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**A SIMULATION STUDY OF PLUTONIUM GAMMA
RAY GROUPINGS FOR ISOTOPIC RATIO DETERMINATIONS**

Raymond Gunnink

June 10, 1974

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A SIMULATION STUDY OF PLUTONIUM GAMMA RAY GROUPINGS FOR ISOTOPIC RATIO DETERMINATIONS*

Abstract

A promising nondestructive method for measuring plutonium isotopic abundance ratios is the analysis of neighboring gamma rays in a spectral grouping whose members belong to different isotopes. We have made a preliminary study of all such peak groupings capable of yielding

this kind of information. This report describes the groups studied and the procedures used, and it contains the results we have obtained using a computer program we developed for predicting the ultimate levels of precision that can be obtained.

Introduction

The complete analysis of plutonium frequently includes measurements for the isotopic ratios of $^{238,239,240,241,242}\text{Pu}$ and for ^{241}Am content. Although these ratios are generally determined by destructive methods, there is increasing incentive for, and some progress toward, measuring plutonium isotopic ratios by nondestructive techniques.

One of the more promising of such techniques involves the measurement of the gamma rays emitted by the radioactive sample. Good results have been obtained for solutions.¹ However, non-uniformly packaged or inhomogeneous samples are much more difficult to measure because of uncertainties in counting efficiency and in gamma-ray

attenuation by the sample matrix or other absorbing materials.

It has been recognized for some time that one way to minimize the influences of these experimental problems is to select a set of neighboring peaks in the spectrum that are due to the different isotopes in the sample and compute the isotopic ratios from the observed peak intensities. Since the energies are nearly equal, the efficiency and attenuation differences are small, and the ratio of isotopic abundances can be directly related to the peak intensities.

We have made a study of all peak groupings that can yield information on plutonium isotopic ratios. To help us in this study, we have developed a computer

*This work was sponsored by the Office of Nuclear Materials Safeguards of the U. S. Atomic Energy Commission.

program for synthesizing the pertinent groupings and simulating the analysis of

spectra taken under a wide variety of counting conditions.

Experimental Approach

If the intensities of two or more gamma rays that have similar energies, and arise from different isotopes can be measured accurately, then the ratio of the isotopic abundances can be calculated from the equation

$$A/B = \frac{I_1 Y_1}{\epsilon_1 Y_1} / \frac{I_2 Y_2}{\epsilon_2 Y_2} = \frac{I_1}{I_2} \frac{\epsilon_2}{\epsilon_1} \frac{Y_2}{Y_1},$$

where

A/B = isotope ratio of interest,

I_1, I_2 = peak intensities of the gamma rays arising from isotopes A and B,

ϵ_1, ϵ_2 = counting efficiencies for the respective gamma rays, and

Y_1, Y_2 = the gamma yield or branching intensities of the respective gamma rays.

Although it may not be possible to obtain accurate absolute values of ϵ and Y individually, it is possible to obtain accurate ratios over a small energy range. The accuracy of the calculated isotopic ratio is then largely limited by

the precision with which the peak intensities are measured.

Gamma-ray energies and emission probabilities have been reported previously,^{2,3} and some preliminary studies have been made that show the type of peak interferences that exist.⁴ In the present study, we have examined the eight peak groupings tabulated in Table 1. Aside from the 43-51 keV peak region, which is nearly always obscured by the prominent 59 keV peak from ²⁴¹Am, these groupings represent all the spectral possibilities for which isotopic ratio measurements can be made. Of these, some will provide better results than others, and some are much more difficult to interpret than others. Furthermore, a rather large variety of counting conditions could be investigated. These might include detector parameters, such as resolution and counting efficiency; source parameters, such as sample size, homogeneity, isotopic content, and age following chemical purification; and counting parameters, such as count rate, length of count, and use of absorbers.

Table 1. Eight peak grouping useful for isotopic ratio determinations.

