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POLONIUM-210 AND PLUTONIUM-238 RADIOISOTOPE

SHIELDING FOR POST-APOLLO MISSIONS

Prepared by:

thay any

Graham L. Hagey AST, Advanced Power Section

Authorized for Distribution:

fer'

Maxime A. Waget ' / Director for Engineering and Development,

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION MANNED SPACECRAFT CENTER HOUSTON, TEXAS August 19, 1966

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POLONIUM-210 AND PLUTONIUM-238 RADIOISOTOPE

SHIELDING FOR POST-APOLLO MISSIONS

By Graham L. Hagey Manned Spacecraft Center

SUMMARY

This paper presents an approach to and the results of a simplified radiation shielding analysis for radioisotope electrical power systems for manned space applications. The analysis is presented for polonium-210 and plutonium-238, the two most promising radioisotopes being evaluated by NASA for post-Apollo electrical power systems.

The results provide typical shield weights and volume geometries as functions of mission configuration and power system variables, and are suitable for mission parametric applications analyses.

INTRODUCTION

Polonium-210 and plutonium-238 are being evaluated by NASA as radioisotope power system energy sources for post-Apollo programs requiring between approximately 3 and 10 electrical kilowatts. Because these radioisotopes emit gamma ray and neutron radiations, shielding is required for protection of equipment and crew personnel.

This study was undertaken to provide typical heat source shielding geometries and weights using simplified assumptions concerning the emitted radiation levels and spectra, and attenuation processes. This information is necessary for parametric applications analyses with respect to configuration/mission interfaces.

SYMBOLS

В	buildup factor for gamma rays
barn	nuclear cross section, 10^{-24} cm ² per nucleus
curie	3.7×10^{10} nuclear disintegrations per sec
\mathbf{E}_{α}	alpha particle kinetic energy, Mev
E _Y	gamma ray kinetic energy, Mev
exp	base of the natural system of logarithms(e), equal to 2.718
ev	electron volts of energy, equal to 1.6×10^{-19} watt-sec
I	neutron source strength, neutrons/sec
k	neutron source multiplication factor the ratio of the number of neutrons present in one fission generation to the number of corresponding neutrons of the immediately preceding generation
Mev	million electron volts of energy, equal to 1.6×10^{-13} watt-sec
N	number of atoms per cm ³ of a shielding material
n	number of neutrons emitted in a multiplying medium
ⁿ o	number of neutrons originally present in a multiplying medium
n, 2n	neutron-neutron reaction in this reaction a nucleus absorbs a neutron and, subsequently, emits two neutrons
n, y	neutron-gamma ray reaction in this reaction a nucleus absorbs a neutron and, subsequently, emits a gamma ray
ppm	part per million by weight

source point to dose point separation distance, cm

rep

R,

roentgen-equivalent-physical -- that dose of ionizing radiation which produces an energy absorption of 93 ergs/cm³ in tissue. In this paper rep is used because the necessary neutron attenuation data are provided in terms of rep; however, rep is no longer used as a radiation unit.

rem

t

α

a,n

Y

μ

Σi

roentgen-equivalent-man -- a biological unit of dose which accounts for the different biological effects of different types of radiations. These varying biological effects are frequently correlated by a coefficient called the relative biological effectiveness (RBE). The rem is equal to the dosage in rads multiplied by the RBE for the type of radiation involved. For fast neutrons an RBE of 10 is generally used; for gamma rays the value is unity.

rad an absorbed radiation dose of 100 ergs/gm in any material

S gamma ray source strength, Mev/sec

time, sec

x, material thickness, cm

alpha particle -- identical to nucleus of helium atom

alpha-neutron reaction -- in this reaction a nucleus absorbs an alpha particle and, subsequently, emits a neutron

gamma ray

gamma ray mass attenuation coefficient, cm⁻¹

gamma ray energy flux, Mev/cm²-sec

arithmetic summation

σ_n microscopic neutron removal cross section, cm² (the product Nσ is the macroscopic removal cross section. cm⁻¹)

4m shield shielding providing 4m steradian protection

POLONIUM-210 RADIATION CHARACTERISTICS

Polonium-210 (Po^{210}) decays by alpha emission with a half-life of 138.4 days to either an excited state of lead-206 (Pb^{206}) or to the ground state of Pb²⁰⁶ as shown in figure 1 (ref. 1). Lead-206 in the excited state decays to the ground state by the emission of a 0.8 Mev gamma ray. The gamma ray is the only intrinsic radiation source requiring shielding as the decay alpha particles do not penetrate the Po²¹⁰ fuel capsule wall.

