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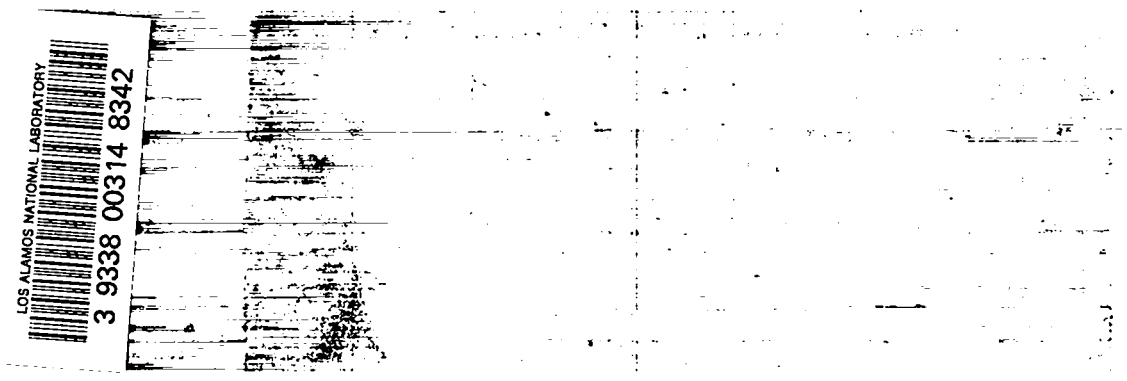
Radiation Characteristics of Plutonium-238



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George M. Matlack
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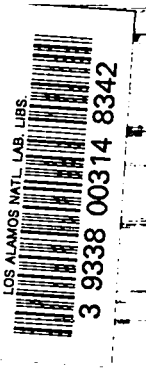

RADIATION CHARACTERISTICS OF PLUTONIUM-238

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ABSTRACT

The primary hazard in the use of ^{238}Pu as a biomedical power source results from the neutron flux caused by spontaneous fission and (α, n) reactions with light elements. Spontaneous fission will cause an activity of approximately 2200 neutrons per gram-second for material containing 81% ^{238}Pu. Neutrons from (α, n) reactions can increase this value to 8,000-10,000 unless every effort is made to produce ^{238}Pu which is extremely free of light element impurities. Most of the gamma radiation is of low energy and easily shielded. A significant fraction of the gamma radiation will, however, be caused by the growth of ^{241}Am, and by the 4n heavy element decay chain resulting from the decay of small amounts of ^{236}Pu. Gamma radiation from spontaneous fission and fission product decay will be negligible.

INTRODUCTION

Biomedical applications of plutonium-238 as a power source are dependent upon the energy output arising from the characteristic radiation spectra associated with this isotope. The natural radioactivity of the pure isotope consists principally of alpha radiation having an average energy of approximately 5.5 Mev. Accompanying this radiation is characteristic gamma energy some of which is significantly penetrating. In addition, this isotope undergoes spontaneous fission which produces a "natural" neutron background.

But this fundamental radiation spectra mentioned above is not all. In addition, plutonium-238 is not produced as a pure isotope, a fact that must be considered in any discussion of its radiation spectra. The "Pu-238" of commerce consists essentially of a mixture of isotopes having a nominal weight distribution as follows,

^{238}Pu	81%
^{239}Pu	15
^{240}Pu	2.9
^{241}Pu	0.8
^{242}Pu	0.1
^{236}Pu	1.2×10^{-4}

Although none of these accompanying isotopes causes significant complications directly, the decay daughters of ^{236}Pu introduce penetrating gamma radiation that must be dealt with. As is shown in this report, it is necessary, or at least highly desirable, to keep this isotope to concentrations less than 1 ppm.

In addition, the average energy of the emitted alpha particles is approximately 5.5 Mev., energy great enough to cause (α, n) reactions with low Z

impurity elements. For this reason, it is desirable that all elements having a Z number less than 14 be kept to a minimum in order to keep the neutron flux to a low background level.

The naturally occurring radiation and the radiation resulting from environment combine to create a biologically hazardous total. This report describes the contribution from each of the above situations and discusses the conditions that must be achieved by which ^{238}Pu may be made biologically acceptable for biomedical uses.