Group I:		Group V:	
94.67	U x ray	203.52	^{239}Pu
97.08	Np x ray	207.98	$^{241}\text{Pu}-^{237}\text{U}(^{241}\text{Am})$
98.44	U x ray		
98.81	^{239}Pu	Group VI:	
99.00	^{241}Am	332.30	$^{241}\text{Pu}-^{237}\text{U}, ^{241}\text{Am}$
99.54	Pu x ray	332.80	^{239}Pu
99.87	^{238}Pu	335.35	$^{241}\text{Pu}-^{237}\text{U}, ^{241}\text{Am}$
101.07	Np x ray	336.06	^{239}Pu
103.00	^{239}Pu	341.48	^{239}Pu
103.03	^{241}Am	344.96	^{239}Pu
103.67	^{241}Pu		
103.75	Pu x ray	Group VII,	
104.23	^{240}Pu	367.02	^{239}Pu
Group II:		368.53	^{239}Pu
110 - 121 keV		368.65	$^{241}\text{Pu}-^{237}\text{U}, ^{241}\text{Am}$
(very complex group including x rays		370.85	$^{241}\text{Pu}-^{237}\text{U}, ^{241}\text{Am}$
and gamma rays from all the isotopes)		375.02	^{239}Pu
123.01	^{241}Am	376.57	^{241}Am
123.67	^{239}Pu	380.16	^{239}Pu
124.52	^{239}Pu	382.70	^{239}Pu
125.17	^{239}Pu		
125.30	^{241}Am	Group VIII:	
129.28	^{239}Pu	632.80	^{241}Am
Group III:		633.19	^{239}Pu
141.64	^{239}Pu	637.97	^{239}Pu
144.19	^{239}Pu	640.15	^{239}Pu
146.05	^{239}Pu	641.37	^{241}Am
146.55	^{241}Am	642.30	^{240}Pu
148.60	^{241}Pu	646.02	^{239}Pu
150.12	^{241}Am	649.50	^{239}Pu
152.77	^{238}Pu	652.19	^{239}Pu
Group IV:		652.88	^{241}Am
160.00	^{241}Pu	654.86	^{239}Pu
160.35	^{240}Pu	658.99	^{239}Pu
161.45	^{239}Pu	662.37	^{241}Am
164.60	$^{241}\text{Pu}-^{237}\text{U}(^{241}\text{Am})$	644.67	^{239}Pu
		742.82	$^{238}\text{Pu}^*$

* The 742 keV peak of ^{238}Pu is not really part of the peak grouping, but for the purpose of the simulator program it is treated as such.

Simulator Computer Program

To begin to assess the effects of even a fraction of these parameters through experimental studies would entail much effort. Furthermore, many of the materials are not readily available. In order to assess the feasibility of making isotopic ratio measurements by gamma-ray spectrometry, a computer simulation code was written. Its purpose was two-fold: first, to indicate the level of precision that one might hope to obtain for isotopic ratios based on spectral information in the various peak groupings, and second, to make sensitivity studies for the various parameters involved in the analysis, to see how variations in their values influence the results. The first study establishes the degree of precision that could be obtained if counting statistics were the determining factor in the analysis of the peak grouping. The second study identifies the analytical parameters most likely to influence the precision of the results and, consequently, those aspects of the experimental design to which the most attention must be given.

The steps in the computer program can be broken into two parts. The first phase of the program involves the synthesis of simulated data for a peak grouping counted under a particular set of conditions. In the second phase, this grouping is analyzed under a selected set of conditions. The input parameters to the program are the following: total net count in the peak grouping, background levels before and after the peak grouping (the code assumes a five-channel background), a reference peak for positioning the remaining peaks in the grouping, the spectrometer gain

and other parameters describing the peak shape characteristics, the isotopic ratios for the system being studied, the gamma rays energies and their emission probabilities, the half-lives of the isotopes, and the number of iterations or replicate analyses.

The individual steps in the program are as follows:

- (1) After reading in the input information, a response curve or data envelope is generated for each of the components represented in the particular peak grouping being investigated. This is done by computing the profile for each peak at the position it holds relative to a reference position, and normalizing its intensity to the gamma emission probability per decay. Procedures for doing this, and the equations used, are described in a previous report.¹ The channel-by-channel addition of these peaks produces sets of idealized spectra, one for each isotopic component, with the intensities of each normalized to an arbitrary unit amount of the isotope.
- (2) A composite spectrum is produced by a linear addition of the component spectra after each has been multiplied by its fractional abundance as specified in the data input.
- (3) The composite spectrum is normalized to the total net count requested.

