 25 was 87%. A total of 1814 g of electrorefined metal was made. The anode heels and cathode metal are usually recycled to the next electrorefining run. The $PuCl_3$ and plutonium collection losses are recycled to the metal conversion flowsheet (see recovery of process wastes, Sec. VI D). Operation of the refining cells in high-quality inert gloveboxes will undoubtedly improve the metal collection yields so that yields of 94 - 95% may be obtained routinely. (Note: Oxidation of the ^{238}Pu anode cylinder before its introduction into the electrorefining cell is believed responsible for the poor yields in some of the runs.) A typical ^{238}Pu product ring is shown in Fig. 10.

D. Ten- to Thirty-Gram ^{238}Pu Metal Casting.

The small scale (1 cm^3) of this operation necessitated development of special casting procedures. The equipment is shown in Fig. 11. The pouring crucible was fabricated from either ThO_2 or MgO-3 wt% Y_2O_3. The end of the casting is broken off to provide a sample. If desired, the melting point of the metal is normally determined during the remelt. The 1/8-in.-o.d. tantalum thermocouple sheath shown in Fig. 11 passes through the

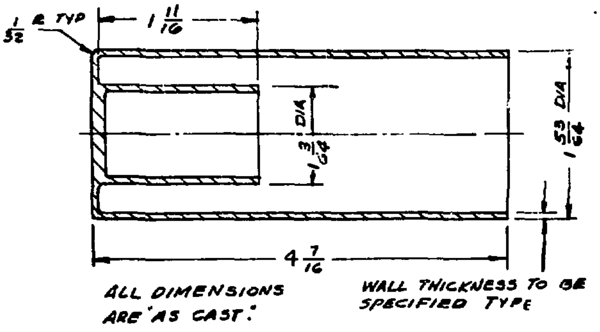


Fig. 8.
Present design of 100-g ^{238}Pu electrorefining crucible.

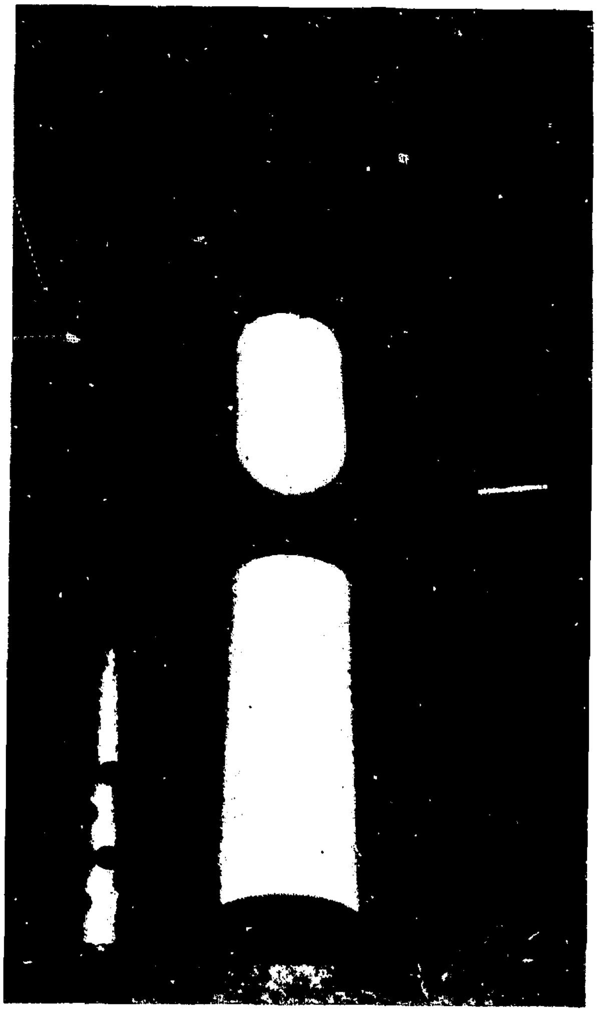


Fig. 9.
Hundred-gram scale electrorefining components.

TABLE IX

PLUTONIUM ELECTROREFINING MATERIAL BALANCE 100-g SCALE, RUN 238-100-6

Description	Pu (g)	Pu (%)
Feed metal	99.2	100
Pu product ring	86.7	87.4
Anode	1.5	1.5
Pu on cathode	2.1	2.1
$PuCl_3$	3.0	3.0
Loss as slag and shot (by difference)	5.9	6.0

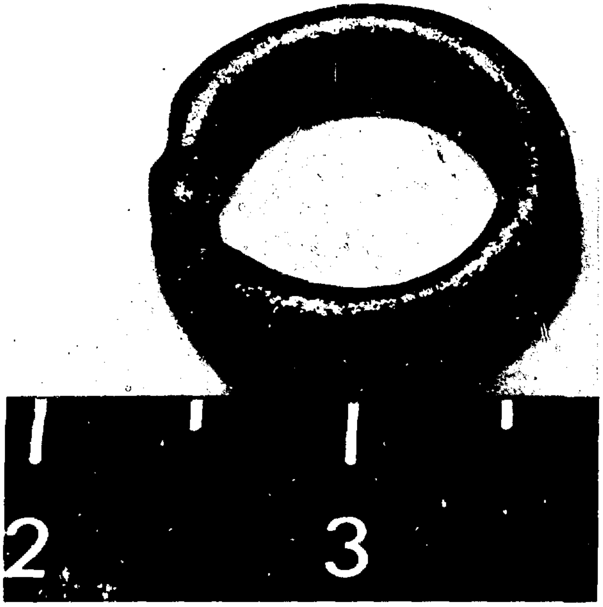


Fig. 10.
Typical 100-g ^{238}Pu product ring.

pouring crucible into the mold crucible containing the molten metal. The melt is cooled at a constant rate to ~500 °C and then heated at a constant rate to ~750 °C to determine the freezing and melting points of the metal. The metal is usually cast in the form of a 0.375-in.-diam cylinder. Plutonium losses in casting 10 to 20 g of metal are 1 to 2 g and are recoverable.

E. Thirty- to Fifty-Gram ^{238}Pu Metal Casting.

The procedure and equipment for 30- to 100-g castings are like those for 10- to 30-g castings. The bomb-reduced or electrorefined metal is placed in a 1.5-in.-diam conical tantalum funnel. The mold crucible is ThO_2 0.625 in. diam by 1.5 in. tall. The usual amount of plutonium is 50 g. With this amount of metal, losses are 1 to 2 g and are recoverable.

F. Preparation of ^{238}Pu-Ga Alloy.

The following procedure is used in preparation of ^{238}Pu-Ga alloys. Ingots of cast electrorefined ^{238}Pu metal and gallium metal are melted in a tantalum crucible. The melt is stirred with a motor-driven tantalum stirrer for 1 h at 750 °C. It is then frozen rapidly by lowering the tantalum crucible into a cooled copper cylinder located below the furnace heat zone. The solid alloy is then annealed at 525 °C for 48 h to eliminate "coring."[19] Upon completion of the annealing period, the furnace is cooled to room temperature.

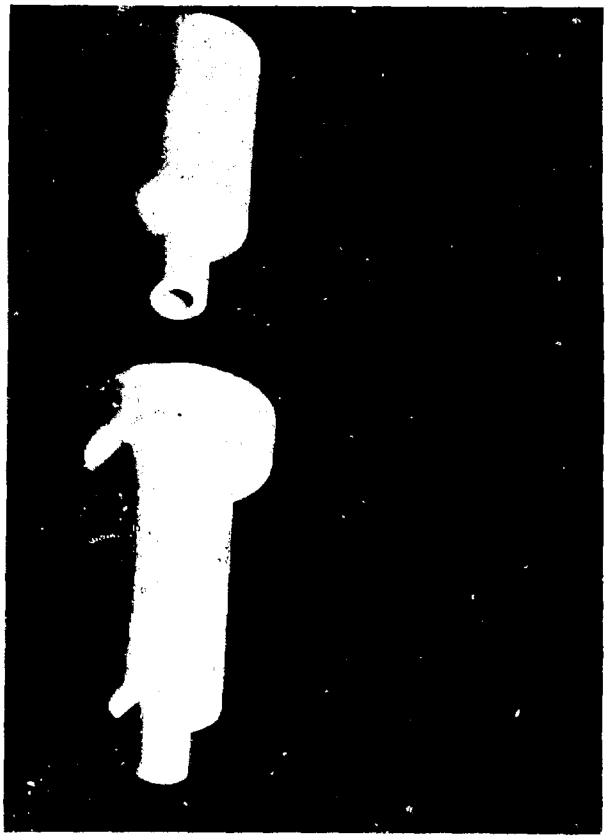


Fig. 11.
Small-scale casting equipment and casting (scale in inches).

G. Preparation of ${}^{238}PuO_2^{16}$ Fuels.

1. **${}^{238}PuO_2^{16}$ Powder.** The feed materials for preparation of ${}^{238}PuO_2^{16}$ are electrorefined ^{238}Pu metal and H_2O^{16} made from ${}^{14}NO^{16}$ by the reaction[20]

$$ {}^{14}NO^{16} + H_2 \xrightarrow[400\,^{\circ}C]{Pt\ cat.} H_2O^{16} + \frac{1}{2} N_2^{14} \quad . $$

The ${}^{14}NO^{16}$ is produced by thermal distillation of natural NO gas[21] which yields process streams of ${}^{14}NO^{16}$, ${}^{14,15}NO^{17}$, and ${}^{15}NO^{18}$. The ${}^{14}NO^{16}$ gas comes off the top of the distillation column and is fed directly to a catalytic platinum furnace[20] and reacted with H_2. The water collected contains small amounts of NH_3. The ammonia and water are separated by distillation under a reduced pressure of argon gas.

The present flowsheet for preparation of ${}^{238}PuO_2^{16}$ powder is shown in Fig. 12. The starting material is

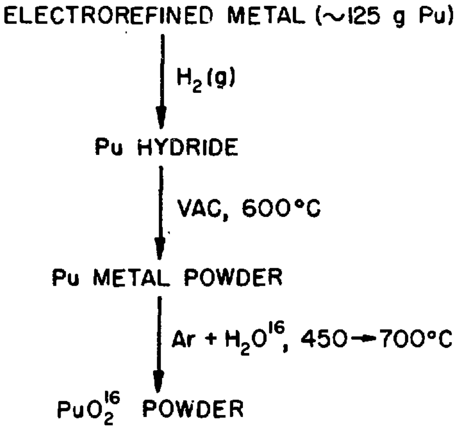


Fig. 12.
Flowsheet for preparation of $^{238}PuO_2^{16}$ Powder.

electrorefined metal. This metal is converted to powder by three cycles of hydriding and dehydriding. The powder is then converted to $^{238}PuO_2^{16}$ by reaction with H_2O^{16}.

The following chemical reactions are involved in this preparation.

$$^{238}Pu\ (solid) + 0.4\ H_2\ (gas) \xrightarrow{\sim 200\ ^\circ C} {}^{238}PuH_{0.8} \quad ,$$

$$^{238}PuH_{0.8} \xrightarrow[vac.]{600\ ^\circ C} {}^{238}Pu\ (powder) \quad ,$$

$$^{238}Pu\ (powder) + 2\ H_2O^{16}\ (gas) \xrightarrow[Ar\ atmosphere]{450\text{-}700\ ^\circ C}$$

$$^{238}PuO_2^{16} + 2\ H_2 \quad .$$

For the last reaction, argon gas saturated with H_2O^{16} at room temperature is passed over the metal powder. Detailed operating instructions for preparation of the oxide powder are given in Appendix A.

2. **$^{238}PuO_2^{16}$ Ceramics.** The present flowsheet for production of $^{238}PuO_2^{16}$ ceramics is shown in Fig. 13. All operations with $^{238}PuO_2^{16}$ powder must be conducted in inert-atmosphere gloveboxes to avoid oxygen isotopic exchange. The sintered ceramic, however, can be handled in air (see below).

The oxide powder is ball-milled for 18 h, then screened through a 200-mesh screen. The powder is pressed at 20 tsi, and the resulting pellet is crushed and rescreened. Union Carbide Corp. Carbowax dissolved in trichloroethylene is added as a pressing aid, and the powder is pressed at 20 tsi. Upon removal from the die, the pellet is cooled to ensure slow vaporization of the Carbowax. The pellet is then sintered at 1550 °C in an ($Ar + H_2O^{16}$) atmosphere.

The above procedure has been used to produce 1-, 30-, and 50-W cylinders of $^{238}PuO_2^{16}$. Approximately a hundred and fifty 1-W, six 30-W, and four 50-W cylinders have been prepared. Processing losses are low: ~1 g of plutonium per 100g of oxide. These losses occur during screening, and the oxide is retained by the glovebox filter.

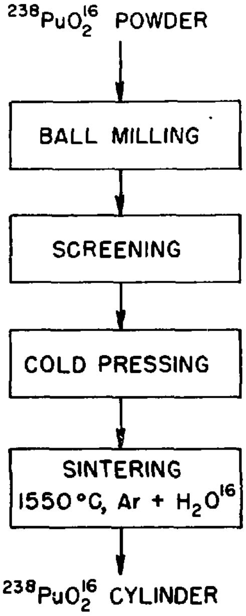


Fig. 13.
Flowsheet for preparation of $^{238}PuO_2^{16}$ ceramics.

H. Preparation of $^{238}PuO_2^{16}$ Cylinders by Oxygen Isotopic Exchange.

Flowsheets for preparation of $^{238}PuO_2^{16}$ cylinders by oxygen isotopic exchange are still under study. The reaction is simply

$$^{238}PuO_2{}^{nat} + H_2O^{16} \xrightarrow{750\ ^\circ C} {}^{238}PuO_2^{16} + H_2O^{nat} \ .$$

This reaction has been studied by workers at Savannah River who prepared oxides with neutron emission rates as low as 5500 n/sec - g ^{238}Pu.[22]

Oxygen isotopic exchange with $^{238}PuO_2{}^{nat}$ powders (flowsheet, Fig. 14) and $^{238}PuO_2{}^{nat}$ pellets (flowsheet, Fig. 15) are still under study. Obviously, the quality of the $^{238}PuO_2^{16}$ depends on the purity of the oxide feed. This purity has presented the principal problem in the preparation, resulting in products of variable neutron emission rate. Powders and pellets prepared using the flowsheet in Fig. 14 have had neutron emission rates 300 to 1500 n/sec - g of ^{238}Pu higher than the measured rate for oxides prepared from electrorefined metal. Chemical methods presently used for preparing $PuO_2{}^{nat}$, anion exchange, oxalate precipitation, and peroxide precipita-

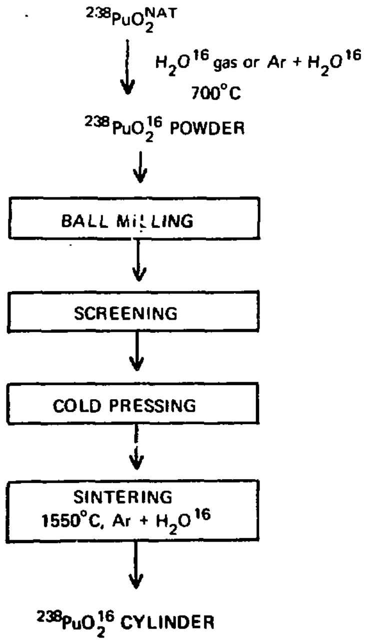


Fig. 14.
Preparation of $^{238}PuO_2^{16}$ powder and ceramics by oxygen isotopic exchange.

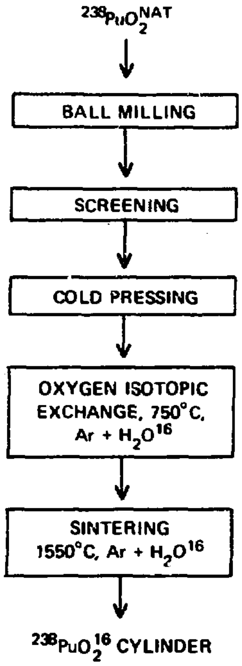


Fig. 15.
Alternate oxygen isotopic exchange flowsheet for $^{238}PuO_2^{16}$ cylinders.

tion, are inferior to electrorefining for routinely producing ^{238}Pu of high chemical purity.

I. Preparation of $^{238}PuN^{15}$ Fuel.

Nitrogen-15 is prepared from $^{15}NO^{18}$ by the reaction[23]

$$^{15}NO^{18} + Cu \rightarrow CuO^{18} + \frac{1}{2} N_2^{15} \quad .$$

The flowsheet for preparation of $^{238}PuN^{15}$ is shown in Fig. 16. Electrorefined metal is again the starting material. The metal is converted to powder by forming and then decomposing the hydride. The powder reacts readily with $^{15}N_2$ to form the nitride. The reaction is simply

$$^{238}Pu \text{ (powder)} + \frac{1}{2}\ ^{15}N_2 \text{ (g)} \xrightarrow[300 - 500\ ^{\circ}C]{} {}^{238}PuN^{15} \quad .$$

V. CHEMICAL AND RADIATION PROPERTIES OF ^{238}Pu FUEL FORMS

A. Electrorefined Metal.

1. Chemical Purity. Five 10- to 20-g lots of electrorefined metal were prepared. Their chemical analyses are given in Table BIII. A typical lot such as LA2 contained less than 50 ppm of detectable metallic impurities, exclusive of neptunium.

Twenty-six 55- to 100-g lots of electrorefined metal have been prepared. Twenty have been analyzed chemically, and the results are given in Table BIV. The chemical purity of the metal from both scales of operation compares favorably, except for iron which is slightly higher in some of the larger scale runs. This small difference is due primarily to different sampling procedures for small and

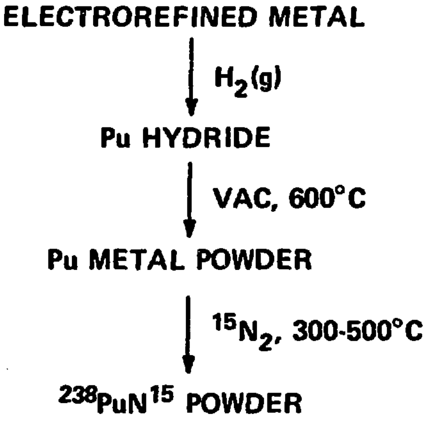


Fig. 16.
Flowsheet for preparation of $^{238}PuN^{15}$ powder.

large castings. The sampling tip is easily broken off the bottom of the smaller castings (see Fig. 11). The samples from larger castings are cut from the top of the casting using stainless steel tools that can contaminate the sample with iron. In the event of segregation of impurities when molten plutonium freezes, most will concentrate at the top of a casting rather than at the bottom.[24] Therefore, all samples listed in Table BIV, unless indicated otherwise, were cut from the top of the casting. In two cases, 100-26 and 100-27, samples were taken at the top and bottom ends of castings, and no significant differences in chemical purity were detected.

Analyses for all the small- and large-scale runs have been averaged, and the results are given in Table X. The total detectable metallic impurities exclusive of neptunium, americium, and uranium were less than 100 ppm. The average neptunium value was 690 ppm. This could be reduced by increasing the $PuCl_3$ concentration in the electrolyte. For example, increasing the $PuCl_3$ concentration from 10 to 20 wt.% would decrease the neptunium in the product from ~30 to ~15% of the feed concentration. However, the rapid growth of ^{234}U in the product, 520 ppm/month for the 80 at.% ^{238}Pu fuel, does not justify such a sacrifice of $PuCl_3$. The average plutonium concentration determined by potentiometric titration was 99.9%. Thus, within the limits of the analysis (±0.5% Pu), the product is pure plutonium.

TABLE X

AVERAGE PURITY OF CAST METAL FROM 10- to 20-g AND 100-g ELECTROREFINING RUNS

(Average of five 10- to 20-g and nineteen 100-g runs)

Element	Impurity (ppm)^a	Element	Impurity (ppm)^a
Li	0.12	Rb	< 0.5
Be	< 0.01	Sr	< 0.1
B	0.2	Y	< 0.1
C	48	Zr	0.3
Na	2	Mo	< 0.5
Mg	7	Cd	< 0.5
Al	8	Sn	1.6
Si	14	Cs	< 2
K	3	Ba	0.7
Ca	3	La	< 1
Ti	0.7	Hf	< 0.5
V	< 1	Re	< 0.5
Cr	1	Pb	0.7
Mn	7	Bi	< 1
Fe	35	^{241}Am	6
Co	< 0.5	^{237}Np	690
Ni	5	^{232}U	.010
Cu	4	^{234}U	115
Zn	< 5	Pu, wt%, by titration	99.9

---

[a] For all tables, ppm impurity = g impurity/10^6 g Pu.

The Am, ^{232}U, and ^{234}U values in Tables BIII, BIV, and X are corrected to the date of electrorefining, because these nuclides grow into the product. For an 80 at.% ^{238}Pu fuel containing 1 ppm of ^{236}Pu and 1 at.% ^{241}Pu, the growth rates are 520 ppm of ^{234}U, 41.2 ppm of ^{241}Am, and 0.020 ppm of ^{232}U per month, where ppm is grams of nuclide per 10^6 g of plutonium.

2. Melting Point. Before this work, the melting point of ^{238}Pu metal was reported to vary from 575 to 615 °C.[4] Measurements on electrorefined metal give a melting point of 639 (±3)°C which is essentially identical to the 640(±2)°C reported for ^{239}Pu.

3. Neutron Emission Rate. The neutron emission rate of ^{238}Pu is composed of contributions from:

spontaneous fission of ^{238}Pu and ^{240}Pu,

reactions of alpha particles with light-element impurities resulting in neutron production, i.e. (α-n) reactions, and

fast fission of ^{238}Pu, ^{239}Pu, and ^{240}Pu.

The contribution from fast fission depends on the geometry and density of the fuel form. It can be calculated and expressed conveniently as a multiplication factor which is applied to the sum of the spontaneous fission and (α,n) neutrons. Multiplication factors for plutonium cylinders of equal length and diameter are given in Fig. 17 for plutonium metal and for PuO_2 at densities of 11.47 and 9.75 g cm^3. The multiplication factor for a metal source varies from 1.07 for 13 g of plutonium to 1.15 for 100 g of plutonium. For 9.75-g/cm^3 oxide sources, the multiplication factor is 1.04 for 13 g of plutonium and 1.08 for 100 g of plutonium.

All of the measured neutron emission rates for electrorefined metal are given in Table BV. The average rate for the 18 samples containing 1 - 2 g of plutonium was 3095 (±46) n/sec-g ^{238}Pu. For a 80.00-wt.% ^{238}Pu fuel, this is equivalent to 2476 n/sec-g of plutonium. The average rate for the 25 samples containing 10 - 20 g of plutonium was 3123 (±59) n/sec-g ^{238}Pu. That for the ten 20- to 30-g samples was 3170 (±50) n/sec-g ^{238}Pu.

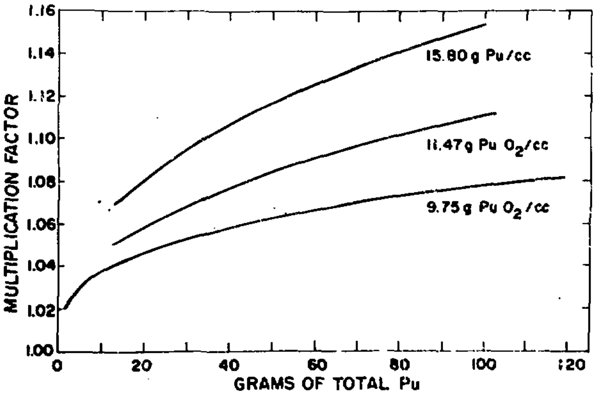


Fig. 17.

Multiplication factors for ^{238}Pu metal and ^{238}PuO$_2^{16}$ 80 at.% enrichment.

4. **Spontaneous Fission Rate.** The spontaneous fission rate of ^{238}Pu can be calculated by correcting the neutron emission rate for:

fast-fission neutrons,

(α,n) neutrons from light-element impurities,

spontaneous fission neutrons from ^{240}Pu, and

thermal-neutron multiplication in the counter.

We used the following criteria in selecting data from Table BV to calculate the spontaneous fission rate.

- Only large (>7-g) samples of plutonium of well-defined geometry were considered. (The small (1- to 1.5-g) samples have large surface area-to-volume ratios and are easily contaminated with oxygen although all metal handling is done in an inert atmosphere).
- Only samples whose gamma spectra showed no impurity elements were considered.
- Only samples that had been characterized by chemical analysis were considered.
- When several samples per electrorefining run were evaluated, the lower neutron emission rates were used.

The data used to calculate the spontaneous fission rate are given in Table XI. Neutron multiplication factors were obtained from Ref. 25. The thermal multiplication factor, which is a characteristic of the neutron counter used, was estimated to be 1%. The purity data from Tables BIII and BIV and the thick-target yields from Table XII were used to calculate the (α,n) contribution. In addition to the impurities shown in Tables BIII and BIV, the metals were assumed to contain 50 ppm of oxygen and 40 ppm of carbon, on the basis of analysis of a few ^{238}Pu metal samples. The (α,n) contributions from carbon and oxygen thus amount to 25 n/sec-g of ^{238}Pu. A spontaneous fission rate of 1.02 x 10^3 n/sec-g ^{240}Pu was used to calculate the ^{240}Pu contribution. Applying these corrections to the measured neutron emission rate gives the ^{238}Pu spontaneous fission values shown in Table XI, which average 2783 (±54) n/sec-g of ^{238}Pu. The earlier literature value is 2586 (±400) n/sec-g ^{238}Pu.[2,3]

5. **Gamma Radiation.** The gamma-ray spectrum of freshly prepared electrorefined metal is mostly the result of ^{238}Pu decay. A typical gamma spectrum is shown in Fig. 18 (A-D) in which all peaks are from ^{238}Pu decay unless otherwise identified. This spectrum was measured on freshly prepared electrorefined metal encapsulated in 0.030-in.-thick tantalum. At the extreme left in Fig. 18 A is a double peak corresponding to the tantalum Kα and Kβ x rays at 58 and 65 keV. The 152-keV peak is the most intense gamma ray emanating from tantalum-encapsulated sources, although its absolute intensity is one tenth that of the 99.6-keV gamma ray. The 208-keV peak is from ^{237}U, the daughter of the α-branch decay of ^{241}Pu. It can always be seen, even in freshly electrorefined metal, because of the short, 6.75-day half-life of ^{237}U. All of the prominent gamma rays in Fig. 18 B, from 700 to 1100 keV, are from ^{238}Pu decay. The peaks in Figs. 18 C and D reflect principally the contribution of ^{236}Pu. The 2614-keV gamma ray is from ^{208}Tl, a daughter of ^{236}Pu. Also seen are single- and double-escape peaks at 2104 and 1593 keV, resulting from the 2614-keV peak, and the ^{40}K background peak at 1460 keV. The 2614-keV photon is always present, and its effect on the gamma-ray dosage as result of aging is discussed later.

The absolute intensities of the principal gamma rays from ^{238}Pu were measured on thin unshielded sources. Electrorefined metal was dissolved in HCl, and americium, neptunium, uranium, and ^{236}Pu daughter activities were removed by separations with anion and cation exchange resins. Two large-volume, >30-cm^3, Ge(Li) detectors were used to measure the gamma rays. Two identical aliquots were examined with each detector under the same geometry used in calibrating the detectors with several standards. Average ^{238}Pu gamma-ray intensities are given in Table XIII. The average relative standard deviation was not greater than 10%. The measured gamma-ray intensities above 700 keV probably are within 10% of the true value, but the accuracy may be somewhat poorer at lower energies because large absorption effects caused difficulties.

TABLE XI

CALCULATION OF SPONTANEOUS FISSION RATE OF ^{238}Pu

E.R. Run	Capsule No.	Pu(g)	Meas'd Neutron Emission Rate (n/sec - g ^{238}Pu)	Multiplication Factor plus Thermalization Factor	Neutrons from Light-Element Impurities (n/sec - g ^{238}Pu)	Neutrons from Spontaneous Fission of ^{240}Pu (n/sec - g ^{238}Pu)	Neutrons from Spontaneous Fission of ^{238}Pu (n/sec - g ^{238}Pu)
LA 2	6	10.64	3070	1.064	48	37	2800
LAR 3	14	14.07	3020	1.071	84	38	2698
LAR 3	29	12.61	2994	1.067	84	38	2684
LA 98	36	13.33	3119	1.068	33	37	2850
100-3	65	26.810	3010	1.082	75	39	2668
100-3	69	11.878	3078	1.067	75	39	2771
100-3	71	11.686	3091	1.066	75	39	2786
100-4	73	24.657	3113	1.080	47	39	2796
100-4	86	13.198	3112	1.068	47	39	2828
100-5	82	7.876	3109	1.058	80	39	2820
100-7	169	13.43	3012	1.071	112	23	2677
100-9	131	12.744	3081	1.068	38	36	2811
100-15	217	18.529	3130	1.075	48	22	2842

6. Properties of a Typical 5-W Electrorefined Metal Source. The properties of a typical 5-W source, 10.65 g of plutonium, 79.8 wt.% ^{238}Pu, Run LA2, are given in Table XIV. The dose rates measured in air through 0.030-in-thick tantalum, 10 cm from the center of the source are 0.31 mRem/h-g ^{238}Pu for neutrons and 0.25 mR/h-g ^{238}Pu for gammas.

B. ^{238}Pu-Ga Alloy.

Adding gallium to ^{238}Pu metal does not significantly affect the radiation properties of the fuel. This can be seen in Table XV which compares the radiation properties of a ^{238}Pu - 3 at.% Ga alloy and its metal feed. The dose rates from the ^{238}Pu metal feed and alloy are essentially the same. (Bomb-reduced metal, Lot LA-7, was used as feed metal in this preparation.)

TABLE XII

SPECIFIC NEUTRON YIELDS FROM LIGHT-ELEMENT IMPURITIES

Element	n/sec-g ^{238}Pu for 1 ppm of Element
Li	5.7
Be	162
B	51
C	0.2
O	0.1
^{17}O	0.62
^{18}O	6.25
F	22
Na	2.7
Mg	2.6
Al	1.2
Si	0.2
P	$<$0.03
S	$<$0.03

Thirty-Watt Pu-Ga Heat Source, LASL 1. Only one large Pu-Ga heat source, made in December 1968 for in-phantom dosimetry studies at Battelle, Pacific Northwest Laboratory (PNL),[26] has been prepared in this program. The properties of this 30-W source are given in Table XVI. It was made before the 100-g scale electrorefining procedures were developed, and several remelts of small sources were required. In spite of excessive handling, the radiation properties of LASL-1 were satisfactory. The chemical purity, given in Table XVII, is not quite so good as that of the metal now being produced routinely on the 100-g scale (see Table X). The radiation properties are compared to those of a typical 5-W electrorefined metal sample in Table XVIII. The neutron emission rates compare favorably. The slightly higher rate of the Pu-Ga alloy is due primarily to the larger neutron multiplication factor of the larger source. Its lower gamma dose rate is due to its increased self-shielding by plutonium atoms. The gamma-ray spectrum was identical to that obtained with freshly prepared electrorefined metal.

