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7/13/81

T.S.
B5584 (1)

DR-5811

UCRL-53145
(ISPO-134)

A Portable Computer to Reduce Gamma-Ray Spectra for Plutonium Isotopic Ratios

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David C. Camp

MASTER

ISPO Task A.63
Development Report

May 15, 1981

Lawrence
Livermore
National
Laboratory

PROGRAM FOR
TECHNICAL ASSISTANCE
TO IAEA SAFEGUARDS

USA
IAEA

Department of Energy
Office of Safeguards & Security

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A PORTABLE COMPUTER TO REDUCE GAMMA-RAY SPECTRA FOR PLUTONIUM ISOTOPIC RATIOS

INTRODUCTION

In response to Task A.63 of the International Safeguards Project Office (ISPO), to upgrade measurement technology used by the International Atomic Energy Agency (IAEA), we have designed and programmed a portable data-reduction microprocessor. The unit allows in-field reduction of gamma-ray spectra and interfaces with the IAEA's multichannel analyzers--the 1000- or 2000-channel memory Silena BS27/N.

In this report, we describe the components used in assembling the microprocessor unit, i.e., the hardware; the software used to control the unit; and the mathematical formulation used to obtain isotopic ratios from the gamma-ray data. We also present a simple overview of the unit's operation and the results of tests on gamma-ray spectra that sought to verify the unit's operating characteristics and to determine the precision and effectiveness of the software developed for data reduction.

DATA-REDUCTION UNIT HARDWARE

The portable data-reduction microprocessor unit (Fig. 1) is contained in a 30.5- x 30.5- x 22.9-cm extruded aluminum case, weighs 10 kg (22 lb), and operates on either 110 V AC (60 Hz) or 220 V AC (50 Hz). It uses an LSI-11/2 microprocessor (Digital Equipment Corporation) and two memories--a programmable read-only memory (PROM) for storing operating software and a random access memory (RAM) for storing data during reduction--to give a powerful yet easy to use data-reduction system. The operator interacts with the unit through a decimal keyboard and a 20-character alphanumeric display. A 7-key function and a 10-digit keyboard (see Fig. 1) provide remote control of the Silena multichannel analyzer (MCA) and the entry of data into the unit. Communication between the unit and the Silena MCA is accomplished by a parallel interface module that transfers data at a rate of 10,000 bytes/s. Thus, a 1000-channel spectrum is transferred in less than 1 s. The hard copy data-reduction results are produced by a 20-character thermal printer.

FIG. 1. The portable data-reduction unit.

CENTRAL PROCESSOR UNIT

The LSI-11/2, the central processor unit (CPU) of the portable data-reduction microprocessor unit, is capable of addressing 32K ($K = 1024$) 16-bit words of memory. A 16-bit buffered parallel input/output LSI-11 bus with asynchronous operation allows the processor and its system components (memory and input/output peripherals) to run at the highest speed possible. An Extended and Floating-point Instruction Sets (EIS/FIS) chip provides fixed-point multiplication, division, and multiple shifting in double-precision arithmetic, as well as floating-point addition, subtraction, multiplication, and division. Thus, the LSI-11/2 is a 16-bit microcomputer with the speed and instruction set of a minicomputer. The processor board (a dual-height module) measures 14.0 x 21.6 cm. The power requirements are +5 V at 1.0 A and +12 V at 0.22 A.

MEMORY

The unit has a programmable read-only memory (PROM) for storing operating software and a random access memory (RAM) for storing data during reduction. The PROM is not volatile--it retains its contents when operating power is removed; thus, programs and data are still available when operating power is restored. The RAM, however, is volatile--when operating power is removed, its contents are lost.

A PROM is identified by the type of integrated circuit (IC) chip it uses; some PROMs use IC chips that can be programmed only once, and others, commonly referred to as erasable and programmable read-only memories (EPROM), use IC chips that can be erased by high-intensity ultraviolet light* and then reprogrammed. The data-reduction system PROM uses an INTEL 2716-type erasable IC chip that has a capacity of 1K 16-bit words. One 8K-EPROM dual-height module holds eight of these chips; so two are necessary to provide 16K of PROM memory. The power requirements for each module are +5 V at 0.5 A and +12 V at 0.02 A.

*A clear quartz window placed over the PROM IC chip allows the ultraviolet light to be directed onto the semiconductor material, thus erasing the program.

The dual-height RAM is a dynamic, high-speed, read/write memory. The programs in the PROM use this memory area to store variables during data reduction. For instance, spectral data from the MCA are stored in the RAM. The RAM module has 32K words of memory, 12K of which are accessed by the CPU. It has its own memory-select switches to configure the memory bank. The power requirements of this module are +5 V at 1.0 A and +12 V at 0.3 A.

LSI-11 COMMUNICATION MODULES

The data reduction system requires both a serial interface module and a parallel interface module. The keyboard, the alphanumeric display, and the printer communicate with the LSI-11 through serial interface channels, and the Silena MCA and the LSI-11 communicate through a parallel interface channel.

To meet the serial communication requirements, we used the dual-height DLV11-J asynchronous serial-line/interface module. This module controls four independent serial input/output channels to the LSI-11 bus, three of which are used by the data-reduction system. The interface transmits and receives data from a peripheral device over Electronics Industry Association (EIA) data-leads-only lines, making the interface RS-232C, RS-422, and RS-423 compatible. Baud rates from 150 to 38.4K can be selected for each channel. The power requirements of the board are +5 V at 1.0 A and +12 V at 0.25 A.

The Silena MCA communicates with the LSI-11 through a 64-line parallel input/output module with TTL logic. This module is capable of connecting 64 external parallel input/output lines to the LSI-11 bus. All input/output lines can be configured as either input or output lines, in increments of eight, by using plug-in components. The power requirements of this module are +5 V at 1.25 A.

This module is ideally suited for communication with the Silena MCA because of its parallel interface--the Mod. 7500-11. This unit, specifically designed for connection to a computer capable of accepting data in a parallel mode, is a circuit card that plugs into the last of the three input/output connectors in the analyzer. The interface logic signals are located at a 25-pin connector on the analyzer's rear panel. The Mod. 7500-11 is unidirectional, only transferring data from the MCA to the computer, and has a data transfer rate of 10,000 characters/s. It sends data from channels 0-1017 of the Silena memory to the computer over eight lines, character by character.