PRODUCTION OF ^{238}PU

Plutonium-238 is produced by the series of nuclear reactions shown in Eq. 1. Kilogram quantities of ^{238}Pu are produced by recycling the ^{236}U, and separating and recycling the ^{237}Np.

$$^{235}U + n \longrightarrow {}^{236}U + n \longrightarrow {}^{237}U \xrightarrow[6.8\ d]{\beta} \quad (1)$$

$$^{237}Np + n \longrightarrow {}^{238}Np \xrightarrow[2.1\ d]{\beta} {}^{238}Pu$$

Heavier plutonium isotopes are produced in amounts which depend on the operating conditions of the reactor. These isotopes are produced through multiple neutron capture by ^{238}Pu. Small amounts of ^{236}Pu are formed also by the reaction

$$^{237}Np\ (n,\ 2n)\ ^{236}Np \xrightarrow[22\ h]{\beta} {}^{236}Pu.$$

This is an important isotope because its short half-life produces a significant amount of radiation. This will be discussed later.

ALPHA AND BETA ACTIVITIES OF PLUTONIUM-238 PRODUCT

The alpha and beta activities found in currently produced conventional plutonium-238 are shown in Table I. This is the product composition whose radiation characteristics are the subject of this report. All values in this report are in terms of one gram of this material.

The ^{238}Pu contributes about 99.9 percent of the total alpha activity. The beta activity from the ^{241}Pu is relatively high, but its low energy of 0.02 Mev. per beta particle contributes very little to the total disintegration energy from the other plutonium isotopes.

NEUTRON RADIATIONS

The neutron emission from plutonium-238 product comes from three sources, spontaneous fission, alpha particle reactions with low Z element impurities, and gamma-ray interactions with impurity beryllium.

TABLE I

Alpha and Beta Activities of Product Plutonium-238

Isotope	Abundance, %(1)	Activity of Pure Isotope, d/sec-g	Contribution to the Activity of 1 gram of Product
^{238}Pu	81	6.35×10^{11}	5.14×10^{11} (α)
^{239}Pu	15	2.27×10^{9}	3.41×10^{8} (α)
^{240}Pu	2.9	8.38×10^{9}	2.43×10^{8} (α)
^{241}Pu	0.8	4.12×10^{12}	3.30×10^{10} (β)
^{242}Pu	0.1	1.44×10^{8}	1.44×10^{5} (α)
^{236}Pu	1.2×10^{-4}	1.97×10^{13}	2.37×10^{7} (α)

Spontaneous Fission. The spontaneous fission rate of isotopically pure ^{238}Pu is 1100 fissions per gram-second with an average of 2.33 neutrons per fission[2,3], resulting in a total flux of 2563 neutrons per gram-second.

In the case of plutonium-238 product having the isotopic abundances listed in Table I, the rate of production of the spontaneous fission neutrons amounts to approximately 2100 per gram-second from the ^{238}Pu isotope and 29 per gram-second from the ^{240}Pu isotope. This total number of 2129 neutrons per gram-second represents the lowest limit possible for neutrons coming from pure plutonium-238 product, assuming no low Z number impurity elements present but having the isotopic composition stated in Table I. The average energy of these spontaneous fission neutrons is 2.0 Mev., with a maximum of 10 Mev. whose distribution is approximately Maxwellian, having a peak at 0.7 Mev.[4]

Neutrons Produced by (α, n) Reactions. Atoms with a low Z nuclear charge are capable of producing neutrons when bombarded by alpha particles providing several conditions are satisfied. First, the energetic threshold for the reaction must not exceed the energy of the alpha particle, which in this case is 5.50 Mev. Second, the coulomb barrier created by the repulsive force between the two positively charged nuclei (the alpha particle and target nucleus), must not be greater than the alpha particle energy. The first condition is a function of the differences between the relative masses of the reacting particles and their products, and of the fraction of energy which must be available to satisfy the conservation of momentum. The second condition imposes an upper limit on the charge of a nucleus which is capable of reacting with an alpha particle of a given energy. The coulomb barrier increases with Z, and for alpha particle interactions reaches 5.5 Mev. at Z = 14, which means that elements beyond silicon would not be expected to give significant neutron yields from alpha particle reactions.

The neutron production from light element impurities can be calculated from the experimental measurements on yields from the pure element, the so-called "thick target" yield.[5] In applying these values to mixtures of an alpha emitter and light elements, allowance must be made for the mole fraction of the light element present, the mole fraction of the alpha emitter, and the shielding factors for all elements in the mixture, according to the relation[6]

$$Y = \frac{\Sigma m_i s_i y_i}{\Sigma m_i s_i}$$

where m_i = mole fraction of the ith element, s_i is the corresponding shielding factor, proportional to $\frac{Z}{\sqrt{Z+1}}$, and y_i is the thick target yield. Table II lists the specific neutron yields for one part per million each of light element impurities in one gram of plutonium metal containing 81% ^{238}Pu.

