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ANALYSIS OF PLUTONIUM GAMMA-RAY SPECTRA

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A PORTABLE MICROCOMPUTER UNIT FOR THE ANALYSIS OF PLUTONIUM GAMMA-RAY SPECTRA*

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ABSTRACT

A portable microcomputer has been developed for the LAEA to perform in-field analysis of plutonium gamma-ray spectra. The unit includes a 16-bit LSI-11/2 microprocessor, 32K words of memory, a 20-character display for user prompting, and a 20-character thermal printer for hardcopy output. Only the positions of the 148-keV Pu-241 and 208-keV U-237 peaks are required for spectral analysis. The unit was tested against gamma-ray spectra taken of NBS plutonium standards and IAEA spectra. Results obtained are presented.

INTRODUCTION

We have developed a portable microprocessor-based data-analysis unit that provides IAEA inspectors with plutonium isotopic ratios from gamma-ray spectra taken in the field. Up to now IAEA inspectors have taken gamma-ray spectra with a portable Silena multichannel analyzer (MCA) and recorded the data on cassette tapes that are later processed at IAEA Headquarters. The spectral analysis problem requires sophisticated data analysis techniques, but considering the practical aspects of inspector use the unit must be portable and simple to operate.

The portable unit (Fig. 1) measures 30.5 x 30.5 x 22.9 cm, weighs 10 kg (22 lb), and operates on either 110 V AC (60 Hz) or 220 V AC (50 Hz). The unit is based on a 16-bit LSI-11/2 microprocessor and two types of memory — a programmable read-only memory (PROM) for storing operating software and a random access memory (RAM) for storing data during analysis — to give a powerful yet easy to use data-reduction system. All components of the portable microcomputer unit are commercially produced.

Fig. 1. The portable data-reduction unit.

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 MCA and the entry of data into the unit. Data transfer between the unit and the Silena occurs via a parallel interface module at a rate of 10,000 bytes/s. Hard copy results are produced by a 20-character thermal printer.

In this paper, we describe each major hardware component and the mathematical formulation used to obtain isotopic ratios from the spectra. We also present results of tests on spectra that indicate the precision and effectiveness of the software developed for data analysis.

HARDWARE

Central Processor Unit

The LSI-11/2, the central processor unit (CPU) of the portable unit, is capable of addressing 32K (K = 1024) 16-bit words of memory. A 16-bit buffered parallel input/output (I/O) bus with asynchronous operation allows the processor and its system components (memory and I/O peripherals) to run at the highest possible speed. 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. Power requirements are +5 V at 1.0 A and +12 V at 0.22 A.

Memory

The memory has 16K words of programmable read-only memory (PROM) for storing operating software and 12K words random access memory (RAM) for storing data during analysis. The PROM provides nonvolatile program storage (retains its contents when operating power is removed); the RAM is volatile (loses contents when power is removed).

For software storage, we use UV-erasable PROM IC chips (EPROM), hence software changes are easily done by erasing and reprogramming the EPROM chips. The microcomputer unit uses INTEL 2716-type IC chips that have capacities of 1K 16-bit words each. One 8K-EPROM dual-height board holds eight of these chips; so two are necessary to provide 16K words of PROM. The power requirements for each board are +5 V at 0.5 A and +12 V at 0.02 A.

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 dual-height board are +5 V at 1.0 A and +12 V at 0.3 A.

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LSI-11 Communication Modules

The microcomputer requires both serial and parallel interface modules. 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 a DLV11-J asynchronous serial-line/interface board. This module controls four independent serial I/C channels to the LSI-11 bus, three of which are used by the microcomputer unit. The power requirements of the board are +5 V at 1.0 A and +12 V at 0.25 A.

The Silena communicates with the LSI-11 through a 64-line parallel I/O module with TTL 1 ic. This module is capable of connecting 64 external parallel I/O lines to the LSI-11 bus. The power requirements of this board are +5 V at 1.25 A.

This module is ideally suited for communication with the Silena MCA's parallel interface -- the Mod. 7500-11. It is unidirectional, only transferring data from the MCA to the computer, and has a data transfer rate of 10,000 bytes/s. Data from channels 0-1017 in the Silena memory is sent to the computer over eight lines, byte by byte. Since each channel consists of 8 bytes (ASCII code), 8144 bytes plus 41 trailing bytes are sent for each spectrum. Thus, the Mod. 7500-11 transfers a spectrum in less than 1 s.

