SSN-97-05
Task No. 8.171

BNL-64032

CONF-970744--

THE APPLICATION OF HIGH-RESOLUTION GAMMA-RAY SPECTROMETRY (HRGS) TO NUCLEAR SAFEGUARDS, NONPROLIFERATION, AND ARMS CONTROL ACTIVITIES

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PRESENTED AT THE

38th Annual Meeting of the
Institute of Nuclear Materials Management
Phoenix, AZ
July 20-24,1997

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ABSTRACT

While well-developed methodologies exist for the employment of high-resolution gamma-ray spectrometry (HRGS) in determining the isotopic composition of plutonium samples, the potential capabilities of such measurements in determining the properties of nuclear materials otherwise remain largely unexploited. These measurements contain information sufficiently detailed such that not only can the isotopic composition of uranium and plutonium materials be determined, but the details of the spectrum obtained will depend reproducibly upon other factors including the total mass, density, chemical composition, and geometrical configuration of the material, and for certain materials, the elapsed time since chemical processing. The potential thus exists to obtain a "gamma-ray fingerprint" for typical containers or assemblies of nuclear material which will then serve to identify that class of item in a later confirmatory measurement. These measurements have the additional advantage that, by comparison with active interrogation techniques which usually require the introduction of some extraneous form of radiation or other intrusive activity, they are totally passive, and thus impose only minimal additional safety or regulatory burdens on the operators. In the application of these measurements to the verification of treaty-limited items, where the information acquired may be sensitive in nature, the use of the CIVET (Controlled Intrusiveness Verification Technique) approach, where a computer-based interface is employed to limit access to the information obtained, may be followed.

INTRODUCTION

Gamma-Ray Spectra of Nuclear Materials

Practically every nuclear material of interest in the nuclear safeguards and nonproliferation areas emits a gamma-ray spectrum which provides a unique "fingerprint" for that material and which also contains information on its isotopic composition and its physical state as well - its mass, density, chemical composition, and geometrical configuration. For certain materials, the intensities of the gamma rays from daughter products which have grown in from the decay of the original material, e.g., ^{241}Am from the decay of 14.7-year ^{241}Pu, provide a useful "clock" which establishes the elapsed time since the last chemical purification of the particular batch of material. Examples of the gamma-ray spectra of a number of nuclear materials are shown in Figure 1.

- ^{235}U. In addition to the well-known 186-keV radiation, widely used to provide a measure of uranium enrichment, several other gamma rays, with energies of 144, 163, 195, 202, and 205 keV, are emitted with intensities approximately a tenth that of the 186 keV peak. Since the absorption of gamma rays by uranium or other heavy materials is highly energy-dependent in this region, the relative intensities measured for these gamma rays can provide information on the configuration of the emitting material and intervening heavy material . This is illustrated in Figure 2, where the relative intensities of the 143, 186, and 205 keV gamma rays of ^{235}U are shown for the case of a thin, non-absorbing source, a thick source, and a thick source with an additional

1 mm uranium absorber (natural or depleted uranium). The intensity patterns of the three gamma rays clearly reflect the configuration of the source material and absorber.

• ^{238}U. In contrast to the low-energy gamma rays emitted by ^{235}U, the gamma rays of ^{238}U (actually emitted by ^{234}U subsequent to the decay of ^{238}U) have high energies, a triplet of peaks at 742, 766, and 786 keV. and a very strong peak at 1001 keV. Since the absorption coefficients of most materials are much smaller for gamma rays in this energy region than, for example, for the gamma rays of ^{235}U, these gamma rays will be far more penetrating than those of ^{235}U, and consequently provide different information on the configuration of the emitting material. It is worth noting that these same high-energy gamma rays are also emitted after the decay of 86-year ^{238}Pu, but with different relative intensities, so that their origin can be uniquely established.

• Low Burn-up Plutonium. The various isotopes of plutonium (with the exception of ^{242}Pu) emit gamma rays over a wide range of energies - from 38 to 769 keV. Accordingly, the isotopic composition can readily be determined (^{242}Pu excepted), with low burn-up plutonium clearly distinguishable from the high burn-up product found, typically, in spent power reactor fuel. Just as for the uranium isotopes, the relative intensities measured for gamma rays of various energies will be heavily dependent upon the configuration of the originating material. The content of ^{242}Pu in a sample of material may be determined, in favorable cases, by isotopic correlations, or alternatively, by measuring radiations from the capture of resonance-energy neutrons in this isotope (1).

