LA-DC-72-473

MASTER

CONF-720519--2

CHARACTERIZATION AND PROPERTIES OF MEDICAL-GRADE ^{238}Pu FUELS

L. J. Mullins, G. M. Matlack, J. Bubernak, and J A. Leary
University of California
Los Alamos Scientific Laboratory
P. O. Box 1663
Los Alamos, New Mexico 87544

ABSTRACT

The ^{238}Pu requirements, 55 - 90g, for an artificial heart or circulatory assist device demand a fuel having minimal radiation properties. The preparation and evaluation of potential ^{238}Pu fuel forms at the Los Alamos Scientific Laboratory has led to the development of four fuel compositions, electrorefined metal, ^{238}Pu - 3at.%Ga, $^{238}PuN^{15}$, and $^{238}PuO_2{}^{16}$. (The latter three fuels are made from electrorefined metal.) Theoretical and experimental studies of these fuels led to the conclusion that $^{238}PuO_2{}^{16}$ is the preferred composition for high temperature application in 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 one to fifty watts.

DISCLAIMER

INTRODUCTION

The successful utilization of plutonium-238 in a radioisotopic 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 under any credible accident. To achieve these objectives, the Division of Isotopes Development USAEC, 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 to minimize fuel-container compatibility problems.

The radiation properties of ^{238}Pu fuel materials were calculated and reported early in this program.[1],[2] Penetrating radiations from ^{238}Pu fuels having the isotopic compositions 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 Pu isotopes and their daughters, and
6) photons that result from alpha particle reactions with light element impurities.

An irreducible neutron emission rate is associated with the spontaneous fission of ^{238}Pu and ^{240}Pu. For the spontaneous fission of ^{238}Pu, the average number of neutrons per fission is 2.33 (± 0.08)[3] and the half life is 5.0 (± 0.6) x 10^{10} yrs.[4] These values give a calculated value of 2586 (± 400) n/sec-g ^{238}Pu. Results obtained in the present study give a value of 2785 (± 55) n/sec-g ^{238}Pu for neutron emission resulting from the spontaneous fission of ^{238}Pu. Using this latter value and the emission rate of 1.02 x 10^3 n/sec-g ^{240}Pu for the spontaneous fission of ^{240}Pu a value of 2260 n/sec-g Pu is calculated for 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 Li, Be, B, C, O, F, Na, Mg, Al, and Si. The (α, n) contribution in conventional grade ^{238}Pu metal can vary from 9900 to 28,000 n/sec-g ^{238}Pu.[1],[5] These neutrons are due to light element impurities in the metal other than oxygen. 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 Pu isotopes, which, in turn, produce excess neutrons. Thus, there is a self-multiplication of the neutron rate. The magnitude of this multiplication will depend on the mass, density, and geometry of the Pu 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 observed from chemically pure ^{238}Pu. As the material ages, more gamma rays appear from daughter products of ^{236}Pu and ^{241}Pu. Gamma rays also arise from (α, n) or (α, p) reactions induced by alpha-particle bombardment of impurity

nuclides. This is particularly evident if the impurities are nitrogen or fluorine.

For medical applications, it is imperative that a fuel's radiation be understood quantitatively, and that it be kept to a minimum. The present program has been concerned with

(1) calculation of the radiation properties of ^{238}Pu fuel materials

(2) preparation and characterization of ^{238}Pu metal of high chemical purity

(3) preparation and characterization of ^{238}Pu compounds and alloys having minimal penetrating radiation

(4) measurement of the spontaneous fission rate of ^{238}Pu

(5) fabrication, characterization, and properties of thirty and fifty watt medical-grade ^{238}Pu fuels.

POTENTIAL MEDICAL-GRADE FUEL COMPOSITIONS

The ^{238} fuel compositions that have been considered for medical applications are listed in Table II. To achieve the minimum weight and volume, unalloyed ^{238}Pu metal would be the preferred form. The physical properties of the metal, however, are complex because it exists in six allotropic modifications between room temperature and its melting point, 640°C However, the metal can be stabilized in the delta phase by the addition of alloying agents such as Al, Ce, Ga, and Sc. The phase transitions for the delta stabilized Ga[6] and Sc[7] alloys are compared in Table III. (The ^{238}Pu - 10 at.% Sc alloy is the fuel used in the French pacemaker device[8].) Of the alloys listed in Table II, only the ^{238}Pu-3at.%Ga alloy has been characterized in the present study. The ceramic forms which have been considered are the carbide, nitride, and oxide. To achieve minimal radiation levels, special isotopic enrichments of carbon, nitrogen, and oxygen are required. (This point will be discussed more thoroughly in following sections.) 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 the prime fuel candidate for most medical applications. $^{239}PuO_2$ has been studied for several years as a fast reactor fuel and a considerable wealth of information has been generated on its chemical and physical properties.

