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Title: Measurement of Plutonium Isotopic Composition - MGA

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IAEA Advanced Plutonium Verification Techniques Training Course
Los Alamos, New Mexico

Measurement of Plutonium Isotopic Composition - MGA

Laboratory Exercises

Objective: To provide students with laboratory experience in gamma-ray assay of plutonium isotopic composition. This module is designed to provide comprehensive instruction in the MGA method and the measurement techniques that produce optimum results. The student will participate in exercises designed to show the main features of MGA.

Equipment:
Canberra InSpector-2000 multichannel analyzer
Canberra GL0515R 500 mm^2 x15 mm planar detector with 7 liter dewar.
Laptop computer
Collimator for germanium detectors
Cables and battery chargers
Various filters

Nuclear Materials:
LAO series
PEO 382 series
HUA5069
PIDIE series
CBNM series
A1 series (##-000)
FZC158

SEE THE APPENDIX TO THIS MODULE FOR SOURCE DETAILS

Instructors: *Los Alamos National Laboratory Technical Staff*

Lesson Plan: *J. Steven Hansen and Doug Reilly*, LANL
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LABORATORY EXERCISES

A. Background

In this module, we will use the Canberra InSpector-2000 Multichannel Analyzer with a high-purity germanium detector (HPGe) and the MGA isotopic anlysis software to assay a variety of plutonium samples. The module provides an understanding of the MGA method, its attributes and limitations. You will assess the system performance by measuring a range of materials similar to those you may assay in your work. During the final verification exercise, the results from MGA will be combined with the $^{240}Pu_{eff}$ results from neutron coincidence or multiplicity counters so that measurements of the plutonium mass can be compared with the operator-declared (certified) values.

B. Exercise 1—Initial Setup of IMCG with MGA

MGA requires an energy calibration of 0.075 keV/Ch. Careful pole zero compensation and amplifier shaping time selection should be made to ensure peak symmetry and good resolution.

1. Physical Setup

a. All IAEA small planar HPGe detectors require negative high voltage. Verify that the IMCA is set for negative HV by removing the two front panel thumb screws, the front panel, and the case. BE CERTAIN THE IMCA IS SWITCHED OFF AND NOT CONNECTED TO THE AC MAINS.

b. Remove the cover labeled "High Voltage Interlock". Verify that the black block labeled "Polarity" is positioned for negative polarity and that the smaller black block labeled "Range" is positioned for "5KV". If they are not, unplug the block(s), rotate them by 180 degress, and plug them in again, taking care not to bend any of the pins. Replace the interlock board, the case, and the front panel.

c. Set up the HPGe detector and collimator. Connect the detector, IMCA, and computer.

Fig. 1. Detector system setup for plutonium isotopic measurements.

2. Setting up the IMCA

The IMCA program is used to acquire and analyze the spectra. This program contains direct links to MGA and MGAU (uranium version of MGA).

a. Switch on the IMCA unit. Start the IMCA program by clicking on the desktop icon. Enter "iaea" for the user name and click continue (there is no password).

b. Select "Germanium Applications", "Measure MGA Sample (Low Energy Spectrum)", and "measure samples".

c. Select the detector and IMCA in the next screen. This should call a parameter file with the correct HV, amplifier gain and PZ. If a parameter file does not exist for your system, press "create file" and follow the instructions on the screen.

d. Select a standard from the CBNM or LAO series (small mass) and ask the instructor to place it in front of the collimator for pole zero and amplifier gain adjustments. Place a 1-mm-thick Cd sheet between the detector and sample.

e. Collect a short spectrum and verify that the 60-keV and 208-keV gamma rays and are within a few channels of 794 and 2773, respectively.

f. Select the correct cadmium filter thickness such that the intensity of the 60-keV gamma ray from the decay of ^{241}Am is approximately the same peak height as the K x-rays in the 94-keV to 104-keV region (plus or minus 50%).

g. If the energy calibration is not correct, remedy it using the energy calibration routine is at ***Analysis/Energy Calibration***. You may place the 59.54-keV gamma ray at channel 794 and the 208-keV gamma ray at channel 2773 for a two-point calibration.

h. Verify that the resolution of the 208-keV gamma-ray peak is better than 875 eV FWHM.