C. $^{238}PuO_2^{16}$ Powder Prepared from Electrorefined Metal (Flowsheet, Fig. 12).

1. Isotopic Composition of H_2O^{16}. The isotopic composition of 23 lots of H_2O^{16} prepared at LASL is given in Table XIX. The average values are 73 ppm of ^{17}O and 22 ppm of ^{18}O, somewhat higher than the desired specification for H_2O^{16} used in the preparation of medical-grade

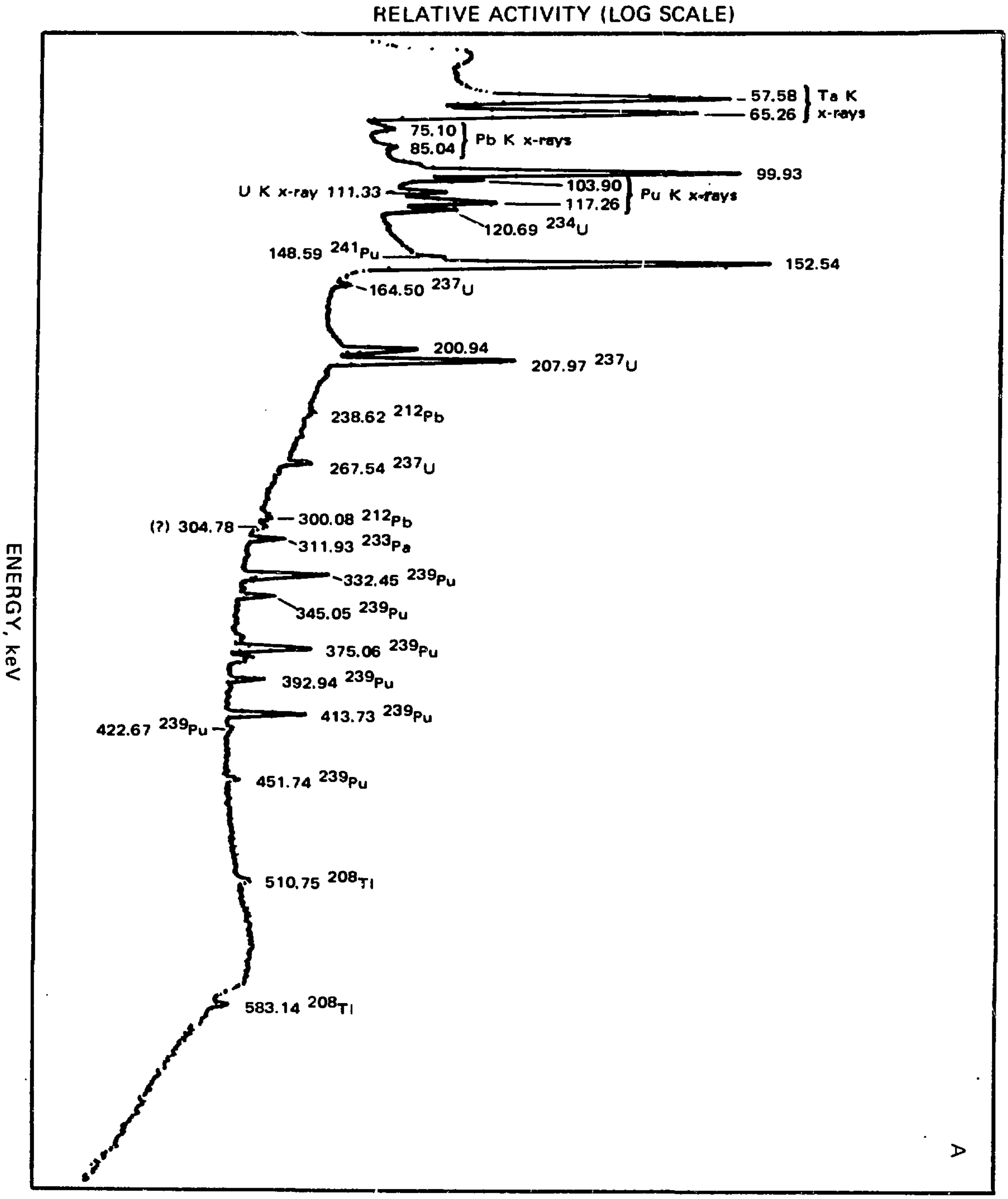
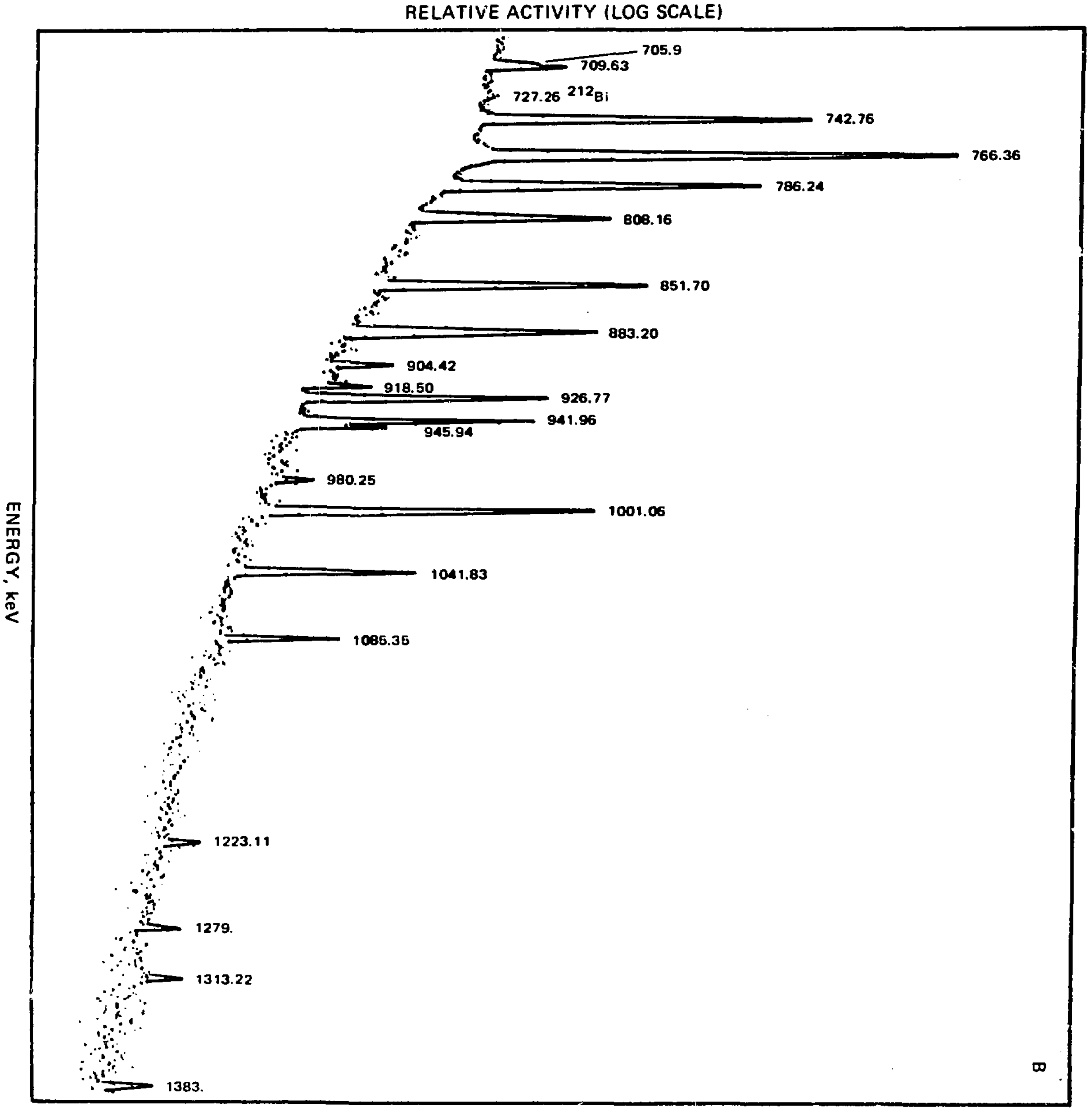
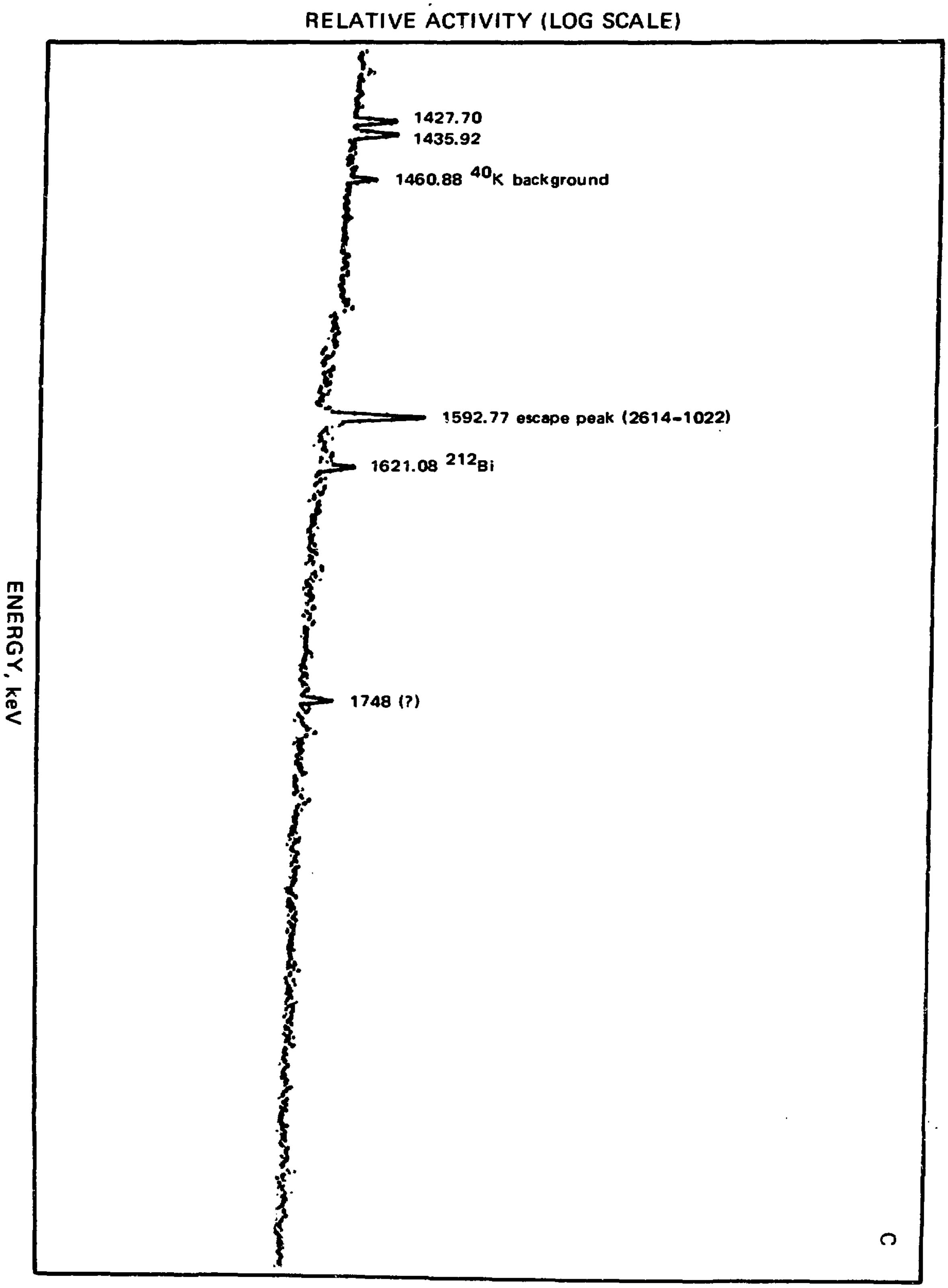


Fig. 18.

Gamma-Ray spectrum of electrorefined ^{238}Pu metal.


RELATIVE ACTIVITY (LOG SCALE)
ENERGY, keV
705.9
709.63
727.26 ^{212}Bi
742.76
766.36
786.24
808.16
851.70
883.20
904.42
918.50
926.77
941.96
945.94
980.25
1001.06
1041.83
1085.35
1223.11
1279.
1313.22
1383.
B


RELATIVE ACTIVITY (LOG SCALE)
ENERGY, keV
1427.70
1435.92
1460.88 ^{40}K background
1592.77 escape peak (2614-1022)
1621.08 ^{212}Bi
1748 (?)
C

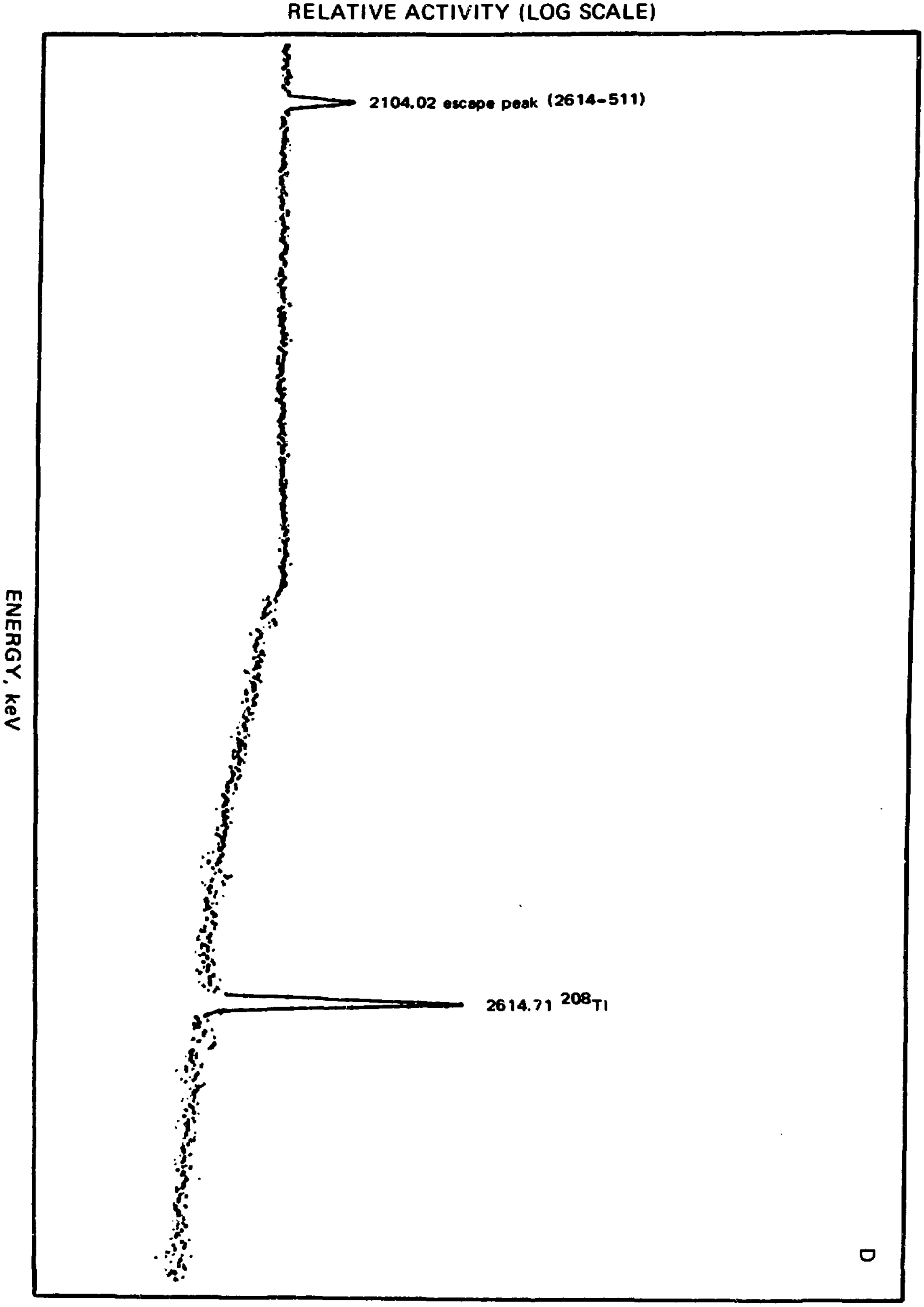

RELATIVE ACTIVITY (LOG SCALE)
ENERGY, keV
2104.02 escape peak (2614-511)
2614.71 ^{208}Tl
D

TABLE XIII

^{238}Pu GAMMA-RAY INTENSITIES AND PRECISION OF MEASUREMENT
(80 at.% ^{238}Pu)

Energy (keV)	Gamma-Ray Intensity (gammas per alpha)	Avg. Std. Dev. (%)
43.5	2.51×10^{-4}	1
99.6	7.66×10^{-5}	1
152.5	8.5×10^{-6}	1
201.2	4.3×10^{-8}	2
707.8	3.3×10^{-9}	7
742.4	5.25×10^{-8}	1
765.8	2.31×10^{-7}	1
785.8	3.3×10^{-8}	2
807.6	8.5×10^{-9}	5
851.3	1.36×10^{-8}	1
882.9	9.4×10^{-9}	2
926.5	5.3×10^{-9}	1
941.8	5.3×10^{-9}	4
1001.1	9.5×10^{-9}	1
1041.8	2.2×10^{-9}	8
1085.1	7.0×10^{-10}	9

fuels. The desired specification is

$$\text{ppm } ^{18}\text{O} + \frac{1}{10} \text{ ppm } ^{17}\text{O} < 20 \text{ ppm} \quad ,$$

$$\text{where ppm } ^{18}\text{O} = \text{g } ^{18}\text{O}/10^{6} \text{ g O} \quad .$$

TABLE XIV

PROPERTIES OF ELECTROREFINED ^{238}Pu METAL, TYPICAL 5-W CYLINDER, *H/D* = 1, RUN LA 2[a]

Melting point (°C)	639 (±3)
Neutron emission rate	
n/sec-g Pu	2450
n/sec-g ^{238}Pu	3070
Dose rates at 10 cm[b]	
Neutron (mRem/h-g ^{238}Pu)	
Calc'd from emission rate	0.31
TLD measurement	0.31
Gamma (mR/h-g ^{238}Pu)	
TLD measurement	0.25

---

[a] Plutonium-238 isotopic composition - 79.8 wt% ^{238}Pu.

[b] Dose rates measured in air through 0.030-in.-thick tantalum.

TABLE XV

RADIATION PROPERTIES OF ^{238}Pu METAL AND ^{238}Pu-3 at.% Ga ALLOY

	^{238}Pu Metal	^{238}Pu-Ga[a]
Neutron emission rate (n/sec-g Pu)	2665	2728
Dose rates at 10 cm[b]		
Neutron (mRem/h-g Pu)		
Calculated from emission rate	0.27	0.27
Thermoluminescent dosimetry	0.24	0.26
Gamma (mR/h-g Pu)		
Thermoluminescent dosimetry	0.21	0.20

---

[a] Five-W sources, 80 wt% ^{238}Pu.

[b] Dose rates in air through 0.030 in. of tantalum.

This amount of ^{18}O and ^{17}O should result in an (α-n) contribution of 115 n/sec-g of ^{238}Pu to the neutron emission rate of $^{238}\text{PuO}_2^{16}$. Most of the work described below was done with H_2O^{16} lots 9, 11, 13, and 29, all of which meet the desired specification. For comparison, the specifications for ^{16}O distributed by Mound Laboratory are ≤150 ppm of ^{17}O and ≤50 ppm of ^{18}O.[27]

2. **Chemical Purity.** The chemical purity of a typical lot of $^{238}\text{PuO}_2^{16}$ powder is compared to that of its electrorefined metal feed in Table XX. Analytical data for nine powder preparations, varying in size from 73 to 138 g of plutonium, are given in Table BVI. The history of the metal feed and size of the oxide preparation are given in Table BVII.

The data show that only trace amounts of impurities are introduced during preparation of oxide powders. The purity of metal and oxide compare favorably. The slight increase in light-element impurities of the oxide should contribute, on the average, ~100 n/sec-g of ^{238}Pu to the neutron-emission rate.

3. **Neutron Emission Rate.** The neutron emission rates of nine $^{238}\text{PuO}_2^{16}$ powders are compared to that of the electrorefined metal feed in Table XXI. An average rate increase of 275 n/sec-g of ^{238}Pu was observed in converting metal to oxide. As mentioned above, light-element impurities, excluding oxygen, should be responsible for ~100 n/sec-g of ^{238}Pu. Available data indicate that the balance, 175, is due to the presence of ^{17}O and ^{18}O in the $^{238}\text{PuO}_2^{16}$. Oxygen isotopic data for preparations LAS 9,

TABLE XVI

DESCRIPTION, COMPOSITION, AND RADIATION PROPERTIES OF 30-W Pu - Ga HEAT SOURCE NO. LASL-1

Wt. of alloy (g)	66.50
Wt. % Ga, chemical analysis	0.94
Wt. of Pu (g)	65.88
Dimensions of alloy casting	
Diam (in.)	0.720
Length (in.)	0.633
Density (g/cm^3)	15.6 (±0.5)
Power (W)	30.3
Power density (W/cm^3)	7.18
Pu isotopic composition (wt%)	
^{236}Pu	0.58 x 10^{-4}
^{238}Pu	80.45
^{239}Pu	16.16
^{240}Pu	2.73
^{241}Pu	0.54
^{242}Pu	0.12
Radiation properties	
Neutron emission rate	
n/sec-g Pu	2535
n/sec-g ^{238}Pu	3151
Dose rates at 10 cm[a]	
Neutron, (mRem/h-g ^{238}Pu)	
Calc'd from emission rate	0.32
Gamma (mR/h-g ^{238}Pu)	
TLD dosimetry, side	0.20
TLD dosimetry, end	0.16

[a] Dose rates in air 10 cm from the center of source, through 0.030 in. of tantalum.

11, and 13 are given in Table XXII, where the ^{17}O and ^{18}O concentrations in the H_2O^{16} used in the preparation are compared to the ^{18}O concentration of the $^{238}PuO_2^{16}$ powder. In each preparation, the ^{18}O concentration is higher in the oxide than in the water, indicating contamination of the oxide by natural oxygen. Although the ^{17}O concentration of the oxide was not measured, it can be calculated by using the ^{17}O concentration of the H_2O^{16} and assuming that the oxide is contaminated by natural oxygen or water vapor. The extent of contamination is shown by the increased ^{18}O content of the oxide. Thus, the calculated ^{17}O concentrations for LAS 9, 11, and 13 are 77.5, 75.9, and 75.4 ppm, respectively. The (α,n) contributions for the ^{17}O and ^{18}O values of Table XXII are 200, 140, and 297 n/sec-g of ^{238}Pu for LAS 9, 11, and 13, respectively. This is equivalent to an average value of ~200 n/sec-g of ^{238}Pu. A detailed analysis of the (α,n) contribution from light-element impurities for the

TABLE XVII

CHEMICAL PURITY OF HEAT SOURCE NO. LASL-1

Element	Impurity (ppm)	Element	Impurity (ppm)
Li	< 0.005	Sr	<0.1
Be	< 0.001	Y	<0.1
B	1	Zr	0.3
Na	< 2	Mc	5
Mg	<10	Cd	2
Al	3	Sn	7
Si	25	Cs	<2
K	6	Ba	<0.1
Ca	< 3	La	<1
Ti	< 1	Hf	<0.5
V	< 0.5	Re	<0.5
Cr	< 0.5	Pb	15
Mn	3	Bi	<0.5
Fe	74	^{241}Am[a]	38
Co	< 0.5	^{237}Np	612
Ni	70	^{232}U	0.048
Cu	5	^{234}U[a]	840
Zn	<10	^{236}Pu	0.58
Rb	< 0.5	F	<2

Total Pu (by titration) - 98.9%

[a] Values corrected to November 1, 1968.

TABLE XVIII

MEASURED RADIATION PROPERTIES OF HIGH-PURITY 5-W ^{238}Pu METAL SOURCES AND 30-W ^{238}Pu-3 Ga SOURCE, LASL 1

	5-W	30-W
Neutron emission rate,		
n/sec-g Pu	2450	2535
n/sec-g ^{238}Pu	3070	3151
Dose rates at 10 cm		
Neutron (mRem/h-g ^{238}Pu)		
Calc'd from neutron emission rate	0.31	0.32
TLD dosimetry	0.30	---
Gamma (mR/h-g ^{238}Pu)		
TLD disometry, side	0.25	0.20
TLD disometry, end	---	0.16

TABLE XIX

ISOTOPIC COMPOSITION OF LASL H_2O^{16} LOTS

Lot	Composition (g O isotope/10^6 g O)	
	^{17}O	^{18}O
6	58	29
7	68	30
8	74	27
9	49	14
10	72	14
11	75	10
12	98	28
13	55	10
14	58	28
15	82	30
16	68	22
17	64	24
18	64	27
19	72	36
20	75	28
21	68	24
22	72	28
23	74	32
29	70	10
30	75	15
31	130	15
51	80	12
52	80	15

LAS 13 powder is given in Table XXIII. The calculated (α,n) contribution in $^{238}PuO_2^{16}$ compared to metal is 331 n/sec-g of ^{238}Pu. The observed or measured neutron difference in emission rates (Table XXI) was 353. The multiplication factors for the small oxide and metal samples should be approximately equal, ~2%.

4. **Gamma Radiation.** The gamma-ray spectrum of a typical lot of $^{238}PuO_2^{16}$ is shown in Fig. 19. The spectrum is identical to that of electrorefined metal except for a gamma ray at 1634 keV which results from the (α,n) reaction with ^{17}O, and whose intensity is about twice that of the ^{238}Pu gamma ray at 1436 keV. Thus, if the metal and oxide had equal densities, the gamma dose rates would be essentially identical. However, the higher density of the metal results in greater self-shielding by the plutonium atoms and lower dose rates.

TABLE XX

CHEMICAL PURITY OF ELECTROREFINED METAL AND $^{238}PuO_2^{16}$ POWDER, LOT LAS 13

	Impurity (ppm)			Impurity (ppm)	
Element	Metal	Oxide	Element	Metal	Oxide
Li	< 0.1	< 0.2	Cu	2	5
Be	< 0.1	< 0.2	Zn	< 5	< 10
B	< 1	< 1	Rb	< 0.5	< 1
C	60	55	Sr	< 0.1	< 0.2
Na	5	20	Y	< 0.1	< 0.2
Mg	5	5	Zr	< 0.1	< 0.2
Al	7	1	Mo	< 0.5	< 1
Si	< 1	20	Cd	< 0.5	< 1
K	< 3	< 1	Sn	< 0.5	1
Ca	< 3	< 6	Cs	< 2	< 4
Ti	1	20	Ba	5	2
V	< 0.5	< 1	La	< 1	< 2
Cr	1	8	Hf	< 0.5	< 1
Mn	1	2	Re	< 0.5	< 1
Fe	55	80	Pb	1	2
Co	< 0.5	< 1	Bi	< 1	< 1
Ni	3	10	Np	664	—

TABLE XXI

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

	Neutron Emission Rate (n/sec-g ^{238}Pu)		
$^{238}PuO_2^{16}$ Lot	Metal	Powder	Difference (Powder - Metal)
LAS 3	3109	3434	325
LAS 6	3156	3495	339
LAS 7	3120	3371	251
LAS 8	3130	3450	320
LAS 9	3074	3293	219
LAS 10	3134	3338	204
LAS 11	3068	3291	223
LAS 12	3067	3309	242
LAS 13	3071	3424	353

TABLE XXII

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

	Oxygen Isotopic Composition (g isotope/10^6 g O)		
	^{17}O	^{18}O	
Prep. No.	H_2O^{16}	H_2O^{16}	$^{238}PuO_2^{16}$
LAS 9	75	10	24
LAS 11	75	10	15
LAS 13	70	10	40

D. Pressed and Sintered $^{238}PuO_2^{16}$ Cylinders.

1. General Discussion. The power density of $^{238}PuO_2^{16}$ powders (~1 W/cm^3 for the bulk powder) is too low to be suitable for use in a circulatory-assist device. A compact must therefore be prepared which has as high a power density as is consistent with fuel stability,* ~3.9 W/cm^3 for the 80 at.% ^{238}Pu fuel or 4.4 W/cm^3 for the 90 at.% ^{238}Pu fuel. We have prepared these compacts in the present program by pressing $^{238}PuO_2^{16}$ powder into a "green" cylinder and sintering the cylinder into a compact. The final sintered cylinders have a height-to-diameter ratio of 1.

Attempts to prepare 30-W cylinders directly from the $^{238}PuO_2^{16}$ powder described above were unsuccessful. Procedures described in Appendix A were developed and led to successful preparation of 30- and 50-W cylinders. These procedures require ball-milling to obtain a satisfactory particle-size distribution and the addition of Carbowax binder as a pressing and sintering aid. Both operations introduce impurities into the product, but continued improvements have allowed us to prepare sources whose neutron emission rates are only about 300 n/sec-g of ^{238}Pu higher than that of an equivalent electrorefined metal source. We have observed that small pellets, 2 W or less, can frequently be fabricated without adding Carbowax binder.

TABLE XXIII

(α,n) CONTRIBUTION FROM LIGHT–ELEMENT IMPURITIES, LOT LAS 13

Element	[$^{238}PuO_2^{16}$(α,n) - E. R. Metal (α,n)] (n/sec-g ^{238}Pu)
Li	0
Be	0
B	0
C	− 4.
Na	+ 41.
Mg	0
Al	− 8.
Si	5.0
^{17}O	47
^{18}O	250.
Sum	+ 331.

2. Ball-Milled Powder.

a. Chemical Purity. The chemical purity of a recent lot of $^{238}PuO_2^{16}$ powder before and after ball-milling is compared in Table XXIV. Analytical data for eight preparations are given in Table BVI.

The milling and screening operations done in stainless steel equipment introduce, on the average, ~150 ppm of iron, ~50 ppm of chromium, ~25 ppm of sodium, ~50 ppm of silicon, ~10 ppm of aluminum, ~20 ppm of nickel, ~50 ppm of copper, and ~70 ppm of tin into the product. Although only sodium, aluminum, and silicon contribute to the neutron-emission rate, we have started work on other milling procedures to reduce product contamination. Mills and balls fabricated from uranium should yield a better product.

b. Particle-Size Distribution. The mean particle size of ball-milled powders that have been successfully pressed into either 30- or 50-W pellets is given in Table XXV. The mean particle size varied from 4 to 9 μ. A representative particle-size distribution curve is given in Fig. 20. The flat distribution curve is typical of powders that yield integral 30- and 50-W cylinders.

c. Oxygen Isotopic Composition. Ball-milling is presently done in an argon-filled glove box that contains ~200 ppm of oxygen. As a result, there is usually about a 50-ppm increase in the ^{18}O concentration of the $^{238}PuO_2^{16}$ powder.

The ^{18}O concentration is reduced to ~15 ppm in the subsequent sintering operation in an atmosphere of (Ar +H_2O^{16}).

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*Fuel stability has yet to be defined. At present, it appears that $^{238}PuO_2^{16}$ cylinders of 85% theoretical density, 9.7 g/cm^3, have adequate thermal shock resistance and radiation stability.

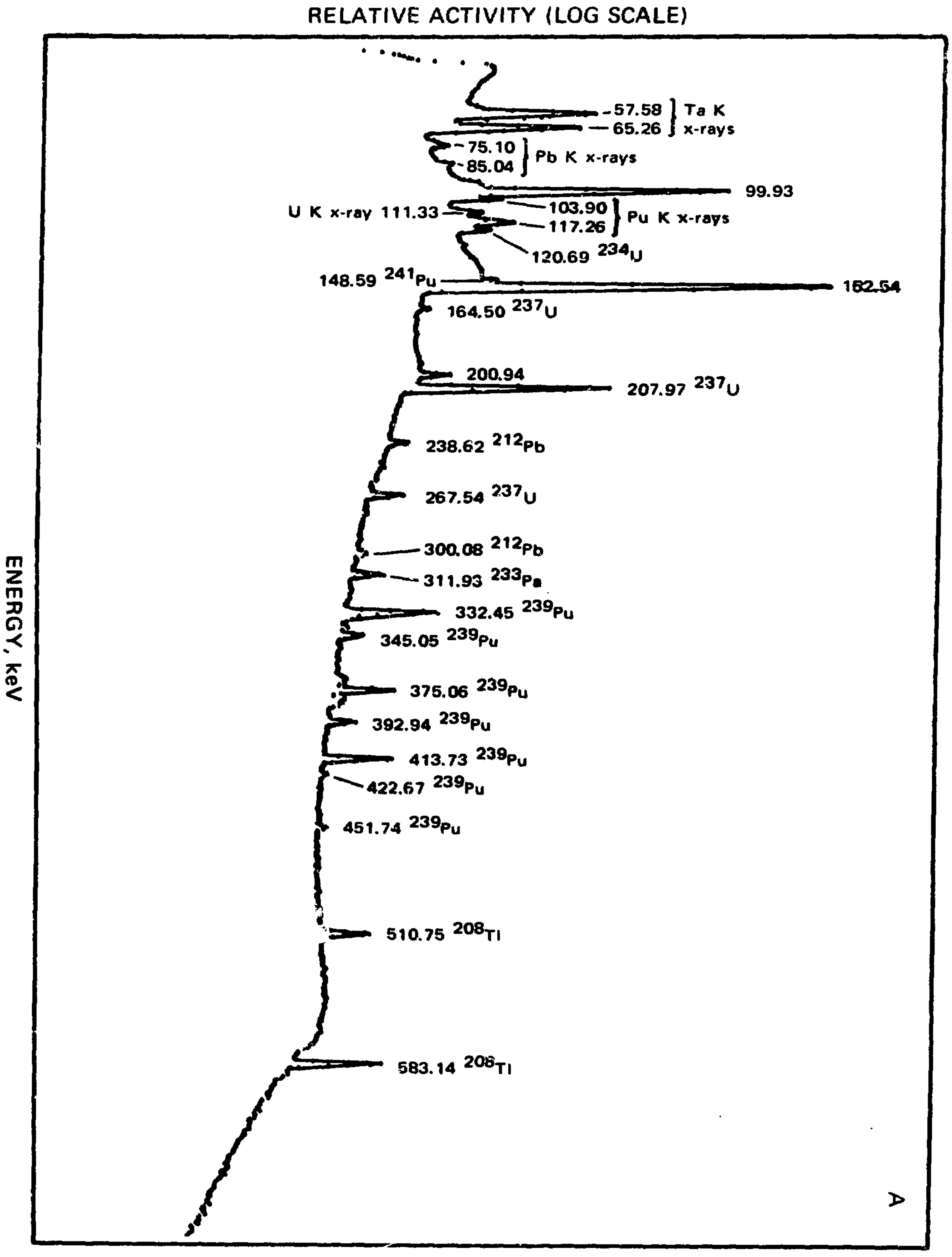
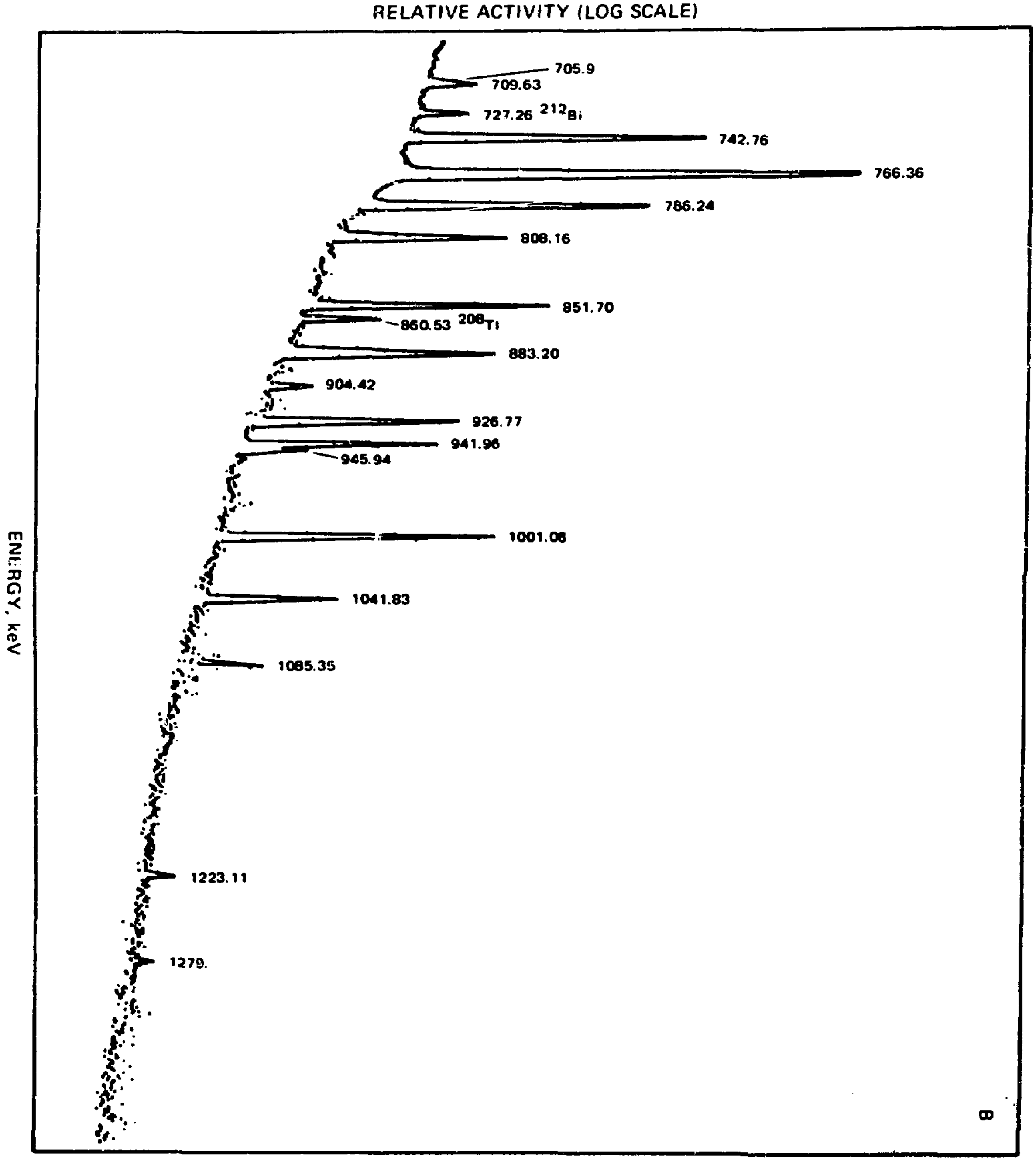
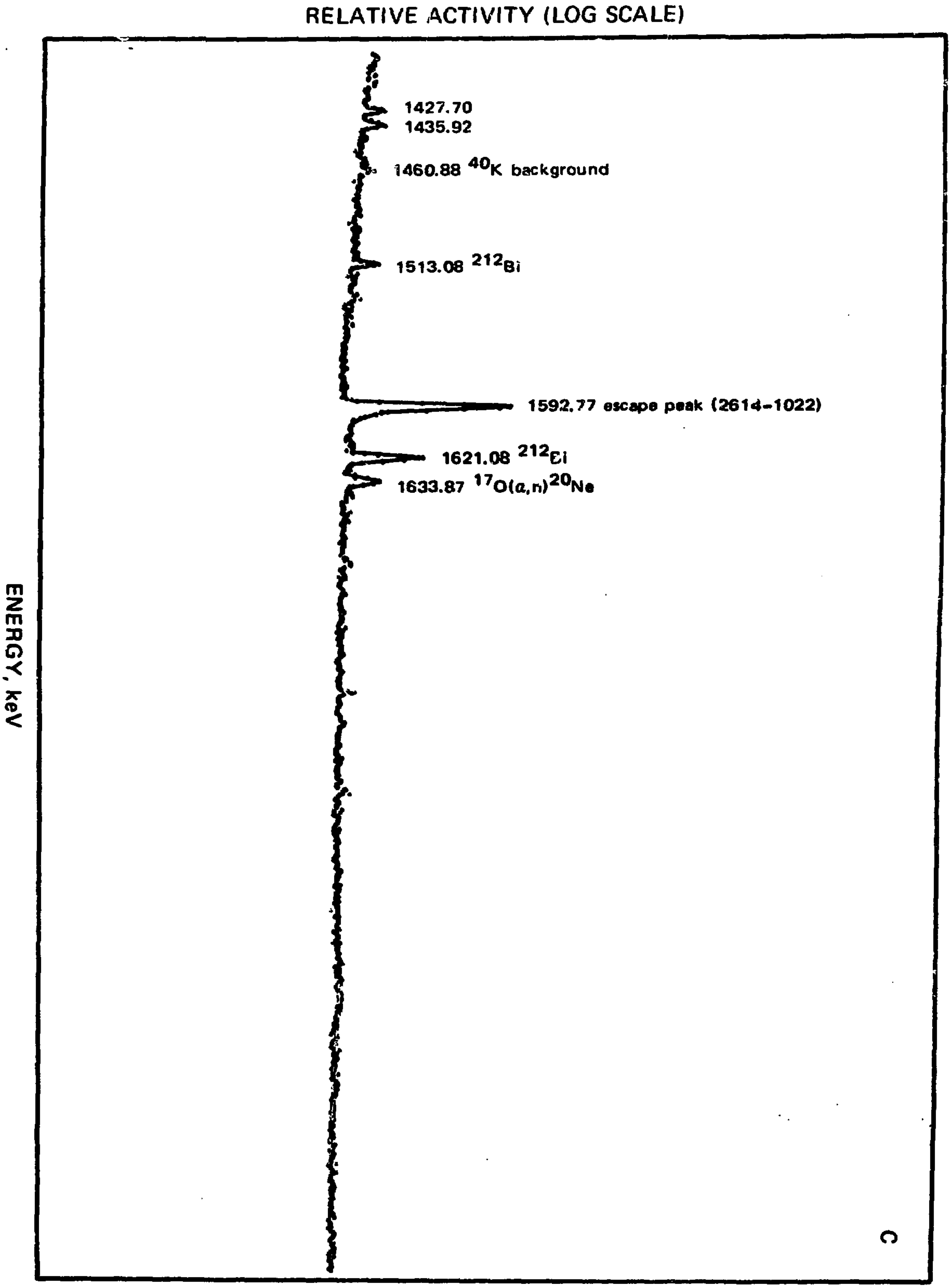


Fig. 19.
Gamma-Ray spectrum of $^{238}PuO_2^{16}$.


RELATIVE ACTIVITY (LOG SCALE)
ENERGY, keV
705.9
709.63
727.26 ^{212}Bi
742.76
766.36
786.24
808.16
851.70
860.53 ^{208}Tl
883.20
904.42
926.77
941.96
945.94
1001.06
1041.83
1085.35
1223.11
1279.
B


RELATIVE ACTIVITY (LOG SCALE)
ENERGY, keV
1427.70
1435.92
1460.88 ^{40}K background
1513.08 ^{212}Bi
1592.77 escape peak (2614–1022)
1621.08 ^{212}Bi
1633.87 ^{17}O(α,n)^{20}Ne
C

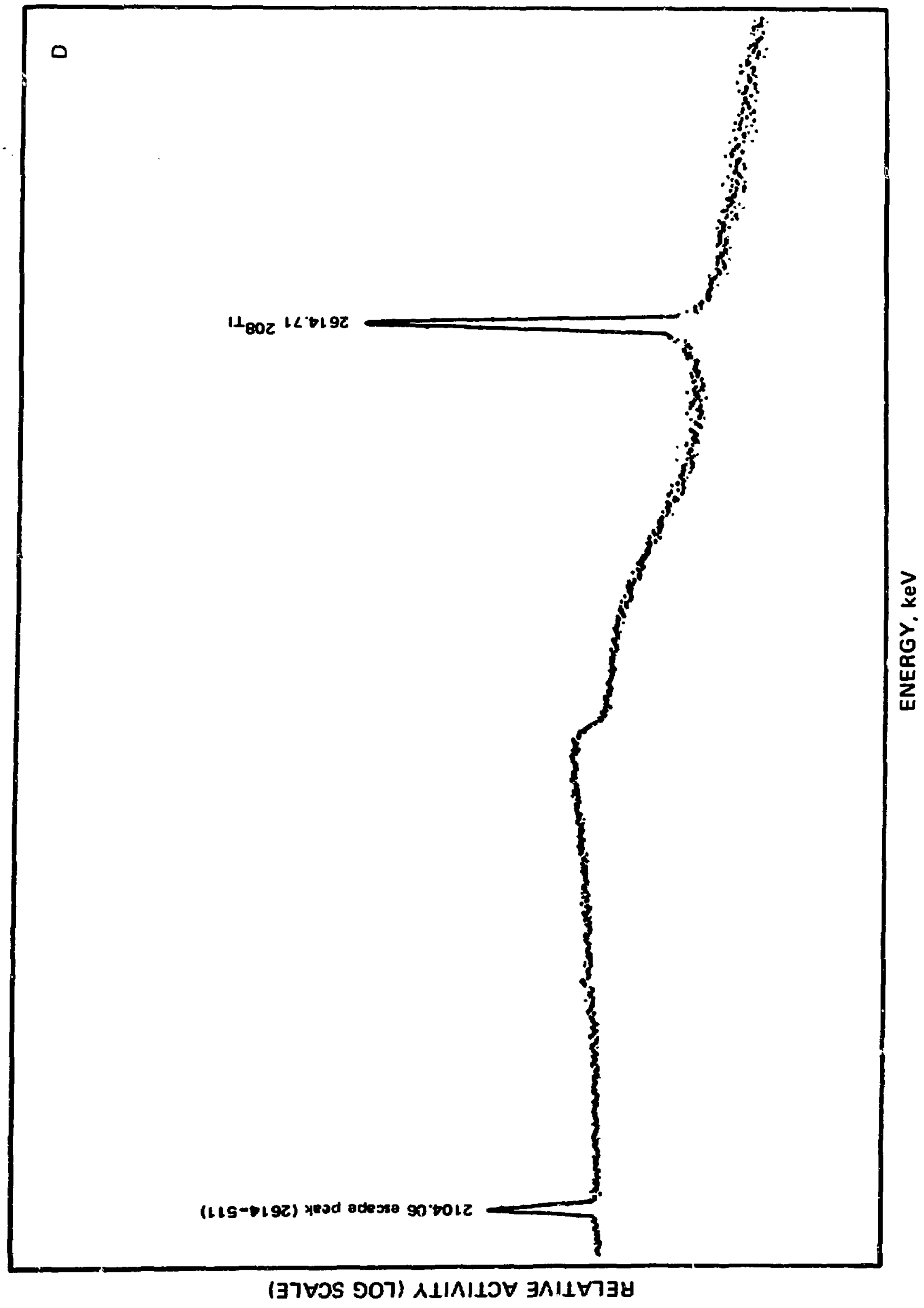

D
2614.71 ²⁰⁸Tl
2104.06 escape peak (2614-511)
ENERGY, keV
RELATIVE ACTIVITY (LOG SCALE)

TABLE XXIV

CHEMICAL PURITY OF $^{238}PuO_2^{16}$ POWDERS, BEFORE AND AFTER MILLING, LOT LAS 11

	Impurity (ppm)			Impurity (ppm)	
Element	**Before**	**After**	**Element**	**Before**	**After**
Li	0.1	0.2	Cu	2	60
Be	< 0.2	< 0.2	Zn	< 10	30
B	< 2	< 1	Rb	< 1	< 1
C	130	140	Sr	< 0.2	< 0.2
Na	9	22	Y	< 0.2	< 0.2
Mg	< 2	< 2	Zr	< 0.2	0.2
Al	10	16	Mo	1	2
Si	< 5	20	Cd	< 1	< 1
K	< 1	< 1	Sn	< 5	200
Ca	< 6	< 6	Cs	< 4	< 4
Ti	1	4	Ba	< 0.4	< 0.4
V	< 1	< 1	La	< 2	< 2
Cr	2	35	Hf	< 1	< 1
Mn	0.4	2	Re	< 1	< 1
Fe	< 2	130	Pb	2	3
Co	< 1	< 1	Bi	< 1	< 1
Ni	4	20	Np	880	860

3. Pressed and Sintered Pellets.

a. Thirty-W Cylinders. The primary objective of the ceramic fabrication program has been the preparation of pressed and sintered 30- and 50-W $^{238}PuO_2^{16}$ cylinders of medical-grade quality. Development of fabrication procedures has gone hand in hand with preparation of sources for AEC contractors. The quality of the sintered pellets has improved steadily, but still does not meet the desired specifications for medical-grade sources. These specifications,[28] for example, call for a neutron emission rate of less than 3300 n/sec-g of ^{238}Pu for a 30-W source. As seen below, the use of the flowsheets shown in Figs. 11 and 12 resulted in 30-W sources having neutron-emission rates of 3608 and 3521 n/sec-g ^{238}Pu.

TABLE XXV

MEAN PARTICLE SIZE OF BALL-MILLED POWDERS

Lot No.	Mean Particle Size (μ)
LAS 6	3.8
LAS 7	4.4
LAS 8	5.7
LAS 9	7.3
LAS 10	6.0
LAS 11	8.0
LAS 12	9.0

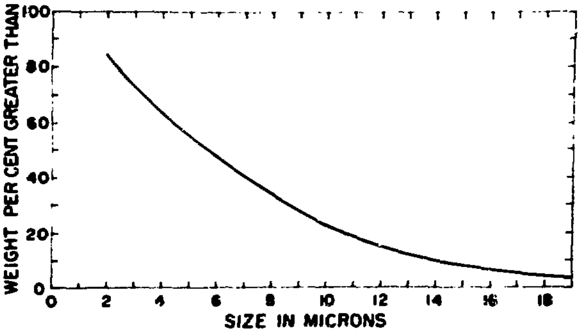


Fig. 20.

Particle-size distribution of typical ball-milled $^{238}PuO_2^{16}$ powder, Lot LAS 8.

Development of 30-W fabrication procedures constituted the first phase of the program. Six 30-W sources were prepared. Small 1- to 2-W control pellets were pressed and sintered with the 30-W sources to provide evaluation samples. Of the six sources made, only two, 30-3 and 30-6, were fabricated as per the flowsheet shown in Figs. 12 and 13. The preparation of 30-1 and 30-5 required recycling through the ceramic fabrication procedure before we could obtain an integral green pellet, free of laminations. In both cases heating of the powder at 1600°C and remilling for 18 h were required. The powder for 30-2 was prepared by the isotopic exchange of PuO_2^{nat} with H_2O^{16}. The natural oxide feed was prepared by dissolving Savannah River oxide in HNO_3-HF, precipitating the peroxide, dissolving the peroxide in HNO_3, precipitating the oxalate, and igniting the oxalate to the oxide. Pellet 30-4 was prepared by reprocessing 30-2 through the ceramic fabrication flowsheet. Thus, the only 30-W pellets typical of the standard process flowsheets are 30-3 and 30-6. The chemical purities of the two pellets and process intermediates are given in Table XXVI. The chemical purity of all the 30-W cylinders is given in Table BVIII.

The sum of the detectable impurities for 30-3 and 30-6, excluding neptunium, uranium, and americium is 517 and 508 ppm, respectively. As already noted, few, if any, impurities are introduced in the preparation of $^{238}PuO_2^{16}$ powder. The present ball-milling and screening operations introduce trace amounts of iron, chromium, sodium, silicon, aluminum, nickel, copper, and tin. The Carbowax and trichlorethylene solvent used are volatilized in the subsequent sintering operation, but the pellet retains measureable amounts of carbon ($\sim$200 ppm). Pellet sintering is done in an atmosphere of ($Ar + H_2O^{16}$) and is preceded by oxygen isotopic exchange at 800 °C (see Appendix A).

The properties of 30-3 and 30-6 are given in Table XXVII. The properties of all six 30-W sources are given in Table BIX, and the plutonium isotopic analysis is given in Table BX. A 30-W cylinder is ~0.85 in. in diameter by ~0.80 in. tall. A typical fuel cylinder is shown in Fig. 21. Source 30-6 was prepared for in-phantom dosimetry studies at Battelle, PNL. The ^{238}Pu enrichment was 86.90 wt.%, and the ^{236}Pu concentration was 0.25 ppm. The dose rates 10 cm from this source, which was encapsulated in 0.064-in. Ta-10 W and 0.025-in. Hastelloy C-276, were 0.35 mRem/h - g ^{238}Pu for neutrons and 0.14 mR/h - g ^{238}Pu for photons. The dose rates for 30-3, 79.82 wt.% ^{238}Pu, were comparable. The neutron emission rates of the 1-W control pellets (H/D = 1) for 30-3 and 30-6 were 3470 and 3312 n/sec-g of ^{238}Pu, respectively. The neutron emission rates for the 30-W cylinders were 3608 and 3519 n/sec - g of ^{238}Pu. For comparison, the neutron emission rate for the 30-W Pu-3 at.%-Ga source above was 3150 n/sec-g of ^{238}Pu. Thus, the neutron emission rates of the 30-W sources were about 400 n/sec - g of ^{238}Pu higher than those of an equivalent Pu-Ga source.

b. Fifty-W Cylinders. Five 50-W cylinders, 50-1 through 50-5, were prepared. The first, 50-1, was prepared to establish the ceramic fabrication procedures for 50-W cylinders. In this preparation, the $^{238}PuO_2^{16}$ powder was made by oxygen isotopic exchange of Savannah River $^{238}PuO_2^{nat}$ with H_2O^{16}. The next three sources, 50-2, 50-3, and 50-4, were prepared from electrorefined metal as per the flowsheets shown in Figs. 12 and 13. The last, 50-5, was made by recycling 50-2 through ceramic fabrication, i.e. ball-milling, screening, pressing, and sintering.

The chemical purity of the control pellets for 50-2, 50-3, and 50-4 is given in Table XXVIII (the chemical purities of all five sources are given in Table BXI). The purity of the 50-W sources is comparable to that of the 30-W sources. The sum of the total detectable impurities for 50-2, 50-3, and 50-4, excluding carbon, neptunium, uranium, and americium is 895, 400, and 236 ppm, respectively. As in the 30-W preparations, most impurities were introduced by the milling and screening. To illustrate this point, the chemical purities of the electrorefined metal feed, $^{238}PuO_2^{16}$ powder, milled powder, and sintered control pellet for preparation 50-3 are compared in Table XXIX. Similar data for all the preparations are given in Table BVI. The sums of the total detectable impurities, excluding carbon, neptunium, uranium, and americium, were 65, 49, 717, and 399 ppm for the metal, $^{238}PuO_2^{16}$ powder, milled powder, and sintered control pellet, respectively.

The ^{18}O concentration in $^{238}PuO_2^{16}$ powders, milled powders, and sintered pellets for 50-2, 50-3, and 50-4 is given in Table XXX. (Data for all the 50-W cylinders are given in Table BXII.) The ^{18}O concentrations in the $^{238}PuO_2^{16}$ powders and sintered pellets are essentially equal to the ^{18}O concentration of the H_2O^{16} used in the preparation.

The neutron emission rates of the 50-2, 50-3, and 50-4 control pellets are compared to those of the metal and oxide powders in Table XXXI. Increases of 216, 213, and 221 n/sec-g of ^{238}Pu were obtained in converting metal to oxide powder. Approximately half of this increase is due to ^{17}O and ^{18}O. Relatively large increases were obtained during ball-milling, mostly due to ^{18}O contamination of the powder during the 18-h ball-milling period. The ^{18}O is removed during the oxygen exchange cycle which precedes sintering, and the neutron emission rates of the sintered pellets compare favorably to those of the starting $^{238}PuO_2^{16}$ powders. Neutron multiplication resulting from the pressing of 50-2, 3, and 4 $^{238}PuO_2^{16}$ powders into 1-W control pellets should yield pellets with neutron emission rates of 3366, 3405, and 3357 n/sec-g of ^{238}Pu. The

TABLE XXVI

CHEMICAL PURITY OF 30-W $^{238}PuO_2^{16}$ CYLINDERS AND PROCESS INTERMEDIATES, (30 - 3 AND 30 - 6)

	ppm of Impurity							
	30 - 3 Cylinder				30 - 6 Cylinder			
Element	**Metal**	**Powder**	**Milled Powder**	**Control Pellet**	**Metal**	**Powder**	**Milled Powder**	**Control Pellet**
Li	< 0.005	< 0.01	< 0.01	0.2	0.008	0.05	< 0.2	< 0.2
Be	< 0.001	< 0.002	< 0.002	< 0.2	< 0.002	< 0.2	< 0.2	< 0.2
B	< 1	1	2	2	< 1	1	1	< 1
C	–	–	–	120	–	15	180	140
Na	2	6	36	22	< 1	11	50	40
Mg	2	40	25	12	5	< 2	8	8
Al	1	8	16	40	10	20	25	40
Si	< 5	< 5	35	60	5	10	40	15
K	< 1	2	4	< 1	4	2	4	2
Ca	< 2	15	12	< 6	20	10	30	8
Ti	< 0.2	2	6	19	< 0.2	2	1	< 0.4
V	< 0.5	< 1	< 1	< 1	< 0.5	< 1	< 1	< 1
Cr	< 0.5	2	40	56	1	1	55	60
Mn	< 0.1	5	5	8	0.3	0.4	3	4
Fe	20	75	220	130	70	20	245	150
Co	< 0.5	1	< 1	< 1	< 0.5	< 1	< 1	< 1
Ni	2	7	40	25	4	7	20	8
Cu	10	20	22	10	2	4	100	10
Zn	< 10	35	25	< 10	< 10	< 10	50	< 10
Rb	< 0.5	< 1	< 1	< 1	< 0.5	< 1	< 1	< 1
Sr	< 0.1	< 0.2	< 0.2	< 0.2	< 0.1	< 0.2	< 0.2	< 0.2
Y	< 0.1	< 0.2	< 0.2	< 0.2	3	< 0.2	< 0.2	< 0.2
Zr	0.5	< 0.2	< 0.2	< 0.2	0.2	0.8	8	4
Mo	< 0.5	< 1	10	2	< 0.5	1	20	18
Cd	< 0.5	< 1	< 1	< 1	< 0.5	< 1	< 1	< 1
Sn	< 0.5	3	15	8	< 0.5	< 5	50	< 5
Cs	< 2	< 4	< 4	< 4	< 2	< 4	< 4	< 4
Ba	< 0.1	< 0.2	1	< 0.4	0.3	3	3	1
La	< 1	< 2	< 2	< 2	< 1	< 2	< 2	< 2
Hf	< 0.5	< 1	< 1	< 1	< 0.5	< 1	< 1	< 1
Re	< 0.5	–	< 1	< 1	< 0.5	< 1	< 1	< 1
Pb	< 0.5	< 1	2	3	1	2	< 1	< 1
Bi	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Np	550	564	494	500	250	210	255	270

TABLE XXVII

PROPERTIES OF 30-W $^{238}PuO_2^{16}$ CYLINDERS, (30 - 3 AND 30 - 6)

	30-3	30-6
Wt. of fuel pellet (g)	74.76	74.69
Wt of Pu, (g)	65.9	65.4
Oxygen/Pu ratio	—	2.01
Pu isotopic compositon		
Wt. % $^{238}Pu^{a}$	79.82	86.90
ppm $^{236}Pu^{b}$	0.70	0.25
Dimensions		
Diam. (in.)	0.846	0.858
Height (in.)	0.803	0.808
Density (g PuO_2/cm^3)	10.11	9.77
Power (w)	29.8	32.2
Power density (w/cm^3)	4.03	4.20
Neutron emission rate		
n/sec-g Pu	2880.	3060.
n/sec-g ^{238}Pu	3608.	3520.
Dose rates at 10 cm		
Neutron (mRem/h-g ^{238}Pu)	0.36	0.35
Gamma (mR/h-g ^{238}Pu)^{c}	0.14	0.17

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[a] g ^{238}Pu/100 g Pu.

[b] g ^{236}Pu/10^6 g Pu.

[c] Measured in air through 0.064-in. Ta-10W and 0.025-in. Hastelloy C-276.

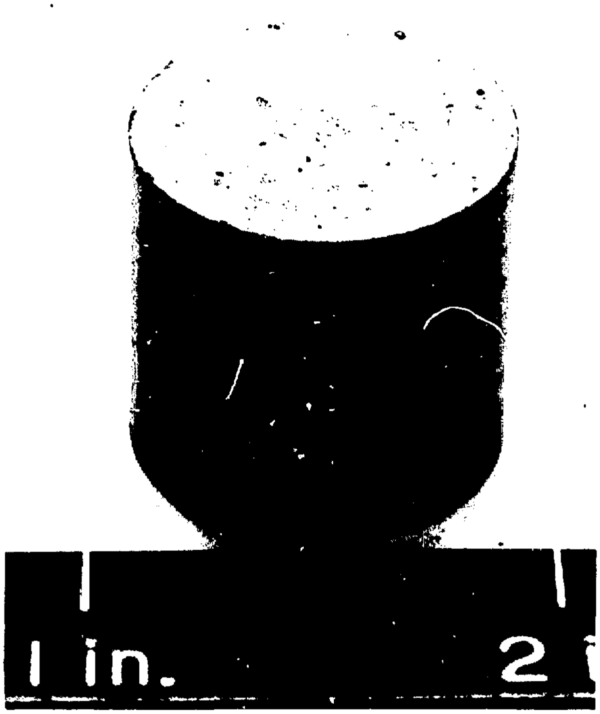


Fig. 21.
Typical 30-W $^{238}PuO_2^{16}$ cylinder, 30-3.

average measured rates were 3380, 3350, and 3334. Thus, the neutron emission rates of the 1-W sintered pellets compare favorably to those of both the $^{238}PuO_2^{16}$ powder and electrorefined metal. The neutron emission rates of the control pellets and process intermediates for all the 50-W preparations are given in Table BXIII.

The properties of 50-W $^{238}PuO_2^{16}$ cylinders 50-2, 50-3, and 50-4 are given in Table XXXII. The properties of all the 50-W sources are given in Table BXIV. Complete plutonium isotopic compositions are given in Table BXV. The 50-W cylinders are 1.00 in. diam. by 1.00 in. tall and have a power density of 3.8 W/cm^3. The physical appearance of a typical 50-W source is shown in Fig. 22. There is no obvious "hour-glassing" or shrinking of the pellet in the middle. The diameter varies less than 0.010 in. over the length of the cylinder. The measured neutron emission rates of 50-2, 50-3, and 50-4 were 3374, 3488, and 3466 n/sec-g of ^{238}Pu, respectively. These rates are compared to the rates of the control pellets in Table XXXIII. Calculations indicate that the neutron-multiplication factor should be 1.06 times greater in the larger sources. Thus, the ratio of neutron emission rates (rate for large source/rate for small source) should be 1.06. The measured ratios were 1.00, 1.04, and 1.04 for 50-2, 50-3, and 50-4, respectively. Thus the measured rates for the 50-W pellets compare favorably to those of the small control pellets and the starting $^{238}PuO_2^{16}$ powder. The measured gamma-dose rates of the 50-W sources were 0.17, 0.14, and 0.16 mR/h-g ^{238}Pu, which is in fair agreement with calculations from measurements of elemental plutonium sources. Neutron emission rates for all the 50-W cylinders and 1-W control pellets are compared in Table BXVI. Note that the neutron emission rates of 50-2, 50-3, and 50-4 are only 24, 138, and 116 n/sec-g of ^{238}Pu higher than the desired specification for medical-grade $^{238}PuO_2^{16}$, which is $<$ 3350 n/sec-g of ^{238}Pu for a 50-W cylinder.

c. Oxygen Isotopic Exchange of Sintered $^{238}PuO_2^{16}$ Pellets. As shown above, $^{238}PuO_2^{16}$ powders undergo oxygen isotopic exchange when they are milled in argon atmospheres containing approximately 200 ppm of O_2.

TABLE XXVIII

CHEMICAL PURITY OF CONTROL PELLETS FOR 50-W SOURCES, (50-2, 50-3, and 50-4)

	Impurity (ppm)		
Element	50-2	50-3	50-4
Li	< 0.2	< 0.2	< 0.2
Be	< 0.2	< 0.2	< 0.2
B	2	1	3
C	100	–	–
Na	60	50	20
Mg	50	10	20
Al	60	20	25
Si	180	100	< 25
K	1	2	< 1
Ca	50	14	< 6
Ti	< 0.4	4	< 0.4
V	< 1	< 1	< 1
Cr	75	50	44
Mn	9	4	2
Fe	270	120	40
Co	< 1	< 1	< 1
Ni	10	10	25
Cu	50	< 0.2	< 10
Zn	20	< 10	< 20
Rb	< 1	< 1	< 1
Sr	< 0.2	< 0.2	< 0.2
Y	< 0.2	< 0.2	< 0.2
Zr	3	4	1
Mo	40	10	48
Cd	< 1	< 1	< 1
Sn	5	< 5	6
Cs	< 4	< 4	< 4
Ba	10	0.2	< 0.2
La	< 2	< 2	< 1
Hf	< 1	< 1	< 1
Re	< 1	< 1	< 1
Pb	< 1	< 1	2
Bi	< 1	< 1	–
Np	240	307	835

TABLE XXIX

CHEMICAL PURITY OF ELECTROREFINED METAL, $^{238}PuO_2^{16}$ POWDER, MILLED POWDER, AND SINTERED CONTROL PELLET, 50-3

	Impurity (ppm)			
Element	Metal	PuO_2^{16} Powder	Milled Powder	Control Pellet
Li	0.1	< 0.2	0.2	< 0.2
Be	< 0.1	< 0.2	< 0.2	< 0.2
B	2	< 1	< 1	< 1
C	–	70	85	–
Na	< 1	1	8	50
Mg	< 1	2	16	10
Al	< 10	< 10	< 10	20
Si	< 5	15	20	100
K	< 3	1	3	2
Ca	< 3	< 10	< 10	14
Ti	4	3	4	4
V	< 0.5	< 1	< 1	< 1
Cr	< 0.5	< 1	45	50
Mn	1	1	3	4
Fe	32	22	180	120
Co	< 0.5	< 1	< 1	< 1
Ni	8	< 1	24	10
Cu	5	2	200	< 0.2
Zn	5	< 10	20	< 10
Rb	< 0.5	< 1	< 1	< 1
Sr	< 0.1	< 0.2	< 0.2	< 0.2
Y	< 0.1	< 0.2	< 0.2	< 0.2
Zr	< 0.1	< 0.2	2	4
Mo	< 0.5	< 0.2	< 1	10
Cd	< 0.5	< 1	< 1	< 1
Sn	6	< 1	100	< 5
Cs	< 2	< 4	< 4	< 4
Ba	2	< 0.2	< 0.2	0.2
La	< 1	< 2	< 2	< 2
Hf	< 0.5	< 1	< 1	< 1
Re	< 0.5	< 1	< 1	< 1
Pb	< 0.5	2	7	< 1
Bi	< 1	< 1	< 1	< 1
Np	300	285	–	307

TABLE XXX

OXYGEN-18 CONCENTRATION OF $^{238}PuO_2^{16}$ POWDERS, MILLED POWDERS, AND SINTERED CONTROL PELLETS, (50-2, 50-3, AND 50-4)

	ppm ^{18}O, (g $^{18}O/10^6$ g O)		
Preparation	**Powder**	**Milled Powder**	**Control Pellets**
50-2	24	75	18
50-3	10	160	10
50-4	10	65	10

However, there is no detectable exchange when 1- to 50-W sintered pellets are handled in the same atmosphere. The exchange experiments described below demonstrate that cylinders as large as 50 W can be handled in air-filled glove boxes saturated with water vapor for short periods without any observable oxygen exchange.

d. Oxygen Isotopic Exchange of 1-W Pellets. The 24-h exposure of sintered 1-W pellets of $^{238}PuO_2^{16}$ at 425 °C to atmospheres of argon plus 150 ppm of O_2, dry air, and air saturated with H_2O did not increase the neutron emission rate. To establish the temperature at which significant exchange takes place in 24 h, we exposed 1-W pellets to air saturated with water vapor at temperatures of 425, 600, 800, 900, and 975 °C. The results, summarized in Table XXXIV, show that no exchange, as evidenced by the neutron emission rates, took place at 425 and 600 °C. Exchange did occur at 800 °C, and it was more pronounced as the temperature was increased.

