Fig 330
7/24/67

MASTER

RFP-954

MEASUREMENTS OF PLUTONIUM 238

THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
P. O. BOX 888
GOLDEN, COLORADO 80401
U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT(29-1)-1106

DISTRIBUTION OF THIS DOCUMENT, IS UNLIMITED.

Fig

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

30 June 1967

RFP-954
UC-41 HEALTH AND
SAFETY
TID-4500

CPSTI PRICES

H.C. \$ 3.00; MN .65

MEASUREMENTS OF PLUTONIUM 238

*John R. Mann
Robert W. Bistline
Valens P. Johnson*

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

THE DOW CHEMICAL COMPANY
ROCKY FLATS DIVISION
P. O. BOX 888
GOLDEN, COLORADO 80401
U. S. ATOMIC ENERGY COMMISSION
CONTRACT AT(29-1)-1106

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

CONTENTS

MEASUREMENTS OF PLUTONIUM 238

Abstract	1
Introduction	1
Experimental	1
Samples	1
Equipment	2
Sample Preparation and Counting	3
Results	3
Conclusions	5
Future Studies	5

MEASUREMENTS OF PLUTONIUM 238

John R. Mann, Robert W. Bistline, and Valens P. Johnson

Abstract. Evaluation has been completed of detectors in use at Rocky Flats for determining detectable plutonium amounts in the body. Specifically, the study included sample measurements of plutonium 238, *in vivo*, with a scintillation spectrometer detector. The findings, using a phantom chest model, showed that four times the maximum permissible lung burden was the minimum which could be detected. The minimum detectable amounts in the liver were less than one permissible liver burden.

INTRODUCTION

Measurements of plutonium *in vivo*^{1,2,3} are made routinely at the Rocky Flats Plant with various detectors. Currently, a study was initiated to determine the minimum detectable amount (MDA) of plutonium 238 (²³⁸Pu) found in the chest with detectors now in use. For the experiment a phantom chest model was set up and three samples of ²³⁸Pu were used.

EXPERIMENTAL

Samples: For the testing, three samples of ²³⁸Pu were furnished by the Monsanto Research Corporation in Miamisburg, Ohio. The contents for each follows:

Sample No. 1: Virgin plutonium 238 – 1 month old, 0.955 microcuries, 10-milliliter volume, plutonium nitrate [Pu(NO₃)₄].

Analysis:	Isotope	Weight Percent
	238	81.51
	239	15.13
	240	2.67
	241	0.609
	242	0.086

Sample No. 2: Aged scrap prior to purification – 1 year old, 0.957 microcuries, 10-milliliter volume, plutonium nitrate [Pu(NO₃)₄].

Analysis:	Isotope	Weight Percent
	238	77.42
	239	18.83
	240	2.91
	241	0.723
	242	0.113

Sample No. 3: Recovered plutonium 238 – 2 months old, 1.014 microcuries, 10-milliliter volume, plutonium nitrate [Pu(NO₃)₄].

Analysis:	Isotope	Weight Percent
	238	79.88
	239	15.91
	240	3.24
	241	0.815
	242	0.154

Gamma spectra of each sample were made using a 2-inch diameter by a 1/2-inch thick sodium iodide, thallium activated [NaI(Tl)] detector with a 5-mil beryllium entrance window, and a 1600-channel, pulse-height analyzer. Typical spectra are shown in Figures 1, 2, and 3.

Sample No. 3 was chosen for the experiments described in this report, since it represented the "cleanest" plutonium that might be encountered in actual practice. This would present the worst possible case for detection of ²³⁸Pu since higher-energy photons from americium or other daughter products would not be available for use as natural tracers.

Earlier experiments established that a 4-inch by 4-millimeter (mm) NaI(Tl) detector placed over either the right or the left side of the phantom chest would not be affected by plutonium placed in the opposite side of the chest.⁴ Only half of the chest cavity was

¹ M. R. Boss and J. R. Mann. "Body Counter Evaluations of Plutonium Exposures." *Health Physics*, 13:259. 1967.

² J. R. Mann and R. A. Kirchner. *Evaluation of Lung Burden Following Acute Exposure to Highly Insoluble Plutonium Oxide (PuO₂)*. RFP-710. Rocky Flats Division, The Dow Chemical Company, Golden, Colorado.

³ E. A. Putzier et al. *Personnel Dosimetry for Radiation Accidents*. International Atomic Energy Agency, Vienna, Austria. 1965.