The fuel forms which have been prepared, characterized, and evaluated are:

Elemental ^{238}Pu

^{238}Pu-3 at.%Ga

$^{238}PuO_2{}^{nat}$

$^{238}PuO_2{}^{16}$

$^{238}PuN^{nat}$

$^{238}PuN^{15}$

COMPARISON OF THE DOSE RATES OF ^{238}Pu FUEL FORMS

The gamma and neutron dose rates of the fuel forms which have been prepared and evaluated are compared in Table IV. 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 Ta 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 the metal. However, the gamma dose rate is approximately doubled as the result of an (α, p) reaction on ^{14}N yielding an excited state of ^{17}O. Replacement of ^{14}N by ^{15}N gives a compound having a gamma dose rate only 10% higher than the metal. (The slightly higher gamma dose rate of the nitride is due primarily to its lower density.) Both the neutron and gamma dose rates of $^{238}PuO_2{}^{nat}$

are significantly higher than the metal. However, $^{238}PuO_2$[16] has dose rates comparable to the metal. Thus, the fuel forms having low neutron and gamma dose rates are ^{238}Pu, ^{238}Pu-3 at.% Ga, ^{238}PuN[15] and $^{238}PuO_2$[16]. Thus so far as radiation is concerned, all of these materials would be suitable for implantable heat sources required to power pacemakers and artificial heart devices.

CHARACTERIZATION AND PROPERTIES OF ^{238}Pu METAL

All of the ^{238}Pu metal of high chemical purity was prepared by electrorefining. Procedures were developed for two scales of operation, 10 - 20 g Pu product, and 50 - 100g Pu product. Details of these processes have been reported elsewhere[9],[10].

Chemical Purity

The chemical purity of a typical lot of electrorefined metal is given in Table V. The total detectable impurities exclusive of Np are less than 100 ppm. If desired, the Np concentration in the product can be reduced by increasing the $PuCl_3$ concentration in the electrorefining electrolyte. However the rapid growth of ^{234}U in the product, (650 ppm/mth for a fuel containing 90 at.% ^{238}Pu), does not justify the sacrifice of $PuCl_3$.

Neutron Emission Rate

The neutron emission rate of ^{238}Pu metal is composed of contributions from spontaneous fission neutrons, $(\alpha - n)$ neutrons and fast fission neutrons. The contribution from fast fission will depend on the geometry and density of the fuel form. This contribution can be calculated and can be 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 having lengths equal to the diameters are given in Fig. 1[11] for Pu metal and for PuO_2 at densities of 11.47 and 9.75 g PuO_2/cc. The multiplication factor for a metal source having a density of 15.8 g Pu/cc varies from 1.07 for 13g Pu to 1.15 for 100 g Pu. The multiplication factor for 13g of Pu in the form of an oxide having a density of 9.75 g PuO_2/cc is 1.04. The multiplication factor increases to 1.08 for 100g Pu in the oxide form.

Measured neutron emission rates of various size samples are given in Table VI. For eighteen small samples containing 1-2 g Pu, the rate was 3095 (± 46) n/sec-g ^{238}Pu. As expected, the rate increased as the sample size increased.

Spontaneous Fission Rate of ^{238}Pu

A critical analysis[10] of all the data on electrorefined metal gives a value of 2785 (± 55) n/sec-g ^{238}Pu for neutrons resulting from the spontaneous fission of ^{238}Pu.

Gamma Radiation

The gamma ray spectrum of ^{238}Pu metal is given in detail in Reference [10].

The absolute intensities of the principal gamma rays from ^{238}Pu, were measured on thin unshielded sources. Electrorefined metal was dissolved in HCl and Am, Np, U, and ^{236}Pu daughter activities were removed by separations with anion and cation exchange resins. Two large volume, > 30 cc, Ge(Li) detectors were used to measure the gamma rays. Two identical aliquots were examined with each detector under the same geometry conditions as that used in calibrating the detectors with several standards. Average values of ^{238}Pu gamma ray intensities are given in Table VII. 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 might be somewhat poorer at lower energies because large absorption effects caused difficulties in interpreting the detector efficiency curve.

CHARACTERIZATION AND PROPERTIES OF ^{238}Pu-3 at.% Ga AND $^{238}PuN^{15}$

The addition of Ga to ^{238}Pu metal does not significantly affect the radiation properties of the fuel[9]. Neutron emission rates and gamma radiations are essentially identical for the metal and alloy.

