A NEW METHODOLOGY FOR DETERMINING FISSILE MASS IN INDIVIDUAL ACCOUNTING ITEMS WITH THE USE OF GAMMA-RAY SPECTROMETRY*

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Abstract

In the safeguards, arms control, and nonproliferation regimes measurements are required which give the quantity of fissile material in an accounting item, e.g., a standard container of plutonium or uranium oxide. Because of the complexity of modeling the absorption of gamma rays in high-Z materials, gamma-ray spectrometry is not customarily used for this purpose. Gamma-ray measurements can be used to determine the fissile mass when two conditions are met: 1. The material is in a standard container, and 2. The material is finely divided, or a solid item with a reproducible shape. The methodology consists of: A. Measurement of the emitted gamma rays, and B. Measurement of the transmission through the item of the high-energy gamma rays of Co-60 and Th-228. We have demonstrated that items containing nuclear materials possess a characteristic "fingerprint" of gamma rays which depends not only on the nuclear properties, but also on the mass, density, shape, etc.. The material's spectrum confirms its integrity, homogeneity, and volume as well. While there is attenuation of radiation from the interior, the residual radiation confirms the homogeneity of the material throughout the volume. Transmission measurements, where the attenuation depends almost entirely on Compton scattering, determine the material mass. With well-characterized standards, this methodology can provide an accurate measure of the contained fissile material.

INTRODUCTION

An increasingly large fraction of the fissile material in existence in the United States and other countries is utilized or stored in the form of accounting items with a standard, reproducible geometry, including, for example, metal parts, reactor fuel elements, containers of plutonium or uranium compounds, and individual components utilized in critical assemblies. In particular, in the United States and the Russian Federation there exist of the order of 100,000 small items utilized in critical facilities, each containing, typically, from about 50 to several hundred grams of fissile material.

While practically all measurements on nuclear materials generate three or fewer individual numbers from which the fissile mass or isotopic composition is determined, high-resolution gamma-ray spectrometry (HRGS), in contrast, is characterized by the wealth of data it produces. Typically, in a given special nuclear material there will exist not one but two or more individual nuclear species, each emitting a gamma-ray spectrum with energies ranging from the x-ray region to some upper

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energy typical of that nuclide. In addition, for most items, the spectrum obtained will depend on the relative orientation of that item with respect to the detector, and carefully selected collimation will provide additional information. Thus, HRGS will provide several arrays of data characterizing a given item. It is useful to consider these arrays as "vectors", in contrast to the individual numbers (scalars) which result from most other measurements. They constitute a highly redundant data set which depends not only on the nuclear properties of the individual nuclides but also on the macroscopic properties of the item--mass, density, shape, etc.

BASIC PRINCIPLES

Gamma-Ray Attenuation

The attenuation of the principal gamma rays from high enriched uranium (HEU) as a function of energy is shown in Fig. 1. While this attenuation is strong for low-energy gamma rays, the sensitivity of HRGS is such that even highly attenuated lines can still be observed and measured with satisfactory precision, typically down to about 5% of their original strength or less. The attenuation, rather than representing an obstacle to the measurement of the material, conveys information on the geometry and other macroscopic properties. Thus it is still possible to measure the 185.7 keV gamma ray through about 1.5 mm, the 1001 keV line through about 2 cm, and the 2614 keV line through more than 3 cm of intervening uranium metal or an equivalent material. In one application the 185.7 keV gamma ray was useful though it was attenuated by a factor of 100.

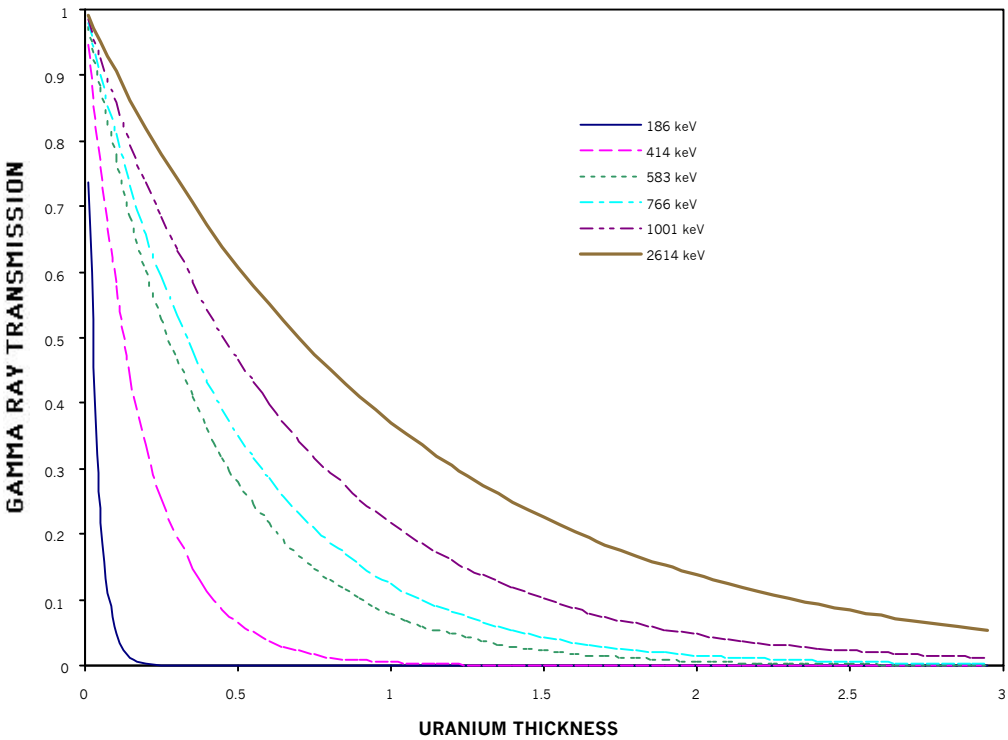

Figure 1. Transmission of Gamma Rays of Various Energies Through Uranium

Depending upon the thickness and density of the fissile material in the item in question, it is possible to obtain or confirm the sectional density (g/cm^2) of this material through a precise measurement of the relative intensities of individual x rays and gamma rays, where the choice of peaks to be measured will depend upon the sectional density. For thin foils or dilute materials, x rays or low energy gamma rays are most useful; for thick materials the higher energy gamma rays of plutonium or of the U-232 impurity in HEU can be utilized.