TABLE XXXI

NEUTRON EMISSION RATES OF $^{238}PuO_2^{16}$ POWDER, MILLED POWDER, AND SINTERED CONTROL PELLETS (50-2, 50-3, AND 50-4)

	n/sec-g ^{238}Pu			
Preparation	**Metal**	**$^{238}PuO_2^{16}$ Powder**	**Milled Powder**	**Control Pellets**
50-2	3084	3300	4000	3387 3374
50-3	3125	3338	4114	3350 —
50-4	3070	3291	3504	3341 3328

Oxygen isotopic exchange studies in air at 1002 °C are summarized in Table XXXV.[29] Three $^{238}PuO_2^{16}$ pellets, 90% of theoretical density, height-to-diameter ratio of 1.00, were used in this study. The fraction of oxygen isotopic exchange was calculated from the observed increase in neutron counting rate.

$$F = \frac{N_t - N_0}{N_\infty - N_0} ,$$

where F = fraction of oxygen isotopic exchange,

$N_\infty - N_0$ = 12, 140 n/sec-g of ^{238}Pu,

N_t = neutron counting rate at time t,

N_0 = neutron counting rate at time 0,

N_∞ = neutron counting rate when PuO_2 is of natural oxygen isotopic composition.

A plot of F vs $t^{1/2}$ is approximately linear, and can be used to obtain a diffusion coefficient, D, for oxygen in PuO_2. An evaluation of D based on the exact solution to the diffusion equation for the 1-W pellets gives $D_{(PuO_2)} = 4 \times 10^{-10}$ cm^2 sec^{-1}.[29] For comparison, the oxygen self-diffusion coefficient in UO_2 is 2.2×10^{-10} cm^2 sec^{-1}.[30]

e. Oxygen Isotopic Exchange of 50-W Cylinders. Oxygen isotopic exchange experiments were done with a 50-W cylinder at 480 and 580 °C. (The surface temperature of a 50-W cylinder resting on a ceramic heat insulator in an air-filled glove box is ~480 °C.) The results at 480 °C are summarized in Table XXXVI. A $^{238}PuO_2^{16}$ cylinder, 50-1, was exposed to atmospheres of Ar, N_2, O_2, dry air, and air saturated with water, for periods of 18.4 to 168 h. The magnitude of the exchange was established by monitoring the relative neutron emission rate of the cylinder, in situ, by means of a shielded probe located outside the glove box. The data show no significant exchange.

The results at 580 °C are summarized in Table XXXVII. Again, cylinder 50-1 was exposed to atmospheres of (Ar-N_2-O_2), dry air, and air saturated with water vapor. We examined both static and flow atmospheres. The counting rate increased less than 2% in all atmospheres. Thus, within the limits of experimental measurement, there was no significant oxygen exchange.

TABLE XXXII

PROPERTIES OF 50-W $^{238}PuO_2^{16}$ CYLINDERS (80 at.%^{238}Pu)

	50-2	50-3	50-4
Wt. of fuel pellet (g)	125.08	125.27	124.68
Wt of Pu (g)	110.	110.	110.
Oxygen/Pu ratio	—	—	1.99
Pu isotopic composition			
Wt % ^{238}Pu[a]	79.72	79.98	79.62
ppm ^{236}Pu[b]	0.76	0.75	0.58
Dimensions			
Diam (in.)	1.006	0.997	1.004
Height (in.)	0.996	1.003	1.013
Density (g PuO_2/cm^3)	9.65	9.75	9.49
Power (W)	49.7	49.8	49.6
Power density (W/cm^3)	3.83	3.88	3.78
Neutron emission rate			
n/sec-g Pu	2690	2790	2760
n/sec-g ^{238}Pu	3374	3488	3466
Dose rates at 10 cm[c]			
Neutron (mRem/h-g ^{238}Pu)	0.35	0.35	0.35
Gamma (mR/h-g ^{238}Pu)	0.17	0.14	0.16

[a] g ^{238}Pu/100g Pu.

[b] g ^{236}Pu/10^6g Pu.

[c] Measured in air through 0.020-in. tantalum and 0.020-in. stainless steel.

E. $^{238}PuN^{15}$ Powder.

Chemically pure ^{238}PuN containing the natural isotopic concentrations of ^{14}N and ^{15}N has a neutron dose rate equal to that of electrorefined metal.[31] The gamma dose rate, however, is higher than that of the metal by a factor of 2.5.[31] This increased gamma dose rate is caused by the reaction $^{14}N\ (\alpha,\rho)\ ^{17}O$. To eliminate this reaction, we prepared $^{238}PuN^{15}$ by reacting electrorefined ^{238}Pu metal (E.R. 238 - 100 - 4) with $^{15}N_2$ gas (99.7% ^{15}N, 0.3% ^{14}N).

1. Chemical Purity. The chemical purity of a typical $^{238}PuN^{15}$ powder is compared to that of its electrorefined metal feed in Table XXXVIII. The purity of both materials is essentially the same.

2. Radiation Properties. The radiation properties of a typical lot of $^{238}PuN^{15}$ are compared to those of its electrorefined metal feed in Table XXXIX. The neutron-emission rates are essentially identical. The small difference in the gamma dose rate is due to decreased self-shielding in the nitride.

3. Gamma Spectrum. The gamma spectrum of $^{238}PuN^{15}$ is essentially the same as that of electrorefined metal, except for a small peak at 871 keV due to 0.3% ^{14}N in the ^{15}N. The $^{238}PuN^{15}$ spectrum is compared to $^{238}PuN^{nat}$ in Fig. 23. The 871-keV gamma ray comes from the first excited state of ^{17}O produced by alpha bombardment of ^{14}N. It has an absolute intensity of 3.0×10^5 photons/sec-g ^{238}Pu. These photons are responsible for the high gamma dose rate of $^{238}PuN^{nat}$.

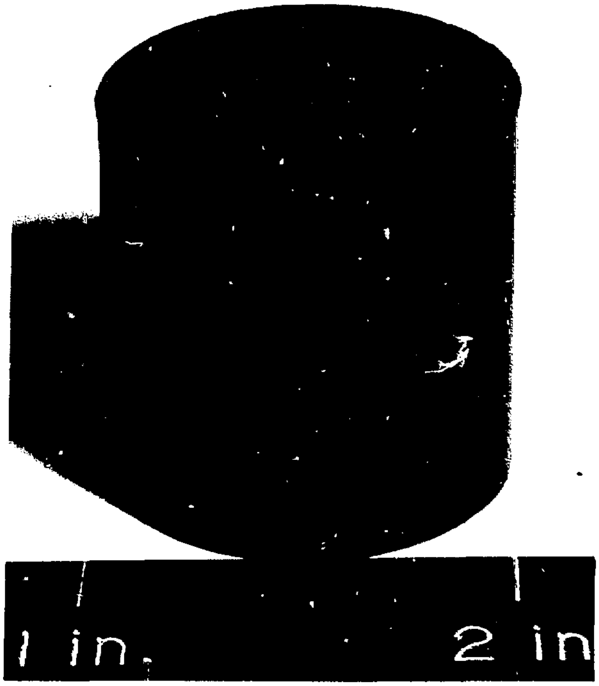


Fig. 22.
Typical 50-W $^{238}PuO_2^{16}$ cylinder, 50-2.

VI. RADIATION PROPERTIES OF PuO_2^{nat}, PuF_4, AND BOMB-REDUCED METAL

A. $^{238}PuO_2^{nat}$.

1. As-Received Oxide Powders. The neutron emission rates of the as-received oxide powders evaluated in this program are given in Table XL. The measured rates of the oxide powders (LA 5 - LA 11) received from Mound Laboratory varied from 51,400 to 129,000 n/sec-g of ^{238}Pu. Their chemical purities are given in Table BXVII. Fluorine, which was detected as an impurity in all the powders by high-resolution gamma spectrometry, was primarily responsible for the high neutron emission rate. Thus, removal of most of the fluorine from Lot LA-6 by heating in air at 900 °C (see below for details) decreased the neutron-emission rate from 59,000 to 16,300 n/sec-g of ^{238}Pu.

TABLE XXXIII

NEUTRON EMISSION RATES OF 1-W CONTROL PELLETS AND 50-W CYLINDERS

	n/sec-g ^{238}Pu	
Source No.	Control Pellets	50-W Cylinders
50-2	3387	3374
	3374	
50-3	3350	3488
	—	
50-4	3341	3466
	3328	

TABLE XXXIV

NEUTRON EMISSION RATES OF SINTERED 1-W $^{238}PuO_2^{16}$ PELLETS EXPOSED TO AIR AND WATER VAPOR

		Neutron Emission Rate (n/sec-g ^{238}Pu)	
Temp (°C)	Pellet No.	Start	After 24 h
425	276	3415	3400
600	274	3425	3440
	275	3450	3425
800	276	3400	3465
	277	3490	3600
900	272	3375	3640
	273	3450	3925
975	274	3440	4165
	275	3425	4000

The neutron emission rates of oxide powders (HBO 907534 - HBO 907551) received from the Savannah River Laboratory are also given in Table XL. They are considerably lower than those reported above, averaging 16,300 n/sec-g of ^{238}Pu. The measured chemical purities are given in Table BXVIII. The amounts of light-element impurities in these powders are quite low, but the neutron emission rates are ~1,000 n/sec-g of ^{238}Pu higher than those measured for PuO_2^{nat} prepared from electrorefined metal, see below. The gamma spectrum of Savannah River oxide Lot HBO 907551 is given in Fig. 24. Nitrogen, fluorine, and sodium impurities can be identified in this spectrum.

The neutron emission rates of oxide powders (HAS 1 - HAS 9,) received from Atlantic Richfield Hanford Co. are also listed in Table XL. Their chemical purities are given in Table BXIX. The ^{238}Pu isotopic composition in these oxides ranged from 86 to 92 at.%. The neutron emission rates were somewhat higher than those of the Savannah River oxides, averaging ~18,000 n/sec-g of ^{238}Pu, or about 3,000 n/sec-g of ^{238}Pu higher than those of LASL-produced natural oxides.

TABLE XXXV

OXYGEN ISOTOPIC EXCHANGE IN AIR, 1-W PELLETS, 1002 °C

	Pellet Number					
	283		284		285	
t, (h)	n/sec-g ^{238}Pu	F[a]	n/sec-g ^{238}Pu	F[a]	n/sec-g ^{238}Pu	F[a]
0	3360	0.0	3370	0.0	3370	0.0
19.7	4090	0.060	4100	0.060	3955	0.048
44.0	4485	0.093	4495	0.093	4305	0.077
70.6	4645	0.106	4695	0.109	4540	0.096
140.0	5085	0.142	5170	0.148	4950	0.130
280.1	5710	0.194	5830	0.203	5585	0.182
703.8	6910	0.292	7005	0.299	6675	0.272

[a] F = fraction of oxygen isotopic exchange

$$= \frac{N(t) - N(^{\circ})}{12{,}140} .$$

TABLE XXXVI

COUNTING RATES OF 50-W $^{238}PuO_2^{16}$ PELLET 50-1, AFTER EXPOSURE TO NATURAL OXYGEN AT 480°C

Atmosphere (vol %)	Exposure Time (h)	Counting Rate (c/min)	
		Initial	Final
99.3 Ar 0.55 N_2 0.17 O_2	18.4	1208.2	1209.1
96.8 Ar 2.4 N_2 0.85 O_2	45.1	1210.1	1214.5
92.3 Ar 5.6 N_2 2.1 O_2	29.5	1213.4	1217.2
Dry air	67.9	1230.5	1231.2
Air sat'd with H_2O	168	1237.0	1240.4

2. LASL Oxide Powders. The neutron emission rates of LASL-produced natural oxide powders are given in Table XLI. The first oxide was prepared by simply heating as-received oxide from lot LA-6 to constant weight and constant neutron activity at 900 °C in air. The neutron emission rate was reduced from 59,000 to 16,300 n/sec-g of ^{238}Pu, mainly through elimination of fluorine by ignition. The second oxide was prepared by igniting a peroxide precipitate obtained from Lot LA-8. The third was prepared by reacting high-purity PuN with air. The nitride was prepared from electrorefined metal. The last oxide listed was prepared by reacting H_2O^{nat} with electrorefined metal using the standard flowsheet shown in Fig. 11, except for the substitution of H_2O^{nat} for H_2O^{16}. The last two oxides are of comparable quality, having neutron emission rates of 15,200 and 15,300 n/sec-g of ^{238}Pu. These data can be used to calculate the neutron contribution from ^{17}O and ^{18}O in natural oxides. The calculation for oxide prepared from electrorefined metal is shown in Table XLII. The ^{238}Pu spontaneous fission value was determined above. The light-element impurity contribution was calculated from the chemical purity analysis and the thick-target yield for (α,n) reactions, Table XII. The neutron multiplication contribution was calculated from the known sample geometry. These calculations give a value of 1.18 x 10^4 n/sec-g of ^{238}Pu for the (α,n) contribution from ^{17}O and ^{18}O in $^{238}PuO_2{}^{nat}$.

TABLE XXXVII

COUNTING RATES OF 50-W $^{238}PuO_2^{16}$ CYLINDER 50-1, AFTER EXPOSURE TO NATURAL OXYGEN AT 580 °C

Atmosphere (vol %)	Manner of Exposure	Exposure Time (h)	Counting Rate (c/min)		% Increase in Counting Rate
			Initial	Final	
99.3 Ar 0.55 N_2 0.17 O_2	Flow	24.5	1254.0	1258.6	0.37
96.8 Ar 2.4 N_2 0.85 O_2	Flow	43.1	1258.6	1267.0	0.67
92.3 Ar 5.6 N_2 2.1 O_2	Static	92.9	1267.0	1271.5	0.36
Same as preceding experiment	Flow	25.9	1271.5	1285.7	1.1
Dry air	Static	96.3	1272.1	1286.8	1.1
Air sat'd with H_2O	Flow	76.5	1286.8	1307.3	1.6

B. $^{238}PuF_4$.

$^{238}PuF_4$ is an intermediate step in the preparation of bomb-reduced metal. Because of its high neutron emission rate, it must be handled very carefully. Dose rates at 10 cm measured through 0.030-in.-thick tantalum are 190 mRem/h-g ^{238}Pu for neutron and 10 mR/h-g ^{238}Pu for gamma. The neutron emission rate is 1.9 x 10^6 n/sec-g of ^{238}Pu.

C. Bomb-Reduced Metal.

The neutron emission rates for 15 castings of bomb-reduced metal are given in Table XLIII. The rates varied from 3295 to 5495 n/sec-g of ^{238}Pu, and averaged 3840 n/sec-g of ^{238}Pu, or 670 n/sec-g higher than the average for electrorefined metal samples of comparable size.

D. Recovery of Process Wastes.

The process wastes generated using the flowsheets shown in Figs. 2, 3, 12, and 13 are recycled, on an available-effort basis, to produce chemically pure ^{238}Pu metal. The peroxide filtrates and bomb-reduction residues are not recycled. All residues from the electrorefining process (Fig. 3), the $^{238}PuO_2^{16}$ powder preparation (Fig. 12), and the ceramic fabrication process (Fig. 13) can be recycled by converting the residues to PuO_2 which serves as feed material to the conversion process, Fig. 2.

A total of 747 g of ^{238}Pu metal has been recovered from process and R&D wasts. (See Table BI, recycle lots 1 - 9, for details of the metal conversion.) Electrorefining of this metal produces chemically pure ^{238}Pu comparable to that produced from as-received oxides.

E. Aging and Fuel Recycling.

1. Growth of ^{236}Pu Daughters in ^{238}Pu. The absolute intensities of major ^{236}Pu-daughter gamma activities, which contribute significantly to the total ^{238}Pu gamma dose, were measured following their growth in two ^{238}Pu samples. Each sample had been analyzed previously for ^{236}Pu content by direct alpha-pulse analysis and then purified by ion-exchange resin separation from uranium, thorium, and other ^{236}Pu daughters and foreign activities. After ^{236}Pu daughters had grown for slightly more than a year, we measured the gamma activities using a Ge(Li) detector calibrated with known amounts of ^{228}Th in equilibrium with its daughters under counting conditions similar to those used for counting the samples.

The measured gamma-ray intensities were compared to those expected as a result of growth from the original ^{236}Pu content (Table XLIV). The expected and measured

TABLE XXXVIII

CHEMICAL PURITY OF $^{238}PuN^{15}$ AND ELECTROREFINED METAL FEED

Element	Impurity (ppm) Metal	Impurity (ppm) $^{238}PuN^{15}$
Li	< 0.005	0.15
Be	< 0.001	< 0.002
B	0.5	1
Na	< 5	8
Mg	2	< 2
Al	2	6
Si	5	15
K	< 1	6
Ca	< 6	12
Ti	0.5	3
V	< 0.5	< 1
Cr	< 0.5	3
Mn	1	0.4
Fe	30	44
Co	< 0.5	< 1
Ni	0.5	4
Cu	3	5
Zn	< 10	< 10
Rb	< 0.5	< 1
Sr	< 0.1	< 0.2
Y	< 0.1	< 0.2
Zr	< 0.5	1
Mo	1	< 1
Cd	< 0.5	< 1
Sn	< 1	1
Cs	< 2	< 4
Ba	0.6	< 0.2
La	< 1	< 2
Hf	< 0.5	< 1
Re	< 0.5	< 1
Pb	< 0.5	4
Bi	< 0.5	< 0.5

TABLE XXXIX

RADIATION PROPERTIES OF $^{238}PuN^{15}$ POWDER AND ELECTROREFINED METAL FEED

	Metal	$^{233}PuN^{15}$
Neutron emission		
Rate, n/sec-gPu	2495	2505
n/sec-g ^{238}Pu	3110	3123
Dose rates at 10 cm in air through 0.030-in. Ta		
Neutron (mRem/h-g ^{238}Pu) (from emission rate)	0.31	0.31
Gamma (mR/h-g ^{238}Pu) (TLD dosimetry)	0.25	0.27

gamma-ray intensities agreed well enough considering the uncertainties introduced by counting errors in measuring the ^{236}Pu content. The agreement showed that literature values for the ^{236}Pu daughter half-life[32] and branching ratios[33] were reliable for calculating growth of ^{236}Pu daughter activities. As the method of calibrating the Ge(Li) detector was similar to that used when measuring absolute intensities of ^{238}Pu gamma rays, the present agreement in results gives added credibility to the reported intensities of the ^{238}Pu gamma rays.

2. Permissable Concentrations of ^{236}Pu and ^{232}U in ^{238}Pu. Proposed specifications for medical-grade ^{238}Pu include the provision that the ^{232}U and ^{236}Pu contents in freshly purified ^{238}Pu shall not exceed concentrations that will produce a total gamma dose over a 10-yr period greater than that produced by 0.300 ppm of ^{236}Pu alone. Concentrations of ^{232}U greater than zero are acceptable if the ^{236}Pu concentration is correspondingly less than 0.300 ppm. The permissable combinations of ^{236}Pu and ^{232}U are given in Table XLV. They can be conveniently represented by

$$^{236}Pu\ (ppm) + 1.79\ ^{232}U\ (ppm) \leqslant 0.30 \quad .$$

This equation is based on the fact that 0.56 ppm of ^{232}U will produce the same total gamma-ray dose in 10 yr as will 1.000 ppm of ^{236}Pu. This ratio is good only for a 10-yr growth period. Different ratios will apply for other growth periods because the rates of daughter growth for ^{236}Pu and ^{232}U differ.

3. Fuel Recycling Considerations. Complete removal of 72-yr ^{232}U and 1.9-yr ^{228}Th is especially desirable because they are the precursors of the short-lived, gamma-emitting daughters, ^{212}Pb, ^{212}Bi, and ^{208}Tl.

Calculations show that the most significant impurity is ^{232}U, and that in any effective purification cycle, 90%, or more, of it must be removed. The presence of ^{228}Th in

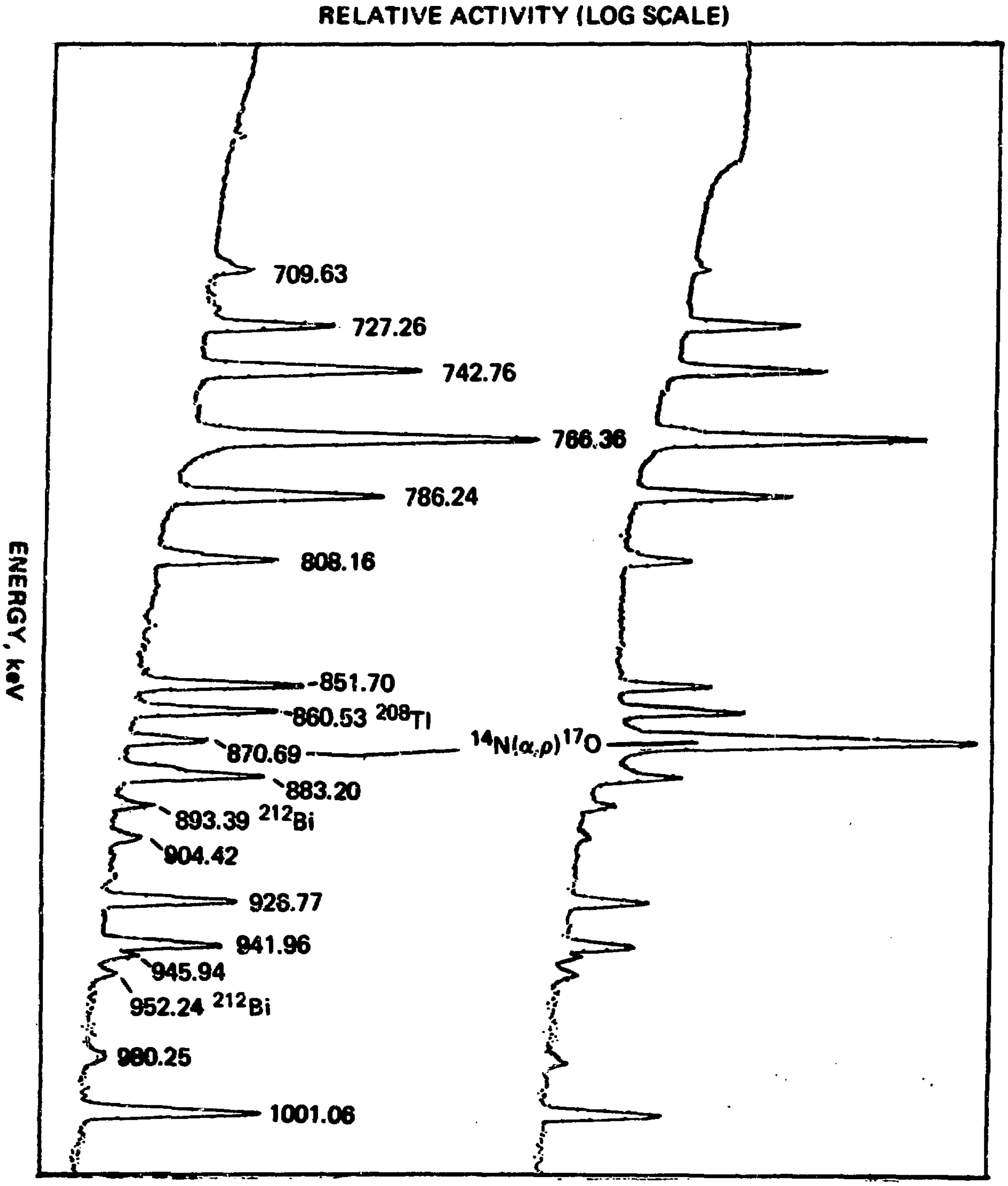


Fig. 23.
Gamma-Ray spectra of ^{238}PuN^{15} and ^{238}PuN^{nat}.

TABLE XL

NEUTRON EMISSION RATES OF AS-RECEIVED $^{238}PuO_2^{nat}$

Lot	Neutron Emission Rate (n/sec-g ^{238}Pu)
LA 5[a]	51,400
LA 6	59,000
LA 9	55,300
LA 10	53,000
LA 11	129,000
HBO 907534[b]	15,900
HBO 907535	15,400
HBO 907538	16,500
HBO 907539	16,300
HBO 907545	16,400
HBO 907546	16,500
HBO 907547	16,600
HBO 907551	16,800
HBO 91129	16,550
HAS 1[c]	20,300
HAS 2	17,400
HAS 3	17,500
HAS 6	16,900
HAS 7	16,700
HAS 8	18,800
HAS 9	19,400

[a] PuO_2, nominal 80 at. % ^{238}Pu, from Mound Laboratory.

[b] $^{238}PuO_2$ nominal 80 at. % ^{238}Pu, from Savannah River Laboratory.

[c] $^{238}PuO_2$, nominal 90 at. % ^{238}Pu, from Atlantic Richfield Hanford Co.

purified materials is less critical. Figure 25 illustrates worst and best cases for ^{228}Th cleanup. The rapidly ascending curve at the left illustrates the increase in gamma dose rate of initially pure ^{238}Pu. The material is allowed to age 6 yr (2.1 ^{236}Pu half-lives), then purified. If all the ^{232}U and ^{228}Th are removed, the gamma dose rate is represented by the line beginning at the 6-yr point and continuing on through a normal growth and decay curve, whose maximum is reached 18 yr after purification, after which it decays with a 72-yr half-life if left undisturbed. The worst case is represented by the curve illustrating complete ^{232}U removal and no ^{228}Th removal. Immediately after this kind of purification, the daughter activity will decrease with an initial 1.9-yr half-life. Even if ^{228}Th is not removed, the gamma dose rate decreases rapidly in the purified material and eventually approaches the maximum which is produced by ^{232}U decay.

Uranium-232 can be separated readily from ^{238}Pu by precipitation of plutonium peroxide. The LASL procedures used for the peroxide precipitation separate 99.9% of the uranium from the plutonium. For example, in a typical experiment, the ratio of U/Pu was 0.130 in the feed solution and 0.130×10^{-3} in the peroxide precipitate.

VII. SHIELDING AND ATTENUATION OF ^{238}Pu GAMMA RAYS BY HASTELLOY, HAYNES-25, TZM, AND Ta-10W

We measured the effectiveness of some possible encapsulation materials for attenuating the gamma rays from ^{238}Pu, using a capsule containing 5.55 g of PuO_2(80% ^{238}Pu) positioned 11 cm from a Ge(Li) detector. The count rate for each gamma ray was measured before and after the various absorbers were placed between the capsule and the detector. We converted the count rates to absolute intensities at the surface of the capsule from measurements of gamma ray-standards substituted for the ^{238}Pu source. The percent of attenuation caused by each material is shown in Table XLVI.

VIII. RADIATION, DOSIMETRY, AND THERMAL-POWER MEASUREMENTS

A. Neutron Emission Rates.

The neutron counter used in these measurements is a water-moderated BF_3 detector, arranged in a well-type geometry that is fixed and reproducible for all samples. The neutron counter was calibrated using two sources, Pu-Be and 10.64 g of electrofined ^{238}Pu metal sealed in welded tantalum containers, shown in Fig. 26. The neutron emission rates from both sources were established by measurements in the Los Alamos Graphite Pile,[34] and gave calibration factors for the neutron counter which agreed within 1.2%. Subsequently, it became the practice to use the electrorefined ^{238}Pu source, Lot LA-2 (Capsule 823-6), as the reference standard for all neutron activity determinations for ^{238}Pu samples.

B. Gamma-Spectrum Measurements.

Routine gamma-spectrum measurements are taken with a 45-cm^3 Ge(Li) detector whose resolution is 1.9 keV FWHM at 1332 keV. The energy range covered is 70 to 2800 keV. Because ^{238}Pu samples are normally

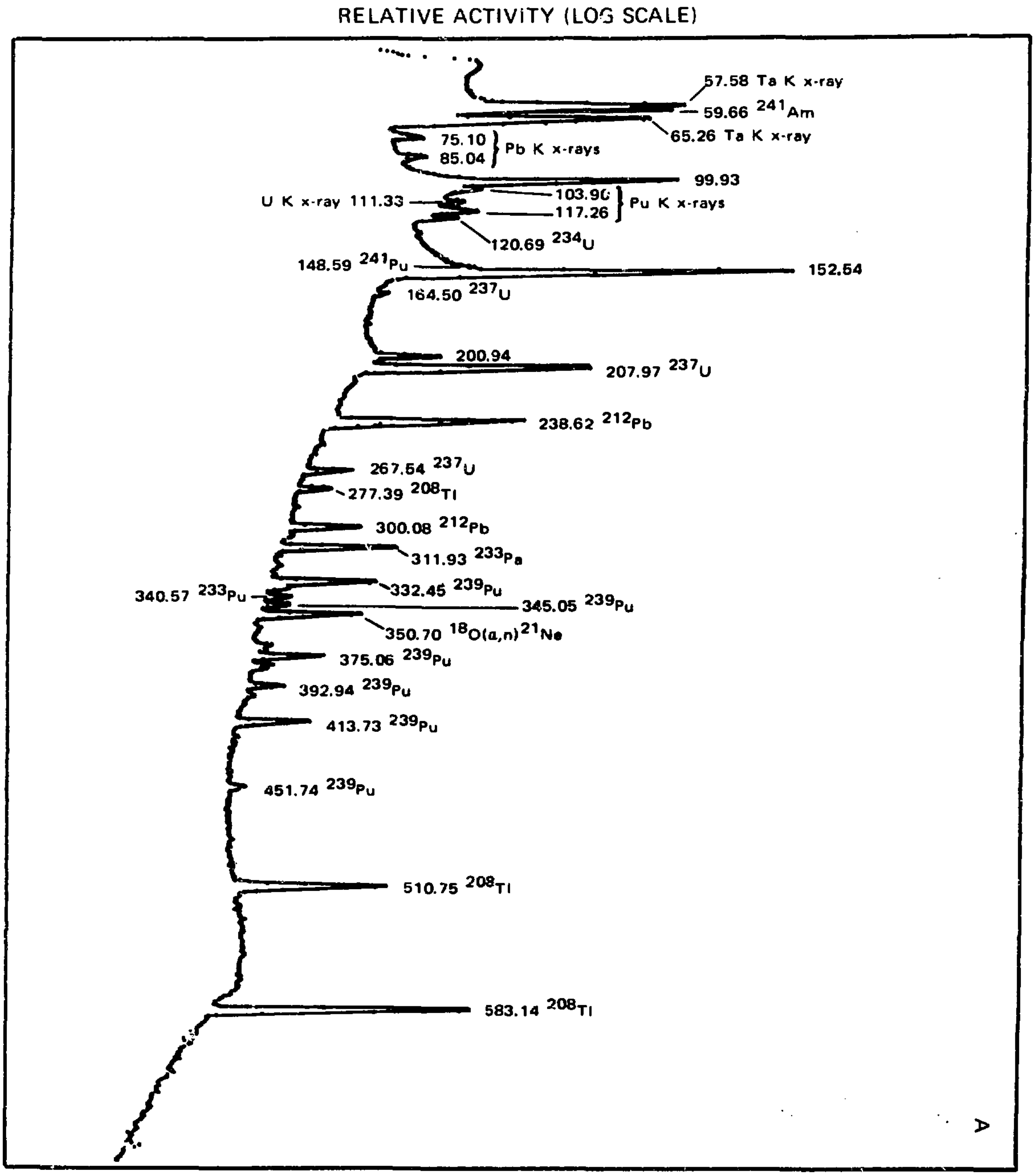
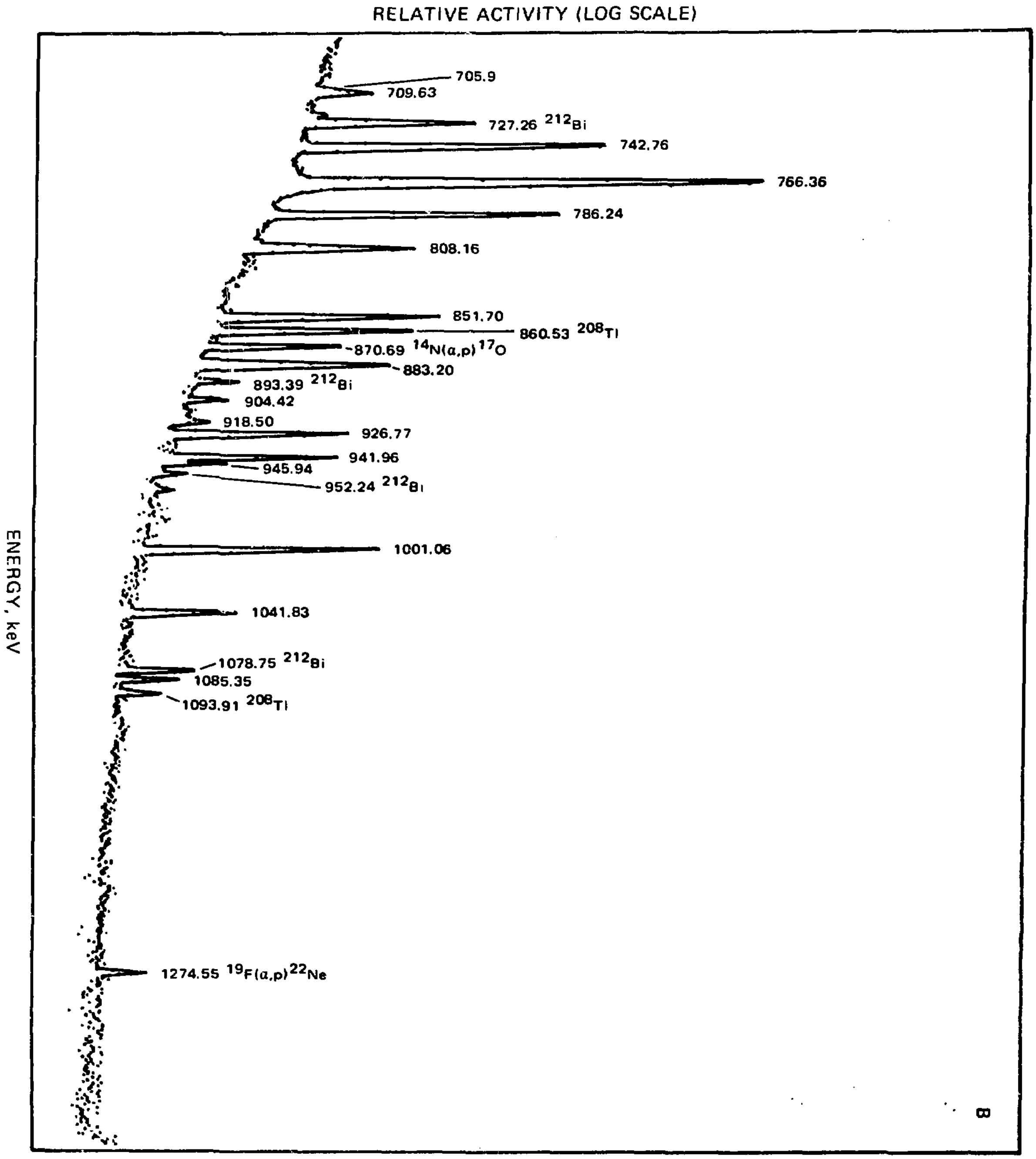
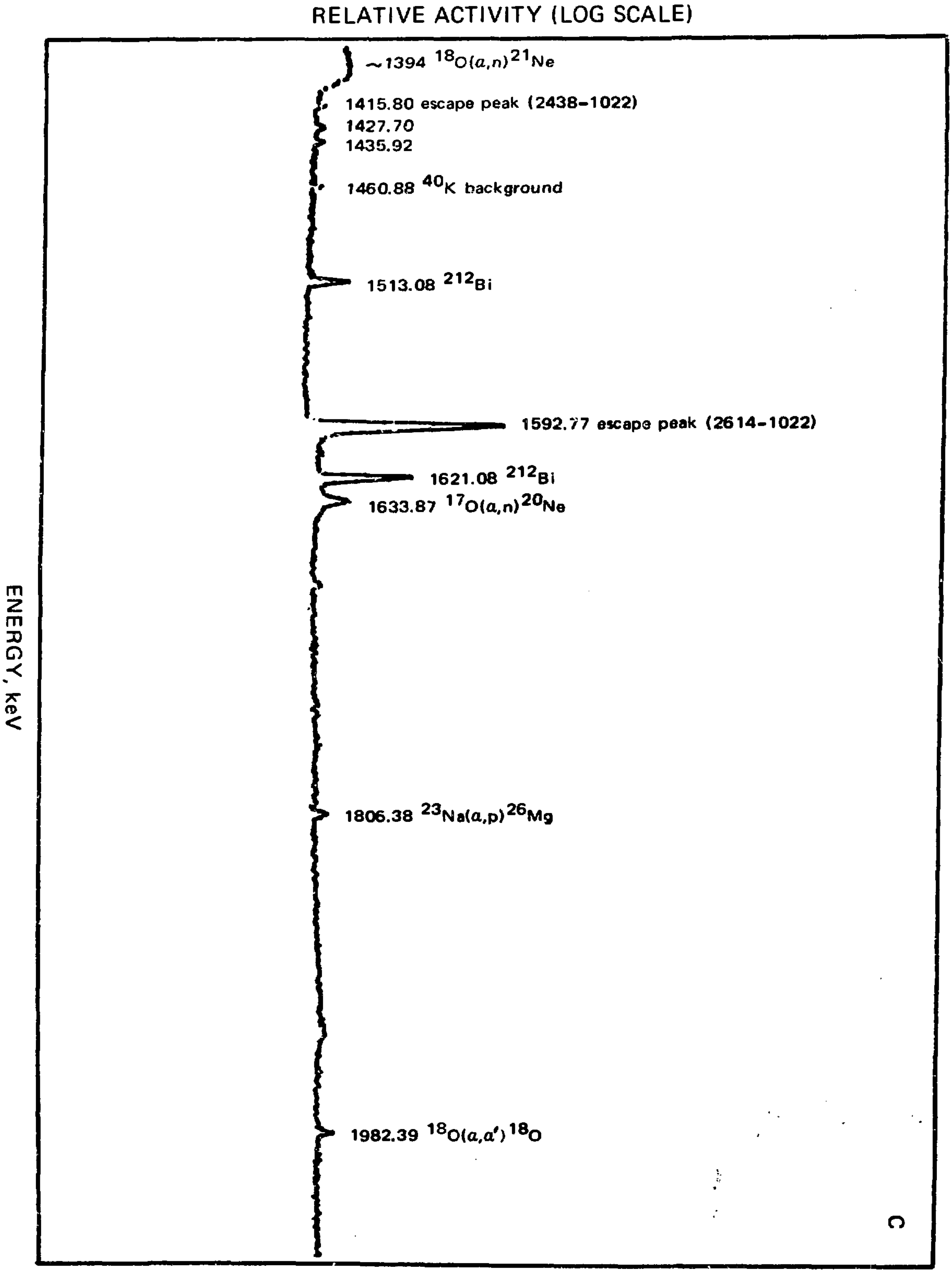


Fig. 24.
Gamma-Ray spectrum of $^{238}PuO_2^{nat}$.


RELATIVE ACTIVITY (LOG SCALE)
ENERGY, keV
705.9
709.63
727.26 ^{212}Bi
742.76
766.36
786.24
808.16
851.70
860.53 ^{208}Tl
870.69 ^{14}N(α,p)^{17}O
883.20
893.39 ^{212}Bi
904.42
918.50
926.77
941.96
945.94
952.24 ^{212}Bi
1001.06
1041.83
1078.75 ^{212}Bi
1085.35
1093.91 ^{208}Tl
1274.55 ^{19}F(α,p)^{22}Ne
B


RELATIVE ACTIVITY (LOG SCALE)
ENERGY, keV
~1394 $^{18}O(\alpha,n)^{21}Ne$
1415.80 escape peak (2438–1022)
1427.70
1435.92
1460.88 ^{40}K background
1513.08 ^{212}Bi
1592.77 escape peak (2614–1022)
1621.08 ^{212}Bi
1633.87 $^{17}O(\alpha,n)^{20}Ne$
1806.38 $^{23}Na(\alpha,p)^{26}Mg$
1982.39 $^{18}O(\alpha,\alpha')^{18}O$
C

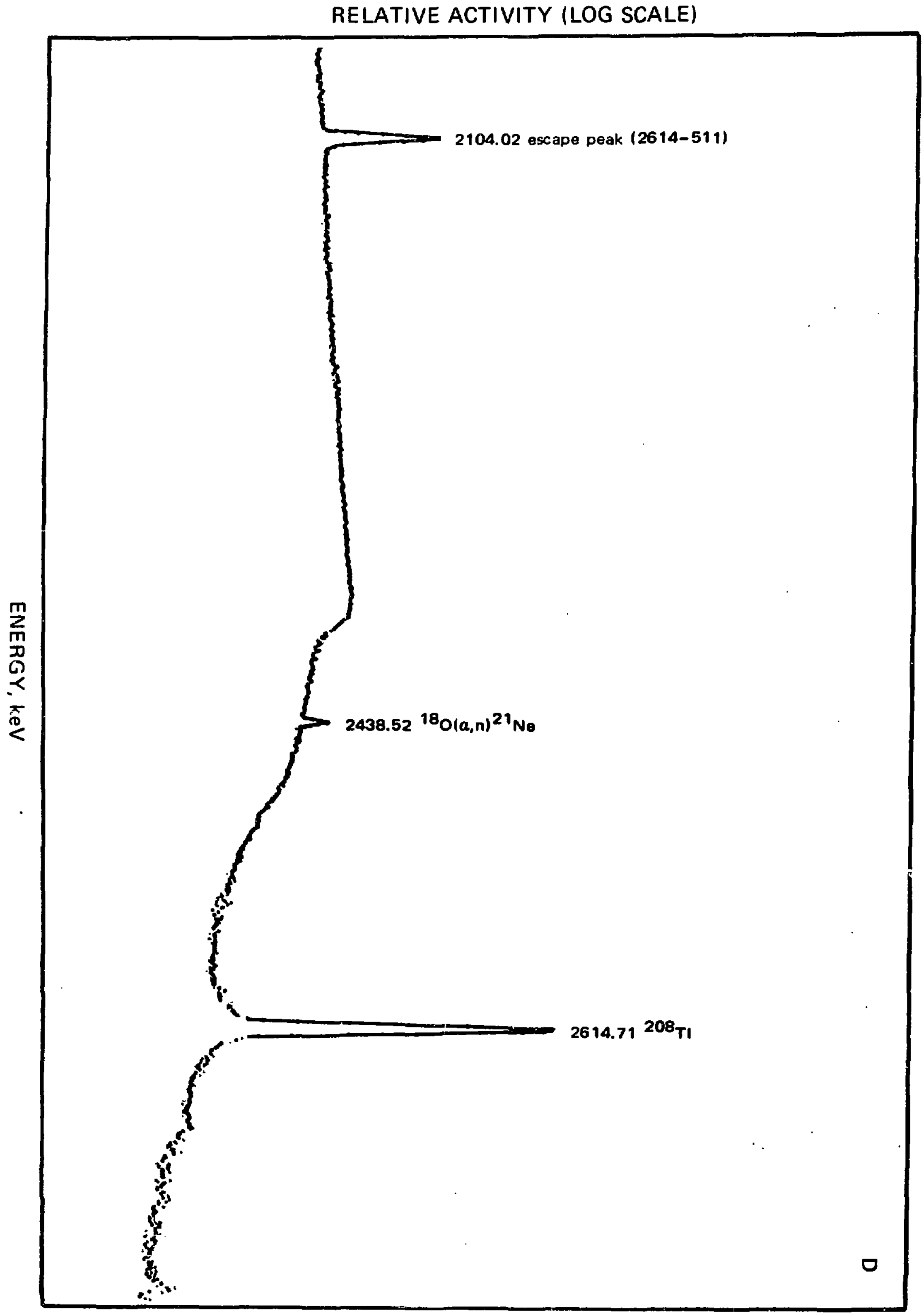

RELATIVE ACTIVITY (LOG SCALE)
ENERGY, keV
2104.02 escape peak (2614–511)
2438.52 $^{18}O(\alpha,n)^{21}Ne$
2614.71 ^{208}Tl
D

TABLE XLI

NEUTRON EMISSION RATES OF LASL $^{238}PuO_2^{nat}$

Method of Preparation	Neutron Emission Rate n/sec-g Pu	 n/sec-g ^{238}Pu
Ignition of as-received PuO_2^{nat}, LA-6	12,960	16,300
Ignition of Pu peroxide, LA-8	12,450	15,640
Reaction of PuN with air	12,100	15,200
Reaction of electrorefined metal with H_2O^{nat} (as per standard flow-sheet for $^{238}PuO_2^{16}$)	12,240	15,300

encapsulated in 0.030-in.-thick tantalum, gamma rays with energies of < 70 keV are absorbed by the capsule and, therefore, not seen in the gamma-ray spectra. The gamma-ray spectra are used to detect light-element impurities that undergo alpha-particle-induced nuclear reactions and to measure $^{17}O/^{16}O$ and $^{18}O/^{16}O$ ratios in oxide materials. All gamma spectra are permanently recorded on standard computer seven-track magnetic tape from a 4096-channel pulse-height analyzer.

TABLE XLII

CALCULATION OF (α,n) CONTRIBUTION FROM ^{17}O AND ^{18}O IN $^{238}PuO_2^{nat}$

	n/sec-g ^{238}Pu
Neutron emission rate	15,300
Contribution from	
(a) Spontaneous fission	2,783
(b) Light-element impurities, other than O	375
(c) Neutron multiplication	225
(d) Thermal multiplication in counter	153
Sum of (a), (b), (c), (d)	3,536
Contribution from ^{17}O, and ^{18}O, by difference	11,764

TABLE XLIII

NEUTRON EMISSION RATES OF LASL BOMB-REDUCED METALS

Lot	g Pu	Neutron Emission Rate (n/sec-g ^{238}Pu)
LA4	22.9	3295
LA5	22.2	3775
LA6	18.2	3345
LA7	25.9	3350
LA8	23.1	3590
LA9	21.3	3150
HBO907546-3	43.8	3600
HBO907546-4	42.3	3620
HAS1	43.8	3700
HAS4-1	36.1	4000
HAS4-2	40.3	3900
HAS6-1	34.5	4590
HAS6-2	41.9	5495
HAS7, 10-1	44.2	4095
HAS7, 10-2	44.4	4070

TABLE XLIV

GAMMA-RAY INTENSITIES OF ^{236}Pu DAUGHTERS

Energy (keV)	Expected γ's/min	Found γ's/min
(49.9 mg ^{238}Pu, 0.66 ppm ^{236}Pu, 1.340-yr growth)		
238	4.47 (± 0.21) x 10^4	4.47 (± 0.04) x 10^4
511	8.67 (± 0.42) x 10^3	9.29 (± 0.31) x 10^3
583	2.83 (± 0.14) x 10^4	2.69 (± 0.05) x 10^4
727	6.16 (± 0.30) x 10^3	5.93 (± 0.33) x 10^3
1620	1.32 (± 0.06) x 10^3	1.2 (± 0.40) x 10^3
2614	3.29 (± 0.16) x 10^4	3.23 (± 0.10) x 10^4
(56.8 mg ^{238}Pu, 0.62 ppm, ^{236}Pu, 1.216-yr growth)		
238	3.99 (± 0.26) x 10^4	3.84 (± 0.04) x 10^4
511	7.74 (± 0.50) x 10^3	9.11 (± 0.37) x 10^3
583	2.53 (± 0.16) x 10^4	2.24 (± 0.03) x 10^4
727	5.50 (± 0.36) x 10^3	5.06 (± 0.66) x 10^3
1620	1.18 (± 0.08) x 10^3	0.81 (± 0.49) x 10^3
2614	2.93 (± 0.19) x 10^4	2.78 (± 0.01) x 10^4

TABLE XLV

PERMISSIBLE COMBINATIONS OF ^{236}Pu AND ^{232}U

(Parts per Million)

^{236}Pu	^{232}U
0.300	0.000
0.250	0.028
0.200	0.056
0.150	0.084
0.100	0.112
0.050	0.140
0.000	0.168

C. Neutron Dosimetry.

^{6}LiF thermoluminescent detectors (TLDs) are placed in a 10-in.-diam. polyethylene sphere with the neutron source outside it as shown in Fig. 27. The neutron dose rate can also be calculated from the neutron emission rate.[1] Results obtained by both methods agree well.

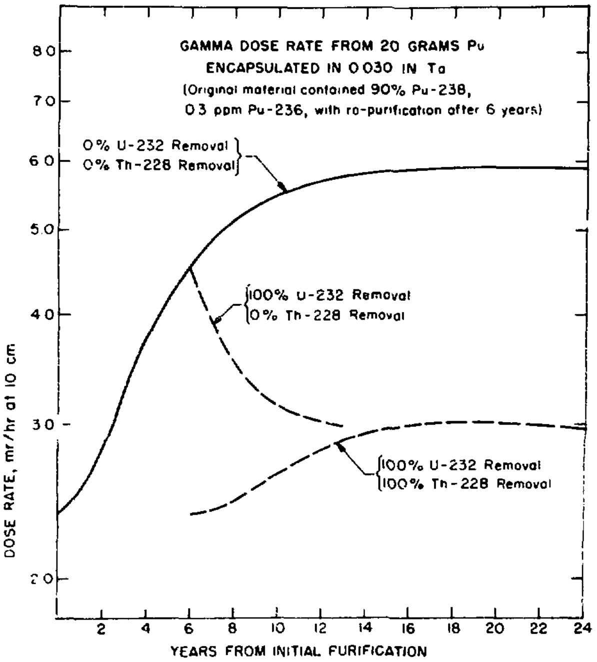


Fig. 25.
Gamma dose rate as a function of time.

D. Gamma Dosimetry.

^{7}LiF thermoluminescent detectors are used for gamma measurements in air 0.8, 2.0, 4.0, 6.0, 8.0, 10.0, 14.0, and 18.0 cm from the center of the source. The experimental arrangement is shown in Fig. 28. A typical data plot is given in Fig. 29. The data fit an inverse-square law between 2 and 14 cm.

E. Thermal Power.

The thermal power of small, 0- to 10-W sources is usually measured in a copper isothermal twin calorimeter. For larger, 10- to 60-W, sources, a water-bath calorimeter similar to the Mound Laboratory Water Bath Calorimeter No. 94,[35] is used. The equipment is shown in Fig. 30.

IX. ANALYTICAL CHEMISTRY METHODS FOR ^{238}Pu MATERIALS

A. General Discussion.

Analysis of ^{238}Pu metal and its compounds often is complicated by the effects of its high specific activity. The intense ^{238}Pu radiation causes radiolytic decomposition of reagents, especially in aqueous solutions, and creates instabilities of the plutonium oxidation states, fading of various colored complex species used in spectrophotometric measurements, and formation of gas bubbles in the solutions. These factors, and the problems in weighing ^{238}Pu materials because of the heat generated by this isotope, were considered and evaluated in developing the following analytical methods.

B. Sample Dissolution.

An important part of several of the methods is quantitative dissolution of the sample in preparation for analysis. Oxide-free metals and nitrides generally are dissolved in mineral acids, notably HCl, at concentrations greater than 3 *N*. Oxide-containing samples are most efficiently dissolved in HCl containing a trace of $HClO_4$ at 300 °C and 3000- to 4000-lb/in^2. pressure.[36] This is accomplished by sealing up to 2 g of the oxide with 7 ml of 12 *M* HCl and 3 - 4 drops of 12 *M* $HClO_4$ in a thick-walled silica tube surrounded by a compensating pressure of carbon dioxide in a tightly closed steel shell. The tube is heated at 300 °C for 2 to 15 h. The silica tube dimensions are selected so that the acid volume never is more than half the volume of the tube. Dissolution of oxides in

TABLE XLVI

PER CENT OF ATTENUATION [100 x (Io-I)/Io] OF ^{238}Pu GAMMA RAYS

Eγ (keV)	0.025-in. Hastelloy	0.063-in. Haynes-25	0.053-in. TZM	0.050-in. TZM	0.040-in. Ta-10W
100	32	71	71	99	99
152	13	38	36	99	90
201	11	36	38	97	76
708	0	1	9	61	5
743	3	7	6	55	11
766	4	8	7	56	11
786	1	7	6	54	10
808	2	9	6	51	10
851	5	7	6	51	7
883	2	6	5	52	8
926	0	8	6	50	6
942	3	4	3	50	6
1001	0	8	7	49	7
1042	2	4	10	47	12
1085	0	16	23	54	0

simmering HNO_3 - HF in a platinum dish also is effective, but less satisfactory. Weighed aliquots of the solutions are taken for various analyses.

C. Plutonium Assay.

Plutonium is measured by controlled potential coulometric titration in solution aliquots containing 1 - 2 mg of ^{238}Pu.[37] The plutonium in 1 N H_2SO_4 is alternately reduced to Pu(III) and coulometrically oxidized to Pu(IV) at a platinum electrode while the electric current required for each quantitative change in oxidation state is integrated. The quantity of plutonium is calculated from the integrated currents. By cycling between the two oxidation states until successive integrated currents are equal, the relative standard deviation of the method is reduced to 0.4 - 0.5%, an order of magnitude greater than that attainable in titrating ^{239}Pu. Bias was successfully eliminated by a chemical calibration of the coulometer. The method was applied with good results to assays of ^{238}Pu metal, nitrides, and oxides.

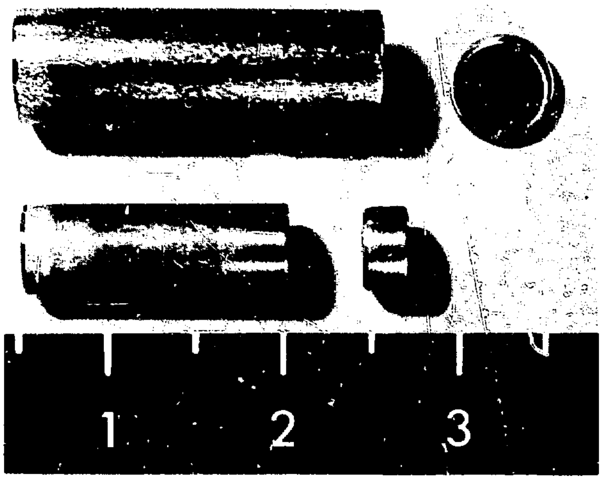


Fig. 26.
Tantalum capsules for electrorefined metal standard (scale in inches).

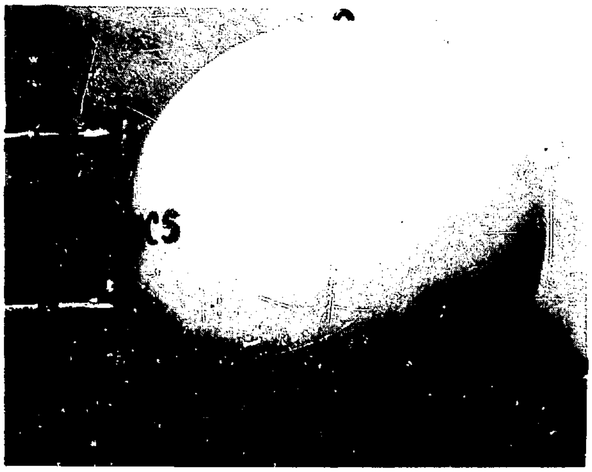


Fig. 27.
Experimental arrangement for neutron dosimetry.

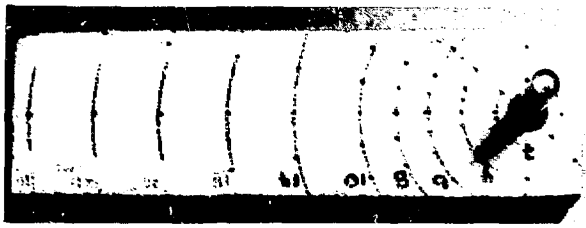


Fig. 28.
Experimental arrangement for gamma dosimetry.

D. Radiochemical Measurement of ^{236}Pu.

An improved radiochemical method is used to measure ^{236}Pu at concentrations as low as 0.01 ppm in ^{238}Pu.[38] The method for ^{236}Pu requires quantitative separation and alpha counting of the ^{232}U daughter from the ^{236}Pu in a 10-mg sample of ^{238}Pu. The sample is absorbed on a cation-exchange resin column from a bromine-hydrobromic acid mixture, and only the uranium is eluted with dilute HBr. The uranium fraction is alpha-counted directly. If the weight of the original sample is accurately known, the ^{236}Pu can be calculated from the ^{232}U content plus the sample weight and growth time. Otherwise, the ^{232}U/^{234}U ratio is measured along with the ^{238}Pu abundance and growth time. The relative standard deviation in the measured ^{236}Pu content is no greater than 5% for growth times of 30 days.

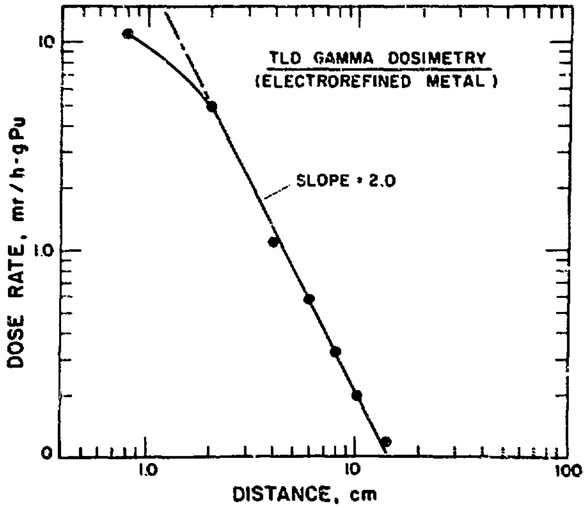


Fig. 29.
Gamma dose rate of electrorefined ^{238}Pu metal, 6-W cylinder (measured through 0.030-in. Ta).

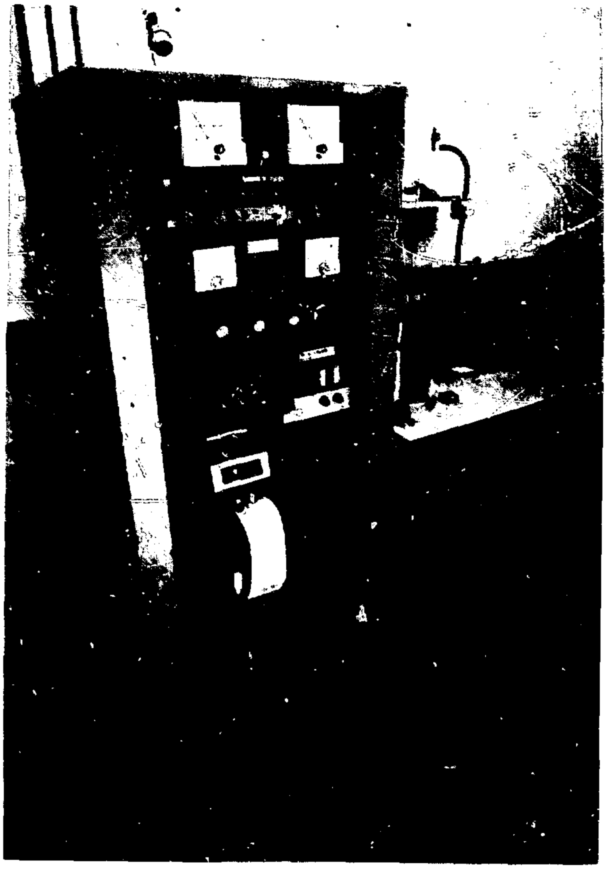


Fig. 30.
Water-Bath calorimeter.

E. Radiochemical Measurement of ^{228}Th.

Analysis of ^{238}Pu for ^{228}Th is done by alpha counting following two ion-exchange separations to remove plutonium and other activities. A 10-mg sample of ^{238}Pu(IV) is absorbed onto a cation-exchange resin column from strong HCl, and most of the neptunium, uranium, americium, and plutonium activities are eluted by washing the column with 6 *M* HCl - 10% HBr solution. Then the ^{228}Th is eluted with 0.5 *M* oxalic acid and loaded onto an anion-exchange resin column from 7.8 *M* HNO_3. The thorium is selectively eluted with 7 *M* HCl, transferred to a glass plate, and alpha counted. The decontamination factor for plutonium removal is about 10^6 in each separation. The relative standard deviation of the method in measuring between 0.01 and 0.9 ppb of ^{228}Th ranged from 12 to 1%, with an average of 6%. Recovery of ^{228}Th is 90 to 95%.

F. Neptunium Measurement.

Neptunium is measured routinely by alpha-counting the ^{237}Np after separation from ^{238}Pu by a double separation procedure that employs both ion exchange and solvent-extraction techniques.[39] A cation-exchange resin column is used to absorb both Np(V) and Pu(III) from a mixture of hydrobromic acid and bromine. The neptunium is selectively eluted with dilute hydrochloric acid. Further decontamination is obtained by reducing the neptunium with $SnCl_2$ and extracting the Np(IV) into a 0.5 *M* thenoyltrifluoroacetone (TTA) in xylene. The last traces of plutonium are left in the aqueous phase. An aliquot of the organic phase is evaporated and alpha counted. As ^{238}Pu cannot be detected in the organic aliquot, the overall alpha decontamination factor is greater than 10^{11}. The lower limit of detection of the method is 10 ppm of ^{237}Np in a 10-mg sample of ^{238}Pu. Measured neptunium levels have ranged between 30 and 20,000 ppm in the refined metal and the various process solution samples analyzed. The relative standard deviation of the method is no greater than 5%.

Total neptunium in ^{238}Pu materials is measured spectrophotometrically[40] following ion separation from the plutonium.[37] Uranium, if present as an impurity, is removed first by extraction from 4 *M* HCl into tri(iso-octyl) amine (TIOA)-CCl_4, whereas neptunium and plutonium are stabilized in their nonextractable (IV) and (III) oxidation states, respectively, by iron(II) and ascorbic acid. Then Np(IV) is extracted from 8 *M* HCl into TIOA-xylene and back-extracted into 0.1 *M* HCl. The neptunium-Arsenazo III complex is formed in 6.1 *M* HCl, and the absorbance is measured at a wavelength of 665 nm. The molar absorptivity of the complex was found to be 100,000, permitting neptunium measurement at concentrations as low as 15 ppm in a 100-mg sample of ^{238}Pu. Relative standard deviations ranged from 3.7% at the 17-ppm neptunium level to 1.1% at the 330-ppm level in plutonium. Recoveries of neptunium from 100 mg of ^{238}Pu averaged 100%, but larger amounts of ^{238}Pu caused low recoveries.

G. Measurement of ^{18}O and ^{17}O in H_2O^{16}.

H_2O^{16} cannot be analyzed directly in a mass spectrometer because high background levels of water would always be present. Analysis for oxygen isotopes requires that the oxygen be introduced as a volatile substance such as O_2 CO, or CO_2. Oxygen is a useful compound to introduce into a mass spectrometer because there is no interference by foreign atoms at the measured masses.[41]

For the routine analysis of H_2O^{16} for the ^{17}O and ^{18}O isotopes, oxygen gas is prepared by electrolysis of the H_2O^{16} to produce H_2 and O_2.[42] The H_2O^{16} is first distilled to remove traces of NH_3 which would interfere with the mass spectrometry. Sodium metal is then added to the distilled water to increase the electrical conductivity and permit reasonable electrolysis rates. The O_2 gas is carefully dried and collected in a 100-cm^3 bulb which is adequate for the 100-atm cm^3 of oxygen usually generated. Oxygen analysis for ^{17}O and ^{18}O is made with a Consolidated Electrodynamics mass spectrometer, Model 21-620A.