Chemically pure ^{238}PuN containing the natural isotopic concentrations of ^{14}N and ^{15}N has a neutron emission rate equal to that of electrorefined metal[9]. The gamma dose rate, however, is higher by a factor of ~ 2.5[9] This increased rate is caused by the reaction $^{14}N(\alpha, p)^{17}O$. To eliminate this reaction, $^{238}PuN^{15}$ was prepared by reacting electrorefined ^{238}Pu metal with $^{15}N_2$ gas (99.7% ^{15}N, 0.3% ^{14}N). The chemical purity of the nitride was the same as the metal feed and the radiation properties were essentially identical. This can be seen in Table VIII where the radiation properties of $^{238}PuN^{15}$ are compared to its metal feed. Neutron dose rates are the same and the small difference in the gamma dose rate is due primarily to decreased self shielding by Pu atoms. The gamma spectrum of $^{238}PuN^{15}$ is essentially the same as the spectrum of electrorefined metal except for a small peak at 871 keV due to the presence of 0.3% ^{14}N in ^{15}N. The $^{238}PuN^{15}$ spectrum is compared to $^{238}PuN^{nat}$ in Fig. 2. 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 in the natural nitride. These photons are responsible for the high gamma dose rate of $^{238}PuN^{nat}$.

CHARACTERIZATION AND PROPERTIES OF $^{238}PuO_2^{16}$ POWDERS

All of the $^{238}PuO_2^{16}$ powders of high chemical purity were prepared by reacting electrorefined metal with H_2O^{16}. The preparation of $^{238}PuO_2^{16}$ powders by oxygen isotopic exchange of PuO_2^{nat} and H_2O^{16} has also been explored by the product is inferior to the oxide made from electrorefined metal[10].

Chemical Purity

The chemical purity of a typical lot of $^{238}PuO_2^{16}$ powder is compared to its electrorefined metal feed in Table IX. The purities compare favorably; only trace amounts of impurities are introduced during preparation of the oxide powder.

Neutron Emission Rate

The neutron emission rates of nine $^{238}PuO_2^{16}$ powders are compared to the electrorefined metal feed in Table X. An average increase in rate of 275 n/sec-g ^{238}Pu was observed in converting metal to oxide. Light element impurities, excluding oxygen, should be responsible for ~ 100 n/sec-g ^{238}Pu. Available data indicate that the balance, 175, is due to the presence of ^{17}O and ^{18}O in the $^{238}PuO_2^{16}$. Oxygen isotopic data for preparation LAS 9, 11, and 13 are given in Table XI 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. 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 the oxide was contaminated by natural oxygen or water vapor. The extent of contamination is given by the increased ^{18}O content of the oxide. Thus, the calculated ^{17}O concentration for LAS 9, 11, and 13

are 77.5, 75.9, and 75.4 ppm respectively. The (α, n) contribution for the values of ^{17}O and the ^{18}O values of Table XI are 200, 1'9, and 297 n/sec-g ^{238}Pu for LAS 9, 11, and 13. This is equivalent to an average value of ~ 200 n/sec-g ^{238}Pu.

Gamma Radiation

The gamma ray spectrum of $^{238}PuO_2{}^{16}$ is identical to that of electrorefined metal except for a gamma ray at 1634 keV which results from a (α, n) reaction with ^{17}O. The spectrum is given in Ref. [10].

CHARACTERIZATION AND PROPERTIES OF $^{238}PuO_2{}^{16}$ CERAMICS

The power density of $^{238}PuO_2{}^{16}$ powders (~ 1 watt per cc 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[1]; ~ 3.9 watts/cc for the 80 at.% ^{238}Pu fuel or ~ 4.4 watts/cc for the 90 at.% ^{238}Pu fuel. These compacts have been prepared 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 hight to diameter ratio of one.

Ceramic fabrication procedures were developed for the preparation of thirty and fifty watt cylinders[10]. These procedures require ball milling and the addition of carbowax binder. Both operations introduce impurities into the product, however continued improvements in the process have resulted in the preparation of sources having neutron emission rates only about 300 n/sec-g ^{238}Pu higher than an equivalent electrorefined metal source. The ^{18}O concentration of the $^{238}PuO_2{}^{16}$ is reduced to ~ 15 ppm during the sintering operation. This value is essentially equal to the ^{18}O concentration of the H_2O^{16} used in preparing and sintering the oxide. The chemical purity of a typical one watt control pellet is compared to the purity of its electrorefined metal feed, $^{238}PuO_2{}^{16}$ powder, and milled powder in Table XII. The data show that most of the impurities are introduced into the oxide during the ball milling operation which was done in stainless steel equipment. Improved milling procedures are presently under study.