As an example, we consider the x rays emitted from a thin HEU sample. For the strong 185.7 keV transition, which occurs in the Th-231 nucleus after the alpha decay of U-235, approximately 9% of events occur through the ejection of a K-shell electron (internal conversion) from the residual thorium atom, with the subsequent emission of a K x ray in practically all cases. Thus, in addition to the 185.7 keV gamma ray (43,000 /g-sec) the source will emit thorium x rays with an intensity of ~4,000 /g-sec. The mean free path for these x rays in uranium is of the order of 0.2 mm, so that the total escaping from the surface will saturate for a sample of ~0.6 mm or more in thickness. The 185.7 keV gamma ray is also strongly absorbed, but with a mean free path of ~0.5 mm. This occurs almost always by photoelectric absorption in the uranium atoms, with the subsequent emission of uranium x rays. Thus, for increasing source thickness, the ratio of the observed intensities of the uranium and thorium x rays will increase from zero in the limit of thin sources to more than 10 in the limit of a thick source (~2 mm of U metal). The same considerations apply to the strong 129.3

keV gamma ray emitted in the decay of Pu-239, where the competing processes of internal conversion and fluorescence will generate x rays from both uranium (the alpha-decay product of Pu-239) and plutonium. Since in these examples, the x-ray and gamma-ray intensities can be measured with high precision, they can provide a sensitive measure of the surface density of the material (g/cm^2) for an item that has a regular and reproducible geometry. This approach could also be applied to the assay of dilute solutions of uranium and plutonium contained in a flat cell of known dimensions.

For low energy gamma rays the same principles will apply. As a second example, we consider the gamma rays emitted from U-235. In the vicinity of the 185.7 keV transition, several satellite lines exist, with intensities, typically, of the order of one tenth that of the 185.7 keV line. We consider two of these lines, at 143.8 and 205.3 keV, respectively. The gamma-ray absorption cross section of uranium changes rapidly with energy in this region, so that the mean free path of the 143.8 keV radiation will be considerably shorter, and that at 205.3 keV, considerably longer than at 185.7 keV. Consequently, the measured relative intensities of these three lines will be a sensitive measure of the source thickness. This is shown in Table 1, where the relative peak intensities are shown for a thin source (original branching ratios in the nuclear decay) and the saturated values for a thick source (~1 mm of U metal, ~ 2 g/cm^2).

Table 1. Observed intensities of the 143.8, 185.7, and 205.3 keV gamma rays of U-235 at the limits of thin and thick sources.

Eγ(keV)	Iγ/I(185.7): Thin	Iγ/I(185.7): Thick
143.8	0.18	0.11
185.7	1.00	1.00
205.3	0.09	0.12

Between the two limiting cases, the relative intensity of the 143.8 keV peak is seen to decrease by 39% and that of the 205.3 keV peak to increase by 33%. Since these ratios can be measured with high precision, they afford a quantitative confirmation of the quantity of material in, for example, the plates and disks which ordinarily constitute the fuel elements in a critical assembly.

For thicker items, higher energy gamma rays may be utilized for the same purpose. For HEU, the most suitable gamma rays are emitted in the decay of U-232 (69 yr), a ubiquitous contaminant found in essentially all HEU in both the United States[1] and Russia[2]. This isotope is produced when U-235 is irradiated in a reactor; the recycling of such material through gaseous diffusion enrichment plants, where there is holdup of a significant fraction of any uranium passing through the cascade, led to the contamination of these plants with U-232 and its subsequent presence in all HEU produced thereafter. Oralloy produced in American enrichment plants typically contains about 100 to 200 parts per trillion of this isotope; comparable material enriched in Russia appears to have a moderately higher concentration. For enriched uranium containing ~80% or more U-235, the gamma rays of U-232 are readily measurable. While the concentration of U-232 will vary from one HEU sample to another, the intrinsic intensity ratios of the gamma rays emitted will be invariant, and since they span a wide energy range, with corresponding mean free paths ranging over nearly two orders of magnitude, they can be used to determine the surface density of items over a wide range of thickness. While other isotopes of interest do not have spectra as rich in peaks as U-232,

their spectra are still useful. For U-238, for example, the mean free paths of the 766 and 1001 keV gamma rays in uranium differ by abut 30%, so that their ratio will depend upon the thickness of the emitting item. For Pu-239 the most energetic gamma ray useful for a determination of the source thickness is at 769 keV, measurable through about 1.5 cm of plutonium metal. For items with more complicated geometrical shapes, it will not usually be possible to define a "source thickness", but the observed gamma-ray spectrum will be highly dependent upon the macroscopic properties of the item, affording a unique signature or "fingerprint" of that particular item, testifying to its authenticity. In earlier work we developed computer algorithms which compare the spectrum of a given item with a previously obtained "template", applying statistical tests to the relative intensