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Nucleonic Instrumentation

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PLUTONIUM DETERMINATION*

by

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January 15, 1963

General Electric Company
Hanford Atomic Products Operation
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PLUTONIUM DETERMINATION

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INTRODUCTION

Scrap and waste materials which potentially contain small quantities of plutonium are accumulated in several plant facilities. For reasons of accountability, nuclear safety, and production economics, it is desirable tc determine quantitatively the plutonium content in various types of sealed containers to permit appropriate storage and processing. The nuclear radiations emitted by plutonium provide one potential method for assaying these sealed containers to determine the plutonium content.

SUMMARY

It is concluded that the plutonium content within a sealed container can be determined accurately if the plutonium is homogeneously distributed in the filler material which is uniformly distributed throughout the container or if no absorption of the radiation takes place within the container.

The 384 kev photon resulting from the decay of Pu-239 appears to provide the best radiation source for quantitative measurements; however, it is believed that an experimental evaluation will be required to determine the useful operating parameters. Such an evaluation would require the use of "standard" containers of known plutonium content and density so the photon energy distribution could be studied as well as source-detector geometry relationships. These "standard" containers could be used on a continuing basis to calibrate any utilized detection system and for any required subsequent developmental effort.

It does not appear feasible to utilize the neutron emission rate as a basis for determining plutonium content of sealed containers since the neutron emission rate can be affected severely by the presence of other elements.

DISCUSSION

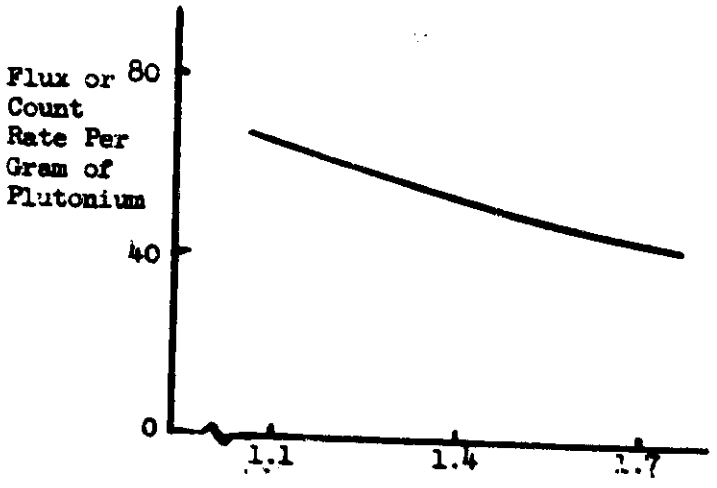
Plutonium is a fissionable material which is both valuable and toxic. For these reasons it is imperative that plutonium be carefully inventoried throughout the production cycle. Material and equipment which are used to process plutonium may become contaminated, and such process materials are saved and placed in sealed cartons or cans for storage. Specific process material, which is known to contain plutonium is stored until reprocessed for plutonium recovery. In other cases the waste material may contain little or no plutonium; thus, reprocessing is economically inadvisable. For nuclear safety, for accountability, and for economic reasons, it is desirable to be able to determine quantitatively the container plutonium content to permit proper storage, reprocessing, or disposal.

Although the sealed containers may be of various sizes and shapes, a feasibility study can be made which is independent of these considerations. For a container in which no self-absorption takes place, the plutonium content could be determined accurately by monitoring the entire surface area. With no self-absorption, the integrated flux through the surface would relate directly to the source strength. From a more practical standpoint, neutrons and photons are attenuated exponentially in traversing matter, and the disposition of the material within the container is of primary importance. In the case of extremely inhomogeneous material,

it is impossible to interpret the leakage radiation to determine plutonium content. However, if the plutonium is homogeneously distributed in the carrier material, which is in turn uniformly distributed throughout the container, the leakage flux is directly proportional to the plutonium content and an accurate quantitative determination is possible. If it is assumed that the detector can discriminate against all other radiations, the flux from a specific source at the detector is a function of the source density, the absorption coefficient of the contained material, and the geometrical relationship which exists between the container and the detector. For a given geometric arrangement, Figure 1 shows the relationship which would exist between the flux at the detector (count rate) per gram of contained plutonium and the specific gravity, which is related to the absorption coefficient of the carrier material.