THIRTY AND FIFTY WATT CYLINDERS

The properties of a thirty-two watt and fifty watt $^{238}PuO_2{}^{16}$ cylinder are compared to a 30 watt metal source in Table XIII. The neutron emission rates of the oxide sources are about 6% higher than the desired specification for medical-grade oxide[10], i.e. < 3300 n/sec-g ^{238}Pu for a 30 watt source and < 3350 n/sec-g ^{238}Pu for a 50 watt source. Continued improvements in the ceramic fabrication procedures should result ultimately in oxide sources having the specified neutron emission rates.

OXYGEN ISOTOPIC EXCHANGE OF $^{238}PuO_2{}^{16}$ CERAMICS

Experiments with sintered cylinders of $^{238}PuO_2{}^{16}$ have shown that no detectable oxygen exchange takes place when one watt and 50 watt cylinders are handled in air atmospheres[10]. This behavior is in marked contrast to the results obtained with $^{238}PuO_2{}^{16}$ powders which must be handled

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

in inert glove boxes containing less than 10 ppm O, to minimize oxygen isotopic exchange. Experiments which are still in progress[12] show that the diffusion coefficient for O in $^{238}PuO_2{}^{16}$ at 1000°C is 4×10^{-10} cm^2 sec^{-1}. For comparison purposes, the oxygen self diffusion coefficient in UO_2 is 2.2×10^{-10} cm^2 sec^{-1}[13].

AGING AND FUEL RECYCLE

Growth of ^{236}Pu Daughters in ^{238}Pu

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

The measured gamma ray intensities are compared to those expected as a result of growth from the original ^{236}Pu content in Table XIV. Agreement between the expected and measured gamma ray intensities was satisfactory considering the uncertainties introduced by counting errors in measuring the ^{236}Pu content. The agreement in results showed that literature values for the ^{236}Pu daughter half life[14] and the branching ratios[15] 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.

Permissable Concentrations of ^{236}Pu and ^{232}U in ^{238}Pu

Specifications for medical grade ^{238}Pu[10] include the provision that the ^{232}U and ^{236}Pu contents in a freshly purified ^{238}Pu shall not exceed those concentrations which will produce a total gamma dose over a 10-year 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 concentrations can be conveniently represented by the equation:

$$^{236}\text{Pu (ppm)} + 1.79\ ^{232}\text{U (ppm)} \leq 0.30$$

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

Fuel Recycle Considerations

Complete removal of 72-year ^{232}U and 1.9-year ^{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, it must be removed to the extent of 90% or better. The presence of ^{228}Th in purified materials is less critical. Figure 3 illustrates a worst case and a best case 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 years (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-year point and continuing on through a normal growth and decay curve, whose maximum is reached 18 years after purification, after which it decays with a 72-year 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-year 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.

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

NON-DESTRUCTIVE TECHNIQUES FOR THE EVALUATION OF ^{238}Pu FUELS

The most valuable single measurement to ascertain fuel radiation quality is the neutron count. This measurement is supplemented by detailed gamma ray spectroscopy which is capable of detecting small amounts of light element impurities such as F, Na, Cl, N, O, P, and Al. Gamma ray spectroscopy is used for routine measurements of ^{17}O and ^{18}O in PuO_2[16].

REFERENCES

[1] Matlack, G. M. and Metz, C. F., "Radiation Characteristics of Plutonium-238", Los Alamos Scientific Laboratory Report, LA-3696 (1967).

[2] Bubernak, J. Matlack, G.M., and Metz, C.F., "Neutron and Gamma Radiations from Plutonium-238", Trans. Amer. Nucl. Soc., 11, 457, (1968).

[3] Hicks, D. A., Ise, J. Jr., and Pyle, R. V., "Probabilities of Prompt-Neutron Emission from Spontaneous Fission", Physical Review, 101, 1016 (1956).

[4] Druin, V. A., Parelygin, V. P., and Khlebnikov, G. I., "Spontaneous Fission Periods of ^{237}Np, ^{238}Pu, and ^{242}Pu", Soviet Phys. JETP, 13, 5, 913 (1961).

[5] Abrahamson, S.G., Carfagno, D. G., and Kokenge, B. R., "Plutonium-238 Isotopic Fuel Form Data Sheets", Mound Laboratory Report, MLM-1564, October 31, 1968.

[6] Ellinger, F. H., Miner, W. N., O'Boyle, D. R., and Schonfeld, F. W., "Constitution of Plutonium Alloys", Los Alamos Scientific Laboratory Report, LA-3870 (1967).

[7] Ballagney, A., Boucher, A., and Carrard, C., "The Pu - Sc System: Diagram and Properties", Plutonium 1970 and Other Actinides, W. N. Miner, ed., 699 - 706, Nuclear Metallurgy, Volume 17, Part II (1970).

[8] Alais, M., Berger, R., Boucher, R., Hixson, J., Laurans, P., and Powneda, A., "The French Nuclear-Powered Pacemaker Program", Nucl. News, 42 - 46, December 1970.