- (4) Each channel count is adjusted to simulate counting fluctuations in accordance with Poisson statistics. This is done by calling a random-number generator to select a number from 0 to 1. This number, used in conjunction with the normal distribution curve, is used to superimpose a statistical variation on the channel counts. Not only are the designated background levels included in this calculational process, but the background counting statistics at the endpoints of the peak grouping (based on a five-channel background) are also folded into the composite spectrum.
- (5) The energy reference peak is now relocated by fitting the top seven channels of the designated peak. This new reference position will differ slightly from the input value as a result of the statistical variations that have been introduced into the channel counts.
- (6) The component gamma-ray response functions are recomputed using the newly determined reference position.
- (7) A least-squares fit is used to fit the idealized component spectra to the composite spectrum.
- (8) The isotopic percentages are computed, based on the fit, and are compared with the input values. Errors are assigned to the values.
- (9) The entire process is repeated many times to assess the reproducibility.

There are several input options that allow for variations to the above procedure. One such option allows a parabolic in-

crement or decrement to be included in the fitting process. Such a degree of freedom may be necessary for large peak groupings where there is some uncertainty in the background interpolation. Another option introduces a nonlinear attenuation into the composite spectrum. In this case, the program can be allowed to iterate in such a way that the attenuation effects are masked out and good ratio measurements are still obtained. Other options allow for a fixed error to be introduced into the FWHM, the amount of peak "tailing," the peak position, or the background. In this way the sensitivity of the results to changes in these parameters can be investigated.

Many of the values used as input constants required some experimental basis — such as peak shape characteristics, total net counts, and background levels. For the purposes of our study, we obtained or estimated these input values from actual spectra we had acquired on several weapons-grade plutonium samples using a small 1 cc Ge(Li) detector and a larger coaxial detector. In general, the study assumed a 1 kg PuO₂ sample, a counting time of 1 h, and a count rate of ~5000 Hz. However, a few analyses were made for a 1 g PuO₂ sample, and different absorbers were used to attenuate the low-energy

Table 2. Approximate isotopic abundances corresponding to different burn up categories.

Burn up (MWd/t)	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu
Weapons grade	0.01	93	6	0.5	0.04
8-10 000	0.10	87	10	2.5	0.3
16-18 000	0.25	75	18	4.5	1.7
25-27 000	1.0	58	25	9.0	7.9
38-40 000	2.0	45	27	15.0	11

Fig. 1. A spectrum of PuO₂ using a large Ge(Li) detector.

gamma rays. A typical total spectrum is shown in Fig. 1.

The study covered five different isotopic blends based on a classification scheme reported by Bishop and Taylor.⁵

The isotopic abundances, corresponding to different fuel burn-up categories, are shown in Table 2. For the purpose of this study, we have assumed an ^{241}Am growth of 1 year.

Results

The results of the simulator program for each of the eight peak groupings listed in Table 1 are shown graphically in Figs. 2-10. These figures indicate the minimum uncertainty that can be expected in the results for each of the isotopic compositions listed in Table 2. Computer plots of the simulated data used for the analysis are shown in the Appendix. They also show how the various com-

ponents contribute to the structure of the composite peak grouping, and they are very useful for visualizing the complexity of the spectral region being studied and how its structure changes with isotopic composition.

Figures 2 and 3 show results using a 1 cc Ge spectrometer system. Its efficiency falls rapidly above 100 keV, but its high resolution (550 eV at 122 keV)

Fig. 2. Reproducibility of results using the 100 keV region. (1 hr equivalent count of 1 kg PuO_2 .)

Fig. 3. Reproducibility of results using the 100 keV region. (1 hr equivalent count of 200 kg/m³ plutonium solution.)

Fig. 4. Reproducibility of results using the 125 keV region.

Fig. 5. Reproducibility of results using the 145 keV region. (Asterisks indicate total net count. Values in parentheses are the average background counts.)

Fig. 6. Reproducibility of results using the 160 keV region. (Asterisks indicate total net count. Values in parentheses are the average background counts.)

Fig. 7. Reproducibility of results using the 208 keV region. (Asterisks indicate total net count. Values in parentheses are the average background counts.)

is absolutely essential if information is to be obtained from the gamma and x-ray peaks in the 100 keV complex. A gain of 0.075 keV/channel was used for this region.