H. Measurement of ^{18}O and ^{17}O in $^{238}PuO_2^{16}$.

In the routine measurement of ^{18}O in PuO_2 depleted in this oxygen isotope, the oxide sample is reacted with graphite at 2000 °C to liberate the oxygen as carbon monoxide which is analyzed mass spectrometrically for ^{18}O. A 50-mg sample is blended with 10 mg of graphite and heated inductively in a graphite crucible at 2000 °C. The carbon monoxide formed is swept from the furnace with helium, collected in a liquid-nitrogen-cooled trap, and then transferred to 6-ft-long gas-chromatographic column filled with Linde 5X molecular sieve. Traces of ^{15}N, which interfere with mass spectrometry of the carbon monoxide, are eluted from the column with helium first, followed by the carbon monoxide which is trapped and transferred to the mass spectrometer. The $^{18}O/^{16}O$ ratio is calculated from the ratio of the peak heights of $^{12}C^{18}O$ to $^{12}C^{16}O$ measured with a CEC 21-620 mass spectrometer. The standard deviation of the method is 4 ppm at the 50-ppm level of ^{18}O in ^{16}O. Comparison of the mass-spectrometry results with the maximum ^{18}O contents predicted from neutron count rates indicates that the method is not biased positively. Mass-spectrometry methods for the measurement of ^{17}O in oxides are still under study.

Measurement of the 351-keV gamma ray from ^{21}Ne produced by the (α,n) reaction of ^{18}O is a possible alternate method for measuring ^{18}O. Spectra of ^{238}Pu metal and some oxides in the 340- to 360-keV region show that the intensity ratios of the 351-keV reaction gamma ray of ^{18}O and the 345-keV gamma ray of ^{239}Pu are related to the ^{18}O contents. Similarly, the spectral region from 1420 to 1660 keV includes the 1634-keV (α,n) reaction gamma ray of ^{17}O and the 1428- and 1435-keV gamma rays of ^{238}Pu which may be used as internal standards. We are studying the experimentally determined ^{17}O and ^{18}O abundances and the neutron emission rates to estimate the reliability of the method. We also investigated the effect of sample size on the measurement of ^{17}O and ^{18}O from the gamma-ray intensity measurements and found it to be insignificant.

I. Measurement of Total Oxygen in $^{238}PuO_2$.[16]

A gravimetric method developed for precise determination of oxygen in refractory oxides was applied to the analysis of $^{238}PuO_2$.[43] One gram of the oxide is ground and mixed intimately with 0.2 g of carbon, and the mixture is pressed into a pellet. This pellet, contained in a graphite crucible, is heated slowly to 2000 °C in an inert-gas-fusion apparatus equipped with an automatic temperature programmer. The oxygen, evolved as carbon monoxide and carbon dioxide, is swept by argon carrier gas into an Ascarite absorption tube in which the carbon dioxide is trapped. The carbon monoxide is oxidized over hot CuO to carbon dioxide which is trapped in a second absorption tube. The oxygen content is calculated from the weight increases of the two tubes. The method is unbiased and precise. The relative standard deviation in measuring 12% oxygen is 0.2% for $^{238}PuO_2$ materials.

J. Nitrogen Measurement.

Nitride nitrogen in plutonium metals or nitrides is measured spectrophotometrically using Nessler's reagent, or titrated with standard hydrochloric acid solution. The sample is dissolved by refluxing in a mixture of hydrochloric and perchloric acids, and the nitrogen is steam-distilled as ammonia into a trap containing boric acid. For samples containing less than 100 μg of nitrogen, Nessler's stabilizer and Nessler's reagent solutions are added to the ammonia in the Boric acid, and the absorbance is measured at 410 nm. For samples containing more than 100 μg of nitrogen, the boric acid solution is titrated with standardized 0.02 *M* HCl using the color change of methyl red-bromcresol green indicator to indicate the endpoint. The relative standard deviation is 3% for the measurement of 50 to 100 ppm of nitrogen and increases to 10% for 20 ppm and 12% for 2 ppm, respectively. For measurements of 500 to 2000 ppm of nitrogen by titration, the relative standard deviation is about 0.7%. This deviation decreases to 0.2% in measuring nitrogen in nitrides.

K. Mass-Spectrometry of ^{15}N.

Nitrogen depleted in ^{15}N was used to investigate the capabilities of a CEC 21-620 mass spectrometer. Measurement of the mass-29 peak permitted calculation of ^{15}N concentrations. Forty-four determinations of ^{15}N concentrations ranging between 2 and 86 ppm showed that the lower limit of reliable measurement was about 5 ppm. The relative standard deviations were 3, 10, and 34%, respectively, for measuring 85, 20, and 4 ppm of ^{15}N in N_2 gas.

L. Spectrochemical Measurement of Trace Impurities in ^{238}Pu Metal, Oxide, Fluoride, Nitride, and Alloys.

Two spectrochemical methods, originally developed for routine analyses of ^{239}Pu, were applied to the measurement of 35 impurity elements in ^{238}Pu metal, nitrate solutions, fluorides, oxides, nitrides, and alloys. These applications require the preparation of standards similar in composition to the materials analyzed, but we used ^{239}Pu as a stand-in for ^{238}Pu in these preparations.

We used a carrier distillation method routinely in the measurement of 25 impurity elements in the ^{238}Pu materials. Each sample was ignited at 700 °C in air in a platinum-lined muffle furnace to form $^{238}PuO_2$ which was ground and then mixed with either Ga_2O_3 or AgCl carrier. Duplicate electrode charges were weighed, vented, and dc arced to yield the spectra for determination of the impurities. The impurity concentrations were evaluated by comparing the sample spectra with spectra of similarly processed standards. The elements determined using the two carriers and the lower limits of reliable determination are listed in Table XLVII. As these elements are usually evaluated by visual comparison of the spectra, the relative standard deviation of the method is about 35%.

Many of the elements determined by the carrier distillation methods and a few other elements also may be measured with excellent lower limits of detection by a graphite-spark excitation technique after separation of the plutonium by anion exchange. This method, being more tedious and time-consuming, is used less frequently than the carrier distillation methods. The sample must be dissolved to provide a 7.5 *M* nitric acid solution containing plutonium in the (IV) oxidation state. The Pu(IV) is absorbed as the nitrate complex onto an anion-exchange resin column, whereas trace impurities pass quantitatively into the effluent. The impurities are concentrated by evaporation of the effluent solution, indium and gallium are added to serve as internal standard and matrix elements, and the solutions are evaporated onto waterproofed graphite electrodes. A high-voltage spark discharge in a pure oxygen atmosphere is used to excite the samples, and conventional methods of photometry, calibration, and calculation are used to evaluate the spectra. The overall relative standard deviation is 20%, and the bias in the measurements is no greater than 20%.[44] The elements determined by this method also are listed in Table XLVII.

M. Carbon Measurement.

A method routinely applied to the measurement of carbon in ^{239}Pu materials[45] was found satisfactory for analyzing ^{238}Pu materials also. The samples, generally

TABLE XLVII

ELEMENTS MEASURED BY SPECTROCHEMICAL METHODS WITH THE LOWER LIMITS OF DETECTION

	Lower Limits of Reliable Detection (ppm)		
	Carrier Distillation Methods Using		
Element	Ga_2O_3 Carrier	AgCl Carrier	Anion Exchange-Cu Spark Method
Li	1	—	0.1
Be	2	—	0.1
B	2	—	—
Na	10	—	0.2
Mg	5	—	0.2
Al	25	—	0.5
Si	20	—	—
P	100	—	—
Ca	25	—	6
V	—	100	1
Cr	5	—	1
Mn	5	—	0.5
Fe	5	—	3
Co	—	10	1
Ni	10	—	1
Cu	5	—	0.5
Zn	20	—	10
Mo	—	20	1
Ag	1	—	—
Cd	20	—	0.5
Sn	5	—	1
W	—	20	—
Pb	5	—	2
Bi	5	—	—
Ta	—	50	—
Ba	—	—	0.1
Cs	—	—	2
Hf	—	—	1
K	—	—	2
La	—	—	2
Rb	—	—	1
Sr	—	—	0.5
Y	—	—	0.2
Zr	—	—	1
Ti	—	—	1

weighing 0.1 g, are ignited in a stream of oxygen at 1000 °C for a minimum of 10 min to oxidize the carbon to carbon dioxide which is measured in a calibrated micromanometer. Metals were found to burn readily under these conditions, but carbon recoveries are low *unless oxide samples are ground to a fine powder before* ignition. For 100-mg samples, the standard deviation is 5 relative per cent for carbon contents greater than 50 ppm, and 5 ppm for carbon concentrations from 10 to 50 ppm. The precision of the method can be improved by using larger, up to 1-g, samples.

N. Fluoride Measurement.

Either of two methods, originally developed for measuring fluoride in ^{239}Pu materials, are used to measure this impurity in ^{238}Pu materials, depending upon their composition. For the metal and other materials that are soluble in dilute sulfuric acid, the fluoride is separated by steam distillation and titrated as HF. For oxides and other insoluble materials, the fluoride is separated by pyrohydrolysis and either titrated or measured with a fluoride-specific electrode.

A metal sample containing no more than 10 μg of fluoride is dissolved in 5 *N* sulfuric acid, and additional acid is added to precipitate most of the plutonium. The fluoride is steam-distilled from the supernatant solution at 135 °C, and determined by titration with 0.001 *M* thorium nitrate using Chrome Azurol S as the color indicator. The standard deviation is approximately 10 relative per cent in measuring 50 to 30 ppm of fluoride and 1 ppm or less in the 1- to 5-ppm range.

Fluoride and chloride are jointly separated by pyrohydrolysis from PuO_2 and other sulfuric acid-insoluble materials. Approximately 1 g of the oxide is heated in a nickel apparatus at 1000 °C in a stream of argon and steam until 8 ml of distillate are collected. The fluoride content of a small aliquot of the distillate is measured with a fluoride-specific electrode. The chloride in the rest of the distillate is measured spectrophotometrically by adding ferric ammonium sulfate and mercuric thiocyanate to form an amount of ferric thiocyanate equivalent to the chloride present. The absorbance is measured at 450 nm. Alternatively, the fluoride in the distillate may be titrated with thorium as described above. Relative standard deviations are approximately 7% for the 5- to 50-ppm concentration range and 10% for the 0.9- to 5-ppm range. For chloride measurement, the relative standard deviations are 5% for the 6- to 50-ppm concentration range and 10% at the lower limit of 4 ppm.

X. PRESENT STATUS OF THE PREPARATION OF MEDICAL-GRADE ^{238}Pu FUELS

The specifications for medical-grade ^{238}Pu metal and $^{238}PuO_2^{16}$ are given in Tables XLVIII and XLIX. These specifications were first presented in Ref. 28. The electrorefined metal presently produced in this program meets all of the specifications of Table XLVIII, provided, of course, that the ^{238}Pu isotopic composition of the oxide feed is greater than 90%.

The $^{238}PuO_2^{16}$ ceramic fuels presently produced do not meet the second and fifth specifications of Table XLIX. The chemical purity of the pressed and sintered oxides is not so good as that of electrorefined metal because impurities are introduced into the product during ball-milling, screening, and pressing. The chemical purity of the oxide powder, however, is as good as that of the electrorefined metal. Planned improvements in the ceramic fabrication procedures are expected to prevent introduction of impurities into the milled powder. The neutron emission rates of the two 30-W pellets fabricated using the present flowsheets were 308 and 221 n/sec-g of ^{238}Pu higher than the specification of 3300 n/sec-g of ^{238}Pu. The neutron emission rates of the three 50-W cylinders were 24, 138, and 116 n/sec-g of ^{238}Pu higher than the calculated specification for a 50-W cylinder. (The neutron emission rate for a 50-W cylinder should be less than 3350 n/sec-g of ^{238}Pu).

TABLE XLVIII

SPECIFICATIONS FOR MEDICAL-GRADE ^{238}Pu METAL (Ref. 28)

Plutonium Isotopic Composition	
^{238}Pu	> 90%
^{236}Pu	< 0.3 ppm with allowable concentration dependent on ^{232}U concentration
Chemical Purity	
Pu	> 99.8 wt %
Impurities (other than Np)	At least as good as achieved through electrorefining
Specific Power	Not less than 0.50 W/g fuel
Neutron Emission Rate (30-W source)	< 3200 n/sec-g ^{238}Pu
Photon Emission Rate	No greater than that achieved through electrorefining

TABLE XLIX

SPECIFICATIONS FOR MEDICAL-GRADE ^{238}Pu OXIDE (Ref. 28)

Plutonium Isotopic Composition	
^{238}Pu	> 90%
^{236}Pu	< 0.3 ppm with allowable concentration dependent on ^{232}U concentration
Chemical Purity	
Plutonium Oxide	> 99.8 wt %
Impurities (other than Np)	At least as good as achieved through electrorefining
Specific Power	Not less than 0.44 w/g fuel
Oxygen-16 Enrichment	Oxygen to be depleted in ^{17}O and ^{18}O so that neutron contribution from these isotopes is less than 100 n/sec ^{238}Pu
Neutron Emission Rate (30-W source)	< 3300 n/sec-g ^{238}Pu
Photon Emission Rate	No greater than that achieved through electrorefining

XI. PERSONNEL RADIATION PROTECTION

LASL's Health Physics Group, H-1, has the primary responsibility for ensuring that all personnel radiation exposures are kept to a minimum. The methods by which this is accomplished are presented in Ref. 46. This manual summarizes radiation limits, protection methods, radiation monitoring, air sampling, personnel monitoring, and waste management. The exposure of all personnel working in the present program has been well below the 5-Rem/yr administrative limit set by LASL.[47]

Personnel exposures to radiation are determined by:

a. film badges worn on the upper chest to measure whole-body exposures.

b. thermoluminescent ring dosimeters worn on the finger to measure extremity exposures.

c. blood counts.

The air is monitored continuously in all ^{238}Pu laboratories. Visual and audio alarms announce any airborne alpha contamination. The plutonium facilities used are described in Ref. 47.

APPENDIX A.

PROCESS OPERATING PROCEDURES

I. CONVERSION OF $^{238}PuO_2$ TO ^{238}Pu METAL (Flowsheet, Fig. 2 in text)

A. Unloading, Sampling, and Batching of the As-Received PuO_2.

Place secondary Savannah River (SR) container in hood. (For a description and drawing of the SR $^{238}PuO_2$ containers see Ref. 48.) Have health physicist check container for alpha contamination.

With health physicist in attendance, carefully loosen cap of secondary SR container one-quarter turn using strap wrenches.

Hand-tighten cap.

Check for alpha contamination.

Place container in transfer can filled with dry ice to cool container before introducing it into air lock.

Introduce container into conversion chemistry line.

Remove primary SR container from secondary container.

Transfer the $^{238}PuO_2$, ~225 g, into four tared stainless steel containers. Each should contain ~56 g of PuO_2. Weigh containers and record oxide weight.

Remove 4 g of $^{238}PuO_2$ from one container. Package this sample as follows:

Encapsulate 1 g in tantalum for nondestructive analysis.

Encapsulate another 1 g in tantalum for nondestructive analysis and identify it as the library sample.

Place 1 g in a glass vial for plutonium isotopic analysis, plutonium chemical titration, and radiochemical analysis.

Place 1 g in a glass vial for spectrochemical analysis.

B. Dissolution of $^{238}PuO_2$.

Place ~56 g of $^{238}PuO_2$ in 2-liter round-bottom flask equipped with heating mantle and reflux condenser.

Add 350 ml of 15.6 M $HNO_3$0.5MHF.

Heat to boiling and reflux the mixture for 4 - 6 h while continuously sparging the solution with argon.

Cool solution to room temperature.

Filter solution through a medium-porosity sintered-glass frit.

C. Precipitation of Plutonium Peroxide.

Dilute solution to 1200 ml.

Add 5 ml of 18 M H_2SO_4.

Cool to 15 °C.

Add 1 liter of 30% H_2O_2. First addition of the H_2O_2 should be done dropwise until there is visual evidence of precipitation. Then the addition rate can be increased.

Digest the precipitate at 10 °C for 1 h.

Filter the PuO_4 on a medium-porosity sintered-glass filter.

D. Disposal of Peroxide Filtrate.

Heat filtrate to destroy the H_2O_2.

When filtrate volume is reduced to ~0.750 liter, take a 15-ml aliquot for radioanalysis.

Evaporate filtrate to dryness and transfer to waste management.

E. Conversion of PuO_4 to PuF_4.

Transfer PuO_4 from glass funnel to platinum boat.

Load platinum boat into nickel furnace tube.

Heat the furnace to 150 °C.

Start caustic scrubbing system.

Admit O_2 at 300 g/h.

Start HF at 350 g/h.

Increase furnace temperature to 600 °C and maintain the above gas-flow rates for 3 h.

Cool furnace to 275 °C and shut off HF gas.

At 250 °C, shut off O_2.

F. Metal Reduction.

To each gram of PuF_4 add 0.45 g of calcium and mix well.

Pour the mixture into a MgO crucible that has been previously packed in a thermite-type reduction bomb.

Place special perforated MgO lid on the crucible and add 0.1 g of I_2/g of plutonium plus 1.0 g of I_2 top dressing in the lid.

After sealing the bomb, evacuate and back fill with argon three times. During this procedure, check the container for leaks.

Place bomb in reduction furnace coil and start Ajax high-frequency converter at 1.5 kW. After 1.5 min, increase power to 2.5 kW. The metal reduction takes ~7 min and is complete when the neutron emission rate drops to background level.

After cooling, open the bomb in an inert-atmosphere glovebox. Separate the Pu^0 from the slag and store the metal for casting. Package residues and transfer them to waste management.

II. PREPARATION OF ELECTROREFINED ^{238}PU METAL (Flowsheet, Fig. 3)

Detailed operating procedures have been written for electrorefining ^{239}Pu.[49] Most are applicable to the present ^{238}Pu process, and personnel should familiarize themselves with them.

A. Electrolyte Preparation.

Dry analytical grade NaCl and KCl in vacuum drying oven at 110 °C for at least 48 h.

Place 37.4 g of NaCl and 47.6 g of KCl in 34-mm-i.d. by 18-cm-long quartz tube. Mix salts thoroughly.

Place 34-mm tube in 50-mm-i.d. by 46-cm-long quartz tube.

Place rubber stopper fitted with quartz thermocouple sheath, vacuum-argon line, radiation shields, and cooling coil in position.

Place assembly in Hevi-Duty electric furnace, Model 3018.

Heat under vacuum, $p \leqslant 25\ \mu$, to 300 °C at ~50 °C/h.

Heat salts to melting at ~400 °C/h.

Hold molten salts under vacuum at ~700 °C for 1 h.

Cool to room temperature.

Remove NaCl salt casting from quartz tube. Casting should slide easily from 34-mm-i.d. tube. Reject casting if it sticks to the quartz.

Store casting in vacuum dissicator

B. Casting of ^{238}Pu Anode Cylinder.

All of the glovebox operations described below are done in argon-filled gloveboxes.

Turn on uranium chip furnace for argon purfication at 700 °C.

Place the 50-g ^{238}Pu button in the tantalum funnel, and load assembly as shown in Fig. A1.

Evacuate casting furnace tube to $< 0.1\ \mu$.

Set furnace controller at 850 °C.

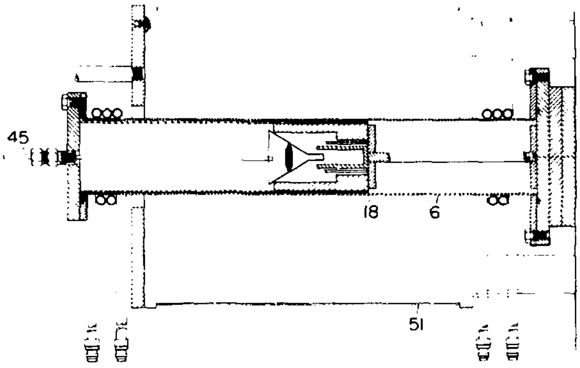


Fig. A1.
Plutonium-238 casting equipment for 50 to 100 g of plutonium.

Heat furnace to 850 °C at full power (~550 °C/h).

Admit purified argon gas to furnace tube to a pressure of 2 - 3 psig.

Lower tantalum thermocouple sheath to bottom of 3/4-in.-o.d. by 0.015-in.-wall tantalum casting crucible and then raise sheath 1/2 in.

Adjust plutonium melt temperature to 850 °C.

Raise tantalum thermocouple sheath out of the melt.

Evacuate furnace tube to $<0.1\mu$.

Hold melt at 850 °C under vacuum for 1 h.

Admit purified argon to furnace tube to a pressure of 2 - 3 psig.

Pass tantalum thermocouple sheath through the tantalum funnel hole several times to ensure that no plutonium is trapped in the funnel.

Shut off furnace power and allow furnace tube to cool to room temperature.

C. Loading of Electrorefining Furnace (see Fig. A2).

All operations described below are done in argon-filled gloveboxes.

Turn on uranium chip furnace for argon purification.

Load two ~50-g anodes into inner cup of $MgO\text{-}Y_2O_3$ refining crucible, part 64.

Load one NaCl-KCl salt casting into refining crucible.

Place refining crucible in tantalum safety crucible, part 23.

Lower safety crucible into nickel can, part 18, which is fitted with a stainless steel adapter, part 19.

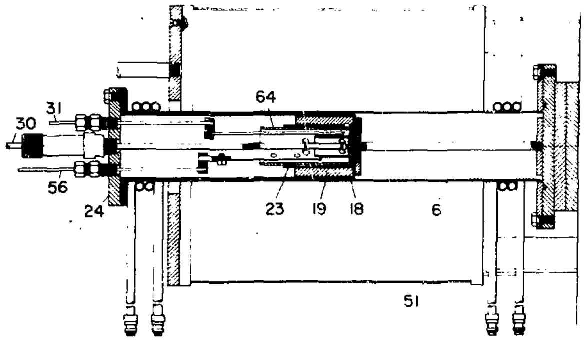


Fig. A2.

Plutonium-238 electrorefining equipment, 100-g scale.

Lower nickel can into furnace tube, part 6.

Place furnace tube head, part 24, on furnace tube. This head is fitted with a tantalum stirrer, part 30; a tungsten cathode cylinder, part 56; a tungsten cathode rod, part 31; a thermocouple sheath, not shown; and a vacuum-argon outlet, not shown. All components are in the raised position when the heat is placed on the tube. The head and nickel can are positioned so that the 1/8-in.-diam thermocouple sheath can be positioned in a 1/4-in.-diam hole in the stainless steel adapter, part 19.

Bolt head in place.

Lower tantalum thermocouple sheath into hole in stainless steel adapter.

Apply vacuum to furnace using Welch mechanical pump.

Fill with purified argon gas to 3 psig. Check for leaks (note--furnace tube should be capable of evacuation to $<50\mu$ with the mechanical pump and should hold the 3 psig of argon without gas-flow indication by the flow meter.

D. Furnace-Heating Cycle.

Evacuate the furnace tube to $p \leqslant 100\,\mu$ using the Welch mechanical pump.

Heat furnace at ~500 °C/h to 400 °C.

Turn off vacuum, admit argon to tube to 2 - 3 psig.

Continue rapid heating to 750 °C.

Adjust melt temperature to 740 °C.

Lower tantalum stirrer; set end 0.03 in. above bottom of refining crucible.

Lower tungsten cathode rod; set end 0.25 in. above bottom of refining crucible.

Lower tungsten cathode cylinder; set end 1.25 in. above bottom of refining crucible.

E. $PuCl_3$ Generation.

Connect positive lead of dc power supply to tantalum stirrer (Note--do not turn on stirrer.)

Connect negative lead of dc power supply to tungsten cathode rod.

Pass 4-A dc current through melt for a total of 200 A/min. Interrupt current every 15 min and measure and record back emf of cell. (Back emf should be ~0.6 V).

F. Pre-electrolysis.

Disconnect positive lead from tantalum stirrer.

Activate tantalum stirrer, and permit it to rotate at ~800 rpm.

Connect positive lead to graphite brushes.

Check back emf. When it is <0.03 V, pass 0.5-A dc current through cell for a total of 10 A/min.

Disconnect negative lead from tungsten cathode rod; raise rod 2 in. above top of refining crucible.

G. Electrorefining.

Connect negative lead to tungsten cathode cylinder.

With stirrer still rotating at ~800 rpm, pass dc current through cell at a value that will permit termination of the electrorefining at a convenient time. Current, however, should not exceed 5 A. For normal operation on an 8-h day, 5-day week, electrorefining is usually started in the afternoon of one day and ended in the morning of the following day. Thus the current is adjusted to end the run in ~16 - 20 h. For example, with 100 g of plutonium in the anode and a refining time of 16 h, the current would be set at 2.1 A; i.e.,

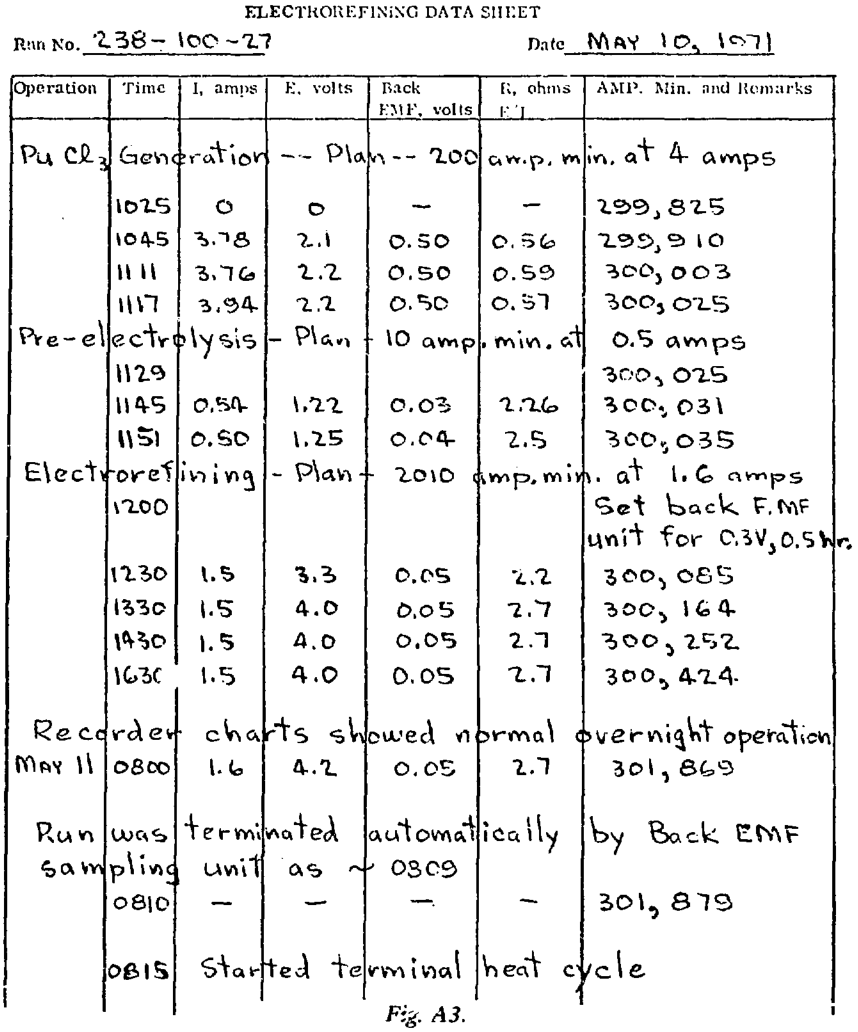
$$\frac{100\text{ g Pu}}{2.97\text{ g Pu/A-h}} \times \frac{1}{16\text{ h}} = 2.1\text{ A} \quad .$$

Turn on automatic back-emf sampling unit; set unit for 0.5-h sampling time; set cutoff for 0.2 V.

For the last few hours of electrorefining, set back-emf sampling unit for 15-min sampling time.

Permit run to continue until back-emf sampling unit automatically terminates it.

The data sheet for a typical electrorefining run (ER 238-200-27) is shown in Fig. A3.



ELECTROREFINING DATA SHEET

Run No. 238-100-27 Date May 10, 1971

Operation	Time	I, amps	E, volts	Back EMF, volts	R, ohms E/I	AMP. Min. and Remarks
PuCl$_3$ Generation -- Plan -- 200 amp. min. at 4 amps						
	1025	0	0	—	—	299,825
	1045	3.78	2.1	0.50	0.56	299,910
	1111	3.76	2.2	0.50	0.59	300,003
	1117	3.94	2.2	0.50	0.57	300,025
Pre-electrolysis - Plan - 10 amp. min. at 0.5 amps						
	1129					300,025
	1145	0.54	1.22	0.03	2.26	300,031
	1151	0.50	1.25	0.04	2.5	300,035
Electrorefining - Plan - 2010 amp. min. at 1.6 amps						
	1200					Set back EMF unit for 0.3V, 0.5 hr.
	1230	1.5	3.3	0.05	2.2	300,085
	1330	1.5	4.0	0.05	2.7	300,164
	1430	1.5	4.0	0.05	2.7	300,252
	1630	1.5	4.0	0.05	2.7	300,424
Recorder charts showed normal overnight operation						
May 11	0800	1.6	4.2	0.05	2.7	301,869
Run was terminated automatically by Back EMF sampling unit as ~ 0809						
	0810	—	—	—	—	301,879
	0815	Started terminal heat cycle				

Fig. A3.
Typical electrorefining data sheet.

H. Final Temperature Cycling.

Upon termination of the electrorefining, stop the stirrer.

Disconnect positive and negative dc leads.

Raise melt temperature to 840 °C.

Gently agitate surface of cathode metal pool by raising and lowering cathode cylinder for ~2 min; set cathode cylinder at normal refining height.

Raise stirrer from melt.

Hold furnace at 840 °C for 4 h.

Raise cathode cylinder from melt.

Shut off furnace.

I. Unloading.

Permit furnace to reach ambient temperature.

Unbolt and remove furnace tube head.

Remove nickel can.

Remove refining crucible, and record total cell weight.

Break crucible and salt away from product ring and anode residue.

Record weights of product, anode, stirrer, cathode, and pre-electrolysis rod.

Load product into casting furnace for final remelt. Casting procedure is same as above.

J. Sampling and Storing of Cast Product.

Cut two 1.5-g samples from top of casting and encapsulate both samples for nondestructive evaluation, i.e. neutron counting, gamma spectrum, and gamma dose rate.

Upon completion of nondestructive testing, submit one of the two capsules for carbon, radiochemical analysis, chemical plutonium, plutonium isotopic, and spectroquantitative analyses (see Fig. A4 for the complete analytical request form).

Have rest of product doubly encapsulated in welded tantalum containers. If metal must be stored in process lines, use special container designed for ^{238}Pu metal (LASL Dwg. 26 Y - 74556) and store metal in inert glove box.

Store tantalum containers in freezer.

III. PREPARATION OF $^{238}PUO_2^{16}$ POWDER (Flowsheet, Fig. 12)

A. General Discussion.

Preparation of $^{238}PuO_2^{16}$ has been carried out on a developmental basis, with only partial optimization of time cycles and equipment design. Procedures and equipment design from earlier ^{239}Pu compound preparations, such as ^{239}PuN, were modified for this work.

The use of a platinum boat in a quartz-Pyrex apparatus provides flexibility for changing conditions. Production work would probably better utilize a possibly less versatile, but also less fragile, apparatus designed for this particular preparation. The present apparatus, with some changes in the platinum boat design, has been used for preparations ranging from 10 to 160 g of plutonium.

Contemplated apparatus improvements include means for closer measurement of material temperature and apparatus pressure during dehydriding cycles.

B. Material Specifications.

^{238}Pu Metal – electrorefined
Platinum Boat – 0.010-in. sheet, $\geqslant$99.98% platinum
Gas Purity
  Helium–99.995% purity
  H_2–99.999% purity
H_2O^{16}
  distilled, no detectable NH_3
  ppm ^{18}O + ppm ^{17}O $\leqslant$ 20 ppm, ppm = g O isotope/10^6 g O.

C. Equipment.

The reaction tube assembly is shown in Fig. A5. It consists of a quartz reaction tube heated by a Hevi Duty Electric Co. furnace, Type M-3012. A platinum boat inside the reaction tube contains the ^{238}Pu. The reaction tube is provided with inlet and exhaust tubes for introduction and removal of gasses. Figure A6 is a schematic of the equipment, which consists of the reaction tube, part 1, also shown in Fig. A5; a multipurpose manifold and mercury-filled manometer (enclosed in dotted lines); a gas-purification system, part 3; a H_2O^{16} bubbler, part 4; and an off-gas system, part 5.

D. Conditioning and Degassing of Equipment.

Pack chip furnace with clean, dry uranium turnings.

After reconnecting furnace, evacuate gas-purification system, part 3, and lines leading to gas cylinders to a pressure of $<$ = 1 x 10^{-2} Torr.

Pressurize system with ~3-psig helium.

Heat chip furnace to 700 °C.

Place liquid N_2 in trap.

Pass helium gas to off-gas system.

Turn on reaction tube furnace.

Evacuate reaction tube and manifold system, part 2. Use heat gun on manifold tubing to accelerate degassing.

After apparent degassing, add several increments of helium and then H_2 to reaction tube held at 500 °C. Reevacuate.

Flow helium through $H_2^{16}O$ at 0.5 atm/liter-min and pass through hot reaction tube.

Flow dry helium through same path.

Conduct final, but short, degassing procedure.

E. Loading ^{238}Pu Metal.

Open reaction tube in good argon atmosphere ($<$10 ppm of O_2)

Place ^{238}Pu metal (30 g or less per chunk) giving equal spacing in platinum boat. Avoid contamination of platinum and boat with silicone grease.

Reseal tube with silicone high-vacuum grease and close stopcocks.

Place tube in furnace and reconnect to manifold.

Degas reconnected joint areas.

F. Preparation of ^{238}Pu Metal Powder.

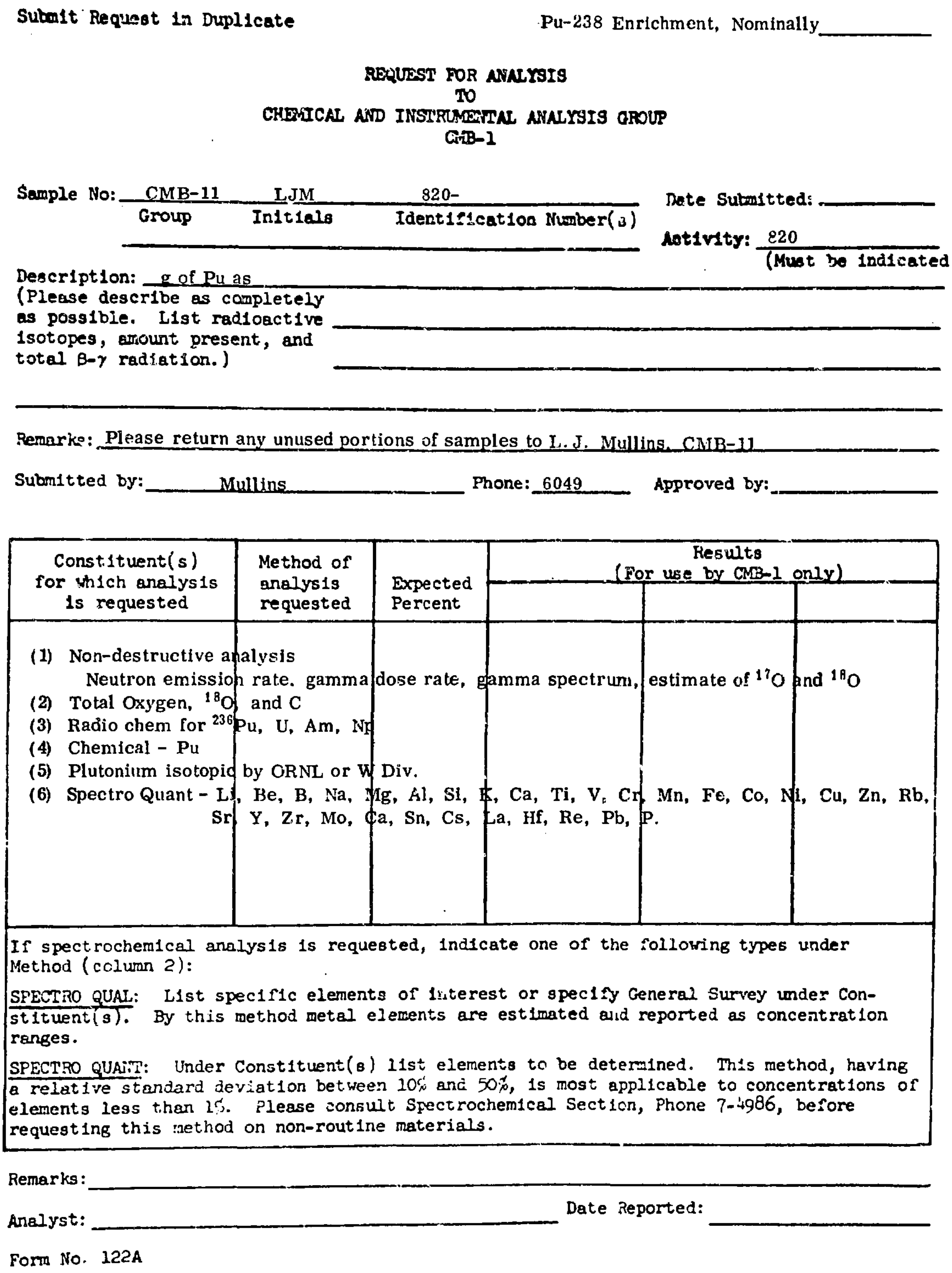
With reaction tube sealed off, purge manifold six times by alternately evacuating and filling with H_2 to 0.5 atm.

Evacuate reaction tube and seal off.

Add H_2 to manifold at ~0.1 atm/liter-min. (This flow rate is used for 120 g of plutonium. Faster rates can be tolerated for smaller batches, but care must be taken to ensure that the plutonium metal does not melt.)

Cautiously admit H_2 to reaction tube at the same rate.

When hydride formation is complete, as indicated by no further consumption of H_2, evacuate system using the roughing pump only, and heat rapidly to about 350 °C (furnace temperature) Heat more slowly (4 °C/min) to



Submit Request in Duplicate

Pu-238 Enrichment, Nominally ____________

REQUEST FOR ANALYSIS
TO
CHEMICAL AND INSTRUMENTAL ANALYSIS GROUP
CMB-1

Sample No: CMB-11 (Group) LJM (Initials) 820- (Identification Number(s))

Date Submitted: ____________

Activity: 820 **(Must be indicated)**

Description: g of Pu as
(Please describe as completely as possible. List radioactive isotopes, amount present, and total β-γ radiation.)

Remarks: Please return any unused portions of samples to L. J. Mullins, CMB-11

Submitted by: Mullins Phone: 6049 Approved by: ____________

Constituent(s) for which analysis is requested	Method of analysis requested	Expected Percent	Results (For use by CMB-1 only)		
(1) Non-destructive analysis Neutron emission rate, gamma dose rate, gamma spectrum, estimate of ^{17}O and ^{18}O (2) Total Oxygen, ^{18}O, and C (3) Radio chem for ^{236}Pu, U, Am, Np (4) Chemical - Pu (5) Plutonium isotopic by ORNL or W Div. (6) Spectro Quant - Li, Be, B, Na, Mg, Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Rb, Sr, Y, Zr, Mo, Ca, Sn, Cs, La, Hf, Re, Pb, P.					

If spectrochemical analysis is requested, indicate one of the following types under Method (column 2):

SPECTRO QUAL: List specific elements of interest or specify General Survey under Constituent(s). By this method metal elements are estimated and reported as concentration ranges.

SPECTRO QUANT: Under Constituent(s) list elements to be determined. This method, having a relative standard deviation between 10% and 50%, is most applicable to concentrations of elements less than 1%. Please consult Spectrochemical Section, Phone 7-4986, before requesting this method on non-routine materials.

Remarks: ____________

Analyst: ____________ Date Reported: ____________

Form No. 122A

Fig. [illegible]4.
Plutonium-238 analytical request form.

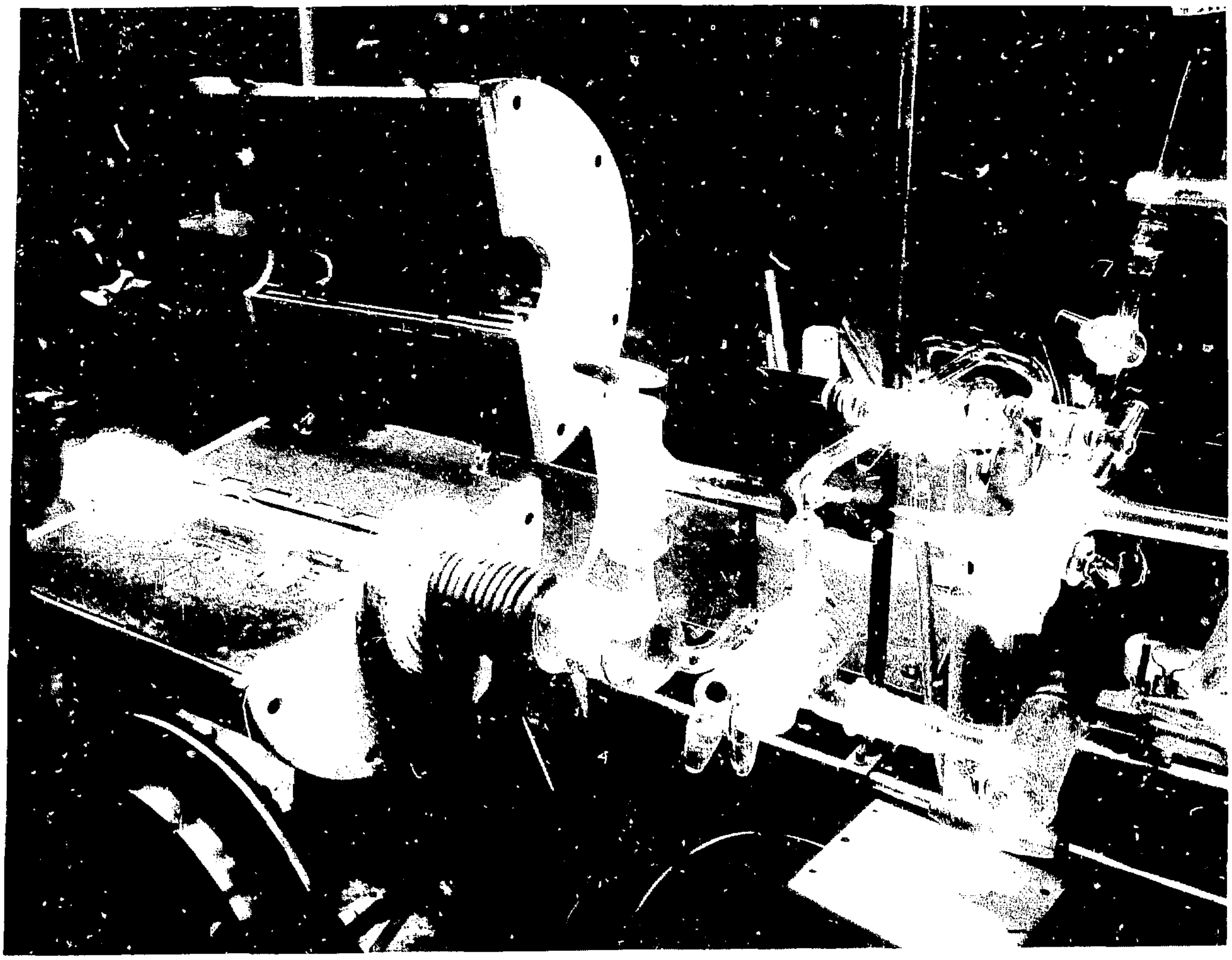


Fig. A5.
Reaction-Tube assembly for $^{238}PuO_2^{16}$ powder preparation.

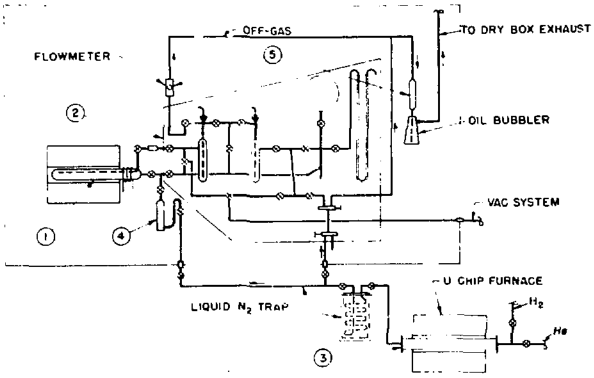


Fig. A6.
Schematic drawing of $^{238}PuO_2^{16}$ powder preparation equipment.

425-475 °C. Present equipment will easily dehydride 0.5 moles of Pu H_{2+x} in ~3 h.

Cut power and start rapid cooling when pressures fall to the 200- to 300-μ range.

For overnight storage add helium, but if hydriding will follow dehydriding the same day, keep the reaction tube cooled with an air blower.

For the second and third hydride formations, use the same techniques listed above.

For the second and third dehydriding cycles, furnace temperatures 5 - 15° higher than for the preceding dehydriding cycle are often needed to maintain the same rate of hydrogen evolution.

After the third dehydriding cycle, add the correct volume of H_2 to the dehydrided metal to provide an approximate overall composition of $PuH_{0.1}$. (This step, while quite necessary for the preparation of PuN, may not be essential for this preparation.)

G. Preparation of $^{238}PuO_2^{16}$ Powder.

Add helium to manifold, and evacuate.

Repeat purging, two to four cycles, as needed to produce a pressure, of $< 1 \times 10^{-3}$.

Slightly pressurize manifold with helium and continue flow through manifold to off-gas system for 5 to 10 min.

Switch helium flow through H_2O^{16} bubbler for an additional 10 min.

Repeat dry helium flow.

Close off-gas valve and fill reaction tube with dry helium to atmospheric pressure.

Flow dry helium to off-gas, switch to H_2O^{16} bubbler, and start He - H_2O^{16} flow over $^{238}Pu\ H_{0.1}$ at 0.5 to 1.0 atm/min.

Raise temperature at 4 °C/min to the maximum dehydriding temperature attained during normal dehydriding.

Continue (He - H_2O^{16}) addition until an approximate fourfold excess of H_2O^{16} has been added. The following emperical formula can be used to establish the duration of a run

$$\text{H of run} = \frac{(0.225)(\text{g Pu})}{\text{He flow rate}} ,$$

where the flow rate is in atmospheres per minute.

Maintain the furnace temperature for 75 - 80 min before stopping $H_2^{16}O$ addition. Raise the temperature at 8 - 9 °C/min to 700 °C and maintain for 45 min.

Flow dry helium over material for 20 min, then evacuate through a liquid N_2 trap for 20 min.

Cool reaction tube and fill with helium for storage until the unloading operation.

H. Sampling and Storage of $^{238}PuO_2^{16}$ Powder.

Unload quartz tube in a high-purity inert glovebox.

At present, samples are still taken because the project is still of an R&D nature.

Seal 5- and 2-g samples in double tantalum capsules. Submit the 5-g sample for a complete evaluation and the 2-g sample for nondestructive analysis only (see Fig. A4 for the analytical request form describing the complete chemical analysis).

Store remaining powder in stainless steel container in inert glovebox if it is to be used in ceramic fabrication within a week. If storage is to be longer than a week, have the oxide stored in welded stainless steel containers and store containers in ^{238}Pu vault.

IV. PREPARATION OF 50-WATT $^{238}PUO_2^{16}$ CYLINDERS (Fig.13)

The procedures given below are for the preparation of a 50-W cylinder of $^{238}PuO_2^{16}$ in which the ^{238}Pu isotopic enrichment is 80 at.%.

A. Ball Milling.

Transfer $^{238}PuO_2^{16}$ powder to the inert-ceramic-fabrication glovebox.

Load ~150 g of $^{16}PuO_2^{238}$ into Fisher Minimill No. 8403, stainless steel ball mill containing forty 0.625-in.-diam stainless steel balls.

Run mill overnight for a total of 18 h.

Unload mill carefully to keep "dusting" to a minimum.

Screen oxide through a 200-mesh stainless steel screen and transfer it to a tared stainless steel container.

Weigh the oxide.

B. Pressing.

Clean 1.0570-in.-diam die (LASL Dwg. 26Y-74572) thoroughly with 400-grit emery paper.

Lubricate die with TCE-graphite mixture; remove excess lubricant with cheesecloth.

Weigh out enough milled powder for the 50-W pellet and three control pellets (see below).

Load $^{238}PuO_2^{16}$ into die.

Press at 20 tsi using 30-ton hydraulic press. Note--for 20 tsi on the 1.057-in.-diam die, set pressure on 4-in.-diam ram at 2793 psi.

Remove pellet from die.

"Slug" pellet through 32-mesh stainless steel screen. Weigh the powder

Weigh out 127.2 g of the powder for the 50-W pellet, 5.5 g for one control pellet, and 2.0 g each for the other two control pellets. (The control pellets provide the samples for evaluation and analysis of the 50-W cylinder.) One of the pellets is stored as a library sample.

Store the powder for the 50-W pellet on the water-cooled chill block for 1 h.

Clean and lubricate die as described above.

Add Carbowax binder dissolved in TCE dropwise to the PuO_2 powder while stirring the oxide in a platinum dish. Five mg of Carbowax are added for each gram of oxide. The concentration of the solution is 30 mg Carbowax/ml TCE.

Load oxide into die.

Press at 20 tsi for 1 min.

Release pellet from die and store on water-cooled chill block for at least 1 h.

Add binder, as above, to the powders for the three control pellets.

Load powder into 0.285-in.-diam die and press at 20 tsi for 1 min.

For a pressure of 20 tsi on the 0.287-in.-diam die, set pressure on 4-in.-diam ram at 206 psi.

Release pellet from die.

Repeat procedure for the second and third control pellets.

C. Sintering.

Remove 50-W cylinder from copper chill block and place in platinum boat.

Place the three control pellets in platinum boat.

Load platinum boat into sintering furnace, and bolt end flange in place.

Start argon flow over the furnace windings. Turn on cooling water.

Start (Ar + H_2O^{16}) flow through furnace at 0.8 atm/min. (This is equivalent to 1 g H_2O^{16}/h.)

Use standard sintering cam on furnace controller, see Table AI for standard sintering cycle.

Turn on furnace power.

When sintering cycle has been completed and furnace has reached ~800 °C, replace (Ar + H_2O^{16}) flow with argon.

When furnace has reached ambient glovebox temperature, unload it as shown below.

D. Unloading, Pellet Measurements, and Sampling.

Unbolt end flange and remove platinum boat from furnace.

Weigh 50-W pellet and control pellets.

Measure diameter of 50-W pellet at top and bottom ends and in middle, measure height, record measurements to 0.001 in.

Weigh and measure dimensions of control pellets.

Calculate density of all pellets from the weights and dimensions.

$$\text{Density (g PuO}_2\text{/cm}^3 = \frac{\text{Wt. (g PuO}_2\text{)}}{12.864\ (\text{Diam, in.})^2\ (\text{Ht. in.})} \ .$$

Seal control pellets in tantalum capsules; submit largest pellet for complete evaluation (see Fig. A4) and the smaller pellets for nondestructive analysis only. One of the smaller pellets is saved as a library sample.

The 50-W pellet is usually encapsulated in a welded inner tantalum container and a welded outer stainless steel container. Tantalum felt is used to prevent the pellet from "rattling" during transport. Because requirements usually vary for each source, complete specifications will be given for each encapsulation.

TABLE AI

SINTERING CYCLE FOR $^{238}PuO_2^{16}$ CYLINDERS[a]

Time, h	°C
2	160
4	380
6	640
8	740[b]
10	760
12	770
14	780
16	780
18	870
20	1130
22	1500
24	1535
26	1535
28	1535
30	1535
32	1350
34	1100
36	900
38	840
40	780
42	630
44	450
46	260
48	Ambient glovebox Temperature

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[a] Furnace atmosphere is flowing argon gas saturated with H_2O^{16} vapor.

[b] For green cylinders contaminated with ^{18}O, the holding time at 740 °C is increased to ~96 h.

APPENDIX B

SUPPLEMENTAL TABLES

TABLE BI

PLUTONIUM MATERIAL BALANCE, CONVERSION OF ^{238}PuO$_2^{at}$ TO BOMB-REDUCED METAL

	Grams of Plutonium				
			Losses		
Lot	Oxide Feed	Samples	Peroxide Filtrate	Slag and Crucible	Pu Metal
LA 1[a]	29.6	1.4	0.1	8.2	19.9
LA 2	29.9	0.8	0.0	5.0	24.1
LA 3	29.7	2.6	0.6	26.5[b]	—[b]
LA 4	30.2	0.8	0.3	0.5	28.6
LA 5	29.6	5.2	0.2	0.1	24.1
LA 6	29.4	3.2	0.2	2.6	23.4
LA 7	30.7	0.6	0.3	1.0	28.8
LA 8	30.9	3.1	0.3	2.2	25.3
LA 9	31.3	4.3	0.2	2.1	24.7
HBO 907534[c]	201.1	6.2	1.3	13.4	180.2
HBO 907535	145.8	5.0	1.6	7.1	132.1
HBO 907538	198.0	6.4	3.3	8.7	179.6
HBO 907539	196.1	5.7	2.6	10.5	177.3
HBO 907546	199.9	5.3	2.6	4.5	187.5
HBO 907547	204.4	3.6	2.1	6.2	192.5
HBO 907551	198.9	5.6	1.6	9.4	182.3
HAS 1,3[d]	96.1	3.6	0.7	3.9	87.9
HAS 2	91.9	5.1	0.7	4.1	82.0
HAS 4	92.5	3.5	0.8	4.3	83.9
HAS 6	87.9	3.0	0.9	3.8	80.2
HAS 7, 10	150.7	8.0	2.4	1.3	139.0
Recycle 1[e]	—	—	4.4	4.1	97.1
2	—	—	2.1	5.5	232.4
3	—	—	1.5	1.4	59.0
4	—	—	0.4	0.2	44.3
5	—	—	0.5	4.6	37.5
6	—	—	2.5	4.2	86.5
7	—	—	3.8	3.7	76.8
8	—	—	2.8	3.1	67.5
9	—	—	0.4	1.3	45.9

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[a] LA lots were received from Mound Laboratory, nominal 80 at. % ^{238}Pu.

[b] Operator error caused reduction failure.

[c] HBO lots were received from Savannah River, nominal 80 at. % ^{238}Pu.

[d] HAS lots were received from Atlantic Richfield, Hanford, nominal 86-92 at. % ^{238}Pu.

[e] Recycle lots were process and experimental wastes generated in this program and processed to produce metal.

TABLE BII

PLUTONIUM MATERIAL BALANCE, ^{238}Pu ELECTROREFINING, 100-g SCALE

	Grams of Plutonium					
Run No.	Feed Metal	Anode Heel	Cathode	Pu Cl_3	Coll'n Loss	Product Metal
238-100-4	84.6	3.6	2.3	3.	11.4	64.3
5	94.0	3.4	1.9	3.	6.4	79.3
6	99.2	1.5	2.1	3.	5.9	86.7
7	77.9	2.5	4.0	2.6	11.9	56.9
8	87.2	2.3	4.6	3.	21.9	55.4
9	105.3	12.3	4.8	3.	22.3	62.9
10	86.1	13.2	2.7	3.	9.6	57.6
11	85.5	2.5	1.8	3.	17.2	61.0
12	81.9	8.3	2.9	3.	12.5	55.2
13	76.4	Stirrer broke, run aborted				
14	94.5	3.1	4.5	3.	6.1	77.8
15	98.6	7.6	2.4	3.	2.7	82.9
16	87.6	13.0	3.8	3.	9.6	58.2
17	104.6	3.9	2.3	3.	18.5	76.9
18	89.2	7.5	3.5	3.	4.7	70.5
19	96.2	3.2	4.5	3.	18.8	66.7
20	105.5	3.1	2.3	3.	16.	81.1
21	100.4	3.1	2.3	3.	20.4	71.6
22	104.2	3.4	2.0	3.	10.5	85.3
23	123.2	9.1	3.5	3.	14.3	93.3
24	107.8	5.7	2.2	3.	5.7	91.2
25	126.1	4.3	2.8	3.	20.6	95.4
26	94.7	4.2	3.5	3.	9.8	74.2
27	99.5	9.1	2.8	3.	4.4	80.2
28	98.4	2.2	2.5	3.	8.7	82.0
29	124.7	3.8	3.6	3.	11.1	103.2

TABLE BIII

PURITY OF CAST METAL FROM SMALL-SCALE, 10- to 20-g ELECTROREFINING RUNS

	Electrorefining Run Imparity (ppm)				
Element	LA-2	LAR-3	LA-4	LA-5	LA-9, LA-8
Li	< 0.02	< 0.005	< 0.01	0.01	0.8
Be	< 0.001	< 0.001	< 0.002	< 0.002	< 0.002
B	< 0.3	0.5	0.3	< 0.3	< 0.3
Na	1	1	4	< 6	< 6
Mg	5	20	6	6	< 4
Al	2	2	6	< 10	2
Si	15	10	55	25	20
K	0.5	< 0.5	< 1	2	2
Ca	6	3	20	10	< 6
Ti	0.4	0.5	1	< 0.8	< 0.6
V	< 0.5	< 0.5	< 1	< 1	< 1
Cr	0.5	1	2	< 2	< 1
Mn	0.6	0.7	0.5	1	0.8
Fe	< 35	< 35	40	20	< 40
Co	< 0.5	< 0.5	< 1	< 1	< 1
Ni	3	< 8	40	8	5
Cu	0.5	4	25	5	1
Zn	< 5	< 5	< 5	< 5	< 5
Rb	< 0.5	< 0.5	< 1	< 1	< 1
Sr	< 0.1	< 0.1	< 0.2	< 0.2	< 0.2
Y	< 0.1	< 0.1	< 0.2	< 0.2	< 0.2
Zr	< 0.1	< 0.1	< 0.2	< 0.2	< 0.2
Mo	< 0.5	2	< 1	< 1	1
Cd	< 0.5	< 0.5	< 1	< 1	< 1
Sn	2	< 0.5	< 1	< 1	2
Cs	< 2	< 2	< 4	< 4	< 4
Ba	< 0.1	0.5	< 0.2	1	< 0.2
La	< 1	< 0.5	< 2	< 2	< 1
Hf	< 0.5	< 0.5	< 1	< 1	< 1
Re	< 0.5	< 0.5	< 1	< 1	< 1
Pb	2	< 0.5	< 0.5	0.5	2
Bi	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
^{241}Am	4	2	4	0	0
^{237}Np	---	537	900	600	865
^{232}U	---	---	0.005	---	0.002
^{234}U	0	0	0	0	0

TABLE BIV

PURITY OF CAST METAL FROM 100-g ELECTROREFINING RUNS

Element	Electrorefining Run Impurity (ppm)											
	100-3	100-4	100-5	100-6	100-7	100-9	100-10	100-11	100-12	100-14	100-15	100-18
Li	0.01	< 0.005	< 0.005	0.06	1	< 0.005	0.02	0.03	1	0.005	0.008	0.1
Be	< 0.002	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.1
B	0.5	< 0.5	< 0.5	< 1	< 1	< 1	< 1	—	< 1	< 1	< 1	2
Na	8	< 5	< 2	2	20	2	—	—	—	< 1	< 1	8
Mg	2	2	24	< 1	8	2	—	—	10	—	5	22
Al	2	2	2	7	10	2	10	10	20	22	10	50
Si	< 5	5	12	45	10	< 5	20	10	6	—	5	< 5
K	< 1	< 1	2	9	10	< 1	—	—	—	—	4	21
Ca	< 20	< 6	1	8	6	< 2	8	7	25	—	20	20
Ti	1	0.5	0.3	< 0.2	0.2	< 0.2	0.3	1	2	2	< 0.2	5
V	< 1	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Cr	< 1	< 0.5	0.5	6	1	< 0.5	2	0.5	2	1	1	< 0.5
Mn	1	1	0.2	3	0.3	< 0.1	0.4	0.3	0.8	0.4	0.3	0.6
Fe	15	30	42	35	20	20	10	15	25	51	70	36
Co	< 1	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Ni	2	0.5	0.5	13	15	2	3	3	7	1	4	5
Cu	3	3	2	3	5	10	5	10	2	2	2	3
Zn	< 10	< 10	< 5	< 10	< 20	< 10	< 20	< 20	< 20	20	< 10	6
Rb	< 1	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Sr	< 0.2	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	0.1	0.3	< 0.1	< 0.1	< 0.1	< 0.1
Y	< 0.2	< 0.1	2	< 0.1	< 0.1	< 0.1	—	4	< 0.1	< 0.1	3	< 0.1
Zr	6	< 0.5	< 0.1	0.4	< 0.1	0.5	0.4	1	0.2	0.2	0.2	< 0.1
Mo	1	1	< 0.5	3	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Cd	< 1	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Sn	< 1	< 1	< 0.5	0.5	< 0.5	< 0.5	< 5	2	5	< 0.5	< 0.5	9
Cs	< 4	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2
Ba	< 4	0.6	< 0.1	0.1	< 0.1	< 0.1	0.8	0.8	1	0.3	0.3	2
La	< 2	< 1	< 0.5	< 1	< 1	< 1	< 1	< 0.5	< 0.5	< 1	< 1	< 1
Hf	< 1	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Re	< 1	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Pb	< 1	< 0.5	< 0.5	1	1	< 0.5	5	4	1	1	1	< 0.5
Bi	< 0.5	< 0.5	< 0.5	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
^{236}Pu	0.50	0.56	0.57	0.52	0.31	0.86	0.93	0.91	0.96	0.28	0.28	0.76
^{241}Am	1	0	2	0	0	0	0	9	17	4	0	0
^{237}Np	940	1630	1300	1870	1270	261	413	448	207	300	250	170
^{232}U	0.008	0.002	0	0.004	0.001	0.007	0.000	0.004	0.001	0.000	0.003	0.020
^{234}U	115	48	0	500	45	150	0	100	0	0	0	0
Pu, wt%	99.9	100.0	100.0	100.0	99.5	—	—	—	—	99.9	100.0	99.8

	Electrorefining Run Impurity (ppm)										
Element	100-19	100-22	100-23	100-24	100-25	100-26 Top	100-26 Bottom	100-27 Top[a]		100-27 Bottom	100-29
Li	0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Be	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
B	2	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 2
C	—	40	45	80	30	40	40	45	50	50	55
Na	3	< 1	< 1	5	5	< 1	< 1	< 1	< 1	< 1	< 1
Mg	21	< 1	< 1	5	4	5	8	6	4	10	< 1
Al	40	10	20	5	10	< 1	< 1	< 1	8	< 1	1
Si	—	< 25	< 25	5	5	20	25	20	25	25	10
K	8	< 0.5	< 0.5	< 1	< 1	10	1	3	4	3	< 0.5
Ca	19	< 3	< 3	< 3	< 3	< 3	< 3	< 3	< 3	< 3	< 3
Ti	2	< 0.2	< 0.2	0.5	2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
V	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Cr	< 0.5	1	8	1	1	< 0.5	< 0.5	2	< 0.5	4	7
Mn	1	< 0.1	2	1	2	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Fe	28	< 2	35	60	50	50	50	50	50	50	25
Co	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 1
Ni	11	1	7	2	4	1	4	< 0.5	< 0.5	< 0.5	< 0.5
Cu	7	< 0.2	3	3	1	4	1	3	3	2	3
Zn	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5	< 5
Rb	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Sr	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Y	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
Zr	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	5	< 0.1	< 0.1
Mo	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Cd	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Sn	3	< 0.5	< 0.5	< 0.5	< 0.5	5	1	5	3	1	7
Cs	< 2	< 2	< 2	< 2	< 0.5	< 2	< 2	< 2	< 2	< 2	< 2
Ba	1	< 0.1	< 0.1	6	4	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1	< 0.1
La	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Hf	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Re	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Pb	< 0.5	< 0.5	< 0.5	1	1	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
Bi	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 0.5	< 1	< 1	—
^{236}Pu	0.76	0.72	0.69	0.67	0.67	0.25	0.24	0.23	—	0.22	0.62
^{241}Am	7	0	11	3	9	28	26	19	—	15	10
^{237}Np	410	293	563	510	923	370	370	950	—	745	910
^{232}U	0.040	0.024	0.035	0.028	0.031	0.005	0.006	0.004	—	0.003	0.020
^{234}U	110	56	165	78	125	270	292	300	—	250	338
Pu, wt%	100.0	100.0	100.0	100.0	—	100.0	99.6	100.1	—	99.4	100.0

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[a] Two samples taken from top.