• ^{241}Am. In addition to the well-known, intense, 59 keV gamma ray emitted by ^{241}Am, less intense, but more penetrating gamma rays with energies up to 722 keV are emitted. In certain instances, where the 59 keV radiation may be largely screened by intervening material, these may be more useful in determining the relative amounts of ^{241}Pu and ^{241}Am in the source, and thus, the age of the material since chemical purification.

• ^{233}U. The gamma-ray spectrum of ^{233}U, with gamma rays ranging in energy up to 366 keV, is shown.

• ^{228}Th. When ^{233}U is produced by the neutron irradiation of ^{232}Th (thorium fuel cycle), inevitably, a certain amount of 70-year ^{232}U is also produced, which, in its decay, produces the 1.9-year isotope ^{228}Th. The decay of ^{228}Th is accompanied by the emission of numerous gamma rays, the most intense with energies of 238, 511, 583, 727, and 2614 keV. These serve both to identify and locate ^{233}U and also may be used, as in the case of ^{241}Am present in plutonium, to determine the elapsed time since the material last underwent chemical separation. Gamma rays from ^{228}Th are also observed in the spectra of some samples of high-enriched uranium, both of U.S. and foreign origin. These gamma rays may serve as an identifier for a particular item or batch of material and may also provide information on the history of the material.

• ^{237}Np. The numerous low-energy gamma rays emitted in the decay of ^{237}Np also provide a unique identifier for this material.

In addition to the gamma rays emitted in the decay of the heavy isotopes cited above, other possibilities exist for the utilization of gamma-ray spectrometry to determine the properties or history of nuclear materials. For example, it is well known that the yield of neutrons produced by the alpha-n reaction in the fluorides of plutonium or uranium-PuF_4, UF_4, UF_6 is high (580 n/sec-g U^{234} in

UF_6). The alpha-n reaction on fluorine produces the 2.9-year isotope ^{22}Na as an end product. Thus, this isotope will grow in over time, and after several half-lives, its decay rate will approach the rate of the alpha-n reaction which produces it. The decay of ^{22}Na is accompanied by two directionally correlated 511 keV gamma rays and another with an energy of 1274 keV. If it is possible to observe these correlated gamma rays from, for example, small cylinders of UF_6, it should be possible to thus determine the elapsed time since the cylinders were filled, or possibly, since the material existed in gaseous form.

The gamma-ray spectra of these materials thus may be utilized in the following areas:

- The detection and identification of certain materials, at a distance which will depend upon the quantity and type of material, amount of shielding, background, etc.

- The establishment of a "fingerprint" which characterizes material of a certain type, quantity, and configuration. This has evident utility for robust and unambiguous confirmatory measurements on items either in storage or undergoing transfer.

- Properties of an individual item or batch of material, such as the elapsed time since processing, or other properties related to its individual history.

APPLICATIONS

Controlled Intrusiveness Verification Technology (CIVET) While high-resolution gamma-ray spectrometry can provide a high level of assurance that a given item containing nuclear materials is what it purports to be, the detailed information provided may also reveal sensitive design information about the item, especially in the case of a treaty-limited item such as an assembled nuclear warhead or its individual sub-assemblies or components. This classic conflict between high-quality verification and risk to sensitive information through intrusive measurements has usually been addressed by the use of low-resolution sensors which blur or reduce spectral detail, thereby reducing transfer of sensitive data but also providing low-confidence verification.

In 1988 Brookhaven National Laboratory (BNL) staff members proposed a novel technical approach for verification of warheads in a dismantlement treaty context which would address this problem, preserving the reliable, high-quality information available from HRGS or other sensors, but at the same time masking this sensitive information from inadvertent disclosure. BNL proposed that a simple but suitably powerful personal computer could acquire and analyze the high-resolution data using algorithms which evaluate the validity of the data, but would display only the outcome of the evaluation to the inspector. The basic CIVET concept is to build sufficient "artificial intelligence" into the computer-based system that the instrument can reach a reliable verification conclusion based on highly intrusive measurements without ever revealing to the instrument operator (or other individual) any of the data on which the conclusion is based. A prototype CIVET system using optical imaging to simulate radiography was first developed and demonstrated to DOE (NN-20) in 1991. Since then, work has focused on development of a CIVET High-Resolution Gamma-Ray Spectrometry (HRGS) system. A prototype system was constructed at BNL and tested on several types of U. S. nuclear weapons in a DoD storage complex in July, 1994.

