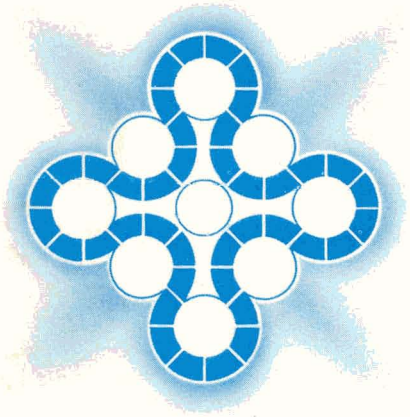
ANCR-1055

UC-15

A RELATIVELY SIMPLE AND PRECISE TECHNIQUE FOR THE ASSAY OF PLUTONIUM WASTE

J.E. Cline



Aerojet Nuclear Company

NATIONAL REACTOR TESTING STATION

Idaho Falls, Idaho — 83401

DATE PUBLISHED—FEBRUARY 1972

PREPARED FOR THE

U. S. ATOMIC ENERGY COMMISSION

IDAHO OPERATIONS OFFICE UNDER CONTRACT AT(10-1)-1375

DISCLAIMER

Printed in the United States of America
Available from
National Technical Information Service
U. S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.95

ANCR-1055

SAFEGUARDS AND NUCLEAR
MATERIALS MANAGEMENT
TID-4500

A RELATIVELY SIMPLE AND PRECISE TECHNIQUE FOR THE ASSAY OF PLUTONIUM WASTE

J. E. Cline

AEROJET NUCLEAR COMPANY

Date Published -- February 1972

PREPARED FOR THE U. S. ATOMIC ENERGY COMMISSION
IDAHO OPERATIONS OFFICE
UNDER CONTRACT NO. AT(10-1)-1375

FOREWORD

STATEMENT OF PURPOSE

This document is intended to be used as a manual for the assay of plutonium waste as it occurs in nuclear fuel fabrication and processing plants. The method proposed is that of passive gamma-ray spectroscopy using differential absorption of the plutonium gamma rays to determine the absorption correction. This is a method recommended by the Technical Support Office of the Office of Safeguards and Materials Management as a result of their involvement in the Plant Instrumentation Program. The document is not intended as a review or critique of all of the techniques of plutonium waste assay currently in use, of techniques in gamma-ray spectroscopy, or of nuclear instrumentation.

ACKNOWLEDGEMENTS

I should like to acknowledge the assistance of the staff of the Technical Support Office, Brookhaven National Laboratory in the preparation of this manual. In particular, that of W. A. Higinbotham who supplied the motivation for writing and E. V. Weinstock who spent much time reviewing the manuscript.

ABSTRACT

A technique is described for use in the assay of plutonium waste. The method is that of passive Ge(Li) gamma-ray spectroscopy using a technique of differential absorption of the plutonium gamma rays themselves to determine the absorption correction. The analytic technique is simple and relatively quick requiring a simple measurement of about 20 minutes to obtain total plutonium content and relative isotopic abundances. Described in the document is the analytic technique, the instrumentation required, the experimental technique and analysis, and a typical analysis. Attempts are made to describe the limitations and uncertainties in the measurement technique.

TABLE OF CONTENTS

FIGURES

TABLES

A RELATIVELY SIMPLE AND PRECISE TECHNIQUE FOR THE ASSAY OF PLUTONIUM WASTE

J. E. Cline

I. INTRODUCTION

Currently, throughout nuclear industry and laboratories, a multitude of techniques are being employed to measure plutonium waste in containers which vary in size from 1 gallon to 55 gallon capacity. The problem is a difficult one because 1) the weight of the total plutonium is very small compared to the total weight of the container, usually about 1 part in 10^4, 2) the matrix in which the plutonium is located causes severe absorption of the gamma rays or neutrons used in the assay, and 3) the counting geometry of most waste containers is considerably less than ideal.

The large number of assay techniques can generally be categorized into four groups: passive gamma-ray, active gamma-ray, passive neutron, and active neutron. Some laboratories use a combination of measurements to assay waste. As a result of the Plant Instrumentation Program (PIP), sponsored by the Nuclear Materials Security Division of the AEC, a study has been made of some of these techniques, each of which has its own set of virtues, faults and complexities. The following document is intended to provide a set of recommendations, based upon the PIP study, for instrumentation and data analysis to provide what is currently felt to be an adequate waste assay. It considers only the passive techniques, primarily gamma-ray scanning, because of their relative ease of use, and represents, within reasonable bounds, the current state-of-the-art in techniques and instrumentation.

II. TECHNIQUES

The principle generally employed in assay of plutonium waste using passive gamma-ray techniques is to measure the emitted intensity of one or more of the higher-energy gamma rays (i.e. 375-keV - 451-keV region) from ^{239}Pu decay and to apply a correction to this intensity to account for some absorption in the matrix. This absorption may be due solely to the matrix material or to both the matrix and the plutonium. Methods currently used to determine the absorption correction involve either 1) external sources, by measuring transmission through the container at one or more gamma-ray energies, or 2) internal gamma-rays, by measuring ratios of the intensities of two or more gamma rays emitted by the plutonium and comparing the observed ratios to the ratios of emitted intensities assuming no absorption. In analyses using either of these two methods, certain assumptions have to be made concerning the plutonium distribution and the matrix itself. The success of the particular analysis is then related to the degree of validity of the assumptions made.

Passive neutron measurements can be made to supplement the gamma-ray measurements. Their value can be of particular significance for those containers where some of the assumptions required for accurate gamma-ray assay of the plutonium waste are in doubt.

The following list of assumptions are most often necessary:

A. Matrix

1. Density of matrix is fairly uniform through barrel. (Otherwise lightly shielded Pu in lower density regions will overpower more heavily shielded Pu.)

2. Fairly uniform atomic number (Z) of matrix throughout barrel. (In most analyses it is necessary to know Z of material doing the bulk of the shielding.)

B. Plutonium

1. No large concentrations of Pu inside dense or thick absorber. (Contribution from this region will be minimal when compared to that of other regions - similar to paragraph A.1.)

2. No large chunks of Pu. (Self absorption will shield most of the Pu and cause a low assay, e.g., a 6-mm diameter sphere of PuO_2 will absorb about 1/2 of the 413-keV radiation emitted by the Pu and about 90% of the 129-keV radiation.)

3. The average density of plutonium in any region of the barrel is fairly low. (The plutonium can be a significant contributor to the absorption. However, even if 200 gms of Pu, considered a rather substantial amount, are evenly distributed over an entire barrel, the attenuation due to Pu is still negligible, i.e. less than 2%, and can be ignored in the analysis.)

Several different analytical approaches have been used by different groups[1-4] using gamma-ray intensity data obtained from either internal or external measurements to determine the absorption correction. Both LASL[1] and GRT[2] apply methods of transmission using NaI(Tl) and external sources. LASL uses the 400.6-keV gamma ray from ^{75}Se and GRT uses the 662-keV gamma ray from ^{137}Cs. An absorption correction to be applied to the observed intensity of the plutonium gamma rays is then based upon the measured transmission of the gamma rays from the external source. Vertical and, often, horizontal collimation of the gamma rays is made to enhance the relative contribution to the total counting rate from Pu located along the center symmetry axis of the container. The techniques either use multiple detectors or employ a simultaneous rotation and translation of the container past a single source and detector. In the technique developed by NUMEC[3], a Ge(Li) detector

is used and a line-source of ^{239}Pu is used for the transmission measurements. The waste containers used by NUMEC are 1-1/2 gallon pasteboard containers and an attempt is made to segregate the different matrix materials into different containers. Absorption corrections were again made on the basis of transmission of the gamma rays from the standard external source through the container. The technique used by ARHCO[4] uses collimated Ge(Li) detectors, and a ^{133}Ba source (355-keV gamma ray) for transmission measurements. At ARHCO, as at NUMEC, the waste is packaged and assayed in small containers ($\sim$2 gallon) prior to being placed in larger barrels. At all of the installations the packages are rotated during counting to obtain an average distribution of the plutonium in the container.

The principal weakness in any technique using external source transmission measurements to determine the absorption correction is the rather high dependence upon uniformity of the matrix and of the plutonium. If the plutonium were located on the surface of a small but highly absorptive piece of the matrix or if the plutonium were in a small and highly absorptive clump, and either of these objects were placed within a light matrix such as paper, analysis based upon transmission measurements would yield highly erroneous results.

It is felt by PIP personnel that the requirements for any analytical approach should be 1) that the technique be relatively simple, in equipment, data taking, and in analysis, 2) that the technique be based upon the assumptions listed above, and 3) that the analysis should be of such a character as to "flag" those containers that severely violate one or more of the assumptions, in particular, those containers that show very high gamma-ray absorption. In general, these latter barrels will have a high plutonium content and will undoubtedly be reclaimed.

For the low plutonium content barrels, any attenuation of the plutonium radiation by plutonium itself can effectively be ignored and all attenuation can be assumed to be caused by the matrix. This simplifies the problem considerably.

III. DETAILS OF SUGGESTED TECHNIQUE

The technique and analytical procedure described below are suggested for several reasons. The primary one is that of simplicity of measurement and of analysis. In this procedure, absorption corrections are determined by differential gamma-absorption using two gamma rays of different energy emitted by the plutonium itself. Since the plutonium to be analyzed will undoubtedly contain a wide variation in relative isotopic concentration of the ^{239}Pu and ^{241}Pu isotopes, most analyses will probably need to measure this isotopic ratio. Both the isotopic ratio and the intensity ratios from a single isotope are much easier to measure accurately using Ge(Li) detectors than NaI(Tl) detectors. Therefore, the use of Ge(Li) or Ge spectrometry is recommended for these measurements.

The measurement itself is relatively simple. A Ge(Li) detector is placed at a suitable distance from the container, rotating at about 1 rpm, and a pulse-height distribution is obtained which represents the gamma rays emitted from the plutonium. A list of the more intense gamma rays emitted by plutonium is given in Table I. A measurement of the

TABLE I

Table of the more intense gamma rays emitted by the isotopes of plutonium normally found in plutonium waste. Column 1 and 2 contain the A, name, and isotope, respectively, of the emitting nuclide, column 3 contains the half life, column 4 is the gamma-ray energy, in keV, column 5 is the uncertainty in the energy, in keV, and the last column is the gamma-ray intensity in units of gamma rays per disintegration. In these waste samples, ^{237}U is in equilibrium with ^{241}Pu and the intensities of the ^{237}U gamma rays are given in units of gamma rays per decay of the ^{241}Pu parent. σ_E = standard deviation in the gamma-ray energy.