TABLE BV

MEASURED NEUTRON EMISSION RATES OF ELECTROREFINED METAL

Run No.	Capsule No.	Pu (g)	Isotopic Comp. (wt%) ^{238}Pu	^{240}Pu	Neutron Emission Rate n/sec-g Pu	n/sec-g ^{238}Pu
LA 2	6	10.639	79.79	2.90	2450	3070
LAR 3	14	14.07	79.48	2.96	2400[a]	3020
	29	12.61	79.48	2.96	2380[a]	2994
LA 4	12	7.38	79.63	2.94	2784[a]	3496
LA 5	10	8.60	79.82	2.98	2680[a]	3358
LA 98	34	15.46	79.71	2.87	2515[b]	3155
	36	13.33	79.71	2.87	2486	3119
100-3	64	27.309	80.07	3.06	2490[b]	3110
	65	26.810	80.07	3.06	2410	3010
	67	10.526	80.07	3.06	2508[b]	3132
	68	13.028	80.07	3.06	2495[b]	3116
	69	11.878	80.07	3.06	2465	3078
	71	11.686	80.07	3.06	2475	3091
100-4	73	24.657	80.21	3.06	2497	3113
	74	29.569	80.21	3.06	2605	3248
	83	14.215	80.21	3.06	2565	3198
	84	15.102	80.21	3.06	2595	3235
	85	11.277	80.21	3.06	2540	3167
	86	13.198	80.21	3.06	2496	3112
100-5	82	7.876	80.08	3.07	2490	3109
100-6	95	14.812	73.02	3.19	2350[b,c,d,e]	3218
100-7	100	16.356	87.03	1.91	2690[b]	3091
	169	13.43	86.99	1.99	2620	3012
100-7	170	9.842	86.99	1.99	2730[b]	3138
	171	10.668	86.99	1.99	2820[b,c,d]	3242
100-8	126	38.542	—	—	2590[b,e]	—
100-9	131	12.744	79.85	2.81	2460	3081
100-10	153	27.060	79.85	2.77	2560[c]	3206
100-11	154	18.486	76.66	3.05	2410[b,e]	3144
	155	16.43	76.66	3.05	2520[b,d,e]	3287
	156	18.22	76.66	3.05	2440[e]	3183
100-12	159	0.936	79.79	2.80	2520	3158
	160	12.205	79.79	2.80	2450	3070
	161	13.966	79.79	2.80	2430	3045
	162	13.51	79.79	2.80	2460	3083
	163	9.283	79.79	2.80	2550[b]	3196
	179	1.272	79.79	2.80	2494[c]	3126
	180	0.9870	79.79	2.80	2407[c]	3017
	181	1.313	79.79	2.80	2412[c]	3023

TABLE BV (cont.)

			Isotopic Comp. (wt%)		Neutron Emission Rate	
Run No	Capsule No.	Pu (g)	^{238}Pu	^{240}Pu	n/sec-g Pu	n/sec-g ^{238}Pu
100-14	215	1.122	86.75	1.98	2680[e]	3089
100-15	216	1.342	87.54	1.85	2750	3141
	217	18.529	87.54	1.85	2740	3130
	218	21.508	87.54	1.85	2750[b]	3141
100-16 100-17	234	1.907	79.99	2.71	2460[b,e,f]	3075
100-18	239	1.364	79.95	2.71	2540[c,d]	3177
100-19	240	1.434	80.02	2.71	2480[f]	3099
100-20	249	1.287	79.38	2.97	2440[e]	3074
	250	60.40	79.38	2.97	2390[e]	3011
100-21	251	1.51	79.93	2.76	2450[c,e]	3065
100-22	304	1.56	79.60	2.75	2550[c,d]	3204
100-23	303	1.43	80.14	2.71	2420[c]	3020
100-24	316	1.465	80.14	2.71	2450[c]	3057
100-25	317	1.504	80.11	2.72	2480[c]	3096
100-26	328-2	1.296	86.64	2.27	2660[c]	3070
	329-2	1.531	86.64	2.27	2640[c]	3047
	333	30.370	86.64	2.27	2760	3186
	334-1	30.028	86.64	2.27	2826[c]	3262
100-27	326-2	1.373	86.40	2.26	2670	3090
	327-2	1.367	86.40	2.26	2740[c]	3171
	330-1	32.246	86.40	2.26	2760	3194
	331	30.777	86.40	2.26	2781	3219

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[a] No gamma-spectrum data available.

[b] N impurity detected in gamma spectrum

[c] Na impurity detected in gamma spectrum;

[d] Cl impurity detected in gamma spectrum.

[e] Chemical analysis data not complete.

[f] Al impurity detected in gamma spectrum.

TABLE BVI

CHEMICAL PURITY OF ELECTROREFINED METAL FEED, $^{238}PuO_2^{16}$ POWDER, MILLED POWDER, AND SINTERED 1-W $^{238}PuO_2^{16}$ PELLETS

	Impurity (ppm)								
	Powder Prep. LAS 3		Powder Prep. LAS 6				Powder Prep. LAS 7		
Element	Metal	Powder	Metal	Powder	Milled Powder	Pellet[a]	Metal	Powder	Milled Powder[b]
Li	< 0.005	0.2	< 0.005	< 0.01	< 0.01	0.2	< 0.03	0.4	0.1
Be	< 0.001	< 0.002	< 0.001	< 0.002	< 0.2	< 0.2	< 0.001	< 0.002	< 0.002
B	< 0.5	3	< 1	1	2	2	< 1	2	< 1
C	—	—	—	—	—	120	—	—	—
Na	< 2	4	2	6	36	22	—	36	25
Mg	24	20	2	40	25	12	—	15	34
Al	2	20	1	8	16	40	10	15	34
Si	12	15	< 5	< 5	35	60	14	10	110
K	2	6	< 1	2	4	< 1	20	20	8
Ca	1	12	< 2	15	12	< 6	7	20	65
Ti	0.3	5	< 0.2	2	6	19	1	2	10
V	< 0.5	< 1	< 0.5	< 1	< 1	< 1	< 0.5	< 1	< 1
Cr	0.5	15	< 0.5	2	40	56	1	2	140
Mn	0.2	2	< 0.1	5	5	8	0.3	0.3	12
Fe	42	10	20	75	220	130	14	20	500
Co	< 0.5	< 1	< 0.5	1	< 1	< 1	< 0.5	< 1	< 1
Ni	0.5	< 5	2	7	40	25	3	3	100
Cu	2	5	10	20	22	10	10	5	165
Zn	< 5	20	< 10	35	25	< 10	< 20	< 20	60
Rb	< 0.5	< 1	< 0.5	< 1	< 1	< 1	< 0.5	< 1	< 1
Sr	< 0.1	< 0.2	< 0.1	< 0.2	< 0.2	< 0.2	0.2	< 0.2	< 0.2
Y	2	< 0.2	< 0.1	< 0.2	< 0.2	< 0.2	4	< 0.2	< 0.2
Zr	< 0.1	2	0.5	< 0.2	< 0.2	< 0.2	1	0.4	0.4
Mo	< 0.5	2	< 0.5	< 1	10	2	< 0.5	< 1	24
Cd	< 0.5	< 1	< 0.5	< 1	< 1	< 1	< 0.5	< 1	1
Sn	< 0.5	< 1	< 0.5	3	15	8	< 2	5	> 200
Cs	< 2	< 4	< 2	< 4	< 4	< 4	< 2	< 4	< 4
Ba	< 0.1	1	< 0.1	< 0.2	1	< 0.4	0.8	2	9
La	< 0.5	< 2	< 1	< 2	< 2	< 2	< 0.5	< 2	< 2
Hf	< 0.5	< 1	< 0.5	< 1	< 1	< 1	< 0.5	< 1	< 1
Re	< 0.5	< 1	< 0.5	—	< 1	< 1	< 0.5	< 1	< 1
Pb	< 0.5	8	< 0.5	< 1	2	3	4	3	20
Bi	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Np	1300	1322	550	564	494	500	415	396	384

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[a] Control pellet for 30-W source, 30-3.

[b] Initial attempts to press a 30-W pellet were unsuccessful. Powder was sintered at 1600 °C for 12 h and remilled.

TABLE BVI (cont.)

	Impurity (ppm)						
	Powder Prep. LAS 8				Powder Prep. LAS 9		
Element	**Metal**	**Powder**	**Milled Powder**	**Pellet**[c]	**Powder**	**Milled Powder**	**Pellet**[d]
Li	0.008	0.5	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Be	< 0.001	< 0.002	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
B	< 1	1	1	< 1	2	2	2
C	—	15	180	140	—	150	100
Na	< 1	11	50	40	< 2	120	60
Mg	5	< 2	8	8	10	20	50
Al	10	20	25	40	25	45	60
Si	5	10	40	15	20	75	180
K	4	2	4	2	< 1	20	1
Ca	20	10	30	8	< 6	20	50
Ti	< 0.2	2	1	< 0.4	< 0.4	< 0.4	< 0.4
V	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Cr	1	1	55	60	6	75	75
Mn	0.3	0.4	3	4	1	3	9
Fe	70	20	245	150	30	200	270
Co	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Ni	4	7	20	8	33	25	10
Cu	2	4	100	10	< 10	80	50
Zn	< 10	< 10	50	< 10	< 20	< 20	20
Rb	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Sr	< 0.1	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Y	3	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Zr	0.2	0.8	8	4	< 0.2	3	3
Mo	< 0.5	1	20	13	< 1	< 1	40
Cd	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Sn	< 0.5	< 5	50	< 5	< 1	10	5
Cs	< 2	< 4	< 4	< 4	< 4	< 4	< 4
Ba	0.3	3	3	1	< 0.2	< 0.2	10
La	< 1	< 2	< 2	< 2	< 1	< 1	< 2
Hf	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Re	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Pb	1	2	< 1	< 1	1	2	< 1
Bi	< 1	< 1	< 1	< 1	< 1	—	< 1
Np	250	210	255	270	195	260	240

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[c] Control pellet for 30-W source, 30 C6.

[d] Control pellet for 30-W source, 50 C2.

TABLE BVI (cont.)

	Impurity (ppm)						
	Powder Prep. LAS 10				Powder Prep. LAS 11		
Element	Metal	Powder	Milled Powder	Pellet[e]	Powder	Milled Powder	Pellet[f]
Li	0.1	< 0.2	0.2	< 0.2	0.1	0.2	< 0.2
Be	< 0.1	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
B	2	< 1	< 1	1	< 2	< 1	3
C	—	70	85	—	130	140	—
Na	< 1	1	8	50	9	22	20
Mg	< 1	2	16	10	< 2	< 2	20
Al	< 10	< 10	< 10	20	10	16	25
Si	< 5	15	20	100	< 5	20	< 25
K	< 3	1	3	2	< 1	< 1	< 1
Ca	< 3	< 10	< 10	14	< 6	< 6	< 6
Ti	4	3	4	4	1	4	< 0.4
V	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Cr	< 0.5	< 1	45	50	2	35	44
Mn	1	1	3	4	0.4	2	2
Fe	32	22	180	120	< 2	130	40
Co	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Ni	8	< 1	24	10	4	20	25
Cu	5	2	200	< 0.2	2	60	< 10
Zn	5	< 10	20	< 10	< 10	30	< 20
Rb	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Sr	< 0.1	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Y	< 0.1	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Zr	< 0.1	< 0.2	2	4	< 0.2	0.2	1
Mo	< 0.5	< 0.2	< 1	10	1	2	48
Cd	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Sn	6	< 1	100	< 5	< 5	200	6
Cs	< 2	< 4	< 4	< 4	< 4	< 4	< 4
Ba	2	< 0.2	< 0.2	0.2	< 0.4	< 0.4	< 0.2
La	< 1	< 2	< 2	< 2	< 2	< 2	< 1
Hf	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Re	< 0.5	< 1	< 1	< 1	< 1	< 1	< 1
Pb	< 0.5	2	7	< 1	2	3	2
Bi	< 1	< 1	< 1	< 1	< 1	< 1	—
Np	300	285	—	307	880	860	—

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[e] Control pellet for 50-W source, 50-3.

[f] Control pellet for 50-W source, 50-4.

TABLE BVI (cont.)

	Impurity (ppm)					
	Powder Prep. 50-5		Powder Prep. LAS 12		Powder Prep. LAS 13	
Element	Milled Powder	Pellet[g]	Metal	Powder	Metal	Powder
Li	< 0.2	< 0.2	< 0.1	< 0.2	< 0.1	< 0.2
Be	< 0.2	< 0.2	< 0.1	< 0.2	< 0.1	< 0.2
B	< 2	< 1	< 1	1	< 1	< 1
C	190	320	45	150	60	55
Na	25	50	< 1	20	5	20
Mg	< 2	30	< 1	10	5	5
Al	65	45	15	3	7	1
Si	20	100	< 25	20	< 1	20
K	25	< 1	< 0.5	< 1	< 3	< 1
Ca	30	6	< 3	< 6	< 3	< 6
Ti	< 0.4	< 0.4	< 0.2	12	1	20
V	< 1	< 1	< 0.5	< 1	< 0.5	< 1
Cr	100	200	6	6	1	8
Mn	8	40	1	3	1	2
Fe	400	500	20	85	55	80
Co	< 1	< 1	< 0.5	< 1	< 0.5	< 1
Ni	56	145	5	10	3	10
Cu	220	3	2	4	2	5
Zn	300	< 10	< 5	< 10	< 5	< 10
Rb	< 1	< 1	< 0.5	< 1	< 0.5	< 1
Sr	< 0.2	< 0.2	< 0.1	< 0.2	< 0.1	< 0.2
Y	< 0.2	< 0.2	< 0.1	< 0.2	< 0.1	< 0.2
Zr	< 0.2	< 0.2	< 0.1	< 0.2	< 0.1	< 0.2
Mo	< 1	< 1	< 0.5	< 1	< 0.5	< 1
Cd	< 1	< 1	< 0.5	< 1	< 0.5	< 1
Sn	100	2	< 0.5	2	< 0.5	1
Cs	< 4	< 4	< 2	< 4	< 2	< 4
Ba	4	5	4	10	5	2
La	< 2	< 2	< 1	< 2	< 1	< 2
Hf	< 1	< 1	< 0.5	< 1	< 0.5	< 1
Re	< 1	< 1	< 0.5	< 1	< 0.5	< 1
Pb	12	2	1	2	1	2
Bi	—	< 1	< 1	< 1	< 1	< 1
Np	—	—	470	—	664	—

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[g] Control pellet for 50-W source, 50-5.

TABLE BVII

COMPOSITION OF METAL FEED FOR $^{238}PuO_2^{16}$ PREPARATION

$^{238}PuO_2^{16}$ Prep. No.	Plutonium Metal Feed		
	ER Run No.	Pu (g)	Total Pu (g)
LAS 3	100-5	73.1	73.1
LAS 6	100-8	38.5	84.2
	100-9	45.7	
LAS 7	100-10	27.1	80.2
	100-11	53.1	
LAS 9	100-12	12.2	127.2
	100-16, 17	115.0	
LAS 10	100-18	54.9	120.6
	100-19	65.7	
LAS 11	100-20	60.2	127.1
	100-21	66.9	
LAS 12	100-22	49.6	137.5
	100-23	87.9	
LAS 13	100-24	86.3	137.7
	100-25	51.4	

TABLE BVIII

CHEMICAL PURITY OF 30-W $^{238}PuO_2^{16}$ CYLINDERS

	Impurity (ppm)					
Element	30-1	30-2	30-3	30-4	30-5	30-6
Li	< 0.5	<0.01	0.2	0.4	0.1	< 0.2
Be	< 1	<0.002	< 0.2	<0.002	< 0.002	< 0.2
B	10	<1	2	1	< 1	< 1
C	—	—	120	—	160	140
Na	20	2	22	30	25	40
Mg	10	30	12	20	34	8
Al	40	75	40	80	34	40
Si	330	220	60	140	110	15
K	—	2	< 1	20	8	2
Ca	50	20	< 6	55	65	8
Ti	—	23	19	13	10	< 0.4
V	—	<1	< 1	<1	< 1	< 1
Cr	35	250	56	80	140	60
Mn	15	10	8	9	12	4
Fe	600	800	130	400	500	150
Co	—	<1	< 1	<1	< 1	< 1
Ni	50	70	25	80	100	8
Cu	20	50	10	225	165	10
Zn	<20	65	< 10	200	60	< 10
Rb	—	<1	< 1	<1	< 1	< 1
Sr	< 5	<0.2	< 0.2	<0.2	< 0.2	< 0.2
Y	—	<0.2	< 0.2	<0.2	< 0.2	< 0.2
Zr	—	<0.2	< 0.2	0.4	0.4	4
Mo	—	18	2	12	24	18
Cd	< 5	<1	< 1	10	< 1	< 1
Sn	20	50	8	110	>200	< 5
Cs	—	<4	< 4	<4	< 4	< 4
Ba	—	4	< 0.4	4	9	1
La	—	<2	< 2	<2	< 2	< 2
Hf	—	<1	< 1	<1	< 1	< 1
			<			
Re	—	<1	< 1	<1	< 1	< 1
Pb	30	5	3	9	9	< 1
Bi	< 1	<1	< 3	<1	< 1	< 1
Np	1974	1020	500	870	384	270

TABLE BIX

PROPERTIES OF 30-W $^{238}PuO_2^{16}$ CYLINDERS

Property	30-1[a]	30-2[a]	30-3[b]	30-4[a]	30-5[a]	30-6[b]
Wt of fuel pellets (g)	72.36	74.08	74.76	74.71	73.88	74.69
Wt of Pu (g)	63.7[c]	65.2[d]	65.9[d]	65.6[d]	65.0[d]	65.4[c]
Oxygen/Pu ratio	2.00	—	—	—	—	2.01
Pu isotopic						
Wt % ^{238}Pu	79.99	80.06	79.82	80.06	79.72	86.90
ppm ^{236}Pu	0.60	0.92	0.70	0.92	0.93	0.25
Dimensions						
Diam (in.)	0.862	0.849	0.846	0.836	0.843	0.858
Height (in.)	0.796	0.770	0.803	0.804	0.784	0.808
Density (g PuO_2/cm^3)	9.51	10.38	10.11	10.34	10.31	9.77
Power (w)						
Calculated	29.0	30.0	30.1	30.1	29.5	32.2
Measured[e]	28.6	—	29.8	29.6	29.3	32.2
Power density[f] (W/cm^3)	3.76	4.20	4.03	4.09	4.09	4.20
Neutron emission rate						
n/sec-g Pu	3240	3380	2880	3790	3360	3060
n/sec-g ^{238}Pu	4050	4222	3608	4734	4215	3521
Dose rates at 10 cm						
Neutron, mRem/h-g ^{238}Pu	0.41	0.42	0.36	0.47	0.42	0.35
Camma mR/h-g ^{238}Pu	0.20[g]	0.19[g]	0.14	0.15[g]	0.14	0.17

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[a] Cylinder not fabricated as per standard flow sheets, Figs. 12 and 13.

[b] Cylinder fabricated as per standard flow sheets, Figs. 12 and 13.

[c] Pu by chemical titration.

[d] Pu by difference (i.e., $(PuO_2)(0.8815) - \Sigma$ chemical impurities.

[e] Measured power corrected to date of preparation of cylinder.

[f] Measured power data used if available.

[g] Measured through 0.068-in. Ta-10W and 0.025-in. Hastelloy, C-276.

TABLE BX

PLUTONIUM ISOTOPIC COMPOSITION OF 30-W $^{238}PuO_2^{16}$ CYLINDERS

Pu Isotope	Weight Percent (g Pu isotope/100g Pu) 30-1	30-2	30-3	30-4	30-5	30-6
236	0.60×10^{-4}	0.96×10^{-4}	0.63×10^{-4}	0.96×10^{-4}	0.93×10^{-4}	0.25×10^{-4}
238	79.99	80.06	79.82	80.06	79.72	86.90
239	16.50	15.82	16.21	15.82	16.07	10.67
240	2.85	2.73	2.92	2.73	2.79	1.95
241	0.53	1.02	0.80	1.02	1.03	0.41
242	0.13	0.37	0.25	0.37	0.39	0.07

TABLE BXI

CHEMICAL PURITY OF 50-W $^{238}\text{PuO}_2^{16}$ CYLINDERS

	Impurity (ppm)				
Element	**50-1[a]**	**50-2[b]**	**50-3[b]**	**50-4[b]**	**50-5[a]**
Li	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Be	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
B	< 1	2	1	3	< 1
C	180	100	---	---	320
Na	3	60	50	20	50
Mg	12	59	10	20	30
Al	300	60	20	25	45
Si	450	180	100	< 25	100
K	2	1	2	< 1	< 1
Ca	400	50	14	< 6	6
Ti	11	< 0.4	4	< 0.4	< 0.4
V	< 1	< 1	< 1	< 1	< 1
Cr	160	75	50	44	200
Mn	15	9	4	2	40
Fe	270	270	120	40	500
Co	< 1	< 1	< 1	< 1	< 1
Ni	25	10	10	25	145
Cu	2	50	< 0.2	< 10	3
Zn	< 10	20	< 10	< 20	< 10
Rb	< 1	< 1	< 1	< 1	< 1
Sr	6	< 0.2	< 0.2	< 0.2	< 0.2
Y	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Zr	7	3	4	1	< 0.2
Mo	22	40	10	48	< 1
Cd	< 1	< 1	< 1	< 1	< 1
Sn	< 1	5	< 5	6	2
Cs	< 4	< 4	< 4	< 4	< 4
Ba	9	10	0.2	< 0.2	5
La	< 2	< 2	< 2	< 1	< 2
Hf	< 1	< 1	< 1	< 1	< 1
Re	< 1	< 1	< 1	< 1	< 1
Pb	3	< 1	< 1	2	2
Bi	< 1	< 1	< 1	---	< 1
Np	1411	240	307	835	390

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[a] Cylinder not prepared as per standard flowsheets, Figs. 12 and 13.

[b] Cylinders prepared as per standard flowsheets, Figs. 12 and 13.

TABLE BXII

OXYGEN-18 CONCENTRATION OF $^{238}\text{PuO}_2^{16}$ POWDER, MILLED POWDER, AND SINTERED CONTROL PELLETS (50-W PREPARATION)

	ppm ^{18}O[a]		
Prep. No.	**Powder**	**Milled Powder**	**Pellets[b]**
50-1	220	280	13
50-2	24	75	18
50-3	10	160	10
50-4	15	65	10
50-5	---	65	---

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[a] g ^{18}O/10^{6} g O.

[b] Analysis of control pellet.

TABLE BXIII

NEUTRON EMISSION RATES OF $^{238}\text{PuO}_2^{16}$ POWDER, MILLED POWDER, AND SINTERED CONTROL PELLETS (50-W PREPARATIONS)

	n/sec-g ^{238}Pu			
Preparation	**Metal**	**Powder**	**Milled Powder**	**Control Pellet**
50-1[a]	—	4573	4836	3609 3596
50-2[b]	3084	3300	4000	3387 3374
50-3[b]	3125	3338	4114	3350 —
50-4[b]	3070	3291	3504	3341 3328
50-5[a]	—	—	4222	3456 3430

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[a] Pellets not prepared as per standard flow sheets, Figs. 12 and 13.

[b] Pellets prepared as per standard flow sheets, Figs. 12 and 13.

TABLE BXIV

PROPERTIES OF 50-W^{238}PuO$_2^{16}$ CYLINDERS

	50-1[a]	50-2[b]	50-3[b]	50-4[b]	50-5[a]
Wt of fuel pellet (g)	123.09	125.08	125.27	124.68	125.37
Wt of Pu (g)	108	110	110	110	110
Oxygen/Pu ratio	—	—	—	1.99	2.00
Pu isotopic composition					
Wt%^{238}Pu[c]	79.81	79.72	79.98	79.62	79.58
ppm^{236} Pu[d]	0.85	0.76	0.75	0.58	0.70
Dimensions					
Diam (in.)	1.008	1.006	0.997	1.004	0.991
Height (in.)	0.978	0.996	1.003	1.013	1.037
Density (g PuO$_2$/cm^3)	9.63	9.65	9.75	9.49	9.56
Power (W)	48.8	49.7	49.8	49.6	49.6
Power density (W/cm^3)	3.82	3.83	3.88	3.78	3.78
Neutron emission rate					
n/sec-g Pu	3165	2690	2790	2760	2851
n/sec-g ^{238}Pu	3966	3374	3488	3466	3582
Dose rates at 10 cm					
Neutron, mRem/h-g ^{238}Pu	0.40	0.35	0.35	0.35	0.36
Gamma, mR/h-g ^{238}Pu	0.20	0.17	0.14	0.16	0.19

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[a] Cylinders not prepared as per standard flow sheets, Figs. 12 and 13.

[b] Cylinders prepared as per standard flow sheets, Figs. 12 and 13.

[c] g ^{238}Pu/100 g Pu.

[d] g ^{236}Pu/10^6 g Pu.

TABLE BXV

PLUTONIUM ISOTOPIC COMPOSITION OF 50-W CYLINDERS

Pu Isotope	Weight Percent (g Pu isotope/100 g Pu)				
	50-1	50-2	50-3	50-4	50-5
236	0.85 x 10^{-4}	0.76 x 10^{-4}	0.75 x 10^{-4}	0.58 x 10^{-4}	0.66 x 10^{-4}
238	79.81	79.72	79.98	79.62	79.58
239	16.03	16.20	15.97	16.36	16.36
240	2.81	2.76	2.73	2.83	2.76
241	0.98	0.96	0.95	0.84	0.94
242	0.37	0.36	0.37	0.30	0.36

TABLE BXVI

NEUTRON EMISSION RATES OF CONTROL PELLETS AND 50-W CYLINDERS

	n/sec-g ^{238}Pu	
Cylinder No.	Control Pellets	50-W Cylinders
50-1[a]	3609 3596	3966
50-2[b]	3387 3374	3374
50-3[b]	3350 —	3488
50-4[b]	3341 3328	3466
50-5[a]	3456 3430	3582

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[a] Cylinders and pellets not made as per standard flow sheets, Figs. 12 and 13.

[b] Cylinders and pellets made as per standard flow sheets, Figs. 12 and 13.

TABLE BXVII

CHEMICAL PURITY OF AS-RECEIVED OXIDE POWDERS
(Mound Laboratory Lots)

Element	Impurity (ppm)				
	LA 5	LA 6	LA 9	LA	LA 11
Li	0.04	0.02	0.1	< 0.01	0.2
Be	< 0.002	< 0.002	< 0.002	< 0.002	< 0.002
B	1	< 0.3	0.5	< 0.3	< 0.3
Na	24	38	28	18	38
Mg	22	25	6	16	4
Al	14	52	9	5	15
Si	45	95	55	45	110
K	6	8	3	1	3
Ca	78	180	N.A.[a]	14	44
Ti	5	4	44	84	12
V	< 1	< 1	< 1	< 1	< 1
Cr	15	60	25	20	15
Mn	30	20	3	6	6
Fe	80	1000	65	40	60
Co	< 1	< 1	< 1	< 1	< 1
Ni	25	70	15	10	15
Cu	24	21	14	4	3
Zn	10	5	5	< 5	< 5
Rb	< 1	< 1	< 1	< 1	< 1
Sr	1	4	< 0.2	< 0.2	< 0.2
Y	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Zr	4	1	< 0.2	< 0.2	< 0.2
Mo	12	4	3	4	2
Cd	< 1	10	< 1	< 1	< 1
Sn	10	10	10	3	3
Cs	< 4	< 4	< 4	< 4	< 4
Ba	6	6	< 0.2	< 0.2	< 0.2
La	< 2	< 1	< 1	< 1	< 1
Hf	< 1	< 1	< 1	< 1	< 1
Re	< 1	< 1	< 1	< 1	< 1
Pb	20	50	25	50	20
Bi	< 0.5	< 0.5	< 0.5	< 0.5	< 0.5
^{237}Np	1990	N.A.[a]	N.A.[a]	3780	3700

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[a] N.A. – No Analysis.

TABLE BXVIII

CHEMICAL PURITY OF AS-RECEIVED OXIDE POWDER
(Savannah River Lots)

	Impurity (ppm)							
Element	**907534**	**907535**	**907538**	**907539**	**907545**	**907546**	**907547**	**907551**
Li	0.2	0.02	0.06	< 0.01	0.2	< 0.1	< 0.2	< 0.2
Be	< 0.002	< 0.002	< 0.002	< 0.002	< 0.002	< 0.2	< 0.2	< 0.2
B	< 1	< 1	< 2	< 2	< 1	< 1	< 2	< 1
Na	4	100	40	20	4	28	< 2	< 2
Mg	15	10	8	10	30	38	< 2	< 2
Al	20	22	12	4	30	16	40	20
Si	< 5	15	45	30	20	20	40	< 25
K	6	2	20	6	6	4	< 1	< 1
Ca	150	62	280	300	160	160	40	100
Ti	2	2	5	13	7	6	0.6	< 0.4
V	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Cr	20	20	15	20	15	25	35	60
Mn	2	2	3	3	3	3	1	1
Fe	55	50	60	60	60	100	140	110
Co	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Ni	15	30	15	20	25	28	35	35
Cu	2	10	2	2	5	32	10	15
Zn	20	< 20	< 10	< 10	20	15	< 10	< 10
Rb	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Sr	5	5	10	10	6	4	5	5
Y	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Zr	3	1	< 0.2	< 0.2	8	< 0.2	< 0.2	< 0.2
Mo	2	< 1	4	4	2	< 1	< 1	50
Ag	< 3	—	—	—	< 3	—	—	—
Cd	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Sn	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Cs	< 4	< 4	< 4	< 4	< 4	< 4	< 4	< 4
Ba	7	7	9	8	7	8	6	8
La	< 2	< 2	< 2	< 2	< 2	< 2	< 2	< 2
Hf	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Re	< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Pb	20	18	25	27	20	20	25	25
Bi	< 1	< 1	< 1	< 1	< 1	< 1	—	—
^{237}Np	725	700	415	380	890	975	958	—
^{236}Pu	1.0	0.86	0.77	0.85	1.1	0.80	0.69	—

TABLE BXIX

CHEMICAL PURITY OF AS-RECEIVED OXIDE POWDER
(Atlantic Richfield, Hanford Co. Lots)

	Impurity (ppm)						
Element	**HAS 1**	**HAS 2**	**HAS 3**	**HAS 6**	**HAS 7**	**HAS 8**	**HAS 9**
Li	0.1	0.1	0.2	0.1	< 0.2	< 0.01	< 0.01
Be	< 0.002	< 0.002	< 0.002	< 0.002	< 0.2	< 0.002	< 0.002
B	2	1	3	< 1	< 1	3	3
Na	66	50	146	29	50	25	15
Mg	36	30	22	8	6	60	15
Al	14	50	12	16	30	35	> 100
Si	40	85	220	6	10	140	350
K	8	10	3	2	30	4	4
Ca	500	10	500	100	< 5	> 100	> 00
Ti	65	5	4	5	2	13	8
V	< 1	1	< 1	< 1	< 1	< 1	< 1
Cr	9	12	2	0.5	< 1	12	40
Mn	2	3	0.5	0.4	< 0.2	2	3
Fe	80	80	34	6	70	45	150
Co	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Ni	21	12	14	8	10	45	60
Cu	2	6	2	< 1	2	5	9
Zn	< 20	20	< 20	< 10	< 10	20	< 10
Rb	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Sr	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Y	< 0.2	< 1	< 0.2	< 0.2	< 0.2	< 0.2	< 0.2
Zr	1	< 0.2	3	< 0.2	< 0.2	7	< 2
Mo	< 1	< 1	< 1	< 1	< 1	4	4
Ag	—	50	—	—	—	—	—
Cd	< 1	< 1	< 1	< 1	< 1	< 1	< 1
					<		
Sn	8	20	< 1	8	< 1	15	20
Cs	< 4	< 4	< 4	< 4	< 4	< 4	< 4
Ba	7	< 0.2	3	3	< 0.2	2	3
La	< 2	< 2	< 2	< 2	< 2	—	< 2
Hf	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Re	< 1	< 1	< 1	< 1	< 1	< 1	< 1
Pb	6	20	11	7	12	20	50
Bi	< 1	< 1	< 1	< 1	< 1	< 0.5	< 0.5
^{237}Np	1560	1997	43	800	1070	285	35
^{236}Pu	0.24	0.32	0.30	0.36	0.28	0.35	0.32

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