Secondary radiation sources come from the interaction of alpha particles with elements in the immediate vicinity of the Po²¹⁰. The most important secondary source is neutrons generated by alpha particle interaction with light element (oxygen, nitrogen, et cetera) Po²¹⁰ impurities. According to Mound Laboratories (refs. 1 and 2) the Po²¹⁰ neutron source strength is approximately 300 neutrons/sec-curie; however, this can be reduced to 100 neutrons/sec-curie, which is equivalent tc 3.1×10^3 neutrons/sec-watt, by carefully controlling fuel form and encapsulation fabrication processes. Additional Po²¹⁰ nuclear properties are presented in figure 1.

PLUTONIUM-238 RADIATION CHARACTERISTICS

Plutonium-238 (Pu^{238}) decays by alpha emission with a half-life of 89.6 years to the excited states of uranium-234 (U^{234}) or to the ground state of U^{234} as shown in figure 2 (ref. 1). Uranium-234 in the excited states decays to its ground state by emitting low-energy gamma rays.

Plutonium-238, in addition, undergoes spontaneous fission with a half-life of 4.9×10^{10} years. The fission process produces both neutrons and prompt fission gamma rays. In addition, the fission products emit additional gamma rays.

In addition to the above Pu²³⁸ intrinsic radiation sources the following secondary radiation sources exist:

(a) Prompt fission gamma rays and neutrons, and fission product decay gamma rays due to the induced fissioning of Pu²³⁸.

Because Pu²³⁸ is fissionable, a neutron flux is generated due to subcritical multiplication. Under the condition of subcriticality, the subcritical multiplication factor or the ratio of neutrons emitted in a multiplying medium to the number of neutrons originally present in the medium is equal to (ref. 3):

$$\frac{n}{n_{o}} = \frac{1}{1 - k} (k < 1)$$
(1)

A Pu²³⁸ source which has a k less than one is safe from the standpoint f criticality; however, for shielding calculations, the number of neutrons emitted from the radioisotope source is n rather than n_0 . For example, in a radioisotope source with k = 0.5, $n/n_0 = 2$; that is, the number of neutrons requiring shielding because of the multiplying medium is two times that originally present.

(b) Gamma rays resulting from the decay of Pu-236 impurity in the Pu-238.

Plutonium-238 is formed by the reactor irradiation of Np²³⁷. It is formed by the Np²³⁷ (n, γ) Np²³⁸ reaction which decays by beta emission to Pu²³⁸. Plutonium-236 is also formed by the reactor irradiation of Np²³⁷; however, it is formed by the Np²³⁷ (n,2n) Np²³⁶ reaction which decays by beta emission to Pu²³⁶. Because the n,2n reaction for Np²³⁷ is a 5 Mev threshold reaction, the yield of Pu²³⁶ in as-produced Pu²³⁸ product is small -- generally about one part per million (ppm) by weight (refs. 1 and 4). Plutonium-236 decays to lead-208 (Pb²⁰⁸) and in the process emits high energy (greater than 1 Mev) gamma rays. The resulting dose rate from Pu²³⁶ decay gamma rays is greater than the gamma ray dose rate from Pu²³⁸ approximately 2 years after the formation of Pu²³⁶, as shown in figure 3 (ref. 1). (c) Neutrons from the α , n reaction with materials in the immediate vicinity of the Pu-238.