Figure 1

Count Rate Per Gram of Plutonium as a Function of Specific Gravity of Material.



From the practical standpoint the following items must be considered:

1. Does plutonium emit a characteristic radiation which can be discriminately detected?
2. Can reasonable count rates be obtained from specified quantities of plutonium in standard containers?

3. Can the system be made insensitive to small variations in the geometrical arrangement?

4. Can corrections be made to account for the specific gravity of the filler material?

The following discussion indicates that a practical system should be possible; however, an experimental evaluation seems in order to establish the operating parameters which should yield the best results.

Radiation Sources

Plutonium is produced in a nuclear reactor primarily by neutron absorption in U-238. The isotope Pu-239 is produced as the U-239 formed by neutron absorption beta decays to Np-239 which subsequently beta decays to Pu-239. Because of the long half-life and large neutron absorption cross section of Pu-239, a considerable number of the atoms produced are, in turn, converted to Pu-240 and Pu-241 by the absorption of additional neutrons. The isotopic content of plutonium produced in a nuclear reactor is thus dependent on the exposure or irradiation history of the fuel element. Since the exposure is controlled, the isotopic composition of the plutonium which would accumulate in the waste and scrap material should be similar in all cases. Table I, which is based on a study by Roesch* summarizes the radiation which is expected from a gram of production quality plutonium.

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* W. C. Roesch, Surface Dose From Plutonium, Geneva Conference Paper, (1958), P/755.

Table I

Important Radiation from Plutonium
Per Gram

Isotope	Neutrons Per Second	Photon Energy Mev	Photons Per Second
Pu-239	0.03	<0.02	6.2×10^{7}
		<0.125	3.6×10^{5}
		0.384	3.2×10^{4}
Pu-240	83	<0.044	5.0×10^{8}
		$\gtrsim 1$	3.4×10^{2}
Pu-241	0	<0.1 (spectrum)	Am-241 (see Note 1)
		<0.334 (spectrum)	U-237 (see Note 2)

Note 1: Over 99% of the Pu-241 transitions are by beta emission to Am-241. The low energy photons are actually emitted as Am-241 decays. Because the half-life of Am-241 is significantly longer than that of Pu-241, the activity continues to increase with time in the range of interest. Thus, this radiation is unsuitable as a source for determining Pu content.

Note 2. About 0.004% of the Pu-241 transitions are by alpha emission to U-237. The half-life of U-237 is 6.75 days; hence, an equilibrium concentration of U-237 will be reached within a few weeks after the separation process. It would be inadvisable to use these radiations to determine Pu content.

High energy photons should provide the best source of radiation since there would be less self-absorption in the material and there would be little or no problem due to scattered radiation. However, a spectrum of high energy photons, such as emitted by Pu-240 as a result of spontaneous fission does not provide a convenient source.

A significant number of neutrons are emitted as a result of the spontaneous fission of Pu-240 and the detection of these might provide a method of determining plutonium content; however, when elements such as fluorine are present, neutrons are also generated by the (α,n) reaction. Fluorine produces about ten neutrons for each 10^{6} alpha particles which are slowed down in it. A gram of the production quality plutonium considered above

would emit in excess of 10^9 alpha particles per second; thus, more than 10^4 neutrons could be produced if these were all slowed down in fluorine. If only one percent of the alpha particles were slowed down in fluorine, more than 10^2 neutrons per second would be generated as compared to the 83 neutrons per second resulting from spontaneous fission of Pu-240. It is obvious that neutron detection could not be utilized to assay for plutonium if fluorine or other such elements are present unless neutron spectroscopy was employed.

It is concluded that the 0.384 Mev photon from Pu-239 provides the most usable source for determining plutonium content in sealed containers.

