SNM Gamma-Ray Fingerprint Monitor

Functional Requirements and Design Specifications

Alan M. Bieber, Jr.
Walter R. Kane

July 1994

SAFEGUARDS, SAFETY, AND NONPROLIFERATION DIVISION
DEPARTMENT OF ADVANCED TECHNOLOGY
Brookhaven National Laboratory
Building 107
P.O. Box 5000
Upton, New York 11973-5000

SSN-94-23
SSN File 5.8.171

SNM Gamma-Ray Fingerprint Monitor
Functional Requirements
and
Design Specifications

Alan M. Bieber, Jr.
Walter R. Kane

July 1994

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED 85

Safeguards, Safety and Nonproliferation Division
Department of Advanced Technology
Brookhaven National Laboratory
Upton, New York 11973-5000

DOE Contract No. DE-AC02-76CH00016

MASTER

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, make any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TABLE OF CONTENTS

I. Introduction	5
II. Scientific Background	5
A. Properties Influencing Gamma-Ray Spectra of Nuclear Materials	5
B. Gamma-Ray Signatures of Materials	6
C. Applications	8
III. Functional Requirements	9
A. Hardware	9
B. Software	10
IV. Design Specifications	11
A. Hardware	11
B. Software	13

I. Introduction

A number of DOE facilities need to perform confirmatory inventory measurements on items of special nuclear material (SNM). The DOE Office of Safeguards and Security (OSS) has tasked the Safeguards, Safety and Nonproliferation Division (SSN) of the Department of Advanced Technology at Brookhaven National Laboratory (BNL) to develop a high-resolution gamma-ray-spectroscopy-based instrument for performing confirmatory inventory measurements on such materials, a "gamma-ray fingerprint monitor" (GRFM). This document is a conceptual design for the SSN GRFM system.

Note that this conceptual design is based on previous experience with measurements of plutonium-bearing materials and comparison of gamma-ray spectrum features, not on actual tests of the procedures or hardware described. As a result, modifications may be necessary when actual prototype hardware and software are tested in realistic circumstances on actual materials of interest.

II. Scientific Background

A. Properties Influencing Gamma-Ray Spectra of Nuclear Materials

The gamma rays emitted from most types and samples of accountable nuclear materials possess attributes which derive from the physical, chemical, and geometrical properties of the individual samples. Accordingly, different types of material, and in many cases, individual samples, can be identified, verified, and tracked. The principal properties which influence the gamma-ray spectra are:

Isotopics. The isotopic composition of most materials can be obtained from the gamma-ray spectrum. In addition, in certain materials, most importantly plutonium, the decay of a fairly short-lived isotope (Pu-241: 14 yr) will produce another isotope (Am-241: 433 yr) whose gamma rays can also be observed and used to determine the elapsed time since processing of the material. Once such a spectrum has been obtained, it is straightforward to predict the gamma-ray spectrum which will be observed at any future date.

Geometrical Properties. Since the gamma rays will usually be heavily absorbed in the material, especially in the case of metallic samples, the spectrum will be strongly dependent upon the shape. For example, in a sphere of plutonium metal, only a small fraction of the low energy gamma rays and x rays will reach the surface and escape, while for a thin foil or shell, a much higher proportion of the low energy radiations will escape. The observed spectrum will thus be characteristic of the type of object being measured.

Physical and Chemical Properties. The effect of the physical and

chemical properties of the material on the gamma-ray spectrum will be manifested chiefly through its density. A given quantity of material in the form of a chemical compound, because of its comparatively low density, will, in general, emit a much higher proportion of low energy radiation than the same quantity in the form of, for example, a metal part.

Quantity of Material. Because of the effects described above, the total quantity of material in a sample cannot usually be derived in a simple way from the gamma-ray spectrum. It can, however, be determined by calibration with the use of objects of a similar type, or in favorable cases, by calculation.

