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September 8, 1976

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September 8, 1976

HIGH-PURITY GERMANIUM DETECTION SYSTEM
FOR THE *IN VIVO* MEASUREMENT
OF AMERICIUM AND PLUTONIUM

MASTER

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CONTRACT E(29-2)-3533

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Printed in the United States of America
Available from the
National Technical Information Service
U. S. Department of Commerce
Springfield, Virginia 22161
Price: Printed Copy \$4.00 Microfiche \$2.25

Printed
September 8, 1976

RFP-2525
UC-41 HEALTH AND SAFETY
TID-4500-R64

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SUBJECT DESCRIPTORS

Body Counter
High-Purity Germanium Detectors
Plutonium
Americium

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Prepared under Contract E(29-2)-3533
for the

Albuquerque Operations Office
U. S. Energy Research and Development Administration

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HIGH-PURITY GERMANIUM DETECTION SYSTEM FOR THE *IN VIVO* MEASUREMENT OF AMERICIUM AND PLUTONIUM

William H. Tyree, Roger B. Falk, Charles B. Wood, and Ronald W. Liskey

Abstract. A high-purity germanium (HPGe) array, photon-counting system has been developed for the Rocky Flats Plant Body-Counter Medical Facility. The newly improved system provides exceptional resolutions of low-energy X-ray and gamma-ray spectra associated with the *in vivo* deposition of plutonium and americium. Described are the operational parameters of the system and some qualitative results illustrating detector performance for the photon emissions produced from the decay of plutonium and americium between energy ranges from 10 to 100 kiloelectron volts. Since large amounts of data are easily generated with the system, data storage, analysis, and computer software developments continue to be an essential ingredient for processing spectral data obtained from the detectors. Absence of quantitative data is intentional. The primary concern of the study was to evaluate the effects of the various physical and electronic operational parameters before adding those related entirely to a human subject.

INTRODUCTION

Inhaled quantities of plutonium and americium are detected and measured with several types of scintillation counting systems at the Rocky Flats Body-Counter Facility. Germanium detector systems have been evaluated in the past, but most have provided limited success. An intrinsic germanium (Ge) counting system, utilizing planar detectors and with more stable operating parameters than earlier lithium-drifted germanium detector types, has been installed to maintain state-of-the-art measurement for inhaled isotopes. The report describes the parameters used in evaluating the current detector system and the performance results.

RADIATION COUNTING CRITERIA

Scintillation detection systems are generally used for the measurement of inhaled plutonium and americium. The method at Rocky Flats Plant involves the detection and quantification of the 59.54 kiloelectron volts (keV) gamma rays from the decay of americium 241 (^{241}Am). If the fraction of the ^{241}Am in any inhaled plutonium (Pu) is known, the amount of plutonium indicated by the americium *tracer* can be determined. When ^{241}Am is absent from or is present as only a small fraction in the plutonium mixture, the measurement of the inhaled plutonium involves the measurement of the L-X rays emitted from the alpha particle decay of plutonium 239 and plutonium 240. The L-X rays are also emitted from the decay of ^{241}Am . The X rays from ^{241}Am have a slightly higher energy than those from the decay of plutonium.

The detectability of any counting system is related to the total effective surface area of the detectors, the efficiency of the detection medium, and the background counting rate. In general a large surface area, nearly 100-percent absorption efficiency at 17 keV (average energy of the L X-ray region and at 59.43 keV photon energies), and excellent resolution are required.

The germanium system provides superior resolution, near 100 percent absorption capability and an intermediate surface area. The high-purity germanium (HPGe) surface area is limited by the present technology for *growing* hyperpure germanium ingots. The fabrication of large surface area detectors, needed in body counting applications, usually requires deep detectors in order to minimize input capacity affecting the resultant resolution figure. Increasing the depth of a large detector, assuming that such an intrinsic detector could be

FIGURE 1. Lucite Cap with Lexan Window Covering the Front of Cryostat.

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built, would increase the high energy response of the detector. In turn, this would significantly increase the background counting rate in the low energy portion of the spectrum. These conflicting characteristics require certain hard choices by the system designer to produce satisfactory low-energy photon detector performance.

DESCRIPTION OF THE DETECTOR SYSTEM

The intrinsic germanium detector system at Rocky Flats consists of two separate arrays of four planar high-purity Ge detectors mounted inside a downward looking cryostat. Each of the planar detectors is 10 millimetres (mm) deep and 36 mm in diameter. Each detector is warranted to have a

Full Width Half Maximum (FWHM) resolution of 800 eV at the 122 keV photon energy of cobalt 57.