Because plutonium dioxide $(Pu^{238}O_2)$ is the recommended fuel form for temperatures greater than 650° C (ref. 5), $Pu^{238}O_2$ was used for shielding calculations. The use of the oxide form, however, increases the neutron source from the existing spontaneous fission of Pu-238 by a factor of 10 (ref. 4). This is due to the a,n reaction with oxygen. This reaction is a threshold reaction and takes place only if the alpha particle energy exceeds the target nucleus reaction threshold. Natural oxygen contains three isotopes; namely 0¹⁶, C¹⁷, and 0¹⁸ in the following abundances: 99.759 percent, 0.337 percent, and 0.204 percent respectively (ref. 1). The a,n reaction thresholds for 0¹⁶, 0¹⁷, and 0¹⁸ are 15.19 Mev, zero, and 0.36 Mev respectively. In the alpha decay of Pu²³⁸ the maximum alpha particle energy is 5.495 Mev and since the 0¹⁶ reaction threshold is greater than 5.495 Mev the a,n reaction will not take place with 0¹⁶. By reducing the 0¹⁷ and 0¹⁸ isotopes in oxyger, that is, using nearly pure 0¹⁶ in Pu²³⁸O₂, the neutron source can be reduced by factors up to a maximum of 10.

Neutron Attenuation

Neutron attenuation processes consist of the gradual degradation in energy of the fast (Mev range) neutrons* by means of elastic and inelastic scattering and, eventually, capture at a reduced energy. The three processes which are important for neutron attenuation are: elastic scattering, inelastic scattering, and capture. The probability of any of these interactions occurring depends upon the interaction material and on the neutron energy (ref. 6).

*The fission and α , n neutrons have varying energies at time of birth. The fission neutron energy spectrum spans the range of approximately 0.1 to 10 Mev with the most probable energy being approximately 1 Mev. The α , n neutrons generally cover a smaller energy range and have a higher, most probable energy. For example, Po²¹⁰ α , n neutrons generated with oxygen have an energy range of approximately 0.8 to 4.8 Mev, and a most probable energy of approximately 2.5 Mev.

Elastic scattering.- Elastic scattering refers to the billiardball type collision of a neutron with a nucleus. An elastic scattering event changes both the direction and energy of the neutron. The energy lost in an elastic scattering collision is inversely proportional to the mass of the target nucleus. A neutron can give all its energy to a hydrogen nucleus by elastically scattering from it, but can lose only a fraction of its energy by elastically scattering from high atomic weight nuclei. Neutrons of any energy can undergo elastic collisions.

<u>Inelastic scattering</u>.- Inelastic scattering occurs when a neutron is temporarily absorbed by a nucleus, which is raised to an excited state and shortly thereafter emits a neutron in a different direction from that of the original neutron. The emitted neutron has less energy than the original neutron, with almost all the difference in energy being emitted in the form of gamma rays. This reaction requires that the neutron have an energy in excess of the energy of the first excited state of the target nucleus. In general, fast neutrons are more readily degraded in energy by being inelastically scattered by heavy nuclei than by light nuclei. For a neutron below about 1 Mev, the scattering will usually be elastic, whereas inelastic scattering becomes increasingly probable as the neutron energy rises.

<u>Capture</u>.- Scattering processes, whether elastic or inelastic, do not eliminate neutrons but only change their direction and reduce their energy. Capture reactions, which absorb or eliminate neutrons, do not compete favorably with scattering reactions for fast neutrons because the capture cross-section is inversely proportional to neutron energy.

When a neutron is captured, the capturing nucleus emits a charged particle or gamma ray. In addition, the newly formed nucleus may also be iadioactive and subsequently emit particles and/or additional gamma rays.

Gamma Ray Attenuation

The three processes which are important for gamma ray attenuation are the photoelectric effect, pair production, and Compton scattering (ref. 6).

<u>Photoelectric effect.</u> The effect occurs when a gamma ray transfers all its energy to one of the atomic electrons in the shielding material and ejects the electric from the atom. The cross section for photoelectric emission decreases rapidly as the energy of the photon increases. <u>Pair production</u>.- In pair production, all the energy of the incident photon is transferred into creating an electron pair; an electron and a positron. The kinetic energy of the pair is equal to the difference between the energy of the photon and the rest-mass energy of the pair, that is, 1.02 Mev (ref. 6). The pair particles, being charged, rapidly give up their energy to the surrounding material. Pair production can only be induced by gamma rays having more than 1.02 Mev of energy. It can occur in the field of either the atomic electrons or the nucleus. The total pair-production cross section thus rises rapidly with atomic number and also with energy.