From the above considerations it is seen that the gamma-ray spectra from objects of a given class, will, in general, have characteristics typical of that object. The gamma-ray spectra obtained from pits of a certain type will have features that are very nearly identical, and will be markedly different from those obtained from, for example, cans of oxide. Objects containing plutonium which are identical physically, however, may exhibit substantial differences in the intensities of the Am-241 lines emitted because of differences in the elapsed time since the material was last processed.

B. Gamma-Ray Signatures of Materials

Plutonium. The isotopes of plutonium emit a number of gamma rays over a wide energy range from 38 to 769 keV. Relatively strong x rays with energies in the vicinity of 100 keV are also emitted. The relative intensities of these radiations will thus depend strongly on the geometry and density of the source. The decay product Am-241 emits gamma rays with energies from 59 to 722 keV. As indicated, the yield of the americium gamma rays will depend upon the elapsed time since processing of the plutonium, and once determined, can be predicted for future measurements, so that it will constitute a "fingerprint" characteristic of a given object, or at least, of a set of identical objects of the same vintage.

For the case in which an isotope (Am-241) is produced by the decay of a parent isotope (Pu-241) with a rate λ_1 and subsequently decays with a rate λ_2 , the number of daughter nuclei at any given time is given by the simple expression

$$N(\text{daughter}) = \frac{\lambda_1}{\lambda_2 - \lambda_1} N_0(\text{parent}) e^{-\lambda_1 t} - e^{-\lambda_2 t}$$

and the rate of gamma-ray emission from the daughter nuclei can be calculated accordingly.

High Enriched Uranium. The gamma rays from HEU are concentrated in the low energy region from 144 to 205 keV, with, in addition, gamma rays of 766 and 1001 keV energy from whatever U-238 remains

in the material. X rays with energies ranging from 89 to 109 keV will be generated by the alpha decay of the uranium and, in addition, by the absorption of gamma rays in the material and subsequent fluorescence of the uranium atoms. U-235 decays to Th-231, and Th-231, in turn, with a 25 hour half-life, to Pa-231, which has a half-life of 32,000 years. Thus, several days after the material is processed chemically the rate of emission of the gamma rays will reach a constant value, and there will be no further changes in the gamma-ray intensity pattern with time. As in the case of plutonium, the intensity pattern of the x rays and gamma rays will depend to some extent upon the geometry and physical properties of the material, but this dependence would not be expected to be as strong as in the case of plutonium. Accordingly, for HEU the gamma-ray spectrum will be characteristic of the particular class of object, but will not be expected to exhibit significant differences from one object to another within a given class. With proper calibration, the gamma-ray yield will provide a measure of the quantity of U-235 present. For samples that are "thick" with respect to the absorption of the strong 186 keV gamma ray, a determination of the yield of this gamma ray provides a value for the enrichment of the material (enrichment meter principle).

U-233. Gamma rays ranging in energy from 42 to 365 keV are emitted from U-233, so that a spectrum typical of a given type of object can be obtained. In addition, a small quantity of the isotope U-232 is always present in all samples of U-233. U-232 decays with a 72 year half-life to Th-228, which has a half-life of 1.9 years. A number of gamma rays are emitted subsequent to the decay of Th-228, with especially strong lines at energies of 238, 583, 727, and 2614 keV. Just as in the case of plutonium, a U-233 sample will thus have a time-dependent gamma-ray spectrum which can give the elapsed time since processing and which, once determined, can be predicted for future measurements.

U-238. The strong 186 keV gamma ray of U-235 is clearly evident in the gamma-ray spectra of normal or depleted uranium, as well as the 766 and 1001 keV gamma rays from the U-238 decay chain. Just as for other materials, the relative intensities of these gamma rays will depend upon the physical and geometrical attributes of a given object. U-238 decays by alpha-particle emission to Th-234, which possesses a 24 day half-life, so that several months are required after processing of the material before the gamma-ray emission rate reaches a steady value.