All of the other spectra are based on results from a detector having an efficiency of 10% at 1.33 MeV relative to a 3 in. $\times$ 3 in. NaI detector. The

resolution of this detector (measured at FWHM) was 1.1 keV at 122 keV and 1.9 keV at 1332 keV. A 1.6 mm cadmium absorber was generally used, and for the higher energy groupings an additional 6.35 mm of lead was also used. The energy scaling for these spectra corresponded to a gain of 0.25 keV/channel.

Fig. 8. Reproducibility of results using the 335 keV region. The upper group of curves is for a 1 h count using a 1.6 mm cadmium absorber. The lower group of curves is for a 4 h count using an additional 6.35 mm lead absorber.

Fig. 9. Reproducibility of results using the 370 keV region. The upper group of curves is for a 1 h count using a 1.6 mm cadmium absorber. The lower group of curves is for a 4 h count using an additional 6.35 mm lead absorber. (Asterisks indicate total net count. Values in parentheses are the average background counts.)

Fig. 10. Reproducibility of results using the 630 keV region.

Conclusions

Some of the conclusions that can be drawn from the results are the following:

- (1) There is no one spectral region that can be used to completely characterize the plutonium isotopic composition of a sample. Instead, a combination of groups must be used. The optimum combination depends on such factors as isotopic composition, age, purity and sample matrix.
- (2) Some groups will yield a more precise ratio than others. However, circumstances may dictate the use of less desirable spectral regions. For example, plutonium that has been recently separated from spent fuel rods will still contain substantial levels of fission products, particularly ^{95}Zr and ^{95}Nb . The quantities that are present before appreciable decay has occurred are sufficient to obscure any useful spectral information above 500 keV. As a result, the peak grouping at 630 keV is useful only when analyzing aged material.
- (3) In the present study, each peak grouping was analyzed without benefit of information available from the analysis of some other grouping. In practice, however, an analytical program involving

all available information should be used, and this should improve the precision of some of the results.

- (4) Before adopting a certain analysis scheme, sensitivity tests should be run to determine which parameters influence the results most. For example, although Fig. 7 indicates that the 203-208 keV peak doublet is quite useful for measuring the $^{239}\text{Pu}/^{241}\text{Pu}$ ratio, simulation runs indicate that the ratio is very sensitive to the characterization of the peak "tailing," an observation that can be made visually by looking at the corresponding figures for this doublet in the Appendix.

By way of additional illustrations, Figs. 11 and 12 show the biases that result when the FWHM and channel positions are varied by 1% in the analysis of data in the 100 keV region.

- (5) It appears that the usefulness of the 335 and 370 keV peak groupings has been largely overlooked. Together, they may provide the best information on the $^{239}\text{Pu}/^{241}\text{Pu}$ ratio and yield the best value for the ^{241}Am content if the 630 keV region is inaccessible.
- (6) The 100 keV region, in principle, contains a wealth of information. However, it is not only extremely

Fig. 11. Sensitivity of results to a 1% change in FWHM using data in the 100 keV peak complex.

Fig. 12. Sensitivity of results to a 1% change in peak position using data in the 100 keV complex.

complex, but it also contains both gamma rays and x-rays. This results in having to contend with two distinctly different peak shapes.¹

An additional complication is the presence of plutonium x-rays coming from alpha-induced x-ray fluorescence.

In its favor, it should be pointed out that the 100 keV grouping is very intense, partially because it resides below the K-shell binding energy of plutonium and consequently is not attenuated by the sample to any greater extent than is a gamma ray of 180 keV.

Furthermore, the quantitative analysis of the 104.23 peak may be the only hope for determining ²⁴⁰Pu content if the 630 keV region is inaccessible.

- (7) The gamma-ray energies and branching intensities reported in Ref. 1 are probably not accurate enough for the type of isotopic measurements that are envisioned. We plan to redetermine the branching intensities for the various plutonium isotopes, giving particular care to obtaining very accurate intensity relationships for the eight peak groupings reported in this study.

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Appendix

The figures in the Appendix are in pairs; the first simply displays the total channel count and background curve for a given peak grouping, and the second shows the component structure making up the total observed spectral region. One such pair of plots is displayed for each of the five isotopic compositions shown in Table 2 of the text.