C. Exercise 2—Understanding the Intensity Advantage of MGA Method

In this exercise, you will acquire a spectrum for a sample of almost pure ^{240}Pu. You recall from the lecture that a principal reason to develop the response function analysis method was to use the high intensity line at 104 keV for the assay of ^{240}Pu. You will collect and keep a spectrum from ^{240}Pu to remind you of this intensity advantage.

The Measurements

a. Remove any cadmium absorbers for this exercise (check inside the collimator).
b. Place the sample FZC158 a suitable distance from the detector and acquire 10,000 counts in the ^{240}Pu peak at 160.3 keV.
c. Print copies of the spectra annotating them as shown below.
d. Place ROIs around the two peaks from ^{240}Pu and determine their peak areas.

e. Record the information in the table below and save the data and spectrum in your course manual.

^{240}Pu Peak Intensity Comparison

Standard	104.24 keV ROI	160.28 keV ROI
FZC158		

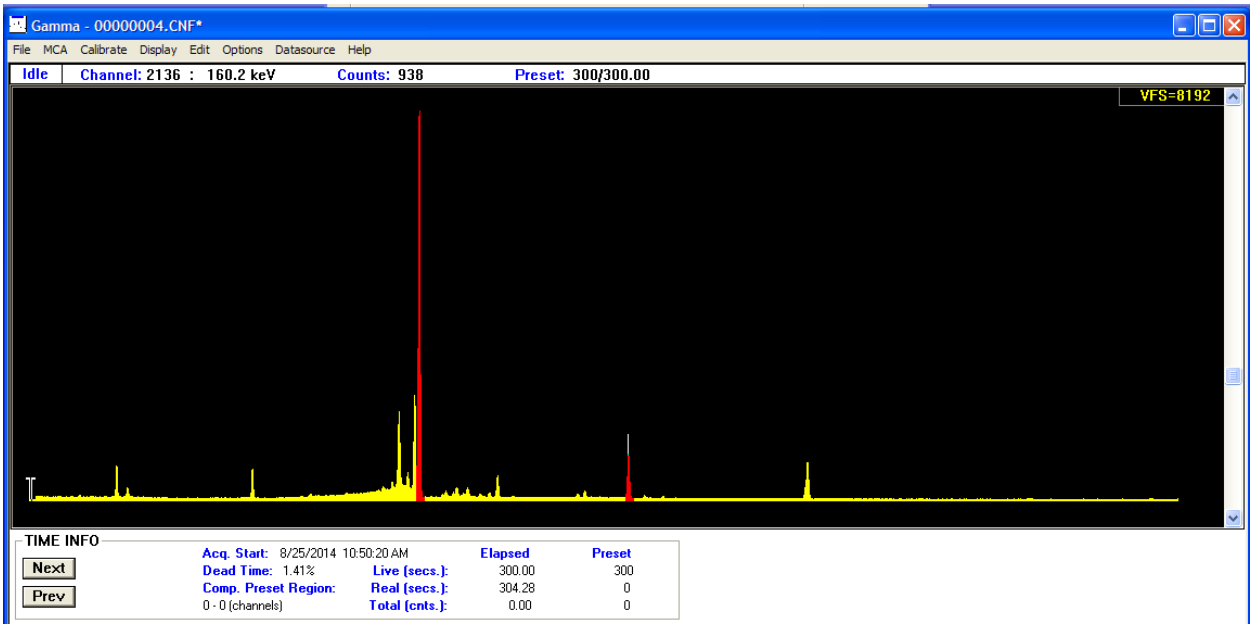

Fig. 2. Spectrum showing two major lines of ^{240}Pu

D. Exercise 3—Assessing the Capabilities of MGA

In this exercise, you will use the MGA program to assay seven samples of differing plutonium isotopic compositions. The goal is to evaluate the system's capability to measure materials similar to those that you might encounter in your work. The parameter of general interest is the $^{240}Pu_{eff}$. This quantity, defined below, when divided into the $^{240}Pu_{eff}$ mass provides the estimate of the total plutonium mass. The fissile mass can then be computed by multiplying the total plutonium mass by the sum of the mass fractions of ^{239}Pu and ^{241}Pu. The three formulas below express these concepts.