Two I/O lines control the data transfer between the 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.

Clock Module

The microcomputer unit has a clock board that provides the time and date and operates in a peripheral slot of the LSI-11 system. When the computer is off, the clock module continues 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, Power Receptacle, and Fan

A 75-W (high-efficiency) switching power supply is used to provide DC power to the microprocessor and peripherals. It is small -- 27.9 cm x 13.1 cm x 4.3 cm, light weight (1.9 kg), and operates over a wide range of input voltages and frequencies. It has two DC outputs: one rated 5 V at 8 A, and the other rated 12 V at 3 A. The AC input specifications are 105 to 130 and 200 to 265 V AC at frequencies 47 to 100 Hz.

A backplane, configured to accept eight 14-cm wide (dual-height) boards, distributes power to the seven circuit-board modules that operate on 5 V and/or 12 V DC. Each board is kept firmly in a backplane connector slot by a rugged but light-weight board 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 boards installed, this assembly weighs approximately 1.8 kg.

A fused connector with voltage selection connects the switching power supply to the AC line power. It eliminates the need to change internal wiring for different AC input voltages, provides a convenient fuse

holder, and lets the user change connections to the power supply with a voltage selection card. The connector also has a built-in filter for reduction of rf interference.

To cool the internal components, 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, was installed.

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 user response or indicate a condition that requires corrective action. The character line is 10.2-cm long, and each individual character is 5 mm by 3.5 mm. The display is 15.7-cm wide by 4.8-cm high by 8.9-cm deep, weighs approximately 200 g, and operates on 5 V DC with current requirements of 500 mA.

A numeric keyboard terminal controls data entry into the microcomputer, and seven function keys provide remote control of the Silena. The terminal receives and transmits ACSII-coded data through one of the DLV11-J serial lines at 300 baud. The ENTER and CLEAR keys allow verification and/or erasure of entries before transmission to the LSI-11. The terminal weighs only 290 g and operates on 12 V DC at a 250-mA maximum.

Hardcopy printout of the results is produced by a quiet, nonimpact thermal printer. It receives information from the LSI-11 through one of the DLV11-J serial lines and prints the full ASCII character set in 20 columns across 58.6-mm-wide thermal paper, at a rate of 1.2 lines/s. A 42-m roll of paper will hold 8,400 lines at 2 lines/cm (approximately 100 spectral analysis outputs). The printer weighs 2.0 kg, and operates on 12 V DC at a maximum current of 1 A.

DATA ANALYSIS SOFTWARE

The objective of the data analysis software is to determine plutonium isotopic ratios from appropriate gamma-ray spectra. This requires determining peak areas pertinent to the isotopes of interest, a simple task if all relevant peaks were well-isolated, single peaks. Unfortunately, the analysis must resolve at least one complicated peak grouping because it provides the only information from which to deduce the Pu-240 isotopic abundance.

Typically, gamma-ray spectra taken by IAEA inspectors cover the 110-220-keV or the 110-450-keV region. The data-analysis software analyzes the following four-peak groupings in the 110-220-keV region: 123-129 keV; 141-150 keV; 160-165 keV; and 203-208 keV. Two others, the 330- and 370-keV peak groupings, are used in the analysis, if the gain is set to include these peaks in the spectrum.

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

$$Y_1 = y_0 e^{\alpha(x_1 - x_0)^2} + A y_0 e^{B(x_1 - x_0)}$$

$$\left[\frac{1 - e^{Co(x_1 - x_0)^2}}{1 - e^{\delta}} \right] \quad (1)$$

where

- y_1 = net data counts
- y_0 = peak height
- α = peak width parameter
- x_1 = channel value of the i^{th} point
- x_0 = peak position
- A, B, C = shape parameters describing the tailing function:
 - $\delta = 1$ for $x_1 - x_0 < 0$
 - $\delta = 0$ for $x_1 - x_0 > 0$.

Fig. 2. Components used to describe a peak shape (see text).

Six parameters — y_0 , x_0 , α , 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. The parameters α , 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.