Z	A	T(1/2)	E (keV)	σ_E	I (γ/disintegrations)
94 Pu	238	86 Y	152.580	0.030	1.270E-05
94 Pu	239	24400 Y	102.930	0.080	2.600E-06
			125.000	0.120	4.800E-07
			129.270	0.030	5.600E-05
			159.600	0.200	2.000E-08
			160.070	0.130	5.000E-08
			161.470	0.050	1.000E-06
			203.550	0.040	4.800E-06
			332.840	0.040	4.300E-06
			345.020	0.030	5.000E-06
			375.040	0.030	1.500E-05
			380.190	0.050	3.000E-06
			382.770	0.050	2.500E-05
			392.700	0.200	2.700E-06
			393.100	0.200	2.700E-06
			413.710	0.030	1.500E-05
			422.620	0.050	1.200E-06
			426.710	0.080	3.300E-07
94 Pu	240	6580 Y	104.150	0.020	1.600E-04
			160.270	0.020	1.040E-05
94 Pu	241	13.2 Y	103.540	0.040	1.010E-06
			148.560	0.020	2.070E-06
			159.960	0.020	7.450E-08
92 U	237	6.75 D in equilibrium with ^{241}Pu	208.000	0.050	5.360E-06
			222.710	0.100	4.600E-09
			234.230	0.100	6.900E-09
			267.530	0.050	1.910E-07
			332.340	0.050	2.990E-07
			335.400	0.050	2.760E-08
			368.630	0.080	1.080E-08
			370.980	0.080	3.150E-08

intensities of these gamma rays in the pulse-height spectrum from a package containing plutonium can reveal the amount of plutonium in the package. If the plutonium were located in the center of the package and were surrounded by no matrix material, then the observed intensity of a strong gamma ray from ^{239}Pu, such as the 413-keV transition would yield directly, through a constant of proportionality, the total content of ^{239}Pu. Similarly the observed intensity of a strong gamma ray from ^{241}Pu decay would yield the total content of ^{241}Pu. The ratio of the observed intensities of two gamma rays from ^{239}Pu, widely separated in energy, such as the 129- and 413-keV transitions, would yield a value which differs slightly from a ratio derived from Table I. This deviation would result primarily from the relatively higher absorption of the lower-energy gamma ray in the walls of the container.

In the case of most containers, however, the plutonium is not so ideally located and is surrounded by a matrix material which may be composed of such diversified materials as paper, gloves, glass, crucibles, etc. but is generally of light ($Z < 20$) material. The matrix causes absorption, often severe, of the gamma rays emitted by the plutonium and the absorption if not taken into account will result in an erroneously low assay. Since the matrix material also preferentially absorbs the lower-energy gamma rays, the observed intensity ratio of two gamma rays (129- and 413-keV) will be lower than in the case of no matrix. In fact, a measurement of this ratio will provide an indication of the absorption effects of the matrix. If certain assumptions are made, an analytic expression can be derived which describes the absorption of the 413-keV transition in terms of this ratio, providing the required absorption correction. The derivation of this expression follows:

A. Algebraic Derivation of Analytical Formulae

Since we choose to ignore the absorption effects due to the plutonium itself, the formulation of the problem becomes much simpler.

Consider a low-energy gamma ray from ^{239}Pu (e.g. the 129-keV gamma) as gamma ray #1. Similarly, let a higher-energy transition such as the 413-keV gamma be gamma ray #2. Then, the intensity of gamma #1 as observed by a detector may be written as:

$$(I_1)_{observed} = (I_1)_{emitted} e^{-\mu_1 x} \qquad (1)$$

where μ_1 is the average attenuation coefficient in the matrix for gamma-ray energy #1, and x is an average distance through the matrix that the plutonium gamma rays travel. For the light elements, Be through Fe, μ_1 does not change much with Z. Furthermore, since in most cases the matrix material is a hydrocarbon, a carbonaceous or a silicate material, it is felt that such a μ_1 can reasonably be defined.

The absorption coefficients used in these equations are the sums of the photoelectric and the Compton coefficients. Elastic scattering is not included since the energy is not altered in the scattering process, and because of the nature of the matrix and plutonium distribution, as many gamma rays of a given energy are scattered into the solid angle of the detector as are scattered out. The units used throughout the following treatment for μ, the absorption coefficient, and for x, the thickness of the absorber, are cm^2/g and g/cm^2, respectively. Units of cm^{-1} and cm would yield equal results but expressing the absorption thickness in cm would not make much physical sense in this example.

Similarly,

$$(I_2)_{observed} = (I_2)_{emitted}\, e^{-\mu_2 x}. \tag{2}$$

Then,

$$\left(\frac{I_1}{I_2}\right)_{observed} = \left(\frac{I_1}{I_2}\right)_{emitted} e^{-(\mu_1-\mu_2)x}, \tag{3}$$

or

$$x = \frac{\ell n\left(\frac{[I_1/I_2]_{em}}{[I_1/I_2]_{obs}}\right)}{(\mu_1 - \mu_2)}. \tag{4}$$

When we put x back into (2), we have

$$(I_2)_{em} = (I_2)_{obs}\, \exp\left[\left(\frac{\mu_2}{\mu_1-\mu_2}\right)\, \ell n\left(\frac{\left[\frac{I_1}{I_2}\right]_{em}}{\left[\frac{I_1}{I_2}\right]_{obs}}\right)\right], \tag{5}$$

or

$$\text{gms Pu (of isotope emitting } \gamma\text{'s 1 \& 2)} = K_1(I_2)_{obs}^{\ x}$$

$$\exp\left[\frac{\mu_2}{\mu_1-\mu_2}\, \ell n\left(\frac{\left[\frac{I_1}{I_2}\right]_{em}}{\left[\frac{I_1}{I_2}\right]_{obs}}\right)\right]. \tag{6}$$

K_1 is a constant that depends upon the detector efficiency effects, counting geometry, and emission branching percentage for gamma ray #2. Equation (6) has the simpler form $\left(\frac{\mu_2}{\mu_1-\mu_2}\right)$,

$$\text{gms Pu (of emitting isotope)} = K_1 \, (I_2)_{obs} \left(\frac{\left[\frac{I_1}{I_2}\right]_{em}}{\left[\frac{I_1}{I_2}\right]_{obs}} \right) \qquad (7)$$

where the constant K_1 may be experimentally determined through the use of standards. The ratio $\left[\frac{I_1}{I_2}\right]_{em}$ is the ratio of measured intensities if no absorption occurs in the matrix. It can be experimentally determined by placing a source inside an empty barrel. This source should be a "thin" source which shows little absorption of the plutonium gamma rays.

For a matrix material that can be approximated by a Z of 8, an approximation that has some physical justification and seems to work, $\frac{\mu_2}{\mu_1-\mu_2}$ is found[5] to be ~1.92 for gamma ray #1 of 129 keV and gamma ray #2 of 413 keV. Finally, then

$$\text{gms } (^{239}\text{Pu}) = K_1 \, (I_{413})_{obs} \left[\frac{\left[\frac{I_{129}}{I_{413}}\right]_{em}}{\left[\frac{I_{129}}{I_{413}}\right]_{obs}} \right]^{1.92} . \qquad (8)$$

This equation works reasonably well if the ^{241}Pu content is less than about 1/5 that of the ^{239}Pu content. If it is greater than this amount, the 148-keV and 267-keV gamma rays from ^{241}Pu content should probably be used together with the appropriate exponent of 4.38. This gives

$$\text{gms } (^{239}\text{Pu}) = K_2 \, (I_{413})_{obs} \left[\frac{\left[\frac{I_{148}}{I_{267}}\right]_{em}}{\left[\frac{I_{148}}{I_{267}}\right]_{obs}} \right]^{4.38} . \qquad (9)$$

Here, the two gamma rays in the ratio are from ^{241}Pu although the analysis is still for ^{239}Pu. This equation should not be used unless the 129-keV transition from ^{239}Pu cannot be seen in the pulse-height spectrum. The reason for this is that the statistical uncertainties in the intensities in the ratio are intensified by the size of the exponent when propagated to an uncertainty in the plutonium assay (see Section B).

B. Uncertainties and Errors

B.1 Propagation of Statistical Uncertainties If we ignore for the moment systematic errors, of which there may be many, we can concentrate upon the propagation of statistical uncertainties through the equations (8) and (9). Equation (8) is of the form:

$$G = K\, I_{\gamma 2} \left(\frac{I_{\gamma 1}}{I_{\gamma 2}} \right)^{A} , \tag{10}$$

where $I_{\gamma 1}$ and $I_{\gamma 2}$ are observed intensities. The relative statistical uncertainty in G resulting from statistical uncertainties in $I_{\gamma 1}$ and $I_{\gamma 2}$ is given by:

$$\frac{S_G}{G} = \frac{\left[\left(\frac{\partial G}{\partial I_{\gamma 1}} \right)^2 S^2_{I_{\gamma 1}} + \left(\frac{\partial G}{\partial I_{\gamma 2}} \right)^2 S^2_{I_{\gamma 2}} \right]^{1/2}}{G} , \tag{11}$$

where S_G = average standard deviation in the quantity G.

From equation (11),

$$\frac{\partial G}{\partial I_{\gamma 1}} = K\, I_{\gamma 2}^{(1-A)}\; A\, I_{\gamma 1}^{A-1} ,$$

and

$$\frac{\partial G}{\partial I_{\gamma 2}} = K\, I_{\gamma 1}^{A}\; (1-A)\; I_{\gamma 2}^{-A} .$$

Inserting these into equation (11), we get:

$$\frac{S_G}{G} = \left[(A)^2 \left(\frac{S_{I_{\gamma 1}}}{I_{\gamma 2}} \right)^2 + (1-A)^2 \left(\frac{S_{I_{\gamma 2}}}{I_{\gamma 2}} \right)^2 \right]^{1/2} . \tag{12}$$

If we have Poisson statistics for $I_{\gamma 1}$ and $I_{\gamma 2}$, then $S_{I_{\gamma 1}} = (I_{\gamma 1})^{1/2}$, etc. Inserting this and the values from equation (8) into (12), we get:

$$\frac{S_G}{G} = \left[\frac{(1.92)^2}{I_{129}} + \frac{(0.92)^2}{I_{413}} \right]^{1/2} . \qquad (13)$$

In the preceding analysis, a factor has been calculated to correct the intensity of the 413-keV gamma ray for absorption. If we had chosen to use the 129-keV transition as the basis of the plutonium determination and to calculate a correction factor to its intensity, we would derive the following formula:

$$G = K_3 \, (I_{129})_{obs} \left[\frac{\left[\frac{I_{129}}{I_{413}} \right]_{em}}{\left[\frac{I_{129}}{I_{413}} \right]_{obs}} \right]^{\left(\frac{\mu_1}{\mu_1 - \mu_2} \right)}$$

$$= K_3 \, (I_{129})_{obs} \left[\frac{\left[\frac{I_{129}}{I_{413}} \right]_{em}}{\left[\frac{I_{129}}{I_{413}} \right]_{obs}} \right]^{2.94}$$

In this case, the relative standard deviation in G is

$$\frac{S_G}{G} = \left[\frac{(1.94)^2}{I_{129}} + \frac{(2.94)^2}{I_{413}} \right]^{1/2} . \qquad (14)$$

From equations (13) and (14) it appears that the statistical uncertainty in the final result is dependent on whether one uses the 129- or the 413-keV transitions as the basis of the calculations. This statistical analysis, however, although exact, takes into account only the measurement and determination of the intensities of the two gamma rays. In practice, however, the non-statistical errors probably control the accuracy of measurements. In any case it is better to use the highest energy gamma ray (such as the 413) since the size of the absorption correction is smaller and the non-statistical errors will undoubtedly be smaller.