Geometrical Considerations

One container for the scrap and waste material is a standard Number 2-1/2 can which is 4 inches in diameter and 5-1/2 inches tall. A typical can might contain about 1600 grams of material with an average density of 1.4 grams per cm^3, which would have an absorption coefficient of about 0.137 cm^{-1} for the 0.384 Mev photons. The flux of photons of this energy through the end of the can would be given approximately by:

$$\phi \approx \frac{S}{2u} \left[1.0 - E(uh)\right]$$

where S is the source strength in photons per cm^3, u is the self-absorption coefficient, h is the height of the can, and the function E is:

$$E(uh) = uh \int_{uh}^{\infty} \frac{e^{-x}}{x^2} dx$$

Assuming one gram of plutonium to be uniformly distributed throughout the can:

$$S = \frac{3.2 \times 10^4}{1.1 \times 10^3} \approx 30 \text{ photons per } cm^3 \text{ per second}$$

where 11[illegible] cm^3 is the volume of the can. The flux is calculated to be about 10^{4} photons per cm^2 per second. This would be a detectable flux level; thus, it is concluded that adequate sensitivity should be available for detecting one gram quantities of plutonium in these particular containers.

A cylindrical container could also be viewed from the side to measure the gamma flux to determine plutonium content. In this case, the flux is given approximately by the relation:

$$\phi \cong S \frac{R^2}{2x} F(\Theta, uz)$$

where R is the radius of the contained material, x is the effective distance between the detector and a line source representative of the material contained within the cylinder, and the F function is:

$$F(\Theta, uz) = \int_0^{\Theta} e^{-uz \sec \Theta'} d\Theta'$$

where Θ is half the angle subtended by the line source at the detector. The flux intensity at the side of the container would be comparable to that at the end, so either geometry could be employed.

In the case of large containers, similar relations would exist; but, because of distances involved, the photon flux per unit area would be less per unit of contained plutonium. Containers other than spheres or cylinders can be approximated by these shapes so simple analytical calculations can be made. In all cases, it must be assumed that the material is uniformly distributed.

It can be shown that it is impossible to make a quantitative determination of the plutonium content of a sealed container if the material is not homogeneous. Consider, for example, a one gram "point" source of

plutonium embedded in a standard 18-inch by 18-inch by 24-inch waste carton containing about forty pounds of waste. The average density of typical waste material would be about 0.145 grams per cm3. The absorption coefficient of this material for the 0.384 Mev photons would be about 0.014 cm^{-1}. The flux as a function of distance, if the point source were embedded in the uniformly distributed filler material, would be:

$$\phi = \frac{Se^{-ur}}{4\pi r^2}$$

Thus, if the gram of plutonium were five cm from the surface, the flux at that surface would be about 100 photons per cm^2 per second. If the plutonium were at the middle of the container, the flux would be about five photons per cm^2 per second at the surface. If the gram of plutonium were located near the far side of the container, the flux would be negligible. Determination of the plutonium content in this case would require that a complete scan of all surface areas be taken and a comprehensive analysis made using the data. Even with all the data it would be impossible to determine the content precisely, since the same surface data could result from a number of distributions.

A situation more difficult to analyze than that discussed would occur if the gram of plutonium were located near one surface of the container with dense shielding material between the plutonium and the nearby container surface. In this case, it would be possible to have negligible flux leakage at that surface because of the shielding and negligible leakage at the far surface because of the distance. The plutonium would be undetectable. This is perhaps an extreme situation, but indicates that system accuracy would be limited by the homogeneity of the contained material.

It is concluded that the reliability of determinations made of non-uniformly filled containers will always be low. Experimental evaluations will be required to determine the geometrical relationships and correction factors for specific gravity of the contained material. The experiments should encompass the range of variations anticipated during normal, routine operation.

REFERENCE

Elliot, F., and G. W. Pearson, A Gamma Monitor for the Rapid Assay of Plutonium in Process Waste Materials, United Kingdom AEC PG Report 278(W), 1962.