The neutron yield from typical plutonium metal containing 81% of the ^{238}Pu isotope can be calculated from the impurity concentrations cited by Grove[7], using the specific yields shown in Table II. The impurity concentrations were given for a minimum and maximum level, and are reproduced

TABLE II

Specific Neutron Yields From
Light Element Impurities

Element	Neutrons Per Second for one Part per Million
Li	4.6
Be	133
B	41
C	0.2
N	0.0 (α, n threshold too high)
O	0.1
F	18
Na	2.2
Mg	2.1
Al	1.0
Si	0.2
P	< 0.03
S	< 0.03

in Table III together with the calculated neutron yield for each element. It should be noted that concentrations for lithium, beryllium and fluorine are not listed; these three elements would contribute significant additions to the total neutron yield from the other elements listed. The minimum level impurities give approximately 6000 neutrons per second for each gram of plutonium metal. With the addition of 2100 neutrons per second from spontaneous fission, the total would be approximately 8000 neutrons per gram-second. The neutron yield from plutonium metal containing the maximum level of impurities is greater than 300,000 per gram-second, a flux too high to be considered for biological applications.

For comparative purposes, calculations were made of the total neutron production from (α, n) reactions on low Z number impurity elements when present in small concentrations such as in "high purity plutonium." These latter concentrations are those found in kilogram lots of plutonium-239 metal purified by the LAMEX process.[8] The impurity levels and corresponding neutron yields are shown in Table IV. Because such extremely low impurity levels are not uncommon for plutonium-239 produced by this process, it can reasonably be assumed that plutonium-238 could be produced routinely with an impurity content not greater than five times that shown in Table IV.

TABLE III

Expected Neutron Yields (α, n) in Typical Plutonium Metal Containing 81% ^{238}Pu

Impurity	Minimum Concentration		Maximum Concentration	
	ppm	n/gram-sec.	ppm	n/gram-sec.
B	100	4100	6400	262,000
Na	600	1300	7600	16,700
Mg	100	200	9000	18,900
Al	100	100	3500	3,500
Total	900	5700	26,500	301,100

TABLE IV

Expected Neutron Yields in High Purity Plutonium Metal Containing 81% ^{238}Pu

Impurity Element	Concentration, ppm	Neutrons/gram-second
Be	0.001	0.1
B	0.5	20.
C	15	3
O	10	1
F	1	18
Na	1	2
Mg	2	4
Al	1	1
	Total neutron yield	49

It is therefore possible by adequate (but realistic) purification to reduce the (α, n) neutron emission rate to a level which is significantly less than the spontaneous fission neutron rate of approximately 2100 neutrons per gram-second. The elements to which particular attention should be given from an impurity standpoint are boron and fluorine.

Although plutonium metal is the material to which much consideration is being given as the power source for certain applications, compounds of plutonium might be more suitable provided they are stable enough to withstand the radiation effects. Table V lists the neutron yields from several light element compounds of plutonium containing 81% ^{238}Pu. These values reflect the neutron production from the light element alone, and do not include those produced from other light element impurities in the compound, or the spontaneous fission neutrons. Spontaneous fission, as explained earlier, will increase the neutron rates in Table V by 2100 per second for each gram of 81% ^{238}Pu. It is clear that plutonium oxide with normal oxygen, and plutonium fluoride, would not be suitable as biological power sources because of their high neutron emission rates. As indicated in Table V, however, the oxide made with low-enrichment ^{18}O, and the nitride would have negligible neutron emission if made with sufficiently high purity.

TABLE V

Neutron Activities From Light Element Plutonium Compounds Containing 1 Gram of 81% Enriched ^{238}Pu

Compounds	Neutrons/sec.
PuN	0
PuO_2 (normal O)	11,300
PuO_2 (^{18}O reduced to 1/100)	113
PuF_4	3,000,000
Pu_2S_3	700

Photo-Neutron Formation. The third mechanism of neutron production is that of photo-neutron formation. This results from the interaction of hard gamma rays with impurity elements. Such gamma rays are present in ^{238}Pu as a result of spontaneous fission and of ^{208}Tl decay (a daughter of ^{236}Pu), although their abundances are only of the order of 500 per gram-second. Hence they should not show a significant neutron production level. Deuterium and beryllium are the only light elements which would produce photo-neutrons under these conditions.