B.2 Non-Statistical Errors and Biases One of the major assumptions in the development of equation (7) is that the matrix material can be approximated by a material of a single atomic number. The exponent, $(\frac{\mu_2}{\mu_1-\mu_2})$, is then evaluated assuming a specific value of the atomic number. In the case of equation (8), Z = 8 was used to obtain $(\frac{\mu_2}{\mu_1-\mu_2}) = 1.92$.

Figure 1 shows the gamma-ray attenuation coefficient, μ, as a function of the gamma-ray energy and of the atomic number of the absorbing material. Perhaps more significant is a plot of the exponent in equation (7), $(\frac{\mu_2}{\mu_1-\mu_2})$, shown as a function of atomic number in Figure 2.

From Figure 2 it may be seen that rather significant errors can be obtained in the measurement if the wrong choice is made for the Z of the matrix material. It is for this reason that better results are obtained if segregation is made of the matrix material by placing only one type of matrix in any given waste barrel. One can then make an estimate from the "known" composition of the average Z of the material doing the absorbing and use the proper exponent from Figure 2. Such procedures should reduce the bias caused by the improper choices for the value of the exponent in equation (7). A similar figure for use with equation (9) is given in Figure 3.

C. Use of Multiple Gamma Rays to Estimate Average Z of Absorbing Material

The use of the analytical formalism developed in Sections A and B above requires some prior knowledge of the nature of the matrix material. In particular some knowledge of the average Z of the absorbing material is required to choose the proper value for the exponent in the analytical equation. In most cases this is not a severe limitation of the techniques since, in general, the material is of low Z and the value of the exponents does not change substantially with Z. Furthermore, the matrix is generally segregated and a reasonable estimate of its nature can be made.

Nevertheless, through the use of the observed intensities of three or four gamma rays it is possible in principle to determine the average Z of the absorbing material. The principal difficulty in the use of this technique lies in the fact that the propagation of statistical uncertainties in the intensity determinations through the analytic expressions often result in a net uncertainty which totally masks the determination of Z. Where anomalously high absorption is observed, however, the method can be used to yield crude estimates of the average Z and to give additional data on the contents of the waste package. Therefore, the following analytical formalism is given.

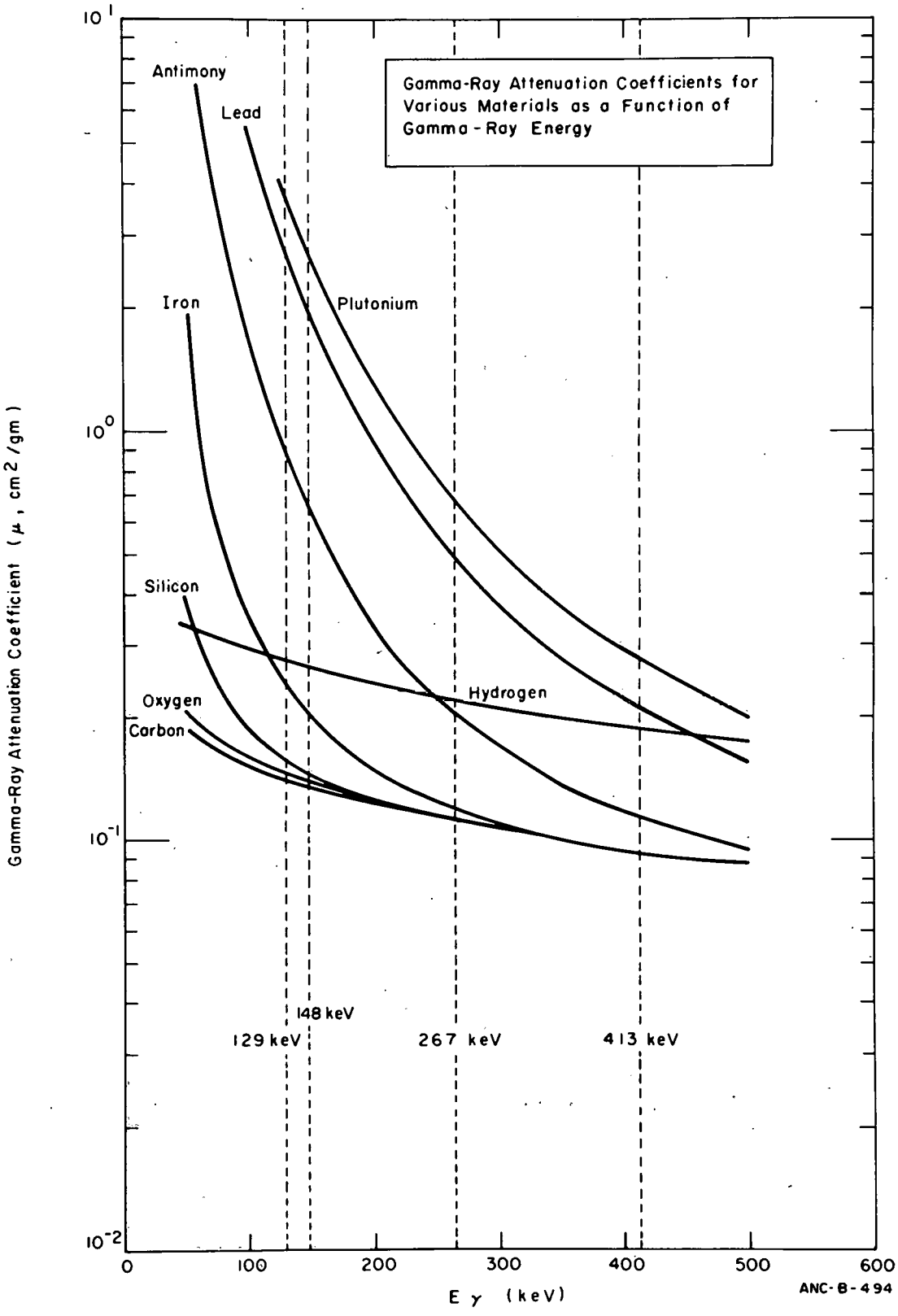


Figure 1 Gamma-ray attenuation coefficients for various materials as a function of gamma-ray energy. The coefficients do not include the contribution from coherent scattering. The values are from reference 5.

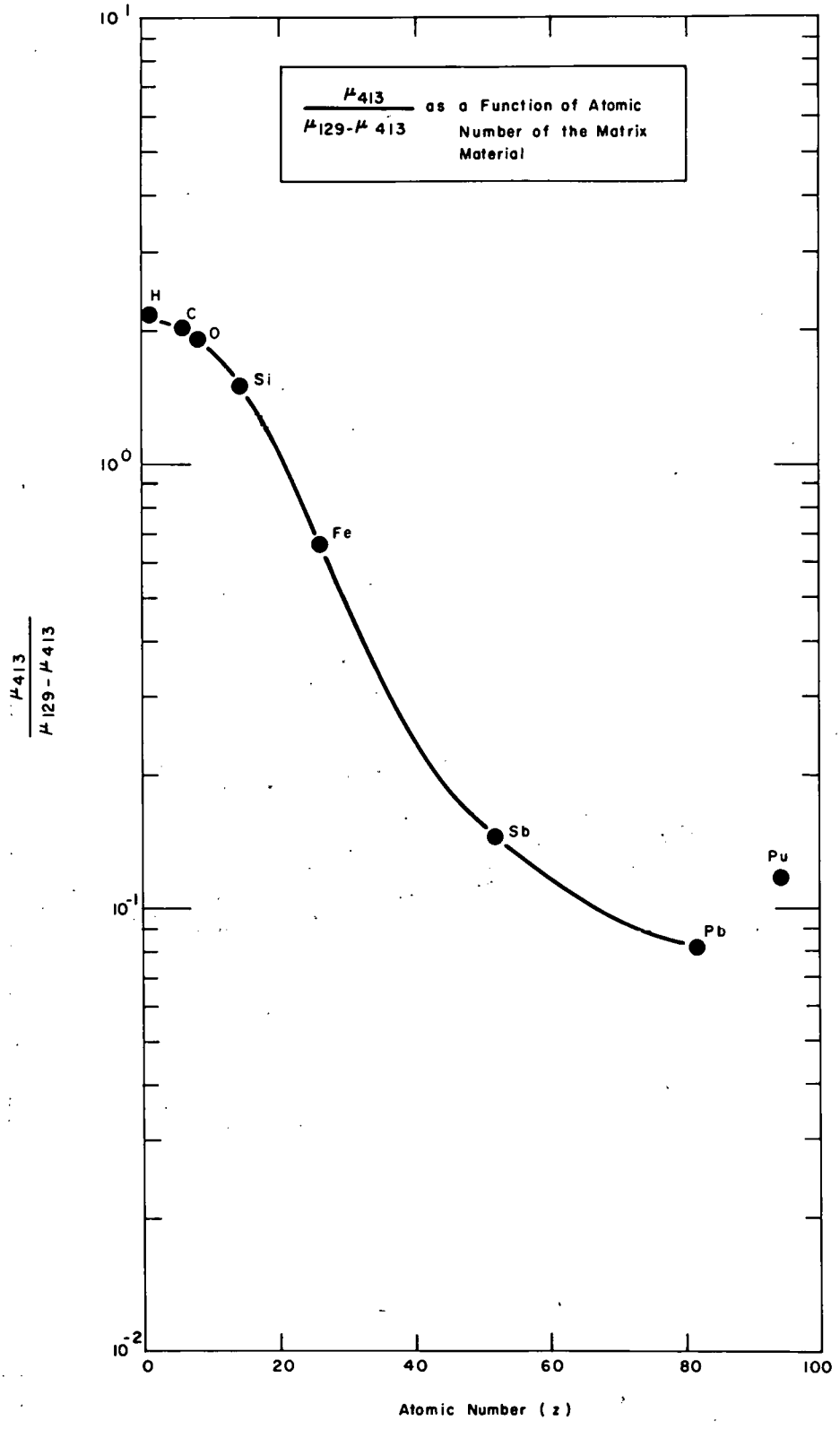


Figure 2 Exponent in equation (7), $\frac{\mu_{413}}{\mu_{129} - \mu_{413}}$, as a function of atomic number.