SPECTRUM ID IS 93PCT PU 100 KEV
 TOTAL COUNTS = $3.00\text{E}+05$

SPECTRUM ID IS 8.PCT PU 100 KEV
 TOTAL COUNTS = 3.00E+06

SPECTRUM ID IS 75PCT PU 100 KEV
 TOTAL COUNTS = 3.00E+06

SPECTRUM ID IS 58PCT PU 100 KEV
 TOTAL COUNTS = 3.00E+06

SPECTRUM ID IS 45PCT PU 100 KEV
 TOTAL COUNTS = 3.00E+06

SPECTRUM ID IS 93PCT 120KEV 1GM
 TOTAL COUNTS = 3.00E+06

SPECTRUM ID IS 87PCT 120KEV 1GM
 TOTAL COUNTS = 3.00E+06

SPECTRUM ID IS 75PCT 120KEV 1GM
TOTAL COUNTS = 3.00E+06

SPECTRUM ID IS 58PCT 120KEV 1GM
 TOTAL COUNTS = 3.00E+06

SPECTRUM ID IS 45PCT 120KEV 1GM
 TOTAL COUNTS = 3.00E+06

SPECTRUM ID IS 93PCT PU 145 KEV
 TOTAL COUNTS = 2.40E+05

SPECTRUM ID IS 87PCT PU 145 KEV
 TOTAL COUNTS = 4.80E+05

SPECTRUM ID IS 75PCT PU 145 KEV
 TOTAL COUNTS = 5.00E+05

SPECTRUM ID IS 58PCT PU 145 KEV
 TOTAL COUNTS = 5.00E+05

SPECTRUM ID IS 45PCT PU 145 KEV
TOTAL COUNTS = 6.00E+05

SPECTRUM ID IS 93PCT PU 160 KEV
 TOTAL COUNTS = 1.20E+05

SPECTRUM ID IS 87PCT PU 160 KEV
 TOTAL COUNTS = 2.40E+05

SPECTRUM ID IS 75PCT PU 160 KEV
TOTAL COUNTS = 3.00E+05

SPECTRUM ID IS 58PCT PU 160 KEV
TOTAL COUNTS = 3.00E+05

SPECTRUM ID IS 45PCT PU 160 KEV
 TOTAL COUNTS = 3.00E+05

SPECTRUM ID IS 93PCT PU 208 KEV
 TOTAL COUNTS = 2.00E+06

SPECTRUM ID IS 87PCT PU 208 KEV
 TOTAL COUNTS = 5.00E+06

SPECTRUM ID IS 75PCT PU 208 KEV
 TOTAL COUNTS = 7.00E+06

SPECTRUM ID IS 58PCT PU 208 KEV
 TOTAL COUNTS = 8.00E+06

SPECTRUM ID IS 45PCT PU 208 KEV
 TOTAL COUNTS = 8.00E+06

SPECTRUM ID IS 93PCT PU 335 KEV
TOTAL COUNTS = 1.00E+06

SPECTRUM ID IS 87PCT PU 335 KEV
TOTAL COUNTS = 1.00E+06

SPECTRUM ID IS 75PCT PU 335 KEV
TOTAL COUNTS = 1.00E+06

SPECTRUM ID IS 58PCT PU 335 KEV
 TOTAL COUNTS = 1.00E+06

SPECTRUM ID IS 45PCT PU 335 KEV
 TOTAL COUNTS = 1.00E+06

SPECTRUM ID IS 93PCT PU 370 KEV
 TOTAL COUNTS = 1.60E+06

SPECTRUM ID IS 87PCT PU 370 KEV
TOTAL COUNTS = 1.00E+06

SPECTRUM ID IS 75PCT PU 370 KEV
TOTAL COUNTS = 8.00E+05

SPECTRUM ID IS 58PCT PU 370 KEV
TOTAL COUNTS = 6.00E+05

SPECTRUM ID IS 45PCT PU 370 KEV
 TOTAL COUNTS = 6.00E+05

SPECTRUM ID IS 95PCT PU 630 KEV
TOTAL COUNTS = 4.00E+05

SPECTRUM ID IS 87PCT PU 630 KEV
TOTAL COUNTS = 4.00E+05

SPECTRUM: ID IS 75PCT PU 630 KEV
 TOTAL COUNTS = 4.00E+05

SPECTRUM ID IS 58PCT PU 630 KEV
TOTAL COUNTS = 4.00E+05

SPECTRUM ID IS 45PCT PU 630 KEV
TOTAL COUNTS = 4.00E+05