$$^{240}Pu_{eff} = 2.52 \times {}^{238}M_f + {}^{240}M_f + 1.68 \times {}^{242}M_f$$

$$\text{Total plutonium mass} = {}^{240}Pu_{eff}\text{mass} / {}^{240}Pu_{eff}$$

$$\text{Total fissile mass} = (\text{Total plutonium mass}) \times ({}^{239}M_f + {}^{241}M_f)$$

The Measurements

a. Select the first standard and record the declared isotopic information in the first line of the table. [Select only one standard from each of the series except for the PIDIE series, for which three standards are measured.]

b. Count the standards for a sufficient time to acquire a precision of better than 0.3% in the 129.3-keV peak area.

c. Enter the required information into the table below. The declared isotopic composition for the standards is provided as an appendix to this module and should be recorded in the first row for each standard.

d. Compute $\mathbf{^{240}Pu_{eff}}$ for the measured results using the formula above and determine the relative percent error between it and the declared values.

e. Figures 3 through 5 show a typical spectrum, relative-efficiency curve, and MGA fit.

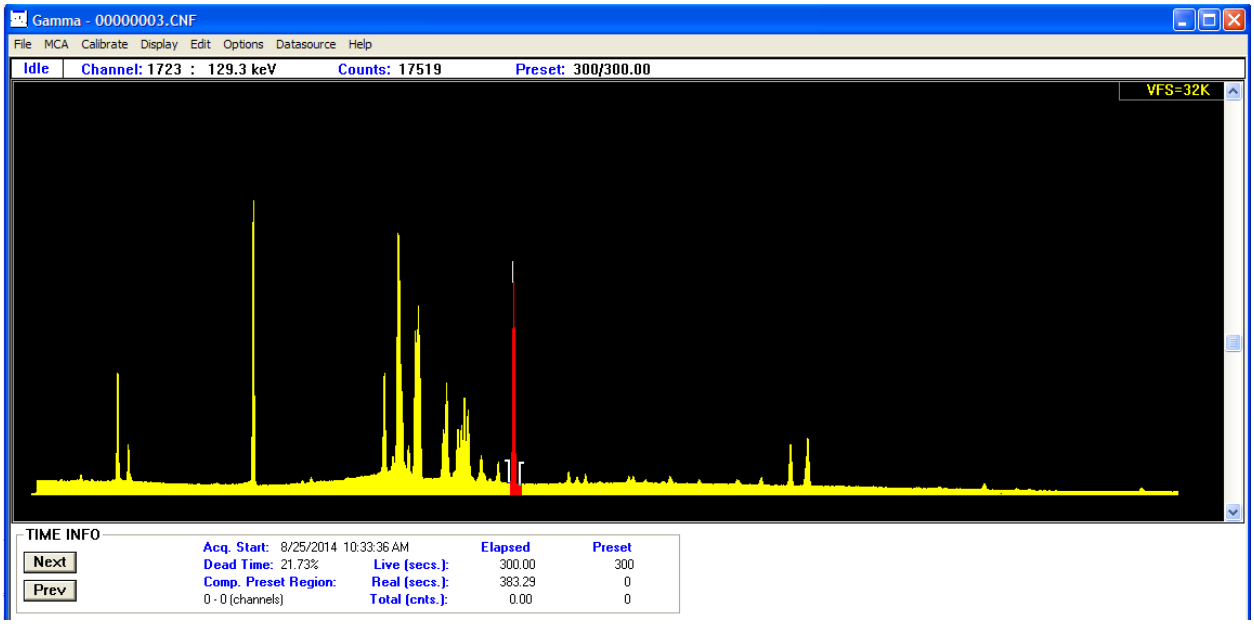

Fig. 3. Spectrum taken with IMCG for MGA

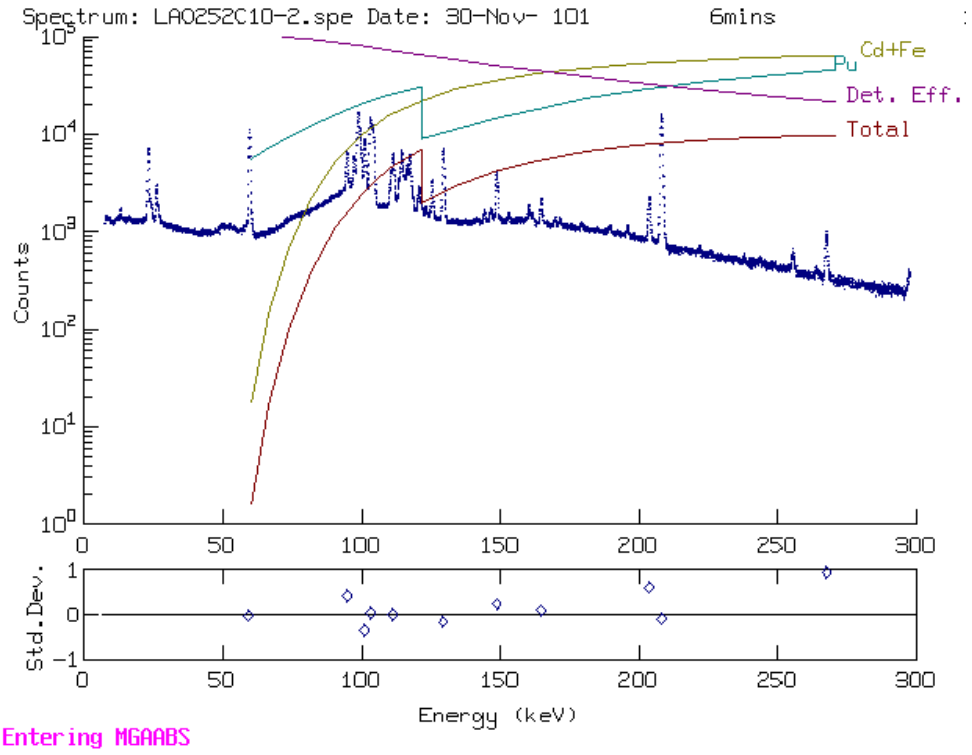

Fig. 4. The components contributing to the MGA efficiency function

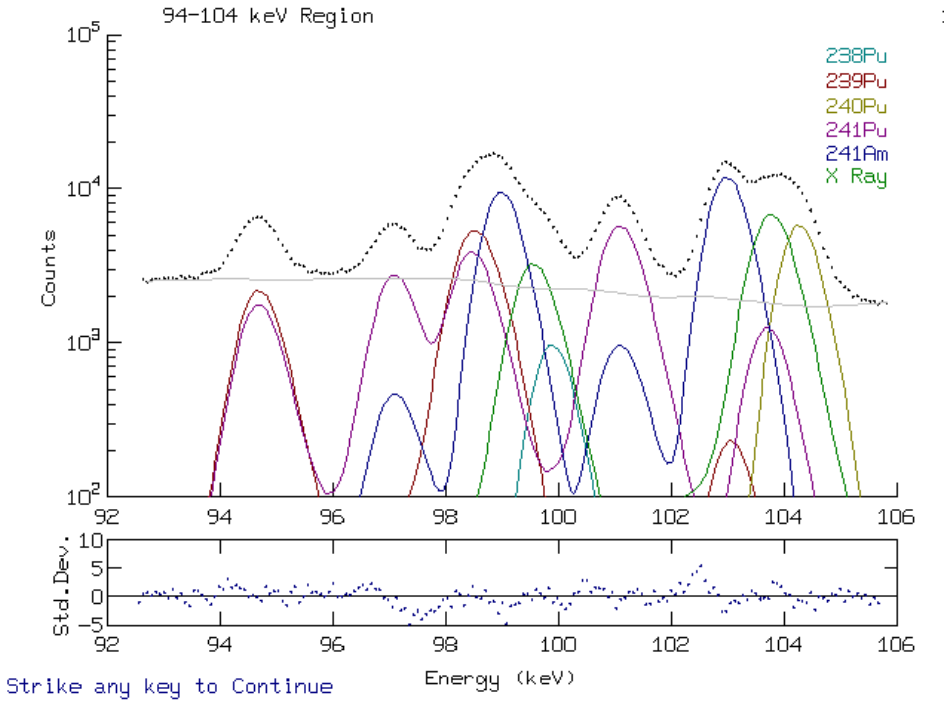

Fig. 5. Response Function Analysis with the MGA Code

Declared and Measured Data Results (MGA in IMCA)