[9] Mullins, L.J., and Leary, J. A., "Plutonium-238 for Biomedical Applications", Nuclear Applications, 6, 287 - 297 (1969).

[10] "Preparation and Evaluation of Medical-Grade Plutonium-238 Fuels", Summary Report for the Period, July 1, 1967 - June 30, 1971, Compiled by L. J. Mullins, Los Alamos Scientific Laboratory Report LA-4940, (in press).

[11] Hanson, G., LASL, N-2, personal communication, memorandum N-2-8345, March 25, 1970.

[12] Jones, W. M., LASL, CMB 11, unpublished work, work still in progress.

[13] Belle, J., "Oxygen and Uranium Diffusion in Uranium Dioxide", J. Nucl. Mat'ls, 30, 3 (1969).

[14] Hoffman, D. C., Ford, G. P., and Lawrence, F. O., J. Inorg. Nucl. Chem. 4, 143 (1957).

[15] Gunnink, R., Niday, J.B., Anderson, R. P., and Meyer, "Gamma-Ray Energies and Intensities", Lawrence Radiation Laboratory Report, UCID - 15439 (1969).

TABLE I

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

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

TABLE II

POTENTIAL MEDICAL-GRADE ^{238}Pu FUEL COMPOSITIONS

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

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

TABLE III

PHASE TRANSFORMATIONS OF PU - 3 AT.% Ga

AND PU - 10 AT.% Sc

Transformation	Temp. of Transformation, °C	
	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

TABLE IV

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

(3/8 in. x 3/8 in Cylinders)

Composition	Dose Rates	
	Gamma mR/h - g ^{238}Pu	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) Pu isotopic composition, 80 wt.% ^{238}Pu.

(b) Dose rates in air through 0.030 in. Ta, 10 cm from the center of the source.

(c) All fuels prepared from electrorefined metal.

TABLE V

PURITY OF CAST METAL FROM TYPICAL ELECTROREFINING RUN

Element	ppm Impurity[a]	Element	ppm Impurity[a]
Li	< 0.01	Rb	< 0.5
Be	< 0.001	Sr	< 0.1
B	< 1	Y	< 0.1
C	40	Zr	0.3
Na	2	Mo	< 0.5
Mg	5	Cd	< 0.5
Al	2	Sn	< 0.5
Si	< 5	Cs	< 2
K	< 1	Ba	< 1
Ca	< 2	La	< 1
Ti	< 0.2	Hf	< 0.5
V	< 0.5	Re	< 0.5
Cr	< 1	Pb	< 1
Mn	< 1	Bi	< 1
Fe	20	Am-241	0
Co	< 0.5	Np-237	500
Ni	2	U-232	0.00
Cu	2	U-234	115
Zn	< 5	Pu, wt%, by titration	99.9

[a] For all tables, ppm Impurity = g Impurity per 10^6 gPu.

TABLE VI

NEUTRON EMISSION RATE OF ELECTROREFINED ^{238}Pu METAL

(80 at % ^{238}Pu)

Sample Size, g Pu	No. of Samples measured	Average Neutron Emission Rate, n/sec-g ^{238}Pu
1 - 2	18	3095 (± 46)
10 - 20	25	3123 (± 59)
20 - 30	10	3170 (± 50)

TABLE VII

^{238}Pu GAMMA RAY INTENSITIES AND PRECISION OF MEASUREMENT

(80 at.% ^{238}Pu)

Energy, kev	Gamma Ray Intensity, gamma 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	$1\ 36 \times 10^{-8}$	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

TABLE VIII

COMPARISON OF THE RADIATION PROPERTIES OF

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

	Metal	$^{238}PuN^{15}$
Neutron Emission Rate, n/sec-g Pu	2495	2505
n/sec-g ^{238}Pu	3110	3123
Dose Rates at 10 cm in air through 0.030 inches 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

TABLE IX

COMPARISON OF THE CHEMICAL PURITY OF

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

Element	ppm Impurity Metal	ppm Impurity Oxide	Element	ppm Impurity Metal	ppm Impurity 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 X

COMPARISON OF NEUTRON EMISSION RATES OF

ELECTROREFINED METAL AND $^{238}PuO_2{}^{16}$ POWDERS

$^{238}PuO_2{}^{16}$ Lot	Neutron Emission Rate, n/sec - g ^{238}Pu 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 XI

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

Prep. No.	Oxygen Isotopic Composition, g isotope/10^6 g O		
	^{17}O	^{18}O	
	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

TABLE XII

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

Element	ppm Impurity			
	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	< 1	< 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	4	4
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	< 2	< 2
Re	< 0.5	< 1	< 1	< 1
Pb	< 0.5	2	7	< 1
Bi	< 1	< 1	< 1	< 1
Np	300	285	--	307