The prototype CIVET HRGS system was intended for verification that items presented for dismantlement under a nuclear arms reduction agreement are actually nuclear weapons and not fraudulent or surrogate materials. The system compares certain characteristics of

the gamma-ray spectrum of an item being verified with stored data from a previously-measured item of the same type which can reliably be accepted as genuine. The stored data, or "templates," are kept under joint custody (of the parties to the agreement) to preclude compromise of the data.

To prevent disclosure of sensitive information, a CIVET instrument must be incapable of clandestine storage or transmission of information gathered in the course of a verification measurement. In practice, this dictates the use of specially designed hardware. In addition, the system software must be designed not only to reach reliable verification conclusions but also to protect sensitive information. Accordingly, the BNL prototype CIVET HRGS system is based on a specially designed computer combined with special software.

The system design had two objectives: (1) to make it possible to verify that the hardware has no clandestine data storage or transmission capability, and (2) to provide the inspected party with high confidence that its sensitive data would not be compromised in the course of verification. The CIVET HRGS computer consists of an IBM-compatible 80C186 CPU, a set of field-programmable gate arrays (FPGAs), programmable logic devices (PLDs), and removable PCMCIA memory cards. All system memory is removable and there is no data storage hardware (disk drives, RAM, or ROM) other than the PCMCIA cards. The first design goal is accomplished by use of only three types of integrated circuits (the 80C186, FPGAs, and PLDs) and elimination of all non-removable storage addresses. The second design goal is accomplished by the use of removable memory cards for all system storage addresses.

The software for the prototype CIVET HRGS system is written in a combination of C, FORTRAN, and assembly language. To minimize the complexity of the system (i.e., to make it possible to verify that it cannot store or transmit data clandestinely), no operating system is used. The system software includes all necessary hard wired interface and control functions. In addition to these functions, the software controls:

(1) acquisition and processing of gamma-ray spectra of check sources to verify the energy calibration;

(2) acquisition and storage of template data (on removable PCMCIA memory cards);

(3) acquisition of data from objects being verified;

(4) comparison of object and template data; as well as

(5) display of the comparison results.

The software uses a simple approach to spectrum data acquisition and reduction. The system has a fixed calibration of .25 keV per channel and an 0.0 keV intercept, so that the channel positions of all peaks are known. Objects are counted for 30 minutes. At the end of this period, the areas of seven peaks (^{235}U at 186 keV, and ^{239}Pu at 129, 375, 414, and 451 keV, and ^{238}U at 761 and 1001 keV) are computed. These peak areas and their associated statistical uncertainties are stored on a PCMCIA card for use as a template, or in system memory for comparison with a template. The comparison algorithm used is a statistical evaluation of the ratios of the areas of the peaks in the template to those in an item being verified.

The only outputs displayed by the system during data acquisition and analysis indicate whether or not acceptable data were collected. Template comparison results are displayed as an overall ratio between the item being verified and the template, the uncertainty in this ratio, and a figure of merit for the

ratio. No sensitive information can be derived from any of the system outputs, but the displays provide enough information to judge whether or not the results are valid.

Two important conclusions can be reached from the measurements carried out in 1994 with the CIVET HRGS system:

- Each particular type of assembled nuclear device has an individual gamma-ray "template" associated with it which differs substantially from that of any other type of device or of a different "mod" of the same device when that modification involves changes in the physics package. The statistical tests applied to the gamma-ray data in the CIVET technology have a very high level of confidence in distinguishing one type of device from another or from some type of surrogate.

- Repetitive measurements on a number of devices of the same type show that the variability in the gamma-ray spectra from one to the next is very small, consistent with known fabrication tolerances.

HRGS measurements carried out more recently have indicated that it is possible to obtain more detailed information on an item by measuring not only the "global" spectrum of the entire item but, with appropriately designed collimation and shielding, to measure spectra of individual regions or components. The data obtained are evidently closely related to the materials and configurations of these components.

In conclusion, the following statements can be made with respect to the utility of high resolution gamma-ray measurements in nuclear safeguards and nonproliferation activities:

- Each type of item containing nuclear materials in a given configuration will have a characteristic gamma-ray "fingerprint" which will serve as a signature for that item.

- With the combined utilization of high resolution gamma-ray spectrometry and the CIVET technology, it is possible to obtain highly reliable data confirming that a particular item is what it purports to be, while at the same time protecting very sensitive data from inadvertent disclosure.