Since each character consists of 8 bits in ASCII code and each channel consists of 8 characters, 8144 characters plus 41 trailing characters are sent for each spectrum. Thus, with a data transfer rate of 10,000 characters/s, the Mod. 7500-11 can transfer the entire spectrum in less than 1 s.

Two input/output lines control the data transfer between the Silena MCA and the computer: one indicates to the computer that a character is on the data line; the other resets this data-ready line once the character has been read. Three lines signal the analyzer status to the computer--data in, data out, and display mode--and six lines transmit operation commands to the MCA from the computer--DATA IN, DATA OUT, REMOTE DATA OUT, DISPLAY, ERASE, and STOP. These commands will be discussed in more detail in the operations section of this report.

TIMING CONTROL MODULE

The data-reduction unit has a timing control module that provides the time and date. This module is a self-supporting time keeper that operates in a peripheral slot of the LSI-11 system. When the computer is off, the timing control module will continue to operate for up to three months, using power from its own rechargeable battery. It is initialized by simple commands from the microprocessor.

POWER SUPPLY, BACKPLANE, FAN, AND POWER RECEPTACLE

All of the modules and peripheral input/output devices in the data-reduction system operate on direct current (DC). Hence, we need a device that converts from alternating current (AC) of varying voltages and frequencies to direct current. A 75-W (high-efficiency) switching power supply fulfills this need. We chose to use a high-efficiency power supply instead of a low-efficiency, or linear-type, power supply because of its small volume and low weight, and its ability to operate over a wider range of input voltages and frequencies. The model installed in the data reduction system is 27.9 cm long by 13.1 cm wide by 4.3 cm high and weighs approximately 1.9 kg. It has two DC outputs: one with a rating of 5 V at 8 A and one with a rating of 12 V at 2 A. The AC input specifications for this model are 105-130- and 200-265-V AC--47-100 Hz.

A backplane configured to accept eight 14-cm-wide (dual-height) modules distributes the power to the seven circuit-board modules that operate on 5 V and 12 V DC. Completely contained in the backplane is the LSI-11 bus. The bus lines are prewired to each module location, and each module is kept firmly in a backplane connector slot by a rugged but lightweight card guide assembly. The backplane with card guide is 27.4 cm long by 14.6 cm deep by 15.2 cm high. With the seven modules installed, this assembly weighs approximately 1.8 kg.

A fused connector with voltage selection connects the switching power supply to the AC line power. This connector eliminates the need to change internal wiring for operation at different AC voltages, provides a convenient fuse holder, and lets the user change connections to the power supply with a voltage selection card. This is a safe way of changing connections to the power supply because the power line cord must be removed from the connector before the fuse or the card can be accessed. The connector also has a built-in filter that protects the equipment from radio frequency interference.

To cool the internal components of the unit, we installed a 11.9-cm-square by 3.8-cm-deep fan that delivers 115 cfm of air and operates on either 120 or 220 V AC. The fan's metal housing acts as an effective heat sink for the bearing, thereby extending its life.

INPUT/OUTPUT PERIPHERALS

A 20-character vacuum fluorescent unit displays messages sent by the LSI-11 through the DLV11-J serial interface at 1200 baud. These messages either elicit a response from the keyboard user or indicate a condition that requires corrective action. Since the flow of information is in one direction only, the LSI-11 must anticipate all transmission timing. The unit is 15.7 cm wide by 4.8 cm high by 8.9 cm deep, weighs approximately 300 g, and operates on 5 V DC with typical current requirements of 500 mA. The character line is 10.2 cm long, and each individual character is 5 mm by 3.5 mm.

A numeric keyboard terminal controls entry of data into the data-reduction program, and a seven-key function provides remote control of the Silena MCA. The terminal, which has an eight-digit display with a seven-segment LED, receives and transmits ASCII-coded data through one of the DLV11-J serial lines at 300 baud. The eight-digit display has an embedded

decimal point that does not use the eight positions and a minus sign that does. The ENTER and CLEAR keys allow verification and/or erasure of entries before they are transmitted to the LSI-11. The unit weighs only 290 g and operates on 12 V DC at a 250-mA maximum.

Hard copy printout of the data-reduction results is produced by a quiet, nonimpact thermal printer. It receives its information from the LSI-11 through one of the DVL11-J serial lines and then prints the full ASCII character set in 20 columns across 58.6-mm-wide thermal paper, at a rate of 1.2 lines/s, regardless of the number of characters per line. On a 42-m roll of paper it prints 8,400 lines of data at 2 lines/cm (approximately 100 spectral analysis outputs). The printer is 22.2 cm long by 11.2 cm wide by 6.85 high, weighs 2.0 kg, and operates on 11/220 V AC. An internal switch, accessible when the paper roll is removed, selects the voltage.

THE PORTABLE DATA-REDUCTION MICROPROCESSOR UNIT AS MINICOMPUTER

Since the portable data-reduction unit has an available input/output channel on the DLV11-J module and an extra slot in the backplane, it could be upgraded to a portable minicomputer if selected internal wiring and switch changes were made. The minicomputer would require a mass storage device (i.e., a floppy disk) and a full alphanumeric terminal. The storage device could be accommodated by inserting an interface card into the remaining backplane slot, and the terminal could be connected to the spare DLV11-J input/output channel. Setting the appropriate switches on the RAM module would provide a full 32K of RAM.

DATA ANALYSIS SOFTWARE

The objective of the data analysis software developed thus far is to determine the plutonium isotopic ratios from the gamma-ray spectral data. This requires determining the peaks appropriate to the isotopes of interest, a task that would be very simple if all the appropriate peaks were well-isolated, single peaks. Unfortunately, the analysis must include

complicated peak groupings because they provide the only information from which to deduce a particular isotopic abundance.

A careful analysis and interpretation of gamma-ray peaks includes the proper delineation of the peak shape, a reproducible description of the background continuum under the peak or peak grouping, and a method for unfolding peak multiplets and removing small interferences. The basic equation for describing peak shapes, developed by R. Gunnink,¹ is written as follows:

$$Y_i = Y_0 \times e^{-\frac{a(x_i - x_0)^2}{2}} + Ay_0 \times e^{-\frac{B(x_i - x_0)}{C}} \times \left[1 - e^{-\frac{C}{2} \times \frac{a(x_i - x_0)^2}{2}} \right]^{\delta}, (1)$$

where

Y_i = net data counts

Y_0 = peak height

a = peak width parameter

x_i = channel value of the i^{th} point

x_0 = peak position

A , B , C = shape parameters describing the tailing function

$\delta = 1$ for $x_i - x_0 < 0$

$\delta = 0$ for $x_i - x_0 > 0$.