<u>Compton scattering</u>. - Both preceding interactions are absorptive processes; the photon disappears entirely. In contrast, the Compton effect is a scattering process which alters the direction and energy of the incident photon but does not eliminate it.

In order to take the Compton effect into account without a very complex calculational procedure, it has become popular to use a socalled Buildup Factor. The Buildup Factor is the ratio of the actual gamma ray flux at a point in a shield to what would be expected if Compton scattering were considered as an absorptive process. The Buildup Factor, in general, decreases with increasing Z and with increasing gamma ray energy, while increasing with the thickness of shielding material.

SHIELDING MATERIALS

The following criteria must be considered in the selection of neutron shielding materials:

(a) High hydrogen density for elastic scattering of neutrons

(b) Inclusion of elements having a large thermal-neutron-capture cross section

(c) Inclusion of elements capable of inelastic scattering -important for neutron energies greater than about 0.5 Mev

- · (d) Ease of fabrication
 - (e) Good temperature stability
 - (f) Good radiation stability

In essence an effective neutron shield must contain components that contribute effectively to each of the neutron attenuation processes (scattering and capture). Thus, although a hydrogenous material such as lithium hydride (LiH) is effective in the elastic scattering energy region, its small removal cross section in the inelastic energy region means that more effective shielding per unit volume can be achieved by including materials such as tungsten, uranium, or lead that have reasonable removal cross sections in this region. Generally the dense, inelastic scattering material is placed between the radioactive source and the hydrogenous material. This arrangement provides the necessary neutron inelastic scattering and degrades the neutron energy so that the hydrogneous material can further degrade the neutron energy by elastic scattering. Fortunately this arrangement also provides the lightest weight shield by placing the dense material as close as possible to the radioactive source. Following elastic scattering by the hydrogenous material the neutrons can be captured at near thermal energies by a thin layer of boron-containing material. Boron is an effective capture material because (1) it has a large capture cross section (3820 barns at 0.025 ev) (ref. 7), and (2) produces alpha particles -- which travel only a few centimeters in air -- rather than gamma rays.

The criteria for selection of gamma ray shielding materials are as follows:

(a) Large attenuation coefficient (probability of energy degradation per unit thickness)

- (b) Ease of fabrication
- (c) Good temperature stability
- (d) Good radiation stability

These criteria point to a derse material such as uranium-238 or tungsten, which as discussed above, also serves in providing neutron inelastic scattering.

For this analysis uranium-238 and LiH were chosen. Because of the small weight required for a boron addition* this factor was not included in the analysis.

*A commercial material called Boral Plate -- consisting of a 1/4-inch aluminur clad sandwich-type plate with a core of boron carbide (B_4C) crystals suspended in cast aluminum -- has an average weight of 3.44 lb per ft².

SHIELD ANALYSIS

Neutron Shielding

The neutron dose rate criteria used was 1 mrem/hr at the dose point. Assuming an average neutron energy emerging from the LiH of 1.0 Mev (ref. 8), 1 mrem/hr is equivalent to a neutron flux of about 10 neutrons/cm²-sec (ref. 9).

The thicknesses of LiH required for Po^{210} neutron flux attenuation as a function of radioisotope thermal power were calculated from reference 10, figures 78* and 2, using the conversion 0.014 mrep/hr per neutron/cm²-sec (ref. 11). The results are shown in figure 4. For source to dose point separation distances greater than 100 cm -- as provided by reference 10, figure 78 -- the mrep/hr obtained for 100 cm were multiplied by the ratio $100^2/R_i^2$, where R_i varies from 150 to 350 cm. The mrep/hr obtained at R_i were then converted to neutrons/ cm²-sec as before, and the thickness of LiH required for attenuation to 10 neutrons/cm²-sec were obtained. The results are shown in figure 5.