The peak width parameter, α , is commonly specified by the full width at half maximum (FWHM = 2.355 σ). This is related to α by $\alpha = -1/2\sigma^2$. FWHM can be expressed in terms of peak energy as

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

The values of K_1 and K_2 can be calculated by determining the FWHM of low- and high-energy peaks in a spec-

trum. 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 $(1 - \exp[-(\alpha(x-x_0)^2)])$ to reduce its contribution to zero at the peak centroid. The parameter C is rather insensitive, and 0.4 appears suitable for all germanium detectors.

Studies have shown that the slope parameter B is detector dependent only, whereas the tail amplitude parameter A is both detector and energy dependent. A simple algorithm, given by $\ln A = C_1 + C_2 \times E$, describes the appropriate relationship between A and the gamma-ray energy. As for α , two or more peaks are used to evaluate the constants C_1 and C_2 . Therefore, the peak shapes for a given spectrum can be determined 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 , which are usually the more intense peaks in the spectrum, to determine the gain and the zero-intercept of the spectrum. Using this information and assuming that the conversion gain is linear, the exact positions of all other peaks to be analyzed can be computed. Good statistical significance is required to determine the tail amplitude parameter, A, accurately, and in most cases, the spectra encountered are not statistically good. In addition, the 148.6-keV peak has interferences on the low-energy side that increase the error in establishing the energy dependence of A. Since changes in A over 100 keV are 1 % or less, the code determines both A and B from the 208-keV peak and treats A as a constant for the spectrum.

If the value of x_0 in Eq. 1 is known (in addition to the other shape parameters), the set of equations for a peak grouping no longer contains unknowns in the exponent. This simplifies the software since the equations are solved by the linear least-squares method rather than by an iterative technique. The only free parameters remaining in the equations are the peak heights. Furthermore, not all of the peak heights in a group need to be free parameters since the relative amplitudes of all peaks belonging to a given isotopic component are determined by the branching intensities in their decay schemes. Therefore, a response envelope for each isotopic component rather than for each peak is computed and the number of unknowns is reduced to the number of isotopes contributing to the peak grouping.

Before the net peak area(s) can be determined, the background beneath the peak(s) must be removed. Instead of using a straight line or a polynomial function to determine the background the following explicit function is used to interpolate background values beneath the peak grouping:¹

$$B_1 = b_n + (b_m - b_n) \times \left(\sum_{j=n}^{j=1} Y_j \right) \times \left(\sum_{k=n}^{k=m} Y_k \right)^{-1}, \quad (3)$$

where B_1 = computed background at channel 1, Y_i = spectrum count of channel i , and b_n, b_m = average background level of the low- and high-energy side of the peak. This function produces smoothed background steps beneath each peak and works equally well for single peaks and complex peak groupings.

After the background is removed from the grouping, a response function for each isotope contributing to the grouping is calculated using peak shape and branching intensity information. This yields a set of linear equations whose number of unknowns equals to the number of isotopic components. Solving for these unknowns requires the inversion of a matrix that yields the necessary peak area information.

The objective is to interpret the spectral data in terms of isotopic ratios. A direct calibration cannot be used since there is no uniformity among the samples or the IAEA's detectors. Therefore, the influence of counting efficiencies and gamma-ray attenuation (due to the sample matrix or other absorbing material) on gamma-ray peak intensities must be accounted for in each individual spectrum. To calculate the ratio of isotopic abundances from the gamma-ray intensities the following equation is used:

$$A/B = \frac{I_1 T_1}{c_1 X_1 T_1} / \frac{I_2 T_2}{c_2 X_2 T_2} \quad (4)$$

where A/B = isotope ratio of interest; I_1, I_2 = peak intensities of the gamma rays arising from isotopes A, B; T_1, T_2 = half-lives of precursors; c_1, c_2 = relative detector efficiency for the respective gamma rays 1 and 2; X_1, X_2 = attenuation factors for gamma rays 1 and 2; and Z_1, Z_2 = branching intensities for gamma rays 1 and 2.