⁴ M. R. Boss and J. R. Mann. *Loc. cit.*

FIGURE 1. The Gamma and X-ray Spectrum of Plutonium 238, Sample No. 1, Virgin Material.

used in the experiments. The calculated activity of ^{238}Pu in one lung which delivers a dose of 0.3 (roentgen equivalent man) rem per week is 0.000 microcuries (μCi). This quantity in one lung is representative, therefore, of a maximum permissible lung burden (MPLB).

The maximum permissible liver burden is equivalent to 0.022 μCi .

The expected activities of the most abundant gamma rays and X-rays from 0.008 μCi of Sample No. 3 are shown in Table I.

TABLE I. Photon Abundance from 0.008 Microcuries of Plutonium 238 in Sample No. 3.

Isotope	Weight Percent	Photon Energy (kiloelectron volts)	Photons per Second
Plutonium 238 (^{238}Pu)	80.21	17	30.
		43	0.077
Plutonium 239 (^{239}Pu)	15.82	17	0.00477
Americium 241 (^{241}Am)	0.0143	60	0.0029

The observed gamma spectra confirmed the expectation that the 17-kiloelectron volt (keV) X-rays from ^{238}Pu were by far the most abundant. It was found later that the phantom attenuated the 17-keV line to an abundance only slightly greater than the 43-keV gamma rays.

Equipment: To simulate *in vivo* measurements in the body counter, a REMAB phantom was used; available from the Alderson Research Laboratories, Long Island, New York. A typical arrangement can be seen in Figure 4. The lungs were filled with a lung-equivalent compound provided by Alderson for chest measurements. Rib bones were in place during the chest measurements since the 17-keV X-rays are much more severely attenuated by bone [half thickness = 0.02 centimeters (cm)] than tissue (half thickness = 0.68 cm). In preparation for liver measurement, the liver cavity was filled with a nitric-acid solution containing a predetermined amount of ^{238}Pu .

Most of the data were obtained with two 4-inch diameter by 4-mm thick NaI(Tl) detectors having 10-mil beryllium entrance windows. A commercially available end

FIGURE 2. The Gamma and X-ray Spectrum of Plutonium 238, Sample No. 2, Aged Scrap.

window, xenon-filled proportional detector and a 500-cm² proportional detector⁵ of Rocky Flats design were also used in the study.

Sample Preparation and Counting: The sample preparation, mixing, and loading into the phantom were carried out in a glovebox. Table II summarizes the sample loads made. The aliquots were kneaded in with the lung compound in plastic bags and then the bags were stuffed into the cavities.

The loads were mixed with a sufficient volume of lung-equivalent compound to fill up the right or left cavity. The phantom was placed in the body counter and the detectors were positioned over the upper, lower, or both portions of one side of the chest. For the liver measurements, the detector was positioned just below and to the right of the sternum. Counting times of 40 or 80 minutes were used. The electronics were set to cover a range of from 1 to 127 kev. In the 17-kev (average) X-ray peak, the counts were integrated from

⁵ W. H. Tyree. *A Large Area Gas Proportional Detector for Low-Energy Photons*. RFP-637. Rocky Flats Division, The Dow Chemical Company, Golden, Colorado. January 6, 1966.

FIGURE 3. The Gamma and X-ray Spectrum of Plutonium 238, Sample No. 3, Recovered Material.

FIGURE 4. The REMAB Chest Phantom in Position for Liver Counts.

9 through 24 kev. Counts in the 43-kev peak were integrated from 32 through 54 kev. Background runs were made using phantom and lung materials, with detectors placed in identical positions as for the plutonium runs.

TABLE II. Phantom Loads Prepared from Sample No. 3.

Load No.	Sample Volume (milliliters)	Activity (microcuries)	Multiple of Maximum Permissible Burden
1	0.75	0.076	9.5 lung
2	0.25	0.025	3.17 lung
3	0.25	0.025	1.14 liver

RESULTS

The first run in the body counter was made with Load No. 1 in the right-chest cavity. The detector was positioned over the upper portion of the chest. Run No. 2 was made with the detector moved to the lower portion of the chest. Runs No. 3 and 4 were made with two detectors operating simultaneously, and with separate readouts. For Runs No. 5 and 6, one of the detectors was positioned below the sternum over the phantom's liver (see Table III).

TABLE III. Summary of Counting Data for 17-Kiloelectron Volt (Average) X-rays.