The $Pu^{238}c_2^{16}$ neutron source strengths were calculated using 3×10^3 neutrons/sec-gm** which is equivalent to 7.4×10^3 neutrons/sec-watt. The additional LiH thickness required for Pu^{233} neutron attenuation was calculated as follows:

$$\frac{I(P_0^{210})}{I(P_u^{238})} = \exp\left(-N\sigma_n x_{LiH}\right)$$
(2)

*Thermal powers up to 125 kW were extrapolated from figure 78. **Savannah River measured 2.1 × 10⁴ neutrons/sec per gm of Pu²³⁸0₂ (ref. 4). Assuming a factor of 7 reduction by use of 0¹⁶ gives 3 × 10³ neutrons/sec-gm. where

$$I(Po^{210})/I(Pu^{238}) = ratio of Po^{210} to Pu^{238}$$
 neutron source strengths, that is, 0.42

 $N\sigma_n$ = neutron removal cross section = 0.15 cm⁻¹ (ref. 1)

x_{LiH} = thickness of LiH

Fquation (2) was solved for x_{LiH} to yield 5.8 cm as the additional LiH thickness required for $Pu^{238}O_2^{16}$ neutron attenuation.

The analytical data of reference 10 were calculated by the moments method solution of the neutron transport equation. A fission energy spectrum of source strength 100 neutrons/sec-curie was used. Recent Monte Carlo neutron attenuation calculations with LiH by Oak Ridge National Laboratory* indicates that reference 10 data are conservative. This conservatism is deemed necessary for a parametric applications

analysis because the Pu²³⁸ source strength does not include neutron subcritical multiplication fissioning. This factor will increase the source neutron flux by a factor of approximately 1.5 to 5, thereby increasing the required LiH shielding. Because the subcritical multiplication is a function of fuel capsule and heat source geometry, inclusion of this term is beyond the scope of a parametric analysis.

Gamma Ray Shielding

Because the LiH neutron shield and intervening spacecraft structure reduce the gamma ray flux, the required uranium shielding is reduced.** Intervening spacecraft structure was parametrically included as 0, 20, and 40 cm equivalent thickness of aluminum. The required thicknesses

*Data to be published.

**Spacecraft structure will also eliminate neutrons; however, no structure neutron attenuation credit was taken.

of uranium to provide 1, 3, and 5 mrem/hr gamma ray dose rates at the dose point were calculated from the following equation:

$$\phi = \frac{SB}{\mu_{\pi R}^2} \exp \left(-\frac{\Sigma \mu_i x_i}{i} \right)$$
(3)

where:

- S = gamma ray source strength; S varies from 2.08 × 10¹¹ Mev/ sec (50 kW) to 5.23 × 10¹¹ Mev/sec (125 kW) (a heat source self-shielding factor of four was assumed)
- B = dimensionless gamma ray Buildup Factor for the composite shield of uranium, LiH, and aluminum
- R = source point to dose point separation distance; R varies from 100 to 350 cm
- µ = mass attenuation coefficient for uranium, LiH, and aluminum: cm⁻¹*
- x. = thickness of shield material for uranium, LiH, and aluminum; cm

Equation (3) assumes a point radioisotopic source. This assumption is valid when the source point to dose point separation distance is equal to or greater than the radioisotope heat source dimensions.

The Buildup Factors for uranium, LiH**, and aluminum were obtained from reference 6 and are presented respectively as figures 6, 7, and 8.

*The mass attenuation coefficients are generally given in units of cm²/gm and are converted to cm⁻¹ by multiplying by the material density in gm/cm³.

**Reference 6 does not provide Buildup Factors for LiH; consequently, the Buildup Factors for water, which has approximately the same hydrogen atom concentration per cm³, were used.

For the composite shield (urenium, LiH, and aluminum) the Buildup Factor used in equation (3) was calculated as the product of the individual Buildup Factors (ref. 8). For Po²¹⁰ and Pu²³⁸ an average gamma ray energy of 0.8 Mev was assumed, and figures 6, 7, and 8 were used to find B as functions of μx . Equation (3) was then solved by iteration for x uranium. The results, for 1, 3, and 5 mrem/hr, are presented respectively as figures 9, 10, and 11.

By using one calculation (equation (3)) for $Pu^{23\delta}$ and Po^{210} uranium thicknesses it was assumed that the $Pu^{23\delta}$ and Po^{210} gamma ray source strengths are equal. This would be the case where the $Pu^{23\delta}$ has 1 ppm $Pu^{23\delta}$, and is approximately 2-year-old fuel on delivery. For this case, the $Pu^{23\delta}$ and Po^{210} gamma ray shielding requirements are approximately equal (ref. 1, figs. VII-9 and VII-12).