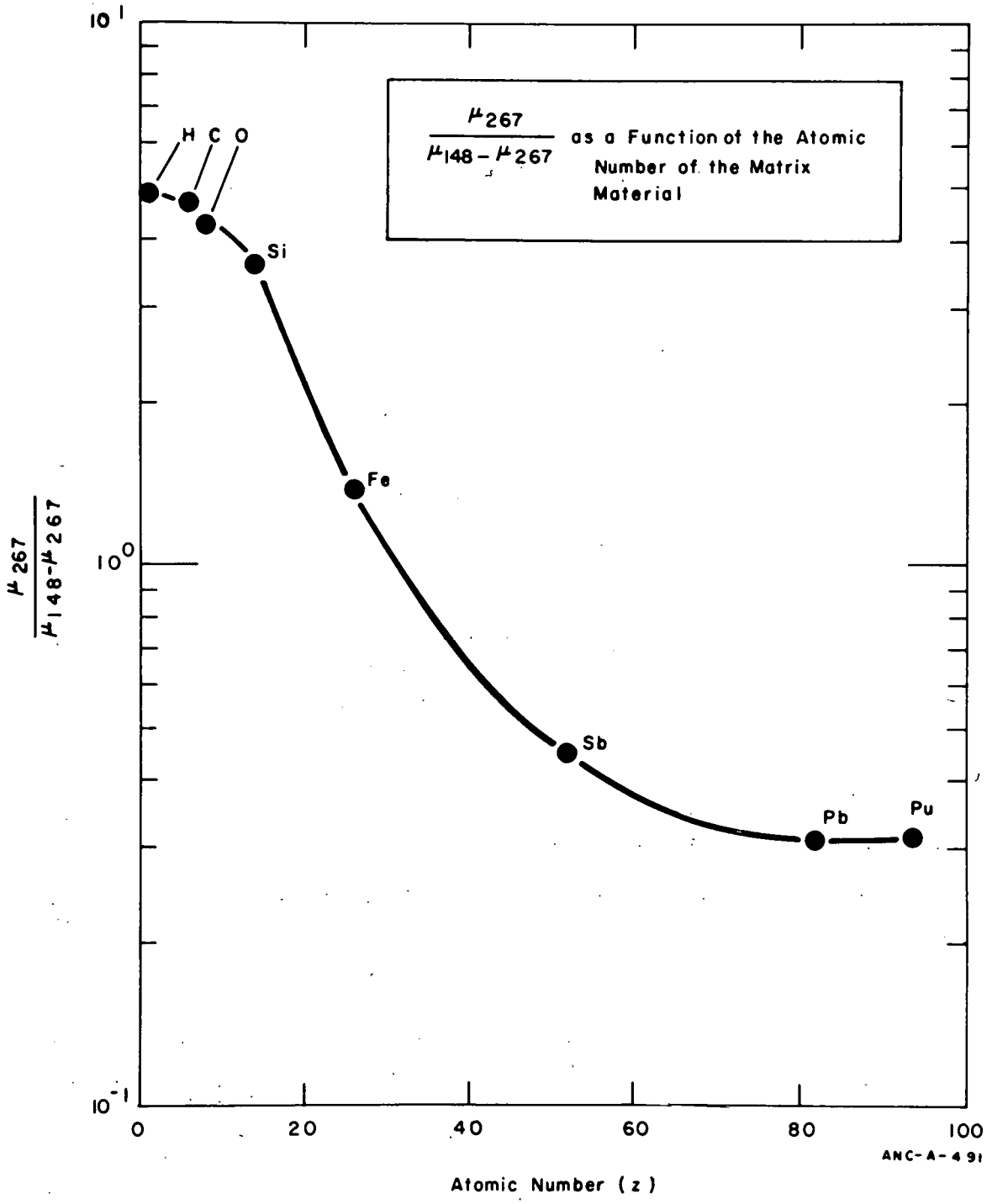


Figure 3 Exponent in equation (9), $\frac{\mu_{267}}{\mu_{148} - \mu_{267}}$, as a function of atomic number.

Consider the following four gamma rays emitted by the plutonium in the waste package:

$$(I_1)_{obs} = (I_1)_{em}\, e^{-\mu_1 x},$$

$$(I_2)_{obs} = (I_2)_{em}\, e^{-\mu_2 x},$$

$$(I_4)_{obs} = (I_3)_{em}\, e^{-\mu_3 x},$$

$$(I_4)_{obs} = (I_4)_{em}\, e^{-\mu_4 x},$$

where μ_1 through μ_4 are functions of Z and represent the attenuation coefficients for gamma rays of energies 1 through 4, respectively, in the matrix of atomic number Z. If these four equations are solved simultaneously to eliminate x, the average distance traveled in the matrix by the gamma rays, equation (15) is obtained.

$$\frac{(\mu_1-\mu_2)}{(\mu_3-\mu_4)} = \frac{\ell n\left[\frac{(I_1/I_2)_{em}}{(I_1/I_2)_{obs}}\right]}{\ell n\left[\frac{(I_3/I_4)_{em}}{(I_3/I_4)_{obs}}\right]}. \qquad (15)$$

The expression on the right side of the equation is an experimentally determined number. The left hand expression can be evaluated as a function of Z for given energies for gamma rays 1 through 4.

The average relative standard deviation in the function F, $\frac{S_F}{F}$, can be calculated by normal error propagation techniques to be

$$\frac{S_F}{F} = \left[\left(\frac{1}{I_1}+\frac{1}{I_2}\right)\frac{1}{\left(\ell n\left[\frac{(I_1/I_2)_{em}}{(I_1/I_2)_{obs}}\right]\right)^2} + \left(\frac{1}{I_3}+\frac{1}{I_4}\right)\frac{1}{\left(\ell n\left[\frac{(I_3/I_4)_{em}}{(I_3/I_4)_{obs}}\right]\right)^2}\right]^{1/2}. \qquad (16)$$

The analytical formalism is similar if three rather than four gamma rays are chosen. The use of four transitions is easier in practice because it enables one to use the stronger lines emitted in the decays of ^{239}Pu and ^{241}Pu. Both isotopes are generally present in relative abundances adequate to permit the use of four gamma ray energies.

The choice of the gamma-ray energies to be used in the analysis is a compromise between 1) the strong gamma-ray peaks in the spectrum,

2) the slope of the function in equation (15), which is highly dependent upon the energies used, and 3) the propagation of statistical uncertainties in equation (16), which again is, in practice, dependent upon the gamma-ray energies chosen. A reasonable compromise choice is as follows: E_1 = 129 keV, E_2 = 413 keV, E_3 = 148 keV, and E_4 = 208 keV. These are usually the four strongest peaks in the gamma-ray pulse-height spectra. If these transitions are chosen, and if the gamma ray attenuation coefficients are obtained from reference 5, a curve for $(\mu_1-\mu_2)/(\mu_3-\mu_4)$ as a function of Z can be calculated and is given in Figure 4. The values calculated are shown as the solid points. The scatter of these points about the smooth curve drawn through them demonstrates the uncertainty in the values of these coefficients. The two values, with their uncertainty flags, shown on the curve are intended only to show the size of the uncertainties in the experimentally determined value from 1) "normal barrels" exhibiting average gamma ray absorption, and 2) "anomalous barrels" exhibiting heavy absorption of the plutonium gamma rays. Values of the intensity ratios

$$\left(\frac{\left[\frac{I_1}{I_2}\right]_{em}}{\left[\frac{I_1}{I_2}\right]_{obs}}\right)$$

for the two cases were 1.2 and 4.0, respectively. For

$$\left(\frac{\left[\frac{I_3}{I_4}\right]_{em}}{\left[\frac{I_3}{I_4}\right]_{obs}}\right),$$

the values were 1.1 and 1.4, respectively. In both cases the number of events in the peak was assumed to be 2000 counts for all peaks. As can be seen from equation (16), the size of the uncertainties is a function of the number of events in the peaks. Therefore, the size of the uncertainty flags as indicated in Figure 4 depends upon the actual measurements made.

As is expected, however, the technique is most useful in estimating Z in those cases where the absorption from the matrix is the greatest. Thus, it is most useful for the "anomalous packages". The values in Figure 4 have been experimentally verified using slab absorbers and should be reasonably valid for homogeneous mixtures of Pu and matrix material.

D. Isotopic Analysis

Isotopic ratio measurements can easily be obtained through the equation

$$\frac{\text{gms } ^{239}\text{Pu}}{\text{gms } ^{241}\text{Pu}} = K_4\left(\frac{I_{129}}{I_{148}}\right)_{obs}, \qquad (17)$$

where K_4 is again an experimentally determined constant. The 148-keV gamma ray from ^{241}Pu decay has been chosen in preference to the 208-keV transition, for ^{237}U equilibrium decay, because of the small energy difference between the 129- and the 148-keV gamma rays. This reduces the error caused by neglecting the relative absorption of the two gamma-ray energies in the matrix. If more accurate results are desired,