The four detectors in a single array are mounted equidistant from the vertical axis of the cryostat and are 5 mm inside the lower face of the cryostat. A beryllium entrance window for each detector of 0.26 mm thickness is mounted as part of the lower face of the cryostat, as shown in Figure 1. Each array has a slip-on cover attached to the lower end of the cryostat protecting the beryllium windows from accidental impact. The cover consists of a layer of 0.127-mm thick Lexan held in place with a Lucite ring attached to the sides of the cryostat. The Lexan was chosen to provide a semirigid layer with minimum attenuation characteristics for the

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FIGURE 2. Interior of Steel Room Showing Arm Holding One Detector Array.

L-X rays produced from the decay of plutonium and americium.

Mechanical Setup:

The cryostats containing the arrays of detectors are mounted over a bed inside a 59-metric ton steel room. The room dimensions are 2.43 by 2.43 by 2.74 metres with 15.24-centimetre (cm) thick walls, floor, ceiling, and door. The interior of the room is covered with a graded shield consisting of lead, tin, and zinc.

The mechanical support arms, holding the detectors, are shown in Figure 2. They provide overhead support for the detection systems with tilt controls for positioning around the patient. The tilt controls are limited in travel to insure safe handling of the cryostats filled with liquid nitrogen. The preamplifiers are mounted as an integral unit to the cryostat to minimize capacity effects on the input circuits in the preamplifier. Liquid nitrogen

filling is provided by permanently installed valve-controlled filling lines from a pressurized storage Dewar flask located outside the Body-Counter Facility.

Electronic System:

Since any combination of analog signals combines the noise components, each detector signal was brought to the analyzing console through its own coaxial cable. This presentation maintained the individual detector resolution figure of between 600 and 650 eV FWHM at 59.54 keV. The effect of the mixer-router on individual resolution was about a 10-percent degradation in the FWHM figure.

The integration and differentiation time constants for each of the pulse amplifiers were set to 6 microseconds. The range of pulses produced by a 10-mm deep detector, even inside a 59-metric ton steel room at 1828.8-metre elevation, included overload pulses having approximately 10 normal pulse widths (see Figure 3). A pulse-stretcher and peak-detection module for each amplifier were included to minimize the effect of baseline distortion on the low energy region from large overload pulses and to eliminate saturated input conditions in the analog-to-digital (ADC) converter. Each of these modules produced a uniform pulse width of 3 microseconds independent of the input-pulse width. The resultant pulse was presented through a coaxial cable to the appropriate input of the mixer-router unit of the pulse height analyzer (PHA). The block diagram of the entire system is shown in Figure 4.

Each group of the PHA memory consists of 256 channels for the storage of the pulses produced from each detector. The PHA has a 4K address memory available for data storage, all of which is accessible from computer control from the cathode-ray tube (CRT) terminal.

Evaluations:

The evaluation of these detectors has included measurement of the effective surface area, resolution determination at the 59.54 keV, the photon energy of ^{241}Am , and the optimization of the ion collection for the 17-keV X-ray region associated with the decay of ^{239}Pu and ^{241}Am .

FIGURE 3. Range of Pulse Shapes from Amplifier.

Surface Area

The effective surface area confirmation tests were conducted using a collimated ^{241}Am source. The tests were made by counting the source for a fixed time period at different positions across the diameter of the beryllium window associated with each detector in each array. These tests were made at 6.35-mm intervals across each detector window.

The tests revealed that within the accuracy of the diameter of the collimator, the active area for each detector was properly specified by the 36-mm diameter physical dimension of each detector. The first tests run on Array No. 2 showed one detector having only about 8 square centimetres (cm^2) of effective surface area. After bias-voltage optimization, the results of the collimation tests

for this detector produced proper surface area. The total area available is about 80.4 cm^2 .

Resolution

The calibration fixture is shown in Figure 5 for small disk sources. The assembly provides a fixed geometry for comparison of different types of sources. The resultant information also provides the raw data for determining the individual detector resolution, FWHM. The lowest resolution figure obtained for any of the detectors was 597 eV FWHM at 59.54 keV. Most of the detectors produced resolution figures of approximately 635 eV. These figures were determined for a specific detector output tied directly to the input of the analog-to-digital converter. The effect of the mixer-router used with the pulse-height

Legend

HPGe = High-Purity Germanium.
 ADC = Analog-to-Digital Converter.
 PHA = Pulse-Height Analyzer.
 CRT = Cathode-Ray Tube.

FIGURE 4. Block Diagram of High-Purity Germanium Detector Counting System.

analyzer (PHA) produced about 10-percent degradation in resolution for any of the detectors.

Collection Efficiency

Since the collection efficiency varies with applied bias voltage, two high-voltage distribution circuits provide the proper operating voltage for each of the individual detectors in the arrays. The operating bias voltage was optimized at 875 volts for the

four detectors in Array No. 1. Array No. 2 had three detectors that provided complete charge collection at 1175 volts. The fourth detector required 1700 volts for proper charge collection of the ions produced from the absorption of the low energy photons in the 17-keV region. All voltages are negative in polarity. A single high voltage power supply with low output impedance was used to minimize noise contribution to the separate voltage-distribution networks.