PARAMETRIC SHIELD WEIGHTS

Using the calculated thicknesses of uranium and LiH, and the heat source shield geometries of figures 12 and 13, parametric shield weights were calculated for Pu^{238} and Po^{210} . Figures 14 and 15 respectively present 4π and shadow shield parametric weights for Pu^{238} . Figures 16 and 17 respectively present 4π and shadow shield parametric weights for Pu^{210} .

The tapered- 4π shield provides maximum required protection in the forward direction, and incrementally reduced protection on the sides and bottom (see fig. 12). The shadow shield provides maximum required protection on the top and two sides; the bottom and two remaining sides are assumed to require no shielding. From a practical standpoint some shielding will probably be required on all six sides to reduce gamma ray reflection and neutron, spacecraft structure activation.

The heat source is surrounded on all sides by 1 inch of superinsulation to reduce heat leak from the heat source, and the LiH is enclosed in a 0.050-inch stainless steel can to prevent hydrogen dissociation loss.

The following example illustrates the use of figures 14 through 17.

P. 238

Radioisotope

Shield geometry	4π.	
Shield parameters	·	
Separation distance	100	cm
Heat source cube size	22	in.
Thermal power	150	kW*
Gamma ray dose rate	3	mrem/hr
Spacecraft aluminum structure	40	cm

Shield weight is calculated from figure 14 as follows:

1950 lb × 1.07 (separation distance) × 1.0 (heat source cube size) × 1.02 (thermal power) × 1.0 (gamma ray ãose rate) × 0.7 (cm of aluminum) = 1950 lb × 0.76 = 1482 lb

CONCLUDING REMARKS

Plutonium-238 shields are heavier than Po^{210} shields when the asproduced Pu^{238} contains approximately one part per million of Pu^{236} , and the Pu^{238} is more than 2-year-old material. The Pu^{238} shield could be lighter -- reduced gamma ray shield material -- if the Pu^{236} is removed or the Pu^{238} is less than approximately 1 year old. Plutonium-238 will, in all cases, require more neutron shield material. For manned missions it is imperative that the neutron sources be reduced as much as possible; for Pu^{238} through the use of $Pu^{238}o_2^{16}$, and heat sources designed for small subcritical multiplication.

It is advantageous to take advantage, where possible, of spacecraft structure for crew personnel shielding. Twenty cm equivalent thickness of aluminum is approximately equal to a factor of three reduction in dose rate.

*Thermal powers were extrapolated from 125 kW to 1.50 kW for plotting purposes in figures 14 through 17.

Experimental shielding studies need to be conducted for Po^{210} and $Pu^{238}O_2^{16}$ with composite layer shields of tungsten, uranium, and LiH. Only by means of such studies, and by theoretical correlations -- Monte Carlo computer studies, for example -- can radioisotope heat sources and shields be properly designed for manned space applications.

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Note: Other radiation - - 70 - 300 neutrons/sec - curie from α , n reaction

Curies per gram......4,500.55 Watts per curie......0.03203

Figure 1. - Decay scheme and nuclear properties of polonium-210.





Note: Other radiation --fission and fission product gamma rays --gamma rays from decay of Pu²³⁶ impurities --fission and α , in neutrons Curries per gram --11.81 (80% Pu²³⁸) Curies per watt --29.1 (80% Pu²³⁸)

Figure 2. - Decay scheme and miclear properties of plutonium-238.

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Figure 5. - Polonium-210 LiH shielding thickness.





Figure 7. - Gamma ray buildup factor for LiH.





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Figure 9. - Polonium-210 and plutonium-238 uranium shielding thickness - 1 mrem/hr, y dose rate.





Figure 10. - Polonium-210 and plutonium-238 uranium shielding thickness - 3 mrem/hr, γ dose rate.



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Figure 11. - Polonium-210 and Plutonium-238 uranium shielding thickness - 5 mrem/hr,y dose rate.

27

a







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Figure 14. - Plutonium-238 parametric shield weights - 4 # shield.

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1



Figure 15. - Plutonium-238 shield weights - shadow shield.

and with



Figure 16. - Polonium-210 shield weights - 4 # shield.

4

NASA-5-65-2082 FEB



Figure 17. - Polonium-210 shield weights - shadow shield.