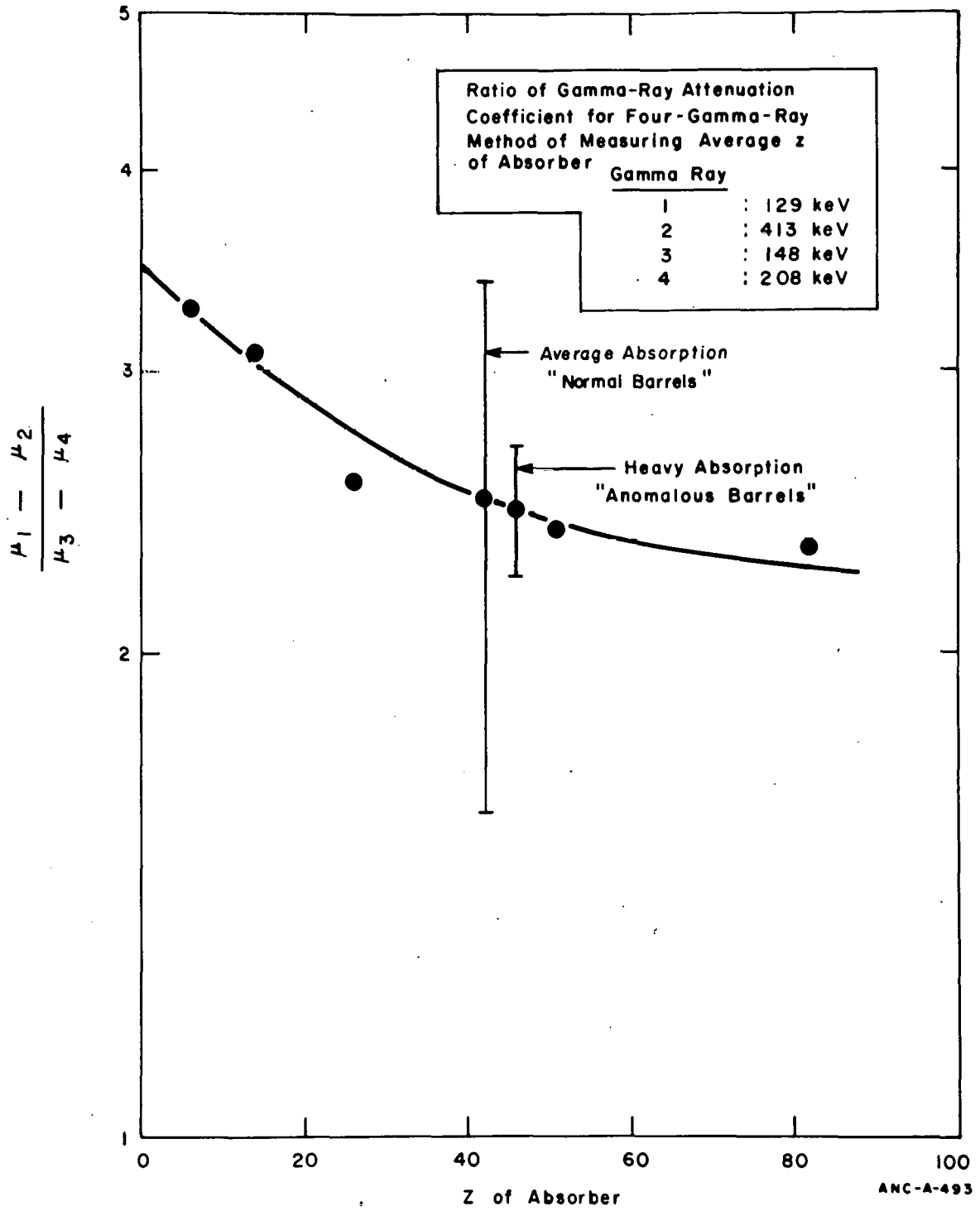


Figure 4 Ratio of gamma-ray attenuation coefficients, $\frac{\mu_{129} - \mu_{413}}{\mu_{148} - \mu_{208}}$, as a function of atomic number. Although the values of the coefficients were taken from reference 5, some scatter in the values about the line drawn through the points is still seen. The value at selected points was also verified using slab absorbers.

this difference in relative absorption must be accounted for. For uniformly distributed plutonium in a uniform 100 pounds of low Z matrix, however, the relative difference in absorption between these two gamma-ray energies is less than 3%. Thus, for "normal barrels", this correction is probably less than the statistical uncertainties in the results.

If the concentration of the ^{241}Pu relative to the ^{239}Pu is sufficiently low, i.e. less than 2 to 3% relative to the ^{239}Pu, the relative ^{241}Pu content can very easily be obtained through the use of the 208-keV gamma rays from ^{241}Pu (actually from ^{237}U) and the 203-keV gamma ray from ^{239}Pu. These gamma rays have been used successfully by Los Alamos[1] and others.[6] The principal advantage in the use of the 203- and 208-keV transitions is the fact that the two are relatively high in energy and are quite close together, thus minimizing and, in most cases, eliminating gamma-ray absorption and detector efficiency corrections. The difficulties in their use arise primarily at relatively high ^{241}Pu concentration where accurate intensity determinations of the 203-keV transition may easily be blocked by the presence of the intense 208-keV transition.

If ^{238}Pu or ^{240}Pu isotopic analyses are required, the problem becomes more difficult. The gamma rays from these two isotopes in "normal" isotopic mixtures of plutonium are quite weak and difficult to analyze. Table I shows a list of gamma-ray energies and intensities for the more intense gamma rays from the isotopes ^{238}Pu, ^{239}Pu, ^{240}Pu and ^{241}Pu. The 152-keV gamma ray from ^{238}Pu may be used for analysis of this isotope and is used in a manner similar to that for the 148-keV from ^{241}Pu in equation (17)

$$\frac{\text{gm } ^{239}Pu}{\text{gm } ^{238}Pu} = K_5 \left(\frac{I_{129}}{I_{152}} \right)_{obs} , \qquad (18)$$

or

$$\frac{\text{gm } ^{241}Pu}{\text{gm } ^{238}Pu} = K_6 \left(\frac{I_{148}}{I_{152}} \right)_{obs} . \qquad (19)$$

The 152-keV peak is very weak and a high-resolution Ge(Li) detector is generally required to resolve it adequately from the 148-keV peak from ^{241}Pu.

Analyses for ^{240}Pu content are somewhat more difficult. The peaks resulting from ^{240}Pu decay are partially obscured by those from both ^{239}Pu and ^{241}Pu decay. Probably the most usable gamma ray for ^{240}Pu analysis is that at 160.27 keV. As can be seen from the data in Table I, however, both ^{239}Pu and ^{241}Pu emit gamma rays having energies within 0.3 keV of the ^{240}Pu gamma ray. Depending upon the relative isotopic

abundancies of the three isotopes, the unresolved contributions to the peak at 160 keV from ^{239}Pu and ^{241}Pu can be comparable to, or can even overpower, that from ^{240}Pu. Therefore, rather large corrections to the intensity of the 160 keV peak are required to account for the contributions from ^{239}Pu and ^{241}Pu if such analyses are to be made. Equation (20) shows the form of such corrections.

$$\frac{\text{gms } ^{239}Pu}{\text{gms } ^{240}Pu} = K_7 \frac{I_{129}}{I_{160} - A_1 \times I_{129} - A_2 \times I_{148}} \qquad (20)$$

where the terms $A_1 \times I_{129}$ and $A_2 \times I_{148}$ represent the contributions to the intensity of the 160 keV peak from ^{239}Pu and ^{241}Pu, respectively. A_1, the ratio of observed intensities of the 160-keV peak from ^{239}Pu and the 129-keV peak is about 1.9×10^{-2}. Similarly, A_2, equal to $\frac{I_{160}}{I_{148}} - {}^{241}Pu$, is about 3.6×10^{-2}. Passive neutron coincidence counting is generally a more accurate method to analyze for ^{240}Pu content.

IV. EQUIPMENT

Various levels of sophistication in equipment can be obtained depending upon the available budget and desires of the individual laboratories. It is here assumed that in most cases the desire is for quick, accurate assay of low-level wastes with a minimum of effort by a semiskilled operator. It is also assumed that it is well known that a high level of automation does not imply a lesser skill on the part of the operator. The equipment listed below represents a spectrometer that is capable of yielding "state-of-the-art" results with an average investment in equipment. The largest variation in costs for various applications is in the analyzer itself.

A. Detector

As was discussed above, a Ge(Li) detector is recommended for these measurements. The larger the detector used, the more sensitive is the measurement, i.e. the smaller the quantity of plutonium which can be assayed and the shorter the time required for a given precision. It is assumed, however, that 15 minutes is a reasonable time in which to make the measurements. Similarly, the sensitivity of a detector is related to the energy resolution through some complex function which varies as $(\text{peak-width})^{-1}$[7] or $(\text{peak-width})^{-1/2}$[8] depending upon whom one consults. In any event, it is important to obtain a reasonably high-resolution detector. The speed with which one can acquire data is also quite important. Therefore, it is important that the detector and the preamplifier be able to be used at counting rates of 10,000-20,000 counts/second. Although this feature is generally a function of the preamplifier and amplifier, it is included here because

generally a detector purchase includes a preamplifier. Finally, the shape of the peaks in the pulse-height spectrum is important to most analytical procedures. Although not absolutely necessary, symmetric peaks are desirable. Asymmetric peaks are generally caused by inferior charge collection characteristics within the Ge(Li) detector itself and can be avoided through the purchase of a detector which does not demonstrate these effects.

With these general comments as background, the following specifications are provided and intended only as a guide for the acquisition of a detector. Several commercial detector suppliers have demonstrated the competence to deliver such detectors. One must, however, be prepared to measure the operating characteristics of the detector received to assure compliance with the specifications.

Detector Size, Volume, Efficiency

Type: either open-ended, or closed-ended coaxial geometry,

Size: >37-mm diameter
>37-mm length

length should not differ from diameter by more than $\pm$ 3 mm.

Drift Depth: undepleted detector core should not be greater than 1-cm diameter.

Sensitive Volume: >35 cc.

Efficiency: The full energy peak efficiency for the 1.33-MeV line from ^{60}Co shall be greater than 7% of that for a 3" x 3" NaI(Tl) detector when both detector are at the same distance and >20 cm from the source.

Detector Resolution The resolution of the detector (FWHM) shall be less than 1.3 keV for 122-keV gamma rays and less than 2.1 for 1.33-MeV gamma rays when used with an amplifier having integrating and differentiating time constants of <2 μsec.

Pulse-Height Spectrum Peak Shape The ratio of the full width at tenth-maximum (FWTM) to the full width at half-maximum for the peak at 1.33 MeV should not exceed 1.95 (a true Gaussian is 1.851). There should be at least 10 channels in the peak (FWTM) when this measurement is made.

Counting-Rate Capability The detector and preamplifier shall be capable of handling a counting rate of 15,000 c/s, using ^{60}Co and including all pulses above 50 keV, with a resolution degradation of <10%.

Cryostat Configuration and Detector Dead Layer The cryostat shall be of the "dip-stick" type and be a "right-angle" configuration. The capacity of the dewar shall be at least 35 liters. (An LD-35 dewar is very adequate.) The detector "dead-layer" shall be no greater than 0.7 mm. (In such a detector, the full-energy peak efficiency curve should have a maximum at about 100 keV or lower.)

Such a detector should be available, including preamplifier, for a price of from $8,000 to $15,000 depending upon the size. It is the feeling of this author that the detector is not the place to save money.

B. Spectrometer - Pulse-Height Analyzer

To acquire sufficient data for an adequate analysis it is necessary to obtain a multichannel analyzer. The most important characteristics of this analyzer are the gain and zero stability of the analog-to-digital converter (ADC) and the counting rate characteristics, again primarily due to the ADC. Thus, the ADC is of prime importance. Two other features of the system are worthy of consideration, the readout system and the self-analysis capability. Both of these latter two considerations are to a large degree subject to the desires and needs of the individual laboratories. If a central computer is available and the results of the assay are not needed immediately, magnetic-tape output is adequate and convenient. Small magnetic cassette units as well as the larger tape decks can be used as data output devices. These tapes may be read at later times and analyzed either on a small computer or a central processor. Punched paper tape may also be used in this manner although for large amounts of data, paper tape is very bulky and slow. If immediate assay results are required, it is possible to obtain self analysis units of varying complexity. The simplest of these contain hard-wired integrating capability to add counts in selectible groups of adjacent channels. Most versatile of the self analysis systems contain small digital processors that, upon command, can perform quick complete, and accurate analyses yielding 1) total gms Pu, and 2) isotopic ratios. The software for these systems can be obtained from the manufacturers or from national laboratories funded to provide such services. At the other extreme of analysis procedures, if infrequent counting and analyses are required, hand analyses may be sufficient, requiring only printed listings of the data.