Run No.	Load No.	Detector Location	Multiple of Maximum Permissible Burdens	Counting Time (minutes)	Gross Counts	Background Counts	Net Counts	Number of Standard Deviations (above background) ^a
1	1	Upper right chest	9.5	40	1333	1163	170	5.
2	1	Lower right chest	9.5	40	1526	1163	363	10.6
3	2	#1 Upper left chest	3.17	40	1297	1218	79	2.3
		#2 Lower left chest	3.17	40	1250	1101	149	4.5
		#1 Plus #2	3.17	40	2547	2319	228	4.7
4	2	#1 Upper left chest	3.17	80	2597	2436	161	3.3
		#2 Lower left chest	3.17	80	2409	2202	207	4.4
		#1 Plus #2	3.17	80	5006	4638	368	5.4
5	3	Liver	1.14	40	1723	1205	518	15.
6	3	Liver	1.14	80	3372	2410	962	19.6

^a A count different from the background count by one standard deviation only suggests an effect. In one that differs from the background by two standard deviations, an effect is reasonably definite. For a difference of three or more standard deviations, an effect is definitely established (see NBS Handbook No. 80, *A Manual of Radioactivity Procedures*, U. S. Department of Commerce, National Bureau of Standards, Washington, D. C., 1961).

FIGURE 5. Phantom in Body Counter with 0.025 Microcuries in Liver, Detector Placed over Liver and below Sternum; 40-Minute Count, Background Subtracted.

All of the pulse-height spectra obtained were similar. In the chest runs, there were no prominent photopeaks either at 17 or 43 keV. The 17-keV photons were attenuated such that the number of counts at this energy became approximately equal to the number in the 43-keV peak. A liver spectrum is shown in Figure 5.

TABLE IV. Body-Counter Sensitivity, Plutonium 238 Versus Plutonium 239.

Location	Counting Time (minutes)	Multiple of Maximum Permissible Burdens	
		Plutonium 238 (MDA)	Plutonium 239 (MDA) ^a
Lower chest	40	4.75 lung	0.47 lung
Upper and lower chest (sum of both detectors)	80	3.85 lung	—
Liver	40	0.65 liver	0.15 lung

^a Measurement made from 1000 parts per million by weight of americium 241, using the 60-kiloelectron volt gamma rays.

Comparison of lung-burden calculations using data from each energy showed almost identical results.

The MDA of ²³⁸Pu was calculated using the Rocky Flats normal distribution of counts obtained on 203 known "cold" subjects. A subject having a count in the energy of interest greater than 2 sigma (σ) above the mean of the distribution is considered to be contaminated. Using 4-inch by 4-mm NaI(Tl) detectors for the body counter, a comparison of the MDA between ²³⁸Pu and ²³⁹Pu is given in Table IV.

Data obtained from the proportional detectors are not shown. Neither detector was able to give evidence of 20 maximum permissible lung burdens in the phantom, so no further experiments were run.

CONCLUSIONS

1. The minimum lung burden that could be detected with the detectors presently in use, on a routine basis, is approximately 4 maximum permissible lung burdens.
2. Of the two types of detectors used, the 4-inch by 4-mm NaI (Tl) crystals are the more sensitive detectors for measuring ^{238}Pu in the lung.

FUTURE STUDIES

Two semiconductor detectors, a lithium-drifted germanium [Ge(Li)] and a lithium-drifted silicon [Si(Li)], were borrowed to obtain data for possible application to *in vivo* plutonium detection. The data show a significant improvement in sensitivity to both ^{238}Pu and ^{239}Pu . An array of practical size would consist of four 10-cm² Si(Li) semiconductor detectors. This would improve the MDA of 4 MPLB obtained with scintillation detectors in the 17-keV region to an MDA of 1.7 MPLB.

Data from the NaI(Tl) detectors show the 43-keV gamma rays to be equally as detectable as the 17-keV X-rays. The semiconductor detectors are capable of resolving the 52-keV X-rays of ^{239}Pu and the 60-keV gamma rays of ^{241}Am . It is therefore reasonable to assume that the 43-keV X-rays of ^{238}Pu would be detectable with an array of Ge(Li) detectors and that this would provide an improvement in sensitivity over the 17-keV X-rays. The extreme attenuation of the lower-energy X-rays by tissue and bone makes a wide variability in sensitivity, due to material distribution and to physiological differences from individual to individual.

Currently, no recommendation can be made to use the semiconductor detectors for routine ^{238}Pu *in vivo* detection without considerably more research and demonstration of applicability. The data appear extremely encouraging however for future use of such detectors, not only for *in vivo* measurement of ^{238}Pu but also for *in vivo* measurement of ^{239}Pu , independent of X-rays and gamma rays from other associated isotopes of plutonium.