Six parameters-- Y_0 , x_0 , a , A , B , and C --characterize a particular gamma-ray peak (see Fig. 2). The data points $Y(i)$ in an overlapping peak multiplet are then considered linear combinations of the contributions from each peak so that

$$Y(i) = \sum_{j=1}^n Y_{ji}, \quad (2)$$

where n is the number of peaks. The parameters a , A , B , and C are needed to describe the peak shape but normally are not of great interest by themselves. It simplifies the fitting process if we predetermine their values from the given spectrum and treat them as fixed rather than free parameters.

FIG. 2. A detail showing the components used to describe a peak shape.

The peak width parameter α is commonly specified by the full width at half maximum (FWHM = 2.355σ). This is related to σ by

$$\alpha = -1/2\sigma^2.$$

FWHM can be expressed in terms of peak energy as

$$[(FWHM)^2 - 0.462] \times (\text{gain})^2 = K_1 + K_2 \times E. \quad (5)$$

The values of K_1 and K_2 can be calculated by determining the FWHM of low- and high-energy peaks in a spectrum. The value 0.462 is a Sheppard's

correction for the error that results from treating all the counts in a given channel as if they were concentrated at the channel center.

The quantities A and B characterize the amplitude and slope of an exponentially rising term that describes the tailing occurring on the low-energy side of the gamma-ray peak. This term is multiplied by $[1 - e^{-Ca(x - x_0)^2}]$ to reduce its contribution to zero at the peak centroid. The quantity C is a rather insensitive parameter, and a value of 0.4 appears suitable for all germanium detector systems.

Studies have shown that slope parameter B is detector dependent only, whereas amplitude parameter A is both detector and energy dependent. A simple algorithm, given by $\ln A = C_1 + C_2 \times E$, describes the approximate relationship between parameter A and the gamma-ray energy. As in the case for α , we use two or more peaks to evaluate the constants C_1 and C_2 in this expression. Therefore, the peak shapes for a given spectrum can be predetermined from two peaks and specified in terms of six parameters: K_1 and K_2 (used to determine α), C_1 and C_2 (used to calculate A), and B.

For this particular application, we use the 148.6-keV peak of ^{241}Pu and the 208-keV peak of ^{237}U . Usually, these two peaks are the most intense peaks in the spectrum, especially for high burnup plutonium. Although the tail amplitude parameter A is generally energy dependent, we must also consider poor 148.6-keV peak statistics, or poor quality spectra, or both. Good statistical significance is required to determine A accurately, and in most cases, the spectra encountered will not be statistically good. The 148.6-keV peak also has interferences on the low-energy side, which increases the error in establishing the energy dependence of A. Consequently, the code determines the tail parameters A and B from the 208-keV peak and treats A as constant for the spectrum.

The 148.6-keV and 208-keV peaks are also used as reference peaks for determining the gain and the zero-intercept of the spectrum. Using this information (assuming that the conversion gain is linear), we can compute the exact positions of the peaks to be analyzed.

If the value of x_0 in Eq. 1 is now also known (in addition to the other shape parameters), the set of equations in Eq. 2 will no longer contain unknowns in the exponents. This simplifies the software since the equations can be solved simply by the linear least-squares method rather than by an iterative technique. The only free parameters remaining in the equations are

the peak heights. However, not all of the peak heights in a group are free parameters since all peaks belonging to a given isotopic component are fixed by a prior calibration (i.e., the branching intensities in their decay schemes have been determined). Therefore, we can compute a response envelope for each isotopic component rather than for each peak and thereby reduce the number of unknowns to the number of components. This occurs before any correction for sample geometry or overall efficiency is made.

Before we can determine the net area of a peak, the background beneath the peak or peak grouping must be removed. This is usually done by interpolating the background, using a straight line or a polynomial function. However, such a procedure is, in principle, incorrect and may lead to erroneous results. Thus, we use the following explicit function to interpolate background values beneath the peak grouping¹:

$$B_i = b_n + (b_m - b_n) \times \left(\sum_{j=n}^{j=i} Y_j \right) \times \left(\sum_{k=n}^{k=m} Y_k \right)^{-1}, \quad (4)$$

where

B_i = computed background at channel i

Y_i = spectrum count of channel i

b_n = average background level of the low-energy side of the peak

b_m = average background level of the high-energy side of the peak.

This function produces smoothed background steps that occur at each peak position. It works equally well for single peaks and complex peak groupings.

Typically, the gamma-ray spectra taken by the IAEA inspectors are in the 110-225-keV region or the 110-450-keV region, depending on the size of the detector. The data-reduction software analyzes four peak groupings in the 110-225-keV region. Two others, the 330-keV peak grouping and 370-keV peak grouping, are used in the analysis if the gain is set to include these peaks in the spectrum. The six peak groupings are shown in Table 1.

The data analysis software analyzes each peak grouping separately. First, the background distribution is removed from the grouping, and then a response function for each isotope contributing to the grouping is generated. The response functions are calculated using peak shape and branching intensity information. This approach yields a set of linear equations whose number of

TABLE 1. Peak groupings analyzed by the data-reduction software.