ADC

Speed To handle the count-rate requirements it is desirable to have a 100-Mhz ADC. If longer counting times can be tolerated, 50-MHz systems may be adequate. A 4096-channel ADC is quite inexpensive and readily available.

D-C Restorer Every manufacturer has his own breed of restorer circuitry. The important feature is that the system (including amplifiers) be compatible so that counting ratios of ~15,000 c/s be handled with less than 10% broadening of the peaks in the pulse-height spectra.

Channel Capacity Memories are currently quite inexpensive. It is therefore recommended that a minimum of 2048-channel, 18-bit capacity (minimum) be required for the system. A 4096-channel capacity is desirable if a need exists to perform analyses for ^{238}U, ^{241}Am, and in some cases for ^{238}Pu, using the higher energy transitions. Digital channel suppression may be used but analog suppression, such as provided by expander amplifiers is not recommended due to the potential drifts and instabilities in these devices.

Linearity The linearity, both differential and integral, is a consideration in the purchase of a multichannel analyzer, but of paramount importance is that it remain constant, whatever it is. Since in this particular application, one does not measure energies precisely (i.e. better than $\pm$0.3 keV) it is not necessary to have an extremely linear system. The deviation from linearity can be measured accurately[9] in any case.

Amplifier An adequate low-noise amplifier can be obtained from several commercial manufacturers. The amplifier should have time constants, integration and differentiation in the range $\approx$2 μsec and have "pole-zero" trimming. Monopolar pulses are normally used since the introduction of bi-polar pulses generally results in a worsening of the resolution by about 50%.

C. Pole-Zero Compensation, D.C. Restorer, High Counting Rate Performance

In the use of high resolution spectrometers, experimental conditions are frequently far from ideal for optimum performance of the electronic systems employed. Perhaps the most difficult class of problems are those which result when high counting rates are encountered. Without the use of special precautions, operation at high counting rates will result in serious shifts of zero and system gain and degradation of resolution. Most of these observed effects are the result of fluctuations in the zero reference baseline at the input of the analog-to-digital converter, produced by random fluctuations. To provide some insight into these effects and the nature of the specialized circuitry employed to correct these problems, Figure 5 is presented. To achieve optimum signal-to-noise ratios in high resolution systems, a monopolar pulse shape with equal integration and differentiation RC time constants is generally employed. As indicated in Figure 5(a) the output pulse shape from such a filter network will exhibit a more-or-less Gaussian shape with equal rise and fall times. Following the pulse there will be a negative undershoot with a long recovery time constant (generally several hundred microseconds). At low counting rates, the pulse will have returned to the original baseline and will have no influence on succeeding events. If, however, at high rates, another pulse (indicated by the dotted line) occurs, the negative tail of the preceding pulse will result in a reduced amplitude measurement by the ADC. At reasonable rates (a few thousand pulses per second) the net effect of this will be an asymmetry of peaks as indicated at the far right of the figure.

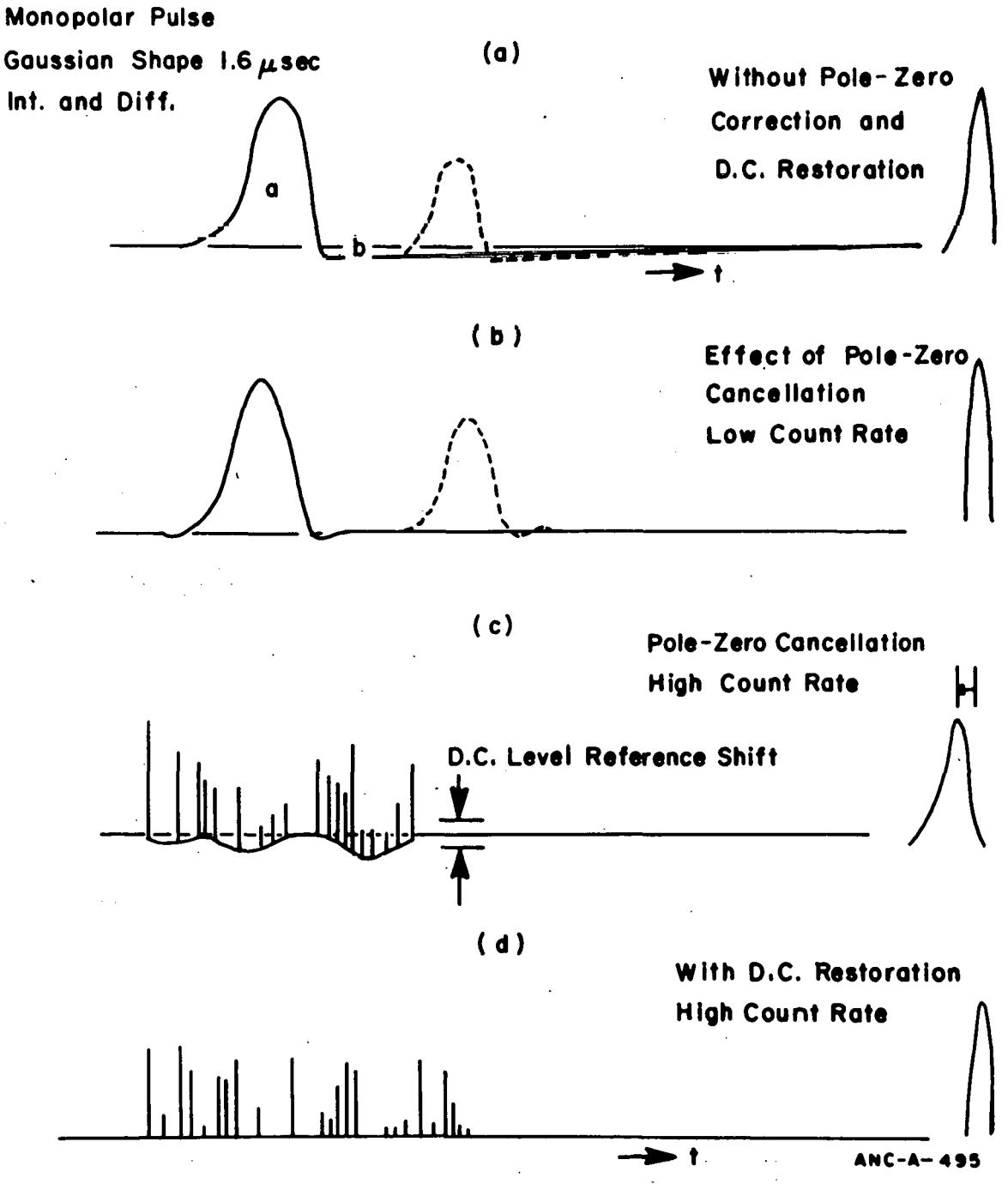


Figure 5 Amplifier pulse shapes with and without pole-zero cancellation and D.C. restoration circuitry.

A circuit technique, which has been termed "pole-zero cancellation", has been developed which quite effectively reduces the undershoot from a mono-polar pulse. The net effect of pole-zero cancellation is shown in Figure 5(b). Here we see that the undershoot returns quickly to the original baseline, thus pulses following will be unaffected. It should be pointed out that pole-zero cancellation must be accomplished on the complete preamplifier-amplifier system to be effective. The use of such networks in low-noise amplifier systems is now quite common, and if adjusted properly in combination with a given detector, will provide considerable reduction in spectrum degradation from undershoot at moderate counting rates.

At high counting rates (in excess of 5000 counts/sec), there are still problems from fluctuations in the baseline as shown in Figure 5(c). In this figure, the time scale has been reduced to show the long-term character of the baseline shift. As a result of residual charge on coupling capacitors within the AC coupled amplifier system, the baseline will vary in a random manner producing a net shift in the zero reference and a general broadening of peaks in the spectrum as indicated on the right side of the figure. This effect can be reduced by the use of DC baseline restoring circuitry. The action of these circuits is to remove the random fluctuations in the zero reference level by means of a diode arrangement. It should be stated that the input time constant on the restorer must be optimized for best performance at high rates. The net effect of this will be that the restorer will restore on noise pulses and a small loss in resolution at low rates will result. The most recent DC restorers have switch selection of this time constant to permit selection of the best operating conditions for a given experiment. The result of restoring action, properly optimized, is illustrated in Figure 5(d). Here it is seen that the zero reference baseline has been maintained at a constant level and the degradation seen without the restorer has been largely removed.

To provide experimental verification of these principles, Figure 6 summarizes the results of studies made on the performance of a laboratory Ge(Li) spectrometer as a function of input counting rate. The spectrometer consisted of a 2.5 cm^2 x 8 mm Ge(Li) detector mounted in a cryostat with a cooled-FET preamplifier, a linear amplifier which includes first order pole-zero cancellation, and a 12-bit ADC. The input of the ADC included a simple restorer circuit with optimized time constants. Results are presented for two cases: 1) without pole-zero cancellation, and 2) with pole-zero cancellation. The gain was adjusted to place the 1.332 MeV peak of ^{60}Co in channel 3750. Although the peaks have been normalized to fall in the same channel, in no case did the peak position change by more than 1 channel below 5000 counts/second and 3 channels at 35,000 counts/second.