The branching ratios are known for each plutonium isotope;² while the half-lives for the plutonium isotopes and americium-241 are those recommended in ANSI 15.22. A self-calibration technique is used to obtain the necessary relative efficiency corrections from peak areas in each spectrum measured. Peak areas for ^{239}Pu , ^{241}Pu , and ^{241}Am are corrected for gamma-ray branching intensities and fitted to the functional form

$$\ln \left(\frac{I_j}{Z_j} \right) = a_0 + a_1 \delta_1 + a_2 \delta_2 + \sum_{j=3}^2 \text{or } 3 \left[a_{j+2} (\ln E_j)^j \right] \quad (5)$$

using a weighted least-squares technique. The value E_j is the peak energy in MeV. The coefficients a_1 and a_2 normalize the ^{241}Pu and ^{241}Am data points to the ^{239}Pu data points and provide 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, a cubic function is used to fit the data (the sum of j is to 3); while a quadratic function is used when only the first four peak groupings are included.

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. A $^{235}\text{U}/^{239}\text{Pu}$ isotopic ratio is determined using the ^{235}U 185.72-keV gamma-ray peak, if there is any statistically significant data in the 185.72-keV region.

Several fits of the relative efficiency data to Eq. 5 are required to determine the a_j coefficients because the 164.58- and 208-keV peak 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 (no more than three iterations are required). 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. The off-diagonal matrix elements of the inverted matrix are significant and must be included in the error determination.

The calculated isotopic ratios and total errors (statistical plus efficiency plus fitting) are printed by the thermal printer. The printout also includes the age of the sample since separation from ^{241}Am , which is calculated on the basis of the $^{241}\text{Am}/^{241}\text{Pu}$ isotopic ratio using the standard parent-daughter relationship. The time required for analysis of four peak groupings is 70 seconds.

DATA ANALYSIS RESULTS

The performance of the portable microcomputer unit was assessed by taking 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 similar 13-cm planar HPGe detectors.

Our measurements included four spectra of each NBS standard: two each taken at conversion gains of 0.11 and 0.34 keV/channel for livetimes of 2,000 and 20,000 s. The intent of these measurements was to obtain a better insight into the variation of results caused by the peak area analysis algorithms and the correction for detection efficiency. All spectra were collected with analyzer deadtimes of less than 20% to reduce possible high count rate effects.

Results obtained for each of the isotopic ratios ^{238}Pu , ^{240}Pu , and ^{241}Pu to ^{239}Pu are plotted in Fig. 3. The ordinate values for each plot are the isotopic ratios obtained by the data-analysis unit (designated by GAMMA) divided by the isotopic ratio calculated from the updated NBS reference values. At the time of our measurements these were:

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

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

The scatter of 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 for sample NBS 948 is 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 (.01%). 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 to 9.37×10^{-6} . This correction is also noted in the ESARDA study³ and in 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 provides a more accurate determination of the peak areas. The intensity and position of the 159.96-keV peak is fixed to the 164.58-keV peak and the weak 160.2-keV ^{239}Pu peak is fixed to the 161.5-keV ^{239}Pu peak. In the case of the 164.58-

^{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 the efficiency correction are small compared with those from determining peak areas for this pair of gamma rays.

CONCLUSIONS

The microcomputer unit provides a sophisticated data-analysis capability for plutonium gamma-ray spectra, yet it is portable and simple to operate. Removing and replacing the programmable EPROMs would allow the unit to carry out other kinds of numerical calculations. One currently unused backplane slot and serial I/O channel would allow the addition of a mass storage device and a full alphanumeric terminal to provide microcomputer capabilities.

The generally good agreement obtained with NBS reference samples lends confidence to the spectral data analysis method used by the data-reduction unit. No systematic errors in the method of determining peaks are indicated, since both positive and negative biases are observed for different isotopic ratios.

Results obtained with the microcomputer unit on IAEA data showed a similar degree of accuracy as the Headquarter-based computer system, but the microcomputer unit shows better precision. The IAEA inspectors have found the unit simple to operate; and find that the quick, in-field analysis of data also aids in judging the quality of the spectra. The IAEA has demonstrated the unit to be very useful for in-field Pu isotopic analysis and feels it will provide important support for inspectors in verifying fissile material contents. Additional tests of the microcomputer using mixed oxides, solutions, plates, and rods are being carried out by the Agency.

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Fig. 3. NBS plutonium isotopic ratio results obtained with the data-analysis unit (GAMA) divided by NBS values. The solid horizontal lines represent the mean values, and the dashed lines the $\pm 1\sigma$ limits of the mean.

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