Standard	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{240}Pu$_{eff}$	Relative Error (%)
PEO382C								
STD11								
LAO								
A1 (##-000)								
ISOSTD								
							Average Error (%)	
							Average Bias (%)	

E. Exercise 4—Assaying Mixed U-Pu Oxides

In this exercise, we will use MGA to assay one mixed U/Pu oxides (MOX) item. We compare the ratio of the two elements as well as the isotopic composition of plutonium. The uranium enrichment is about 1%. This presents a significant challenge to measure the enrichment.

The Measurements

a. Measure HUA5069 and enter the measured isotopic information in the line below the reference values.

b. Count the item for a sufficient time to acquire a precision of better than 0.3% in the 129.3-keV peak area.

c. Record and save the spectrum and response-function analysis for comparison with the pure plutonium samples taken in the last exercise. What differences exist due to the presence of the uranium in the MOX? Note these on the spectrum for future reference.

Declared versus Measured Data Results

Lot ID	Ref. Date	Pu Mass (g)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{235}U/U %	U/Pu
HUA5069	8/89	114.0	0.060	87.276	11.665	0.813	0.186	1.106	1.073	5.960

F. Exercise 5 - THE WALL THICKNESS PROBLEM

1. Background

To function properly, MGA requires high-quality peak information in the highly-convoluted 100-keV region. MGA performs best when peak shapes can be approximated with a Gaussian peak with a short, low-intensity tail. Thick attenuators not only decrease the count rate in the low-energy region, but also produce conditions that favor long, high-intensity tailing. If attenuation becomes too extreme, the analysis fails. This exercise attempts to study and document this failure mechanism.

2. Physical Setup

a. Identical to the previous exercises with the cadmium absorber in place.

b. Set of 6 - 8 steel attenuators each with a thickness of 1/8 inch (3.175 mm)

3. Measurements

a. Conduct the measurements with one of the large LAO or PEO standards.

b. Place a ROI around the 208 keV peak and accumulate counts for a fixed period of real time for each measurement. Observe any peak shape discrepancies such as low energy tailing.

c. Beginning with zero iron attenuators, increase the thickness of the iron attenuators until the analysis package no longer provides a reliable result. Maintain the dead-time at approximately 30% by positioning the source appropriately.

d. Fill in the table with the required information for each thickness.

e. Record in the comments section any warning or error messages.

f. When failure occurs record the error message in the comment section.

Effects of Attenuation on Assay Results

Source:_____		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	$^{240}Pu_{eff}$
DECLARED ISOTOPICS								
Attenuator	Thickness(cm)							
None	0.0							
Fe	0.3175							
Fe	0.6350							
Fe	0.9525							
Fe	1.270							
Fe	1.588							
Fe	1.906							
Fe	2.224							

At Failure:
Attenuator Material -- Fe
Measured Thickness of Attenuator (Cm)

G. Exercise 6—Effect of Deadtime on Assay Results

Select a high-mass sample and make 100-s measurements using real time. Begin at 20% deadtime and increase to greater than 90%, recording the effects of increased deadtime in the table below.

Effects of Deadtime on Assay Results

Sample	DT%	Real Time (sec)	104 keV Area	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	$^{240}Pu_{eff}$	$^{240}Pu_{eff}$ Sigma	208 keV FWHM
	20%	100									
	30%	100									
	40%	100									
	50%	100									
	60%	100									
	70%	100									
	80%	100									
	90%	100									

H. Exercise 7 - THE HEAVY METAL ABSORBER PROBLEM

1. Background

MGA uses the many peaks in the 60-260 keV range to calculate the relative efficiency curve. It assumes that the transmission through the absorbers in this energy range varies smoothly without sudden discontinuities (see Figure 4). When an absorber with the absorption edge above 60 keV is used (Ta, W, Pb…) then the transmission through the absorber will have a discontinuity at the absorption edge (similar to the Pu transmission curve in Figure 4). Since MGA does not know about this jump of the transmission curve, its calculations of the efficiency curve may be incorrect, which may lead to poor results.