D. Pile-Up Effects at High Rates

In the system indicated in Figure 6, at high rates some asymmetry remains on the high side of the peak. This results from pile-up effects which are the result of two pulses arriving at the input of the ADC within a very short time. Events associated with pile-up generally remove that event from its normal position in the pulse-height distribution

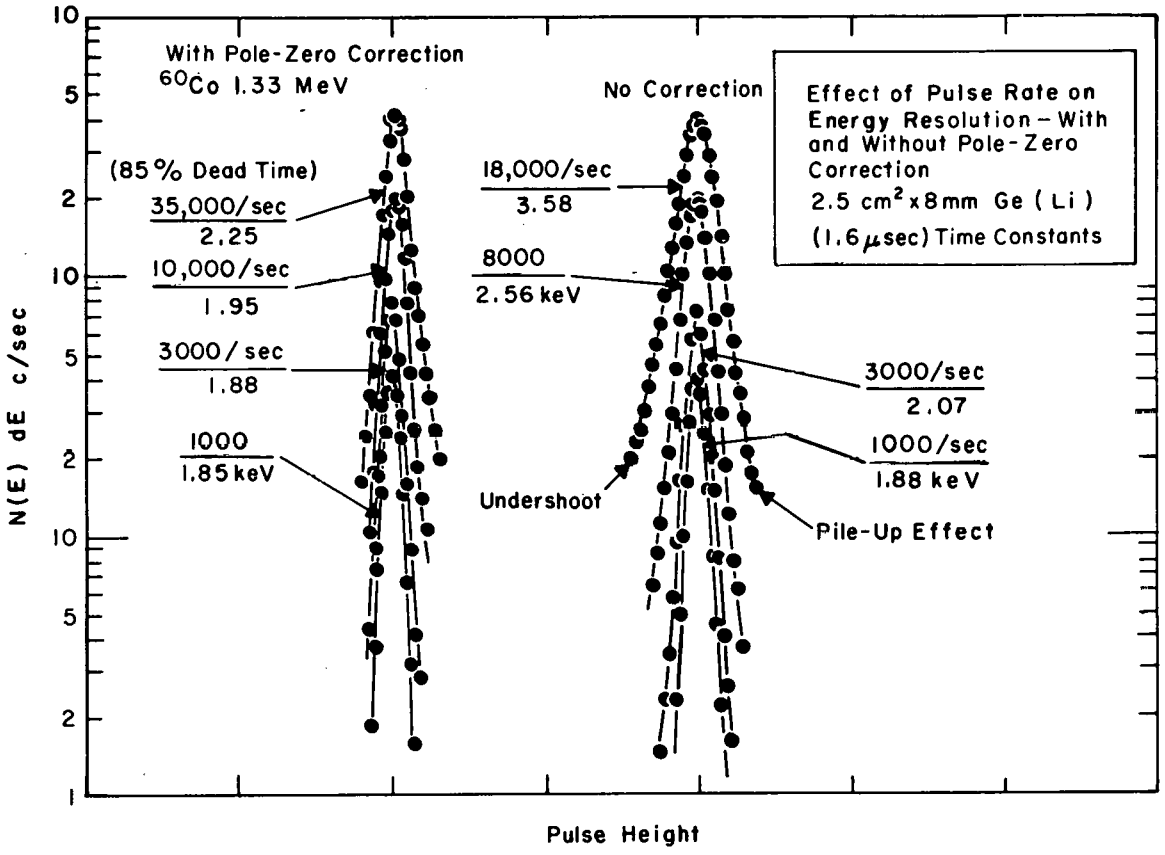


Figure 6 Pulse-height distributions of 1.33 MeV peak from ^{60}Co as a function of counting rate with and without pole-zero compensation. The effects of pulse pile-up are not affected but the peak width is highly affected through the use of pole-zero circuitry.

and place the pulse at some other pulse height. These pile-up problems are called random pulse summing effects and their magnitude is proportional to the square of the counting rate.

The effects of pulse pile-up have been measured in a series of efficiency measurements made at several different counting rates. The results are presented in Figure 7. These data have been corrected for counting losses in the amplifier and a 1-Mhz scaler used to record the counting rate. A fit can easily be made to the data, verifying the relationship and yielding a resolving time of 13.6 μsec. This value is consistent with the observed pulse shape. A width (across the base of the pulse) of approximately this magnitude was observed on an oscilloscope for pulses from the amplifier. The spectrometer used in these measurements incorporated "pole-zero" pulse shaping and baseline restoration. The amplifier time constants were 1.6 μsec integrating and differentiating and the pulse was monopolar.

From these data, it may be seen that some substantial losses can occur in the full energy peaks at counting rates above 10,000 counts/second. These must be taken into account if accurate measurements are to be made at such rates. The effect can be reduced in magnitude if

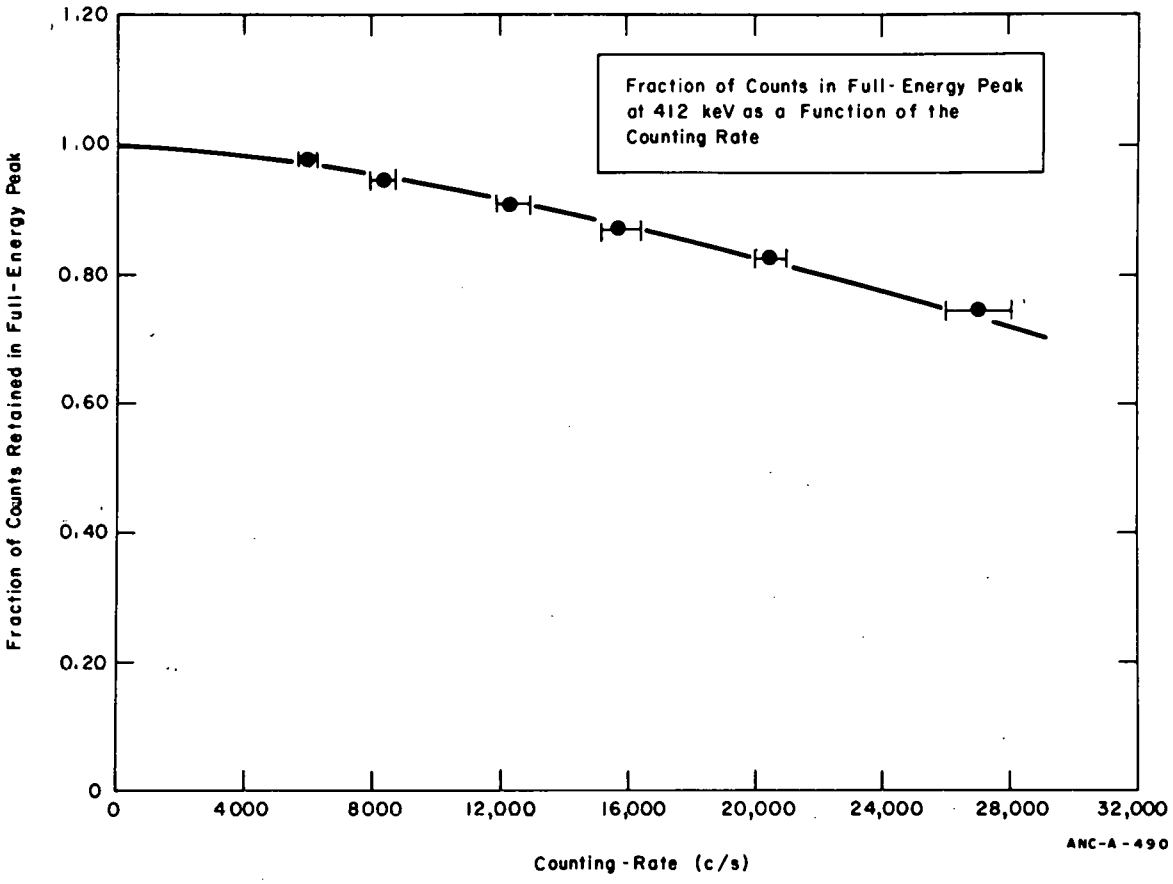


Figure 7 Fraction of counts retained in a full-energy peak as a function of detector counting rate. The pulses are lost as a result of pulse pile-up.

shorter time constants or time constant[10] switching are used in the amplifier.

V. MEASUREMENTS AND ANALYSIS

A. Experimental Arrangement of Detector

The Ge(Li) detector should be located as far from the source as is practically possible so as to reduce problems caused by poor counting geometry. A poor geometry as used here is one in which the various portions of the source are not equal distances from the detector. The horizontal axis of the detector should intersect the geometric center of the barrel. The barrel should be rotated at a rate not less than 1 revolution per length of the count. A distance from the center of the barrel to the detector equal to the length of the barrel will yield a counting rate of $\sim$0.5 c/s/gm (^{239}Pu) for the 413-keV peak, using a large ($\sim$50 cc) Ge(Li) detector. This means 5% statistics can be obtained on a 0.5 gm barrel in 1 hour. If poorer accuracy but higher precision is permitted, the detector can be moved closer to the source.

The detector should be housed in a three-sided lead cave to shield against background radiation. The lead should be at least 2 inches thick. In addition, the detector cap itself should be encased with about 40 mils of some metal such as cadmium. This encasement should include the sides as well as the front of the cap. The purpose of this absorber is to absorb the 60-keV transition from ^{241}Am and ^{237}U in the Pu source, reducing the extraneous counting rate in the detector. If the sides are not shielded, air-scattered low-energy radiation as well as direct radiation will be observed through the sides. Cadmium is a reasonable choice as a preferential absorber for 60-keV radiation; 40 mils has a transmission at 60-keV of 0.005, at 129-keV of 0.51 and at 413-keV of 0.91. In addition, it is readily available.

B. Data Analysis Techniques

Many procedures are currently available for analyzing the data from Ge(Li) spectrometers for gamma-ray energies and, more importantly for this experiment, for gamma-ray intensities. The procedures generally fall either into the categories of 1) hand analysis, or 2) automated or semi-automated analysis using digital computers. The basic difference between the two so far as intensity determinations are concerned lies in the fact that in hand analyses, the peaks are integrated by adding the counts in the channels of the peak and subtracting a suitable background, whereas in automated analyses the peaks are fitted with an analytic function together with a suitable background and the areas are computed.

The principal advantage of hand analyses techniques is its simplicity and ability to integrate asymmetric peaks more easily. The major disadvantages are 1) the time consuming nature of the analyses, a major problem when much data must be analyzed, and 2) a possible lack of consistency in background determinations, potentially a very serious problem. Advantages of automated computer analysis of data are obvious, 1) speed of analysis, 2) ease of analyses, and 3) internal consistency of analysis. Major disadvantage of computer data reduction lies in the difficulty of analyzing asymmetric peaks or in the cost for small numbers of analyses. Several computer routines have been written for the analysis of asymmetric peaks. A typical routine has been described by Lederer.[11] In this technique the data are fit with a Gaussian function and a modified exponential function describing the low-energy "tailing" on the peak. If the detector used to obtain the data does not give asymmetric peaks, a routine which simply uses a Gaussian function[12] may be more easily used.

Regardless of the analysis technique used, be it hand or automated analysis, the problem of how to choose an appropriate background or base under the peak must be faced. There are, again, several different techniques being employed by different groups. Many techniques use a line, either linear or quadratic, drawn between the channels on the low-energy side of the peak in question and those on the high-energy side. Others use an extension of the spectrum shape on the high-energy side under the peak. There are advantages and disadvantages to either group of approaches but regardless of which technique is used, the most important aspect is that of consistency. Most of the area determination

techniques work quite well for prominent peaks providing a reasonable baseline is chosen in a consistent fashion and, in the case of hand analysis, the same number of channels are chosen in each analysis for the same peak.