Isotope	Energy (keV)	Intensity	Isotope	Energy (keV)	Intensity
<u>Group 1</u>			<u>Group 2</u>		
^{239}Pu	122.994	1.00×10^{-5}	^{239}Pu	141.567	3.2×10^{-7}
^{239}Pu	123.62	1.97×10^{-7}	^{239}Pu	144.211	2.83×10^{-6}
^{239}Pu	124.51	6.13×10^{-7}	^{239}Pu	146.077	1.19×10^{-6}
^{239}Pu	125.21	7.11×10^{-7}	^{241}Am	146.557	4.61×10^{-6}
^{239}Am	125.292*	4.08×10^{-5}	^{241}Pu	148.567*	1.87×10^{-6}
^{239}Pu	129.294*	6.26×10^{-5}	^{241}Am	150.11	7.40×10^{-7}
<u>Group 3</u>			<u>Group 4</u>		
^{241}Pu	159.955	6.74×10^{-8}	^{239}Pu	203.537*	5.60×10^{-6}
^{239}Pu	160.19	6.20×10^{-8}	$^{241}\text{Pu}-^{237}\text{U}$	208.0*	5.33×10^{-6}
^{240}Pu	160.28	4.02×10^{-6}	^{241}Am	208.0	7.91×10^{-6}
^{239}Pu	161.45	1.20×10^{-6}			
$^{241}\text{Pu}-^{237}\text{U}$	164.58*	4.53×10^{-7}			
^{241}Am	164.58	0.67×10^{-7}			
^{241}Am	165.93	2.32×10^{-7}			
<u>Group 5</u>			<u>Group 6</u>		
$^{241}\text{Pu}-^{237}\text{U}$	332.401	2.98×10^{-7}	^{239}Pu	367.050	8.65×10^{-7}
^{241}Am	332.401	1.49×10^{-6}	^{239}Pu	368.546	9.03×10^{-7}
^{239}Pu	332.853	5.06×10^{-6}	$^{241}\text{Pu}-^{237}\text{U}$	368.603	1.05×10^{-8}
$^{241}\text{Pu}-^{237}\text{U}$	335.438	2.39×10^{-8}	^{241}Am	368.603	2.17×10^{-9}
^{241}Am	335.438	4.96×10^{-6}	$^{241}\text{Pu}-^{237}\text{U}$	370.933	2.71×10^{-8}
^{239}Pu	336.104	1.134×10^{-6}	^{241}Am	370.933	5.23×10^{-7}
^{239}Pu	341.522	6.62×10^{-7}	^{239}Pu	375.042*	1.57×10^{-5}
^{239}Pu	345.011*	5.59×10^{-6}	^{241}Am	376.595	1.38×10^{-9}
			^{239}Pu	380.166	3.03×10^{-6}
			^{239}Pu	382.751	2.59×10^{-6}

*Indicates that a peak is used for determining the relative efficiency curve. The ^{238}Pu 152.68-keV gamma-ray peak and the ^{235}U 185.72-keV peak are analyzed individually.

unknowns is equal to the number of isotopic components. Solving for these unknowns requires the inversion of a matrix that yields the necessary peak-height or area information.

The objective of the data analysis is to interpret the spectral data in terms of isotopic ratios. We cannot use a direct calibration since there is no uniformity among the samples or the IAEA's detectors. Therefore, we must consider the influence of counting efficiency and gamma-ray attenuation (due to the sample matrix or other absorbing materials) on gamma-ray peak

intensities. To minimize these factors, we select sets of neighboring gamma-ray peaks from a spectrum emitted by different isotopes and then calculate the ratio of isotopic abundances from the gamma-ray intensities of the selected sets, using the following equation:

$$A/B = \frac{I_1 T_1}{\epsilon_1 X_1 Z_1} \bigg/ \frac{I_2 T_2}{\epsilon_2 X_2 Z_2}, \quad (5)$$

where

A/B = isotope ratio of interest

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

T_1, T_2 = half-lives of precursors

ϵ_1, ϵ_2 = detector efficiency for the respective gamma rays

X_1, X_2 = attenuation factors for the respective gamma rays

Z_1, Z_2 = branching intensities or gamma yields of the respective gamma rays.

The branching ratio Z and the half-life T are factors characteristic of radioactive decay, and in most cases are known. For the plutonium isotopes, we used the gamma-ray intensities published by R. Gunning et al.² The half-lives used for the plutonium isotopes and the americium-241 isotopes are those recommended in ANSI B.22. The influence of efficiency and attenuation, physical factors that are functions of energy, is minimized by taking ratios of gamma-ray peak intensities for pairs of gamma rays that are nearly equal in energy. If the energies are close, attenuation and efficiency differences are small, and the factors essentially cancel each other.

However, a convenient pair of peaks for a particular isotopic ratio is not always available and does not use all the information available in the spectral data. To solve this problem we determine the energy dependence of the overall efficiency for the gamma rays emitted by the sample. This includes the energy dependence of the gamma-ray self-attenuation in the sample itself, the attenuation factors in any other materials between the sample and the detector efficiency, and all geometric factors.

A self-calibration technique is used to obtain the necessary relative efficiency corrections from peak areas in each spectrum measured. Areas for

^{239}Pu , ^{241}Pu , and ^{241}Am (peaks denoted by asterisks in Table 1) are corrected for gamma-ray branching intensities and fitted to the functional form

$$\ln \left(\frac{I_i}{Z_i} \right) = a_0 + a_1 \delta_1 + a_2 \delta_2 + \sum_{j=3}^{2 \text{ or } 3} \left[a_{j+2} (\ln E_i)^j \right], \quad (6)$$

using a weighted least-squares technique. The value E_i is the energy of the peak in mega electron volts. The coefficients a_1 and a_2 normalize the ^{241}Pu and ^{241}Am data points to the ^{239}Pu data points and obtain the $^{241}\text{Pu}/^{239}\text{Pu}$ and $^{241}\text{Am}/^{241}\text{Pu}$ isotopic ratios. If the 330- and 370-keV peak groupings are included in the spectrum, we use a cubic function to fit the data (the sum of j is to 3); when only the first four peak groupings are included, we use a quadratic function. For a complete analysis, the indicated peaks of these four groupings must be in the spectrum.

We determine the $^{238}\text{Pu}/^{239}\text{Pu}$ isotopic ratio and the $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio from the ^{238}Pu 152.68-keV gamma ray and the ^{240}Pu 160.28-keV gamma ray after correcting their peak areas for branching intensity and relative detection efficiency. If ^{235}U is present in the sample, the $^{235}\text{U}/^{239}\text{Pu}$ isotopic ratio is determined in the same way, using the ^{235}U 135.72-keV gamma-ray peak. The presence of ^{235}U is established by determining whether there is any statistically significant data in the 185.72-keV region.

Several fits of the relative efficiency data to Eq. 6 are required to determine the a_j coefficients because the 164.58- and 208-keV peak gamma-ray areas must be corrected for contributions by ^{241}Am . These corrections cannot be made in a rigorous manner since they require previous knowledge of the $^{241}\text{Am}/^{241}\text{Pu}$ isotopic ratio. Thus, we obtain an approximate answer for this ratio after the first fit, make corrections to the peak areas, and repeat the fit. This iterative procedure continues until there is no statistically significant change in the results of the fit. It works because the corrections are generally never more than 7%, even for a 16-year-old sample abundant in ^{241}Am .

The errors for the isotopic ratios are determined from the errors for the a_j coefficients and from the goodness of fit. The error values assigned to the coefficients are determined from the inverted matrix of the weighted equations used in the least-squares analysis. The equations are weighted

according to the statistical errors on the peaks used to determine the relative efficiency. In this particular case, we found that the off-diagonal matrix elements of the inverted matrix were not insignificant and had to be included in the error determination.

The calculated isotopic ratios and total errors (statistical plus efficiency plus fitting) are printed out by the thermal printer. The printout also includes the age of the sample since separation from ^{241}Am . This age is calculated on the basis of the $^{241}\text{Am}/^{241}\text{Pu}$ isotopic ratio, using the standard parent-daughter relationship. A reproduction of a typical printout is shown in Fig. 3.

OPERATION INSTRUCTIONS

In this section we provide brief, step-by-step instructions for using the data-reduction microprocessor unit to produce isotopic ratio results from spectral data. (A more comprehensive set of instructions is provided in the unit's operations manual.) We assume that the user is familiar with the Silena MCA and that the Silena MCA is operationally functional, adjusted for the detector being used, and has the correct gain calibration. The accuracy of the final isotopic abundance ratios is directly dependent on the quality of the data. Good results cannot be obtained from poor spectral data.

OPERATING STEPS

First Step

Check the voltage selection card and fuse in the power receptacle. The number on the left-hand side of the voltage selection card should match the available AC voltage. If it does not match, change the voltage selection and the fuse. Pull the small lever marked FUSE PULL outward to force the fuse out of its holder. Remove the voltage selection card and orient the PC board so that it shows the desired voltage on the top left side of the card. Re-insert the card. The contact strips on the card energize the proper terminals on the power supply. Return the fuse lever to normal position and install the correct fuse. Slide the transparent cover back over the voltage selection

TYPICAL DATA ANALYSIS PRINTOUT RESULTS COVERING THE 120- TO 220-keV REGION

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7:59 15/1/81
SPEC # 14; LT = 1000 s

GAIN = 0.1064 keV/CH

CHNL ZERO E = 109.457

FWHM (208 keV) = 0.856

152-keV peak area =
7583 ± 4.4%

186-keV peak area =
0 ± 0.0%

238PU/239PU = 0.002129
± 0.000098

240PU/239PU = 0.189621
± 0.006330

241PU/239PU = 0.024388
± 0.000300

241AM/241PU = 0.083424
± 0.002162

235U/239PU = 0.000000
± 0.000000

Time since Sep. =
606.41 ± 15.12 days

Peak En. Counts %ER
122.93 1589 6.4
123.62 270 Fixd
124.51 854 Fixd
125.21 794 Fixd
125.26 6301 2.0
129.29 86634 0.4

Peak En. Counts %ER
141.57 545 15.4
144.21 4434 2.2
146.08 1868 Fixd
146.56 1237 7.8
148.56 106729 0.3
150.11 200 Fixd

Peak En. Counts %ER
159.96 3591 Fixd
160.19 79 Fixd
160.28 3822 3.0
161.45 1524 5.5
164.58 25468 0.1
165.93 127 59.2