2. Physical Setup

a. Similar to the previous exercises with the cadmium absorber in front of the detector.

b. Set of 3 - 4 Ta and/or W attenuators each with a thickness of 0.1 mm.

3. Measurements

a. Conduct the measurements with one of the Pu standards. Adjust the Cd absorber thickness such that the 60-keV peak height is about half of those in the 100-keV region. Set the measurement time to 100s. Record the results into the table below.

b. Add one sheet of Ta filter in front of the Cd absorber (between the Cd absorber and the Pu source). Notice the change of the 60-keV peak relative to those in the 100-keV region. Record the results into the table. Discuss the results and output messages.

c. Repeat with additional sheets of Ta (and/or W). Discuss the results.

d. If time permits, conduct one or two other measurements using different Pu standards. Record the results into the table. Discuss the results and output messages.

Effects of heavy metal filter on Assay Results

Source:_____		^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	$^{240}Pu_{eff}$
DECLARED ISOTOPICS								
Attenuator	Thickness(mm)							
None	0.0							
Ta	0.1							
Ta	0.2							
Ta	0.3							
Ta	0.4							
W	0.1							
W	0.2							
W	0.3							
W	0.4							

I. Exercise 8 - THE HOLE IN ABSORBER PROBLEM

1. Background

In the previous exercise, the discontinuity in the absorber transmission curve changes the efficiency of the 60-keV relative to those in the 100-keV region in a way that MGA cannot account for and may give poor results. A Cd absorber with an appropriate hole in the middle can also alter the efficiency curve in such a way that MGA may not be able to determine the correct efficiency curve and thus may give poor results.

2. Physical Setup

There are several Cd absorbers with various holes with different sizes in the center.

3. Measurements

a. Conduct the measurements with one of the Pu standards. Tape 2 Cd absorbers with the same or different hole sizes together (with the total thickness of about 2 mm or more) with the holes aligned in a way such that the 60-keV peak is reasonable relative to the peaks in the 100-keV region. Measure the item for 100s. Record the results into the table below. Discuss the results and output messages.

b. Readjust the alignment of the 2 Cd absorbers such that the height of the 60-keV peak is increased or decreased by a factor of 1.5–2. Re-measure the item and record the results into the table. Discuss the results and output messages.

c. If time permits, conduct one or two other measurements using different Pu standards. Record the results into the table. Discuss the results and output messages.

Effects of absorber with hole on Assay Results

Source:_____	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{240}Pu$_{eff}$
DECLARED ISOTOPICS							
Hole info:							
Source:_____							
DECLARED ISOTOPICS							
Hole info:							

J. Exercise 9 - THE SLOPPY INSPECTOR PROBLEM

1. Background

A sloppy operator may not set up the measurement carefully or correctly even if he/she has the correct equipment. One such case to be discussed here is when the Cd filter does not fully cover the detector crystal and allows some of the gamma rays entering the detector unfiltered (similar to those entering the detector through the hole in the previous exercise).

2. Measurements

a. Conduct the measurements with one of the Pu standards. Use a normal Cd absorber such that the 60-keV peak is about ¼ to ½ of those in the 100-keV region when the absorber fully covers the detector.

b. Now slide the absorber to one side so that some of the gamma rays enter the detector without passing through the absorber. Adjust the absorber (and perhaps the source and detector positions) such that the 60-keV peak is about the same height as those in the 100-keV region.

c. Measure the item for 100s and record the results into the table below. Discuss the results and output messages.

Effects of Sloppy setup on Assay Results

Source:_____	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	^{240}Pu$_{eff}$
DECLARED ISOTOPICS							
Sloppy measurement							
Source:_____							
DECLARED ISOTOPICS							
Sloppy measurement							

REFERENCES

1. R. Gunnink, "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances": Vol. 1 Methods and Algorithms, Lawrence Livermore National Laboratory, UCRL-LR-103220, Vol.1.
2. R. Gunnink and W.D. Ruhter, "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances": Vol. 2 A Guide to Using MGA, Lawrence Livermore National Laboratory, UCRL-LR-103220, Vol.2 [1990].