C. Analysis of Typical Data, Example

A typical spectrum from some plutonium waste is shown in Figure 8. The position of the 129.3-, 148.6-, 152.7-, 160.1-, 267.5- and 413.7-keV peaks are as indicated. The backgrounds used in the integrations to obtain the total counts in the peaks are also shown in the figure. The choice of such backgrounds is subject to conventions used at any particular laboratory. What is felt to be a reasonable convention and the one that has been adopted here is to extend linearly the spectrum shape above the high energy side of the peak under the peak itself. Again, whatever convention is used, the important feature is that of consistency. The peaks in Figure 8 were integrated both by adding up the counts in the individual channels, and by fitting a Gaussian function to the data and calculating the area of the Gaussian, using a computer. In this case, the results from the two techniques did not differ by more than 2%. This difference results primarily from incorrect description of the distribution in the wings of the peaks by Gaussian functions. The difference is a bias rather than an inaccuracy and is completely negligible if a single method is always used. Much more serious differences would occur in the two types of analyses if the peaks were asymmetric, generally having low energy "tailing" effects. For this reason, it is most advantageous to use detectors evidencing symmetric peak shapes.

Table II lists the results of the peak integrations from automated analysis for the data in Figure 8. The numbers given are for total counts in the full-energy peak for a 4000-second counting period. Normally, a counting period of this length would not be used.

TABLE II

Table of gamma-ray energies and relative intensities (in units of counts in the full-energy peaks) for the relevant peaks in the data of Figure 8.

E_γ (keV)	Area (counts)	Nuclide Assignment
129.3	32200	^{239}Pu
148.6	129400	^{241}Pu
152.6	3600	^{238}Pu
160.1	11900	Combination of ^{240,239,241}Pu
267.5	10800	^{237}U (in equilibrium with ^{241}Pu)
413.7	4600	^{239}Pu

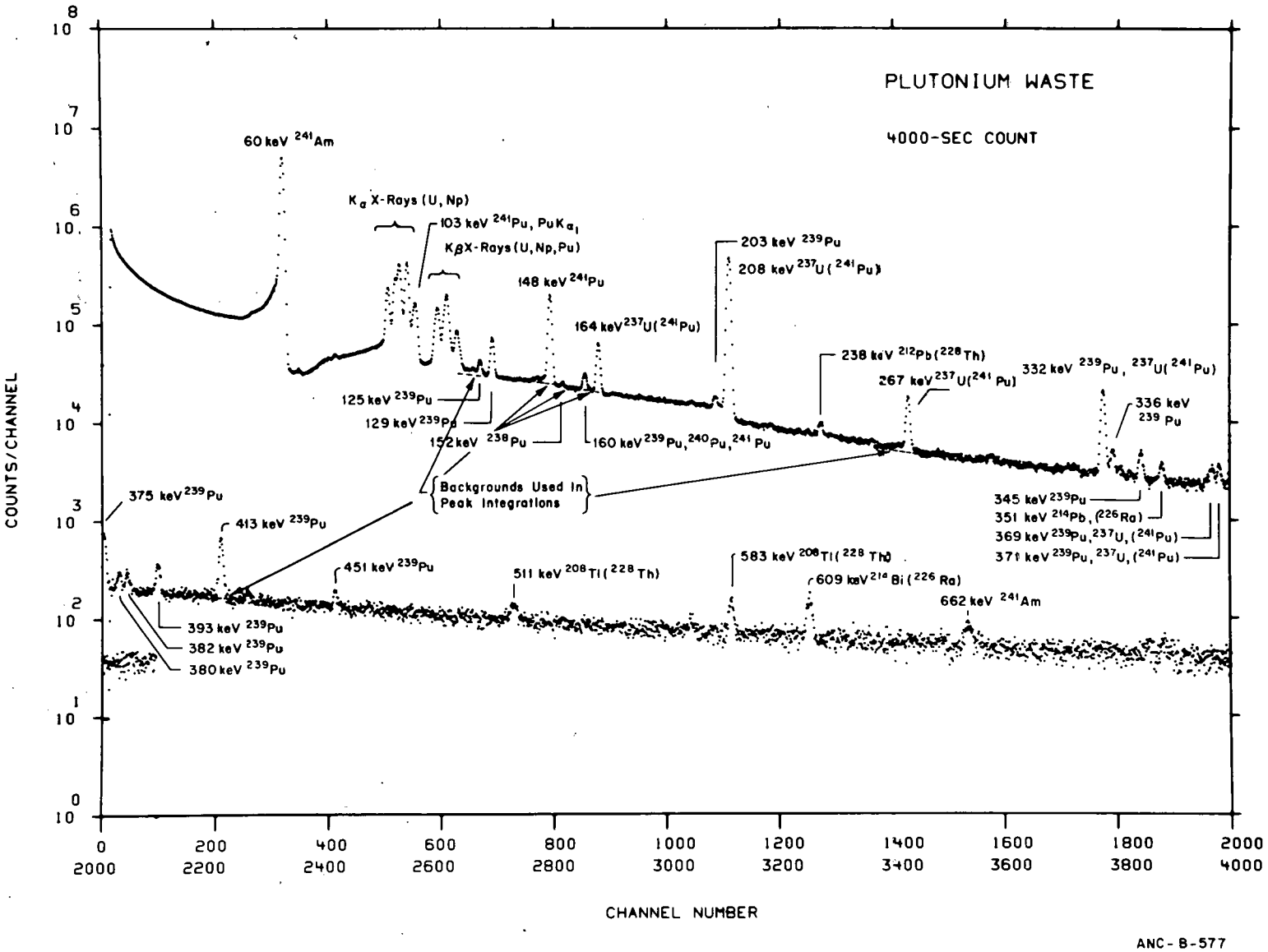


Figure 8 4096-channel pulse-height spectrum of the gamma rays from a waste barrel containing 0.9 grams of plutonium.

^{239}Pu Analysis Using equation (8), with the experimentally measured parameters for the detector and configuration used, the equation becomes:

$$\text{gms } ^{239}\text{Pu} = 8.84 \times 10^{-2} \left(\frac{I_{129}}{\text{counting time}}\right)_{obs} \left(\frac{7.4}{\frac{I_{129}}{I_{413}}_{obs}}\right)^{1.92}$$

$$= 8.84 \times 10^{-2} \left(\frac{3.22 \times 10^{4}}{4 \times 10^{3}}\right)\left(\frac{7.4}{\frac{32200}{4600}}\right)^{1.92}$$

$$= 0.751 \text{ grams } ^{239}\text{Pu}.$$

Isotopic Analyses

(a) ^{241}Pu From equation (17) and parameters for this detector,

$$\left(\frac{\text{gm } ^{239}\text{Pu}}{\text{gm } ^{241}\text{Pu}}\right) = 90.4 \left(\frac{32200}{129400}\right) = 22.5.$$

(b) ^{238}Pu

$$\left(\frac{\text{gm } ^{239}\text{Pu}}{\text{gm } ^{238}\text{Pu}}\right) = 32.6 \left(\frac{32200}{3600}\right) = 291.6$$

(c) ^{240}Pu

$$\frac{\text{gm } ^{239}\text{Pu}}{\text{gm } ^{240}\text{Pu}} = K\left[\frac{I_{160} - 1.25 \times 10^{-3} * I_{129} - 3.6 \times 10^{-2} I_{148}}{I_{129}}\right]^{-1}$$

$$= 1.31 \times \frac{32200}{11900 - 40 - 4658} = 5.84$$

The analysis summary is given in Table III.

TABLE III

Assay of the plutonium waste from the data of Figure 8.

Isotope	Grams	%
^{238}Pu	0.0020±0.0005	0.2
^{239}Pu	0.75 ±0.05	82.2
^{240}Pu	0.13 ±0.03	14.0
^{241}Pu	0.033 ±0.002	3.6
TOTAL	0.913 ±0.100	100.0

The ^{240}Pu relative isotopic concentration has by far the largest uncertainty because of the necessity to correct the observed intensity of the 160-keV gamma ray for the presence of ^{241}Pu and ^{239}Pu.

D. Identification and Analysis of Anomalous Barrels

Barrels that severely violate the basic assumption used to devise the analytical procedures can usually be identified by an unusually low value of the ratio $\frac{I_{129}}{I_{413}}$. In the example chosen in the analysis above this ratio was 7, as compared with 7.4 for no absorption at all. For barrels that exhibit an extraordinary amount of absorption, usually from high Z material, this ratio has been observed to be as low as 25% of this value. As a general rule, if the value of the ratio drops below about 60-70% of its value with no absorption, the barrel should be denoted as anomalous and set aside for other procedures. The other procedures can include profiling measurements using collimated counters to determine the location of the bulk of the plutonium, or transmission measurements to determine the absorption as a function of position in the barrel and to locate the heavy mass. It is possible to estimate the Z of the absorbing material using procedures outlined in Section III.C above. However in most cases, the absorption will be caused by a mixture of high and low Z material and the correction factor will be difficult to estimate. For the anomalous barrels studied at this laboratory, the value of the exponent in equation (8) which yielded proper analyses (verified by subsequent recovery of the plutonium in the 55-gallon barrels) was about 1.0. These particular anomalous samples had values for the ratio inside the bracket in equation (8) ranging from 2 to 4.5. The plutonium in these barrels was found to be concentrated in relatively small clumps, resulting in some self absorption by the plutonium.

REFERENCES

1. LA-4705-MS, Nuclear Safeguards Research & Development Program Status Report, January-April 1971.

2. WASH-1177, Proceedings of the Symposium on Passive Gamma-Ray Assay, USAEC Report, p. 5 (June 1971).

3. A. J. Dukat, J. Gonser and D. B. James, Trans. Am. Nucl. Soc. 13, #2, 749 (1970).

4. ARH-1993, W. H. Zimmer, Atlantic Richfield and Hanford Co., (March 1971).

5. Values of the total absorption coefficients are taken from LASL 2237. These coefficients will vary slightly according to the reference used.

6. T. Dragnev and A. Stefanescu, IAEA, Vienna, private communication.

7. J. E. Cline, IEEE Trans. Nucl. Sci., Vol. NS-15, #3, 198 (1968).

8. J. A. Cooper, Nucl. Instr. and Meth. 82, 273 (1970).

9. R. J. Gehrke, J. E. Cline and R. L. Heath, Nucl. Instr. and Meth. 91, 349 (1971).

10. V. Radeka, Brookhaven National Laboratory Report, BNL-16245 (October 1971).

11. C. M. Lederer, UCRL 18948

12. R. G. Helmer, R. L. Heath, M. Putnam and D. H. Gipson, Nucl. Instr. and Meth. 57, 46 (1967).