Peak En. Counts %ER
203.54 6180 1.6
208.00 234120 0.2
    
```

FIG. 3. Reproduction of a printout produced by the 20-column thermal printer.

card and the fuse. Plug the line cord into the power receptacle and then plug the line cord into the AC power source. DO NOT TURN ON POWER YET.

Second Step

The printer also has a voltage selection switch that must be set correctly before the power can be turned on. To set this switch, withdraw the front panel assembly of the printer just as it is withdrawn when the paper roll is changed (see operations manual). Inside the printer housing, on the second printed circuit card from the left-hand side, is a blue switch mechanism labeled 110V 220V. Set the switch to the available voltage. No fuse changes are required.

While the front panel assembly of the printer is out, check the quantity of paper on the paper roll. Approximately 30 cm of paper are used for a data analysis printout. The operations manual provides detailed instructions for changing the paper roll.

Third Step

Connect the flat ribbon cable to the data-reduction unit and to the Silena MCA. Do this without power applied to either unit to prevent accidental damage to components on either interface board. Since the connectors are keyed, there is no confusion about how the connection is made.

Fourth Step

Switch the power ON to both units, but to the data-reduction unit first: the Silena MCA will NOT function properly when connected to the data-reduction unit unless the power to the data-reduction unit is on. The Silena MCA can now be operated in the normal manner by using the pushbuttons on its front panel or the identically labeled function keys on the data-reduction keyboard. The function keys are activated by pressing a key to activate the function keyboard light and then pressing the ENTER key.

When the function keys are controlling the MCA, the pushbuttons become inoperative, but they can be reactivated by pressing the RESTART switch on the keyboard. The RESTART switch releases the microprocessor unit's control of the MCA and causes the time and date to be displayed. If an error is made in the sequence of commands to the MCA, the message COMMAND ERROR is flashed on the display. An example of such an error would be issuing the DATA IN command while the MCA is in DISPLAY. The MCA must be in STOP before the DATA IN command can be issued.

Fifth Step

When the power is turned on to the microprocessor unit or the RESTART switch is pressed, the time and date are displayed. To correct the time or the date, press the INIT key and the ENTER key consecutively. This activates the software, which prompts the user for the correct time and date and sets the internal clock board. The operations manual provides more detailed instructions for entering the time and the date.

Sixth Step

Data must be in the analyzer memory before the analysis of a spectrum can begin. Before transferring the data in the Silena MCA memory to the data-reduction unit, make sure that the analyzer is in the display mode. Then press the START key and the ENTER key. When this command is given, the analyzer will go to the DATA OUT mode and back to DISPLAY in less than a second. When the analyzer is in the DATA OUT mode, data are being transferred from the Silena MCA memory to the data-reduction unit memory. When the transfer is complete, the time and date shown on the display are printed. The spectrum number and livetime are also printed automatically if they are passed from the analyzer during the data transfer. If this information is not transferred--which is possible since not all Silena MCA models have PROM IC's programmed to transfer spectrum number and livetime--the user is prompted and he enters it on the data-reduction keyboard. (Instructions for this type of entry are given in the operations manual.)

Seventh Step

Once the data transfer is complete, the message "148 PEAK POS" will appear on the alphanumeric display. Determine the nearest channel address corresponding to the peak channel position of the ^{241}Pu 148.6-keV gamma-ray peak and enter the channel number value on the numeric keyboard. If the value shown on the keyboard display is incorrect, press the CLEAR key and enter the correct value. The entered value will appear on the alphanumeric display for approximately 5 s, and then the message "208 PEAK POS" will appear. Enter the nearest channel address of the ^{237}U 208-keV gamma-ray peak. The entered value will appear on the alphanumeric display. The software checks the entered peak positions for validity. If all requirements are met, the program continues with the analysis of the spectral data, and the results are printed out. The end of the analysis is indicated by a display of the current time and date.

Eighth Step

You may turn off power to both units in either order--data reduction unit first and MCA second or MCA first and data-reduction unit second. However, if you are only turning off power to the data-reduction unit, you must disconnect it from the MCA if the MCA is to operate normally. We recommend that power to the data-reduction unit not be switched off intermittently. Such action may result in damage to the switching power supply.

ERROR DIAGNOSTICS

We have incorporated several checks into the software to detect situations that might cause errors or poor results. If certain conditions are not met by the spectral data or the information entered by the user, the following error messages are flashed on the alphanumeric display.

BAD PEAK POSITION. If statistically significant data do not exist near the peak positions indicated by the user in response to the peak-position

queries, the error message BAD PEAK POSITION is flashed on the display. This indicates that either an incorrect value was entered for a peak position or no data were transferred from the analyzer to the microprocessor. Instead of continuing, the program returns to the beginning and waits for the user to take corrective action and restart the analysis.

BAD ZERO; CHECK POS. If statistically significant peaks are found, the software proceeds to determine more accurate peak positions from which the gain and channel-zero energy of the spectrum are calculated. The results, along with the FWHM, are printed out, and the gain and zero are checked to ensure that the spectral information required for the isotopic analysis--i.e., the first four peak groupings--is included in the spectrum. If the channel-zero energy is greater than 121.5 keV, the message BAD ZERO; CHECK POS is flashed on the display. This indicates that channel-zero energy precludes analysis of the 120-keV peak grouping, which is required by the software. This condition may exist because of an incorrect peak position entry or an improper energy calibration of the amplifier/analyzer.

BAD GAIN; CHECK POS. If the product of the spectrum gain and maximum channel in the spectrum (1000 or 2000, depending on the analyzer) is less than 210 keV, the message BAD GAIN; CHECK POS is displayed. The microprocessor software cannot analyze the spectrum if the 208-keV peak region is not included. So, again, either the correct peak position must be entered or the spectrum retaken with the proper energy calibration of the amplifier/analyzer.