GAMMA RADIATION

The gamma activity from plutonium-238 product is intense, but mostly of an easily-shielded form. Table VI lists the more prominent sources of x-rays and gamma rays.

TABLE VI

Gamma Radiation From Plutonium-238 Product

Isotope	Energy, Mev.	Photons/gram-sec.
^{238}Pu	0.017 (L x-ray)	6.7×10^{10}
	0.043	2.0×10^{8}
	0.099	4.6×10^{7}
	0.150	5.2×10^{6}
	0.203	2.0×10^{4}
	0.760	2.6×10^{5}
^{239}Pu	0.017 (L x-ray)	3.4×10^{7}
	0.052	2.4×10^{4}
^{240}Pu	Less abundant than ^{239}Pu	
^{241}Pu	0.145	$\leq 4 \times 10^{4}$
^{236}Pu	0.048	$\leq 7 \times 10^{7}$

Gamma rays will also result from the radioactive decay of daughter nuclides of ^{236}Pu and ^{241}Pu. The ^{236}Pu is the precursor of the 4n decay chain which ends with stable ^{208}Pb and is illustrated in Eq. 2, shown on the next page.

The rate-controlling step in the sequence, as far as the formation of gamma-emitting daughters is concerned, is the growth of 1.91 year ^{228}Th. The amount of gamma activity from lead, bismuth, and

thallium daughters increases with time after purification of the plutonium-238 product, reaching a maximum in 18 years.

$$^{236}Pu \xrightarrow[\alpha]{2.85\ y} {}^{232}U \xrightarrow[\alpha]{72\ y} {}^{228}Th \xrightarrow[\alpha]{1.91\ y}$$

$$^{224}Ra \xrightarrow[\alpha]{3.64\ d} {}^{220}Rn \xrightarrow[\alpha]{52\ s} {}^{216}Po \xrightarrow[\alpha]{0.16\ s} \qquad (2)$$

$$^{212}Pb \xrightarrow[\beta]{10.6\ h} {}^{212}Bi \xrightarrow[\alpha\ (34\%)]{60\ m} {}^{208}Tl$$

$$^{212}Bi \xrightarrow[(66\%)]{\beta\ 60\ m} {}^{212}Po \xrightarrow[\alpha]{0.3\ \mu s} {}^{208}Pb\ (Stable)$$

$$^{208}Tl \xrightarrow{\beta\ 3\ m}$$

The principal gamma rays of these three elements, and their abundances, are shown in Table VII in which the percentages refer to the number of photons per 100 disintegrations of ^{228}Th. This convention is used since these short-lived daughters are nearly always seen in equilibrium with ^{228}Th, and because of the branching decay of ^{212}Bi.

TABLE VII

Principal Gamma Ray Abundances From ^{236}Pu Daughters

Nuclide	Gamma, Mev.	% Abundance
^{212}Pb	0.239	82
^{212}Bi	0.727	6
^{208}Tl	0.277	3
	0.511	8
	0.583	30
	0.860	3
	2.62	34

Of these gamma rays, three contribute significant amounts to the total gamma ray flux from plutonium-238 product. These are the 0.239 Mev. gamma from ^{212}Pb, and the 0.583 and 2.62 Mev. gammas from ^{208}Tl. The intensity growth of these gamma rays as the plutonium-238 product ages after purification is shown in Table VIII.

TABLE VIII

Growth of ^{212}Pb and ^{208}Tl Gamma Activity*

Time After Purification, years	^{212}Pb, 0.239 Mev., photons/sec.	^{208}Tl, 0.583 + 2.62 Mev., photons/sec.
0.1	330	260
1.0	27,000	21,000
2.0	90,000	70,000
5.0	330,000	260,000
10.0	570,000	450,000
18.0 (max.)	660,000	510,000

* For 1 gram of plutonium-238 product containing 1.2 ppm ^{236}Pu.

If one compares these values with the gamma activity from ^{238}Pu shown in Table VI, it is seen that after five years the intensity of gamma rays from ^{208}Tl will have increased to the same order of magnitude as the 0.760 Mev. gamma from ^{238}Pu. Because the 2.62 Mev. gamma ray from the ^{208}Tl is the most energetic, gamma shielding requirements should be based on it. If future ^{238}Pu products contain less than 1 ppm ^{236}Pu, shielding requirements will be less.

Gamma rays result also from the spontaneous fission process, in the form of prompt gammas emitted during the fission event, and as gamma rays emitted from the radioactive fission products. The total energy release through the prompt gamma emission is 7 to 9 Mev., with an average of 10 photons, for each fission.[9] The average energy is therefore less than 1 Mev. The numbers and approximate energies of the prompt gammas are shown in Table IX, on the next page.[1] Compared to gamma radiation from other sources in the plutonium-238 product, it is unlikely that these should be considered as contributing a significant dosage. The fission product gamma rays would be expected to show an increase in their abundance as the plutonium-238 power source ages. However, because the average half-life of the fission products is short compared to the spontaneous fission half-life, a steady state will be reached in less than a day after purification. The energies and abundances of the fission pro-

TABLE IX

Prompt Fission Gammas In

Plutonium-238 Product

Energy Range, Mev.	Photons Per Gram-Sec.
0.0-1.0	4600
1.0-3.0	1700
3.0-5.0	150
5.0-7.0	25

duct gammas are shown in Table X.[1] Like the prompt gammas, these will not affect gamma dosage rates significantly.

TABLE X

Gamma Rays From Fission Products

In Plutonium-238 Product

Energy Range, Mev.	Photons Per Gram-Sec.
0.1-0.9	5900
0.9-1.8	1000
1.8-3.0	400

Americium-241 will also be a source of gamma radiation. It will be recalled (see Table I) that the plutonium-238 product contains 0.8 percent ^{241}Pu. This nuclide has a 13 year half-life, decaying by beta emission to ^{241}Am, which, in turn, has a half-life of 458 years, and decays by both alpha and gamma emission to ^{237}Np. The ^{241}Pu activity is the second most intense of the plutonium isotopes present but does not create a hazard because of the very low maximum energy of its beta particle, which is 0.02 Mev. The ^{241}Am gamma activity increases as the sample ages, and reaches relatively high rates in several years, as shown in Table XI.

This maximum amount of gamma activity is the same order of magnitude as the 0.05-0.10 Mev. gammas from the ^{238}Pu, and might constitute a personnel hazard from an unshielded 1-gram source. However, the shielding required for the

TABLE XI

Gamma Activity From Growth of ^{241}Am

in Plutonium-238 Product

Time, years	0.060 Mev., photons/gram-sec.
0.5	8.6×10^6
1.0	1.7×10^7
2.0	3.4×10^7
5.0	7.6×10^7
10.0	1.4×10^8
70.0 (max.)	3.0×10^8

gamma radiations from the ^{238}Pu and ^{236}Pu daughters will also be adequate for the ^{241}Am radiations.

RADIATION DOSAGE

The principal radiation hazard from ^{238}Pu results from its gamma ray and neutron emissions. Hazards from the alpha particle and beta particle flux are not likely to exist, because any container with a wall thickness greater than 4 mg./cm.^{2} will absorb both the ^{238}Pu alpha particles and the ^{241}Pu beta particles. Aging of the ^{238}Pu will lead to increasing amounts of 2.25 Mev. beta particles from the ^{212}Bi daughter of ^{236}Pu. A container wall thickness of approximately 1000 mg./cm.^{2}, or greater, will absorb these beta particles. This corresponds to approximately 0.06 inch of tantalum.

Estimation of the radiation dosage from neutrons can be made first on the basis of the spontaneous fission rate, since this represents the irreducible minimum dosage. Neutrons arising from (α, n) reactions between ^{238}Pu and light element impurities will increase the dosage in direct proportion to the ratio of (α, n) neutrons to spontaneous fission neutrons. The average energy of the neutrons from spontaneous fission is 2 Mev., and the dosage equivalent for neutrons of this energy is 1 millirem per hour for a flux of 8 neutrons per second per square centimeter. If a value of 2150 neutrons per gram-second (for 81% ^{238}Pu) is assumed, the neutron dosage at a distance of 1

centimeter will be $\frac{2150}{4\pi \times 8}$ = 21 millirem per hour from 1 gram of 81% ^{238}Pu. A continuous 24-hour exposure would result in a dose of 3500 millirem per week, approximately 35 times the maximum permissable of 100 millirem per week. This is for one gram of pure plutonium, containing 81% ^{238}Pu, and emitting only spontaneous fission neutrons. If the same weight of plutonium contained enough light element impurities to triple its neutron emission rate from 2150 per second to 6450 per second, its neutron radiation level would correspond to slightly in excess of 10,000 millirem per week at 1 centimeter distance for a continuous exposure, a dose which is 100 times the maximum permissable level.