TABLE XIII

COMPARISON OF THE PROPERTIES OF A 30 WATT

ELECTROREFINED METAL SOURCE TO 32 AND 50 WATT $^{238}PuO_2$[16] CERAMIC SOURCES

	^{238}Pu - 3 at % Ga	32 Watt $^{238}PuO_2$[16]	50 Watt $^{238}PuO_2$[16]
Power, watts	30.3	32.2	49.8
Power density, watts/cc	7.18	4 20	3.88
Pu isotopic composition			
wt. % ^{238}Pu	80.45	86.96	79.98
ppm ^{236}Pu	0.58	0.25	0.75
Neutron emission rate,			
n/sec - g ^{238}Pu	3150	3519	3488
Dose rates at 10 cm			
Neutron, mRem/h - g ^{238}Pu	0.31	0.35	0.35
Gamma, mR/h - g ^{238}Pu	0.16[a]	0.14[b]	0.14[c]

(a) Measured in air through 0.030 in. Ta

(b) Measured in air through 0.064 in Ta-10W and 0.025 in. Hastelloy C-276

(c) Measured in air through 0.020 in Ta and 0.025 in Hastelloy C-276

TABLE XIV

GAMMA RAY INTENSITIES OF ^{236}Pu DAUGHTERS

(49.9 mg ^{238}Pu, 0.66 ppm ^{236}Pu, 1.340 yr growth)

Energy, kev.	Expected γ's/min.	Found γ's/min
238	$4.47 (\pm .21) \times 10^4$	$4.47 (\pm .04) \times 10^4$
511	$8.67 (\pm .42) \times 10^3$	$9.29 (\pm 31) \times 10^3$
583	$2.83 (\pm .14) \times 10^4$	$2.69 (\pm .05) \times 10^4$
727	$6.16 (\pm .30) \times 10^3$	$5.93 (\pm .33) \times 10^3$
1620	$1.32 (\pm .06) \times 10^3$	$1.2 (\pm .40) \times 10^3$
2614	$3.29 (\pm .16) \times 10^4$	$3.23 (\pm 10) \times 10^4$