CHECK PEAKS. Another error may occur in determining the FWHM parameter as a function of energy, expressed by Eq. 3. If the peak shape analysis results in a value of K_1 less than 0, the message CHECK PEAKS is flashed on the display. This error occurs if the 148.6- and 208-keV calibration peaks have very poor counting statistics, if the incorrect peak positions have been entered, or if either peak has a channel that has overflowed. Channel overflow is most likely in the 208-keV peak and results in a

distorted peak shape. When a channel overflows, a spectrum cannot be analyzed. Since we presently have no correction for overflow, the user must guard against it.

POOR RESOLUTION. If the FWHM of the 208-keV peak exceeds 1.15 keV, the message POOR RESOLUTION is flashed on the display. This warns the user that the spectral resolution is not good and that the results are questionable. Although the data will be analyzed, the user should take action to improve the energy resolution. Such action might involve reducing the detector count rate or adjusting the amplifier pole and shaping the time constant (see the Silena operations manual for more details). An energy resolution of 1.15 keV at the 208-keV peak is a reasonable value for a large coaxial germanium detector; however, for a smaller planar detector, an energy resolution of 0.8 keV can be attained. Since the software is written for all types of germanium detectors, we use an upper limit energy resolution of 1.15 keV at the 208-keV peak for all spectra.

DATA ANALYSIS RESULTS

We assessed the performance of the data reduction unit with spectra of NBS plutonium isotopic reference materials--NBS-SRM 946, 947, and 948--and with spectra taken by the IAEA. Both sets of data were taken with a Silena MCA and a 13-cm planar HPGe detector.

Our measurements included four spectra of each NBS standard: two taken with a conversion gain of 0.11 keV/channel, for livetimes of 2,000 and 20,000 s, and two taken with the same set of livetimes and a conversion gain of 0.34 keV/channel. The intent of these measurements was to obtain a better insight into the variation of results caused by the peak area analysis algorithm and the correction for detection efficiency. All spectra were collected with analyzer deadtimes of less than 20% to reduce the effects of a high count rate. Figure 4 shows a 20,000-s spectrum of NBS sample 947 taken with our Silena system.

FIG. 4. The 110- to 210-keV gamma-ray spectral region of NBS 947 taken with a Silena BS 27/N analyzer.

The results obtained for NBS standards 946, 947, and 948, for each of the three isotopic ratios— $^{238}\text{Pu}/^{239}\text{Pu}$, $^{240}\text{Pu}/^{239}\text{Pu}$, and $^{241}\text{Pu}/^{239}\text{Pu}$ —are plotted in Figs. 5, 6, and 7. The ordinate values for each plot are the isotopic ratios obtained by the data-reduction unit (designated by IAEAPU) divided by the isotopic ratio calculated from the updated NBS reference values. The half-lives used in the gamma-ray spectrum analyses were used to update the NBS isotopic abundances. At the time of measurement these were:

	NBS 946	NBS 947	NBS 948
$\%^{238}\text{Pu}$	0.23	0.28	0.01
$\%^{239}\text{Pu}$	84.32	76.94	91.72
$\%^{240}\text{Pu}$	12.23	18.58	7.92
$\%^{241}\text{Pu}$	2.64	3.00	0.32
$\%^{242}\text{Pu}$	0.58	1.20	0.03

The first two data points for each sample are from the spectra with the lower conversion gain. The first data point of each pair is from the spectrum with the shorter livetime accumulation.

The scatter of the data for the $^{238}\text{Pu}/^{239}\text{Pu}$ isotopic ratio results is relatively small from samples NBS 946 and NBS 947, with standard deviations ranging between 1.9 and 2.3%. The larger variation in the results, obtained

FIG. 5. $^{238}\text{Pu}/^{239}\text{Pu}$ isotopic ratio results obtained with the data-reduction unit (IAEAPU) divided by NBS values.

FIG. 6. $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio results obtained with the data-reduction unit (IAEAPU) divided by NBS values.

for sample NBS 948, is mainly due to the poor counting statistics of the 152-keV gamma-ray peak of ^{238}Pu , an isotope that has a very low abundance in NBS 948. (Our initial results for the $^{238}\text{Pu}/^{239}\text{Pu}$ ratio showed a bias of approximately 2.0%. The results shown here reflect a reduction in the ^{238}Pu 152-keV gamma-ray branching ratio of 2.0%, from 9.56×10^{-6} to 9.32×10^{-6} . This discrepancy is also mentioned in the ESARDA study³ and the Mound Laboratory work.⁴)

The $^{240}\text{Pu}/^{239}\text{Pu}$ ratio is obtained by analyzing the complex multiplet at 160-keV. This multiplet consists of a peak at 159.96 keV from ^{241}Pu , a peak at 160.19 keV from ^{239}Pu , and the ^{240}Pu peak at 160.28 keV. Since the energy separation of the ^{241}Pu peak and the ^{240}Pu peak is less than the energy resolution of the detector at this energy, fitting each peak separately, with many free parameters, can produce poor results. The method of generating isotopic component response functions described earlier reduces the number of free parameters to the number of isotopic components. The

FIG. 7. $^{241}\text{Pu}/^{239}\text{Pu}$ isotopic ratio results obtained with the data-reduction unit (IAEAPU) divided by NBS values.

intensity and position of the 159.96-keV peak is fixed to the 164.58-keV peak and the two ^{239}Pu peaks are fixed to each other. In the case of the 164.58- and the 159.96-keV peaks, we apply a small correction to the 164.58-keV peak intensity for the ^{241}Am contribution. For older NBS standards, this correction is slightly less than 7%. The standard deviations of the results ranged from 1.4 to 3.0%.

The results obtained for the $^{241}\text{Pu}/^{239}\text{Pu}$ isotopic ratio are in excellent agreement with the NBS reference values. This ratio is determined primarily by the ^{239}Pu 203.5-keV and ^{237}U 208-keV peak pair. The observed scatter in the results arises mostly from variations in the determination of the 203.5-keV peak area and from the correction of the 208-keV peak for the contribution from ^{241}Am . Errors from efficiency correction are small compared with those from determining peak areas for this pair of gamma rays.

The generally good agreement obtained with NBS reference values lends confidence to the spectral data analysis method used by the data-reduction unit. There does not appear to be any systematic error in the method for

determining peak areas since both positive and negative biases are observed for different isotopic ratios. The results suggest that small corrections, on the order of 2-3%, are required in published values for the branching intensities of the ^{238}Pu 152-keV gamma-ray peak.

The IAEA provided us with 14 gamma-ray spectra, "expected" isotopic percentages, and isotopic results it had obtained using codes on the ND6620 system (see Appendix). The 14 spectra were taken with data accumulation livetimes ranging from 1,000 to 4,000 s. The conversion gains for these spectra were 0.16-keV/channel or 0.34 keV/channel. The observed count rates produced analyzer deadtimes from 11 to 25%. Figure 8 is a plot of one of these spectra. Note that the effects of pileup are most noticeable on the

FIG. 8. The 108- to 260-keV gamma-ray spectral region of a plutonium sample taken by the IAEA.

high-energy side of the 208-keV peak. A considerable amount of forward scattering, indicated by the long up-sloping plateaus below each peak grouping, is also evident in these spectra. Both these effects increase the uncertainty in the determination of the background.