APPENDIX 1: Important Information on Plutonium Standards

Lot ID	Ref. Date	Pu Mass (gm)	Isotopic Composition [mass fractions, %]						$^{240}Pu_{eff}$ (%)	$^{239}Pu + ^{241}Pu$
			^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am		
A1-86	1970	10.009	0.012	94.138	5.584	0.248	0.018	0.000	5.645	94.386
A1-87	1970	9.997	0.012	94.138	5.584	0.248	0.018	0.000	5.645	94.386
A1-88	1970	10.001	0.012	94.138	5.584	0.248	0.018	0.000	5.645	94.386
A1-89	1970	10.001	0.012	94.138	5.584	0.248	0.018	0.000	5.645	94.386
A1-90	1970	10.101	0.012	94.138	5.584	0.248	0.018	0.000	5.645	94.386
A1-91	1970	10.001	0.012	94.156	5.574	0.240	0.018	0.000	5.634	94.396
A1-92	1970	10.000	0.010	94.535	5.243	0.198	0.014	0.000	5.292	94.733
PEO382A	5/80	19.98	0.024	89.60	9.68	0.589	0.110	0.30	9.925	90.189
PEO382B	5/80	74.91	0.024	89.60	9.68	0.589	0.110	0.30	9.925	90.189
PEO382C	5/80	149.8	0.024	89.60	9.68	0.589	0.110	0.30	9.925	90.189
PEO382D	5/80	299.6	0.024	89.60	9.68	0.589	0.110	0.30	9.925	90.189
STDSRP12-1	11/97	874.7	0.0536	87.333	11.847	0.5448	0.2224	0.5318	12.356	87.878
STDSRP12-2	11/97	859.5	0.0536	87.333	11.847	0.5448	0.2224	0.5318	12.356	87.878
LAO250C10	09/83	60	0.058	82.16	16.18	1.27	0.334	0.124	16.89	98.34
LAO251C10	10/83	172	0.065	81.94	16.35	1.30	0.344	0.135	17.09	83.24
LAO252C10	10/83	322	0.057	82.11	16.22	1.27	0.344	0.129	16.94	83.38
CBNM Pu93	6/86	~6	0.0117	93.412	6.313	0.2235	0.0395	0.1047	6.412	93.636
CBNM Pu84	6/86	~6	0.0703	84.338	14.207	1.0275	0.3576	0.2173	14.985	85.366
CBNM Pu70	6/86	~6	0.8458	73.319	18.295	5.4634	2.0772	1.1705	23.916	78.782
CBNM Pu61	6/86	~6	1.1969	62.526	25.406	6.6793	4.1925	1.4452	35.466	69.205
PIDIE6-2	1/88	<1	0.0226	89.338	10.099	0.446	0.094	0.2651	10.31	89.78
PIDIE6-4	1/88	<1	0.109	77.80	19.71	1.827	0.560	1.554	20.925	79.627
PIDIE6-6	1/88	<1	0.935	66.466	23.834	5.247	3.519	3.738	32.10	71.713
FCZ158	1978	0.6952	0.016	0.955	93.77	0.699	4.56	0.001	101.47	1.654
Lot ID	Ref. Date	Pu Mass (g)	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{241}Am	$^{235}U/U$ %	U/Pu
HUA8971	8/89	235.6	0.059	87.092	11.796	0.845	0.209	0.638	1.022	2.657
HUA5062	8/89	242.5	0.066	87.110	11.801	0.847	0.1770	1.287	0.727	1.624
HUA5069	8/89	114.0	0.060	87.276	11.665	0.813	0.186	1.106	1.073	5.960
HUA5065	8/89	306.0	0.066	87.111	11.801	0.846	0.177	1.306	0.769	1.434
HUA5301	8/89	367.8	0.050	87.370	11.642	0.754	0.184	0.543	0.225	2.191

NOTE: ^{240}Pu effective = 2.52 x $^{238}M_f$ + $^{240}M_f$ + 1.68 x $^{242}M_f$