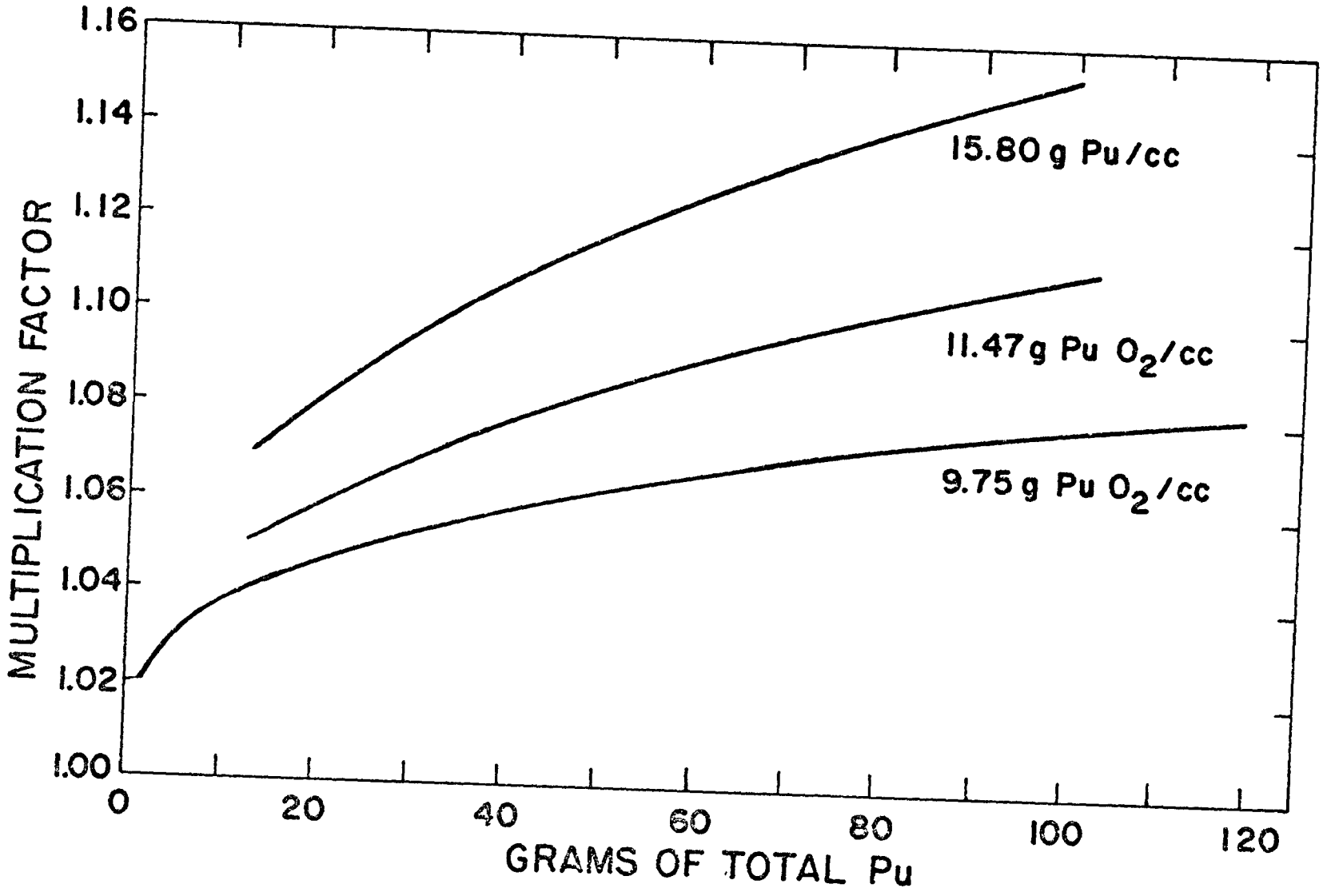

Fig. 1 Neutron multiplication factors for ^{238}Pu metal and $^{238}PuO_2$[16]