Np-237. Gamma rays with energies between 86 and 237 keV, and x rays, are emitted in the decay of Np-237. Accordingly, the relative intensities of these radiations will depend upon the geometry and physical properties of the emitting object.

Ac-228. Ac-228 decays to 1.9 year Th-228. As indicated, strong gamma rays with energies of 238, 583, 727, and 2614 keV are emitted in the decay of Th-228. When the thorium is purified chemically and the radium removed, the gamma rays of Th-228 will

initially decay with a half-life of 1.9 years but then as the Th-228 is replenished through the decay of Th-232 and Ra-228 they will gradually increase again in intensity, reaching an equilibrium value after several Ra-228 half-lives of 5.7 years. From measurements on these gamma rays it is thus possible to determine the elapsed time since processing, and a fingerprint characteristic of this time will be present in the spectrum of any thorium-containing object.

Fission Products. The gamma rays from various fission products have been studied extensively and are well characterized. If necessary, it should be straightforward to determine the quantity of fission products present in a given sample and their relative age.

C. Applications

Confirmatory Measurements. It is clearly possible, with appropriate measurements, to confirm relevant nuclear properties of a given object containing SNM. This could take the form of shipper-receiver measurements where they are made on identical systems at the shipping and receiving facilities, or at one facility where it is necessary to establish certain properties of the material.

Item identification and Tracking. For items containing plutonium and possibly certain other nuclear materials as well, it should be possible to assign a unique identity to a given item, or at least to a class of items of identical design and approximately the same vintage. For other materials, it should be possible to assign a given gamma-ray spectrum to an item of given design and configuration. In the case of plutonium and certain other materials, after a gamma-ray spectrum is initially established, it will be possible to predict the spectrum at any future time.

Item Monitoring. With a gamma-ray detector of sufficient resolution and efficiency, it should be possible to monitor the radiations emitted by a substantial number of items in a storage area and determine in a short time if any items are moved or removed from the area.

As an example, we consider a storage area containing 50 items, each of which contains 4 kg of Pu. A detector situated at an average distance of 10 meters from the items will receive about 10 414 keV gamma rays/cm²-sec from each item. A high-purity germanium (HPGe) detector with reasonable efficiency (50%) will then register of the order of 100 414 keV events/sec for each item, and 5000 such events/sec for all 50 items. If one item is removed, in 10 minutes the deficit in the 414 keV peak area will be 6000 counts out of a total of 3 million. The statistical fluctuation (3 sigma) of 3 million counts will be 5000 counts, so that the peak area deficit due to the removal of one item should be clearly evident in about 10 minutes.

It is possible that an adversary might attempt to defeat the system by introducing an additional source of gamma rays at the same time the SNM-containing item is removed. To protect against such a scenario several Pu gamma rays, for example those at 375, 414, and 451 keV, along with several energy regions in which no Pu gamma rays are found should be monitored. This strategy requires a detector with an energy resolution considerably better than a NaI scintillation detector.

III. Functional Requirements

In the following description of the functional requirements for a GRFM system, requirements for the system are distinguished from desired capabilities. Requirements are capabilities that the system must incorporate in order to perform its measurement function. Desired capabilities would enhance the system's performance or usability, but need not necessarily be incorporated in the final design, and if not incorporated would not prevent the system from achieving its design goals. Requirements are stated in the form "...the system must...", while desired capabilities are stated in the form "...the system should..."

A. Hardware

1. Measurement Capability

The fundamental requirement for the hardware of a GRFM system is that it must be capable of acquiring and storing a gamma-ray spectrum from an item of SNM with sufficient accuracy and precision that high confidence can be placed in the resultant fingerprints and comparisons between them.

2. Ruggedization

The GRFM system should be capable of operation in a normal industrial environment such as would be encountered in SNM storage areas.

3. Maintainability

The hardware should be designed to minimize the need for routine maintenance or adjustment. The system should be designed such that any unavoidable routine adjustments can be made with as little disassembly or opening of enclosures as possible.