FIGURE 5. Calibration Fixture.

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DETECTOR PERFORMANCE

Resolution:

With a system resolution of approximately 650 eV FWHM for each detector, an energy calibration of about one keV per channel in the PHA results in over 80 percent of the photoelectric events being

accumulated in a single channel. With 80 to 90 percent of the photopeak contained in a single channel, an optimum relation is achieved between the positive count and the background count in that channel. Figures 6 and 7 show spectra of ^{241}Am and ^{239}Pu , respectively, without absorbers for typical detectors (10 cm² and 10 mm deep) in the array at an energy calibration of 1 keV per channel.

FIGURE 6. Detector Response for Americium 241.

Electronic Constraints:

During the initial period of system evaluation, several background rejection schemes were tried to further reduce the background continuum produced from each detector. These schemes included pulse-shape discrimination and anti-coincidence operation using a large sodium iodide

thallium [NaI(Tl)] scintillation detector. The additional constraints on the pulses required by the ADC did not significantly reduce the background spectra beyond that available through careful adjustment of the pole-zero response of the amplifier output circuits. Both timing systems produced poorer detection capability when compared with presenting the pulses directly into the mixer-router tied directly to the ADC.

FIGURE 7. Detector Response for Plutonium 239.

Radiation Background:

A comparison of background data obtained from the detector system mounted inside a 59-metric ton steel room is shown in Figure 8. The counting time was 4000 seconds. The lower curve shows the results from the summation of detected events from 8 detectors (two 4-detector arrays) without a human subject present in the steel room. The

upper curve shows the scatter effect from a human subject (without radiation exposure), who was added to the interior of the steel room.

PROGRAMING

An example of a typical presentation from a single counting period using an ^{241}Am source with the

FIGURE 8. Background Data with and without a Human Subject.

system is shown in Figure 9. The multiple spectra analysis required for data reduction necessitates the use of a computer system with flexible programing capability.

Data processing for the HPGe detector counting system is provided by a Data General-1220 computer and a dual disk system. Software calls for this system, use a BASIC language compiler operating a Real Time Operating System (RTOS). The BASIC compiler is listed in the system as a directory, and direct conversion to machine language is not required by the programmer. The BASIC language has call-routines capability allowing the use of previously written assembly programs to manipulate the four thousand (4K) channel pulse-height analyzer (PHA). Several other compilers are available with the disk operating system including assembly, Fortran, and Algol.

Programs are constructed to prompt the user by presenting various questions which are, in turn, answered by the user. This approach is shown in flow-chart form in Figure 10. The user may choose to store a spectrum, subtract a background from a spectrum, or connect several programs to perform a complete data analysis sequence. Several statistical programs are available for operations on the data, including programs for curve fit and data smoothing. Graphics capability has been programmed to allow visual display of spectra before or after data manipulation.

The dual disk-drive capability offers a large storage capacity, and the ability to store over 300 complete spectra on a single disk cartridge. Operating programs may be stored on one disk and the other may be used for spectrum storage.

The high-speed tape reader and cathode-ray terminal provide a rapid input-output interface

FIGURE 9. An Eight-Detector Presentation for Americium 241.

with the computer. The reader also can be used to input data from other systems for evaluations. Program development constitutes a major effort for getting the detector system operational for routine counting and in general, consists of the following parts:

1. Setting up the readout sequences of detector data output.
2. Initiating a sequence of 8-detector data output formatting.
3. Providing statistical analysis programs.
4. Providing graphical formatting for data presentation.

Figure 11 shows the operating area for the system centered around the PHA, computer, and CRT input-output terminal.

CONCLUSIONS

Two HPGe high-resolution detector systems have been installed in the Rocky Flats Body-Counter Facility. These systems are providing better than 650 eV FWHM at the 59.54-keV photon energy of ^{241}Am for each detector. Each detector has a

physical area of 10.18 cm². Tests with a collimated ^{241}Am source confirmed that the effective detection surface area and physical area were identical.

Optimization of the collection efficiency for the 17 keV X-ray region of americium and plutonium was performed for each detector. Amplitude variations from photon interactions anywhere across the diameter of any of the HPGe detectors for the 59.54 keV gamma ray produced a maximum change of 130-eV shift in peak energy. A computer is an essential ingredient for the successful operation of the system. The programing provides rapid readout of all the channels used in the PHA for detector presentation and formatting of the desired data. BASIC language is used for the interactive language between the CRT control system, computer, and the PHA.

Fine tuning of the detection parameters for the *in-vivo* detection of ^{241}Am at low parts per million will continue, with the ultimate goal of the direct detection of plutonium 239 without the presence of americium 241. Detailed calibrations are in process to define the ^{241}Am system sensitivity and detectability for both the americium tracer method and the method using the L-X rays from plutonium 239, and these will be reported in the future.

FIGURE 10. Programing Flow Diagram.

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FIGURE 11. Console for System.

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