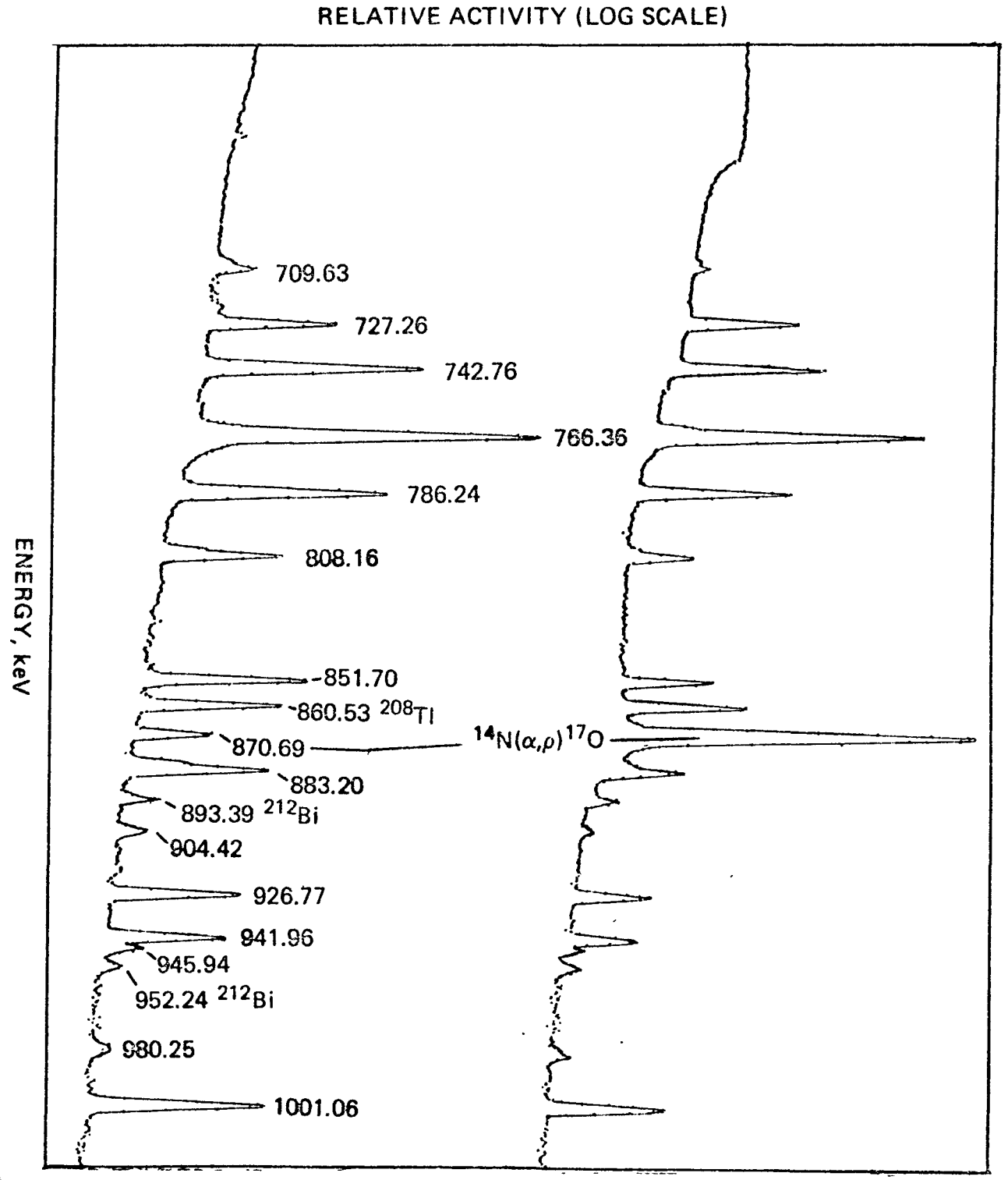

Fig. 2 Comparison of the Gamma Spectrum of $^{238}PuN^{15}$ and $^{238}PuN^{nat}$.

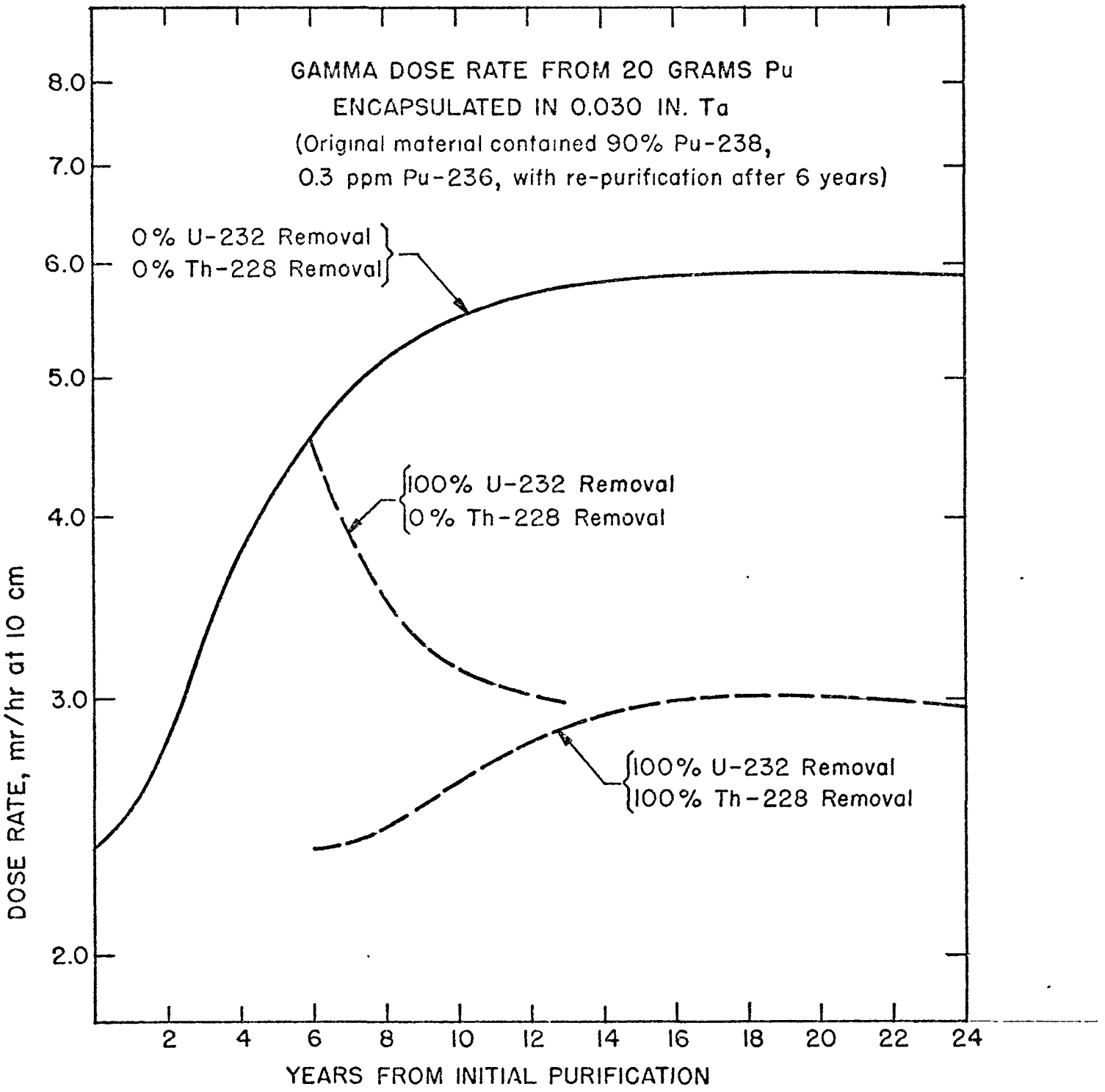

Fig. 3 Gamma dose rate as a function of time.