4. Power Supply

The system power supply (AC power adapter, rechargeable batteries and battery charging system) must be capable of powering the system for up to eight hours without shutdown, and the battery-charging system

must be capable of completely recharging totally-discharged batteries within 12 hours.

5. Enclosures and Packaging

System enclosures and packaging must be rugged enough to withstand air transport and vehicular transport over rough roads, wide temperature variations, and dirty and/or dusty environments. In addition, they should be designed to minimize access to system internals.

6. User Interface

The user hardware interface (displays and controls) should be designed to allow maximum visibility for all system displays and to minimize the possibility of accidental manipulation of any system controls. The number of user-operable controls or adjustments should be minimized to the extent possible.

7. Portability

The entire system must be small and light-weight enough to be hand-carried in one trip by no more than two persons. The detector must not require continuous cooling during transport.

B. Software

1. Measurement Capability

The system software must have the capability to determine the presence of Pu-239, U-233, U-235, or U-238, and must be capable of constructing gamma-ray fingerprints and comparing these fingerprints such that high confidence can be placed in the results. The software should be capable of computing a figure of merit for any comparison.

2. User-Friendliness

The system software should be designed to minimize user input through use of menus and defaults wherever possible and should use clear, simple language in all prompts and displays.

3. Error Detection and Recovery

The system software should be designed so that errors in user input and, to the extent possible, hardware errors do not result in abnormal termination of the program. When an error is detected, a message should be displayed informing the user of the nature of

the error and the actions needed to attempt to correct it.

IV. Design Specifications

In the following design statements, the functional requirement or desired capability (presented above in section III) which the design statement addresses is referenced parenthetically at the end of each section.

The GRFM system will be composed of the following hardware:

- a portable high-resolution gamma-ray spectroscopy system (the Canberra Inspector system) made up of an intrinsic germanium detector and its associated preamplifier and high-voltage power supply, a spectroscopy amplifier, and an 8192-channel multichannel analyzer;
- a laptop computer system consisting of a microprocessor, random-access memory (RAM), read-only memory (ROM), a keyboard, a liquid-crystal character display, a floppy disk drive, and a hard disk drive; and,
- a rechargeable battery power supply.

The system will use specially-written software to control fingerprint construction and comparison. Commercial software appropriate for the hardware (the Canberra GENIE-PC system in the case of the Canberra Inspector system) will be used to control data acquisition.

A. Hardware

1. Detector, Preamplifier, and High-Voltage Power Supply

The detector will be either a 200 mm² by 10-16 mm deep planar HPGe detector (for use with Pu-bearing materials) or a closed-end coaxial high-purity ("intrinsic") germanium detector with a photopeak efficiency (relative to NaI) of at least 15% (for use with U-bearing materials). The detector preamplifier will be a cooled field-effect transistor (FET) preamp. The high-voltage power supply will be a miniaturized, stabilized, low-power unit. The preamplifier and high-voltage power supply will meet or exceed all specifications for currently-available state-of-the-art commercial portable HRGS systems (III.A.1. and 4.).

The specifications for the Canberra Inspector system are as follows:

High-Voltage Power Supply:

Voltage: 0 to 5000 Volts
Current: 100 μ Amps
Voltage Adjustment: Continuous via software
Controls: Computer disable, enable and voltage
readback hardware interlock for bias
inhibit (via software)

2. Spectroscopy System

The spectroscopy system will be composed of a spectroscopy amplifier and an 8192-channel multichannel analyzer. The spectroscopy system will meet or exceed all specifications for currently-available state-of-the-art commercial portable HRGS systems (III.A.1.).