The estimation of the gamma radiation hazard is more complex than it is for the neutron hazard because of the many different gamma rays and the fact that daughter growth causes an increase with time in the intensity of some of the gamma rays. The most abundant gamma rays in either fresh or aged material fortunately are of low energy so that shielding can be achieved without difficulty. Table XII shows the dose rates for each gamma ray from the plutonium isotopes in 81% ^{238}Pu. Two situations are illustrated, one for the bare source (without correction for self-absorption) and the other for the source encapsulated in a tantalum container with walls 0.03 inch thick. Practically all of the radiation hazard from unshielded material is caused by the ^{238}Pu, and amounts to 5000 roentgens per hour at 1 centimeter for 1 gram of 81% ^{238}Pu. When a tantalum container with 0.03-inch walls is used, the dosage is reduced to about 50 milliroentgens per hour.

The gamma radiations from the radioactive daughter products are not as easily shielded, except for the americium activity, because these gamma rays are more energetic than are most of the gamma rays from the plutonium. Their contributions to the gamma ray doses from 1 gram of 81% ^{238}Pu, at a distance of 1 centimeter, are shown in Table XIII. These values were calculated for a ^{238}Pu source which has aged 5 years from the final chemical purification, with the assumption that all impurity elements were removed. In the unshielded case, the added dosage from the daughters is 8 roentgens per hour, which is insignificant

TABLE XII

Gamma Ray Doses From Plutonium Isotopes

In 1 Gram of Plutonium Containing 81% ^{238}Pu

Nuclide	Energy, Mev.	r./hr., at 1 cm., unshielded	r./hr., at 1 cm., with 0.03" Ta shield
^{238}Pu	0.017	5000	0.00002
	0.043	2.2	0.0004
	0.099	0.6	0.002
	0.150	0.1	0.015
	0.203	0.0006	0.0002
	0.760	0.03	0.03
^{239}Pu	0.017	2.4	------
^{241}Pu	0.145	0.0004	0.00006
^{236}Pu	0.017	2.	------
	0.048	2.	0.0003

TABLE XIII

Gamma Ray Doses From Daughter Products

In 1 Gram of Plutonium Containing 81% ^{238}Pu

Nuclide	Energy, Mev.	r./hr., at 1 cm., unshielded	r./hr., at 1 cm., with 0.03" Ta shield
^{241}Am	0.017	8	----
	0.060	0.6	0.02
^{212}Bi	0.727	0.003	0.003
^{212}Pb	0.239	0.01	0.006
^{208}Tl	0.277	0.0005	0.0004
	0.583	0.01	0.01
	0.860	0.002	0.002
	0.511	0.003	0.003
	2.62	0.04	0.04

in comparison to the 5000 roentgens per hour from the unshielded plutonium isotopes. With the 0.03-inch Ta shield, the radiation from the daughters is reduced to 84 milliroentgens per hour. This amount, added to the shielded plutonium contribution of 50 milliroentgens per hour, gives a total gamma ray dosage of 134 milliroentgens per hour at 1 centimeter distance for 1 gram of the plutonium product.

HAZARD FROM HELIUM GENERATION

A non-radioactive hazard may result from an increase of internal pressure within a hermetically sealed power source, caused by the generation of helium from the alpha decay of the ^{238}Pu. Table XIV shows the volume of helium gas which would be generated by a 1-gram plutonium-238 product source.

TABLE XIV

Volume of He at STP in 1-Gram Plutonium-238 Product

Time, years	Vol., cc.
0.1	0.074
0.5	0.37
1.0	0.74
2.0	1.6
5.0	3.7
10.0	7.4

SUMMARY

1. The neutron radiation from present day ^{238}Pu product would be prohibitive from a biological standpoint unless low Z-number impurity elements are kept to a very low level. Experience in producing high purity ^{239}Pu by the LAMEX process indicates however, that satisfactory low levels of concentrations of these elements can be reached.

2. The gamma radiation from present day ^{238}Pu product, although intense, is largely of low energy

content and easily shielded. The gamma radiation coming from the decay products of the ^{236}Pu isotope is quite energetic and may require shielding. Lowering the ^{236}Pu concentration to approximately 0.1 ppm would reduce this problem to one of negligible concern.

3. A possible non-radioactive hazard may arise from the pressure generated by the helium produced by the alpha decay of the ^{238}Pu.

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