- Each of the nuclear materials present in an assembled nuclear device not only emits its own characteristic gamma-ray spectrum but also functions as an absorber for the gamma rays emitted by other materials in its vicinity. For this reason, any removal of material, or substitution of one material for another, will cause a change in the characteristic gamma-ray signature of the device, even though low-energy gamma rays emitted from some regions of the device may not be detectable from the exterior.

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ACKNOWLEDGMENT

This work performed under the auspices of the U.S. Department of Energy, Contract No. DE-AC02-76CH00016.

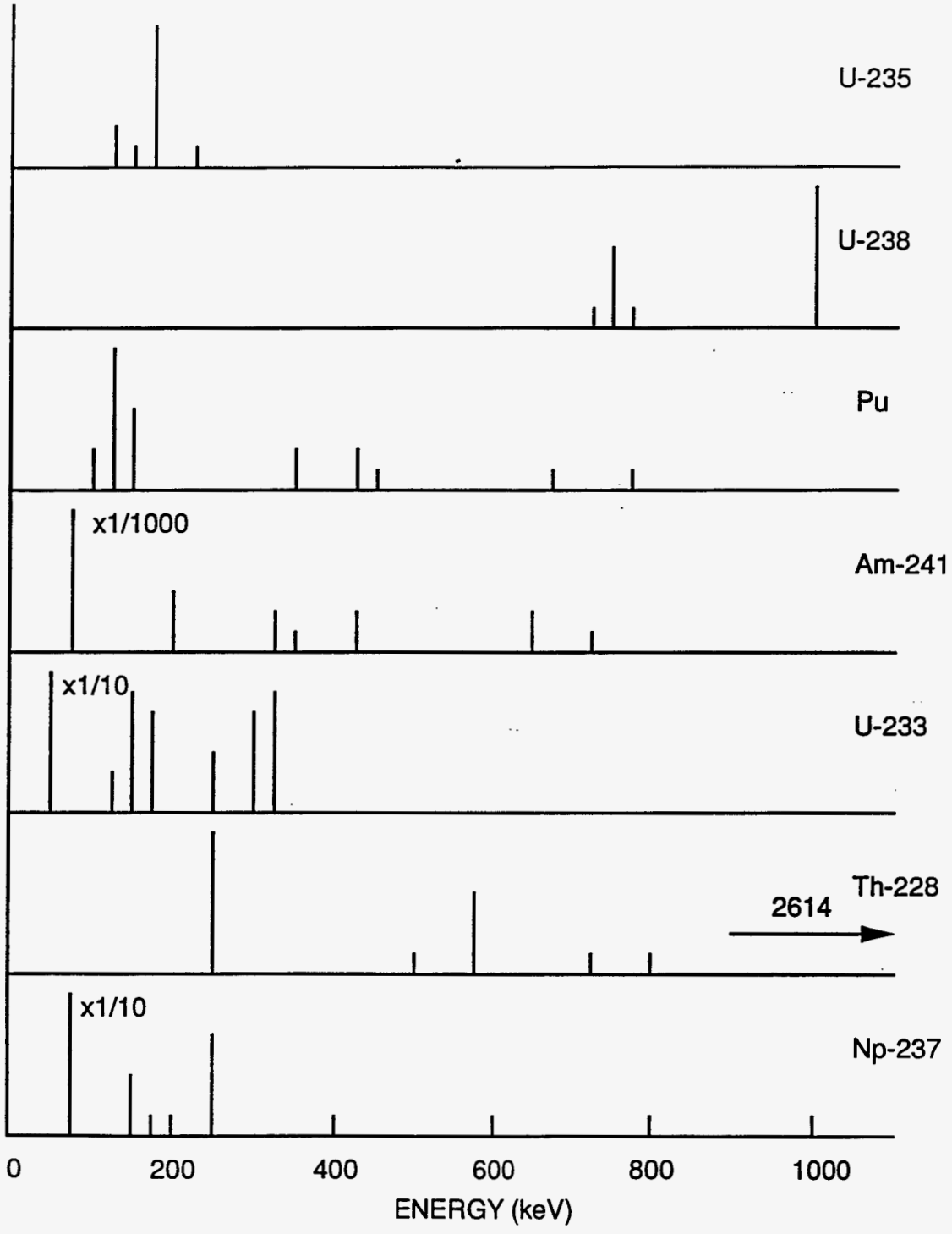

Figure 1. Gamma-Ray Spectra of Important Nuclear Materials

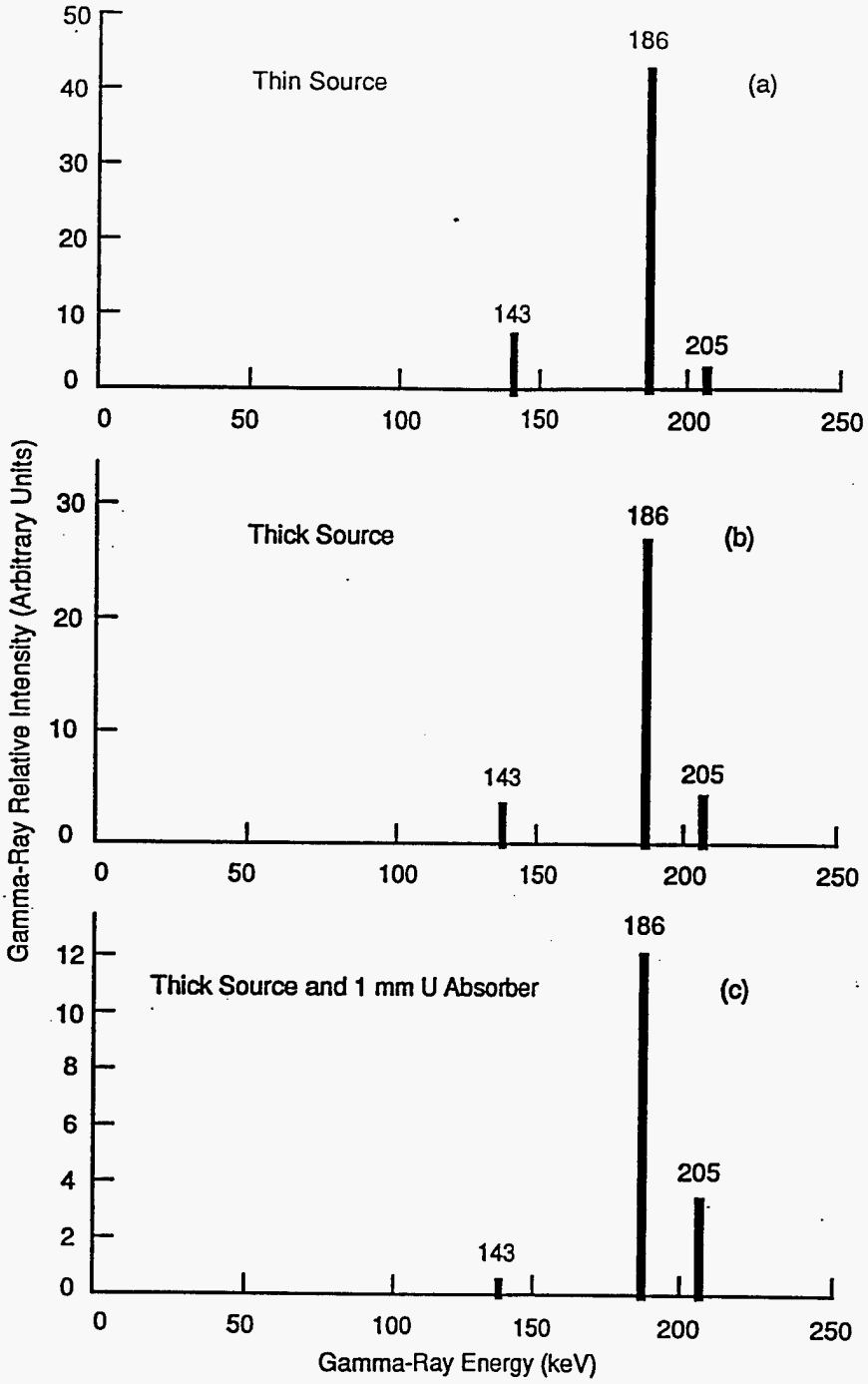

Figure 2. Relative Intensities of Low Energy Gamma Rays of ^{235}U for a Thin Source (a), Thick Source (b), and Thick Source with a 1 mm U Absorber (c).

Report Number (14) BNL--64032
CONF-970744--

Publ. Date (11) 199707
Sponsor Code (18) DOE/NN, XF
UC Category (19) UC-900, DOE/ER

DOE