The specifications of the Canberra Inspector spectroscopy amplifier and analog-to-digital converter are as follows:

Amplifier:

Gain: 2 to 1500
Shaping Time Constants: 1 or 4 μ Sec
Noise: 3.5 μ Volt RMS for gains ≥ 100
Temperature Coefficient: Gain $\leq \pm 0.0075\%/^{\circ}\text{C}$
DC level $\leq \pm 7.5 \mu\text{Volt}/^{\circ}\text{C}$

Analog-to-digital converter:

Type : 100 MHz Wilkinson
Integral Nonlinearity: $< \pm 0.025\%$ of full scale
Differential Nonlinearity: $< \pm 0.9\%$

3. Microprocessor and Memory

The spectroscopy system will be controlled by a laptop computer system. The laptop will have an 80386 or better processor, color LCD display, at least 8 Mbytes RAM, a 3 1/2" 1.44 Mbyte floppy disk and a 100 Mbyte or larger hard disk (III.A.1., 6., and 7.).

4. Power Supply

The power supply will be composed of rechargeable battery packs for the laptop and Inspector and 110/220 volt AC recharging units. The battery packs will be capable of operating the system continuously for at least 8 hours, and the power supply will be capable of recharging completely discharged batteries in 12 hours or less. The battery charger will incorporate protection against low or high input voltages, and surges or spikes on input power (III.A.1., 4., and 7.).

B. Software

All software used in the GRFM system will be completely documented. For each code module (subroutine, function, or procedure) in the software, the documentation will include the following:

- a description of the purpose and overall function of the module;
- a full, commented source code listing;
- a flow chart or logic diagram;
- a detailed explanation of the operation of the module;
- a complete listing of all variables used in the module, with a description of each including its format, expected and allowed values, and domain; and,
- a listing of all modules which reference or are referenced by the module, with a complete description of all parameters or arguments passed between the modules.

1. Data Acquisition and Processing

a. Data Acquisition

Data acquisition will be controlled by commercial software appropriate for the specific hardware used. In the case of the Canberra Inspector system, the appropriate software is the Canberra GENIE-PC program. Spectrum data will be stored on the system hard disk drive for use by the fingerprint construction and comparison routines.

b. Data Processing

For each spectrum, processing will begin with computation of the area and the uncertainty in the area under each statistically-significant U or Pu peak of interest. Standard analysis algorithms will be used for these computations. Peak shape will be analyzed by computation of the values for FWHM, and peaks will be rejected if these values are inappropriate. For Pu and enriched U, the system will then compute the isotopic composition of the material. The full fingerprint for an item will consist of the peak areas, uncertainties, and FWHMs plus the isotopic composition, if computed. (III.B.1.).

2. Data Storage and Retrieval

The system software will have the capability to store and retrieve both raw spectrum data files and fingerprint files on the laptop's hard or floppy disks. This will allow data to be acquired at one location and then used at another, or for data to be acquired over a period of time, without stopping to process each spectrum (III.B.1.).

3. Fingerprint Comparison

The fingerprint comparison algorithm used will be based on that used in the Controlled Intrusiveness Verification Technology (CIVET) High-Resolution Gamma-ray Spectroscopy (HRGS) system, developed by BNL for use in verification of nuclear warheads. The comparison algorithm basically computes weighted ratios of values in two data sets and performs a least-squares fit of the data to the expected value of 1.0. The ratios are weighted in inverse proportion to their uncertainties, so that the most precise values are weighted most heavily. Since this approach uses ratios, the data on peak areas can be combined with data on isotopics in one comparison process. The outputs of the algorithm are the fitted average ratio, confidence limits for the fitted ratio, and a goodness-of-fit statistic derived from the summed residuals. Two data sets are considered to compare if the confidence limits for the fitted ratio include 1.0 and the goodness-of-fit statistic is acceptably small. The exact values for the width of the confidence limits on the fitted ratio and the limit on the goodness-of-fit statistic depend on the magnitude of the overall experimental error (a combination of counting statistics and non-statistical experimental errors such as errors in reproducing counting geometry). This overall experimental error cannot be evaluated without testing of the actual instrument. Based on the results of tests of the CIVET HRGS, it appears likely that a 2-sigma (95%) confidence limit and a goodness-of-fit statistic of less than 2.0 are probably appropriate initial values.