The results obtained for the $^{238}\text{Pu}/^{239}\text{Pu}$, $^{240}\text{Pu}/^{239}\text{Pu}$, and $^{241}\text{Pu}/^{239}\text{Pu}$ isotopic ratios using the data-reduction unit and codes on the ND6620 system are plotted in Figs. 9, 10, and 11. The ordinate values for each plot are the isotopic ratios obtained from the data-reduction unit divided by the "expected" isotopic ratio results. The IAEA's "expected" isotopic abundances were:

	<u>Group I</u>	<u>Group II</u>	<u>Group III</u>
% ^{238}Pu	0.140	0.1335	1.378
% ^{239}Pu	75.34	75.24	57.68
% ^{240}Pu	20.947	20.93	24.44
% ^{241}Pu	2.908	3.906	11.29

FIG. 9. $^{238}\text{Pu}/^{239}\text{Pu}$ isotopic ratio results obtained with the data-reduction unit (IAeAPU) (●) and with ND6620 codes (○) divided by IAEA "expected" results.

FIG. 10. $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio results obtained with the data-reduction unit (IAEAPU) (●) and with ND6620 codes (○) divided by the IAEA "expected" results.

There is considerable scatter in the data-reduction unit results as well as in the ND6620 results. In general, our results are in agreement with the ND6620 results, which leads one to believe that the observed differences are due to the quality of the acquired data or to incorrect "expected" isotopic percentages but not to the method of spectral analysis. The only really significant bias between our results and the ND6620 results is the $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio for spectra 14 and 15. These spectra are of sample III--the higher burnup material. It is not clear what causes the difference. Since both of these spectra were taken at rather high count rates (25% deadtime), one possibility might be the pileup of 159.96-keV events into the 160.28-keV region.

Comparison of the results obtained from spectra taken with a conversion gain of 0.34 keV/channel and spectra taken with a conversion gain of 0.16 keV/channel indicates that little or no information is gained by including the 330-keV and 370-keV regions. In most cases the errors arising from the

results are greater because poorer counting statistics in the 300-keV region make the determination of the relative efficiency curve less reliable.

If the 300-keV region is chosen, then more attention should be given to the detector-absorber arrangement to improve the counting statistics at these energies. But even if this is done, longer counting times will be required. With the iterative method, used in the data-reduction unit for determining the relative efficiency curve, convergence is less likely if the ^{241}Pu gamma rays in the 300-keV region are included because significant ^{241}Am contribution must be subtracted from the ^{241}Pu peaks. We recommend that gamma-ray spectra extending through the 300-keV region not be taken.

If these 14 spectra are typical of the in-the-field spectra taken by IAEA inspectors, then our results indicate that the portable microprocessor unit will perform as well in the field as the ND6620 system does at its home base. Of course, the unit still requires extensive laboratory testing, and perhaps field trials, before we can make a complete evaluation of its performance. The unit was developed not only to reduce data in the field but also to aid

FIG. 11. $^{241}\text{Pu}/^{239}\text{Pu}$ isotopic ratio results obtained with the data-reduction unit (IAEAPU) (●) and with ND6620 codes (o) divided by IAEA "expected" results.

the user in taking better quality spectra. It was built to be flexible for future use: we could easily upgrade its hardware and software for additional calculations and use it as the central processing unit of a lightweight minicomputer system.

ACKNOWLEDGMENTS

We acknowledge the contributions of M. DeCarolis, D. Rundquist, G. Nelson, A. Ranaivo, and others, of the IAEA, for their assistance in this task. We thank J. Skalyo, Jr., of ISPO, Brookhaven, for his advice and guidance during the course of this task. We thank K. Dewitt, R. Gunnink, and A. Prindle, of Lawrence Livermore National Laboratory, for their assistance and advice in this work.

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APPENDIX

This appendix contains the isotopic results provided by the IAEA. They are listed with the errors obtained using the ND6620 system and its spectral analysis software. The data are in the form of isotopic ratios (open circles) in Figs. 9, 10, and 11. The results given in parentheses are the "expected" results. Each numbered spectrum was sent to LLNL and analyzed by the IAEAPU code. The filled-in circles in Figs. 9, 10, and 11 indicate the $^{238}\text{Pu}/^{239}\text{Pu}$, $^{240}\text{Pu}/^{239}\text{Pu}$, and $^{241}\text{Pu}/^{239}\text{Pu}$ isotopic ratios.

TABLE A1. Isotopic composition of samples.

Spectrum number	Sample number	keV/ channel	Dead- time	% ²³⁸ Pu	% ²³⁹ Pu	% ²⁴⁰ Pu	% ²⁴¹ Pu	Ratio ²⁴¹ Am/ ²⁴¹ Pu	Age (y)
1		.148	16%	.130 ± .005 (.140)	75.2 ± 0.7 (75.34)	21.1 ± 0.7 (20.947)	2.85 ± 0.04 (2.908)	0.240 ± 0.006	4.5 ± 0.1
2		.148	17%	.126 ± 0.005 (.140)	74.4 ± 0.6 (75.34)	21.9 ± 0.6 (20.947)	2.92 ± 0.04 (2.908)	0.230 ± 0.007	4.3 ± 0.1
3		.148	20%	.122 ± .007 (.140)	75.1 ± 1.1 (75.34)	21.3 ± 1.1 (20.947)	2.88 ± 0.06 (2.908)	0.224 ± 0.014	4.2 ± 0.2
4		.161	11%	.133 ± .006 (.140)	76.1 ± 0.7 (75.34)	20.2 ± 0.7 (20.947)	2.87 ± 0.03 (2.908)	0.250 ± 0.006	4.6 ± 0.1
5		.337	19%	.120 ± .008 (.140)	76.6 ± 1.3 (75.94)	19.9 ± 1.3 (20.947)	2.73 ± 0.08 (2.908)	0.264 ± 0.013	4.9 ± 0.2
6		.337	20%	.127 ± .007 (.140)	74.9 ± 1.4 (75.34)	21.5 ± 1.4 (20.947)	2.80 ± 0.08 (2.908)	0.264 ± 0.014	4.4 ± 0.2
7		.337	20%	.108 ± .008 (.140)	74.0 ± 1.3 (75.34)	22.4 ± 1.3 (20.947)	2.84 ± 0.09 (2.908)	0.29 ± 0.09	5.3 ± 0.3
9		.337	20%	.135 ± .011 (.140)	72.4 ± 1.5 (75.34)	24.1 ± 1.5 (20.947)	2.66 ± 0.09 (2.908)	0.235 ± 0.011	4.4 ± 0.2
10	a	.337	20%	.132 ± .005 (.1335)	76.3 ± 0.8 (75.24)	19.9 ± 0.8 (20.93)	3.00 ± 0.06 (3.006)	0.26 ± 0.01	4.8 ± 0.2

TABLE A1. Continued.

Spectrum number	Sample number	keV/ channel	Dead- time	% ²³⁸ Pu	% ²³⁹ Pu	% ²⁴⁰ Pu	% ²⁴¹ Pu	Ratio ²⁴¹ Am/ ²⁴¹ Pu	Age (y)
11	b	.161	22%	.129 ± .005 (.1335)	75.9 ± 0.7 (75.24)	20.5 ± 0.7 (20.93)	2.83 ± .05 (3.009)	0.37 ± .03 ^c --	-- --
12		.161	22%	.135 ± .005 (.140)	75.9 ± 0.8 (75.34)	20.6 ± 0.7 (20.947)	2.85 ± 0.03 (2.908)	0.252 ± 0.004 --	4.7 ± 0.1 --
13		.161	19%	.129 ± .005 (.140)	75.9 ± 0.7 (75.34)	20.6 ± 0.7 (20.947)	2.79 ± 0.03 (2.908)	0.254 ± 0.004 --	4.7 ± 0.1 --
14		.161	25%	1.33 ± .10 (1.378)	58.2 ± 4.3 (57.68)	24.5 ± 5.1 (24.44)	10.8 ± 0.8 (11.29)	0.086 ± 0.005	1.7 ± 0.1
15		.343	25%	1.51 ± .07 (1.378)	57.9 ± 2.4 (57.68)	22.6 ± 2.9 (24.44)	12.3 ± 0.5 (11.29)	0.068 ± 0.004	1.4 ± 0.1
16		.343	25%	.132 ± .06 (.1335)	76.3 ± 0.6 (75.24)	19.9 ± 0.6 (20.93)	2.92 ± 0.05 (3.009)	0.244 ± 0.010	4.5 ± 0.2

^aUranium-235/total Pu ratio measured to be .024 ± .005 (.017), without correction for the 184.55-keV and 188.23-keV gamma rays of ²³⁹Pu, which may interfere with the 185.718-keV gamma-ray peak of ²³⁵U.

^bUranium-235/total Pu ratio measured to be .031 ± .005 (.017), without correction for the 184.55-keV and 188.23-keV gamma rays of ²³⁹Pu, which may interfere with the 185.718-keV gamma-ray peak of ²³⁵U.

^cThis value is doubtful because the 125-keV peak was not available for analysis. Thus, the ²⁴¹Am analysis is based primarily on the ²⁴¹Am 146.557-keV gamma ray, which appears as a single peak with the 146.077-keV gamma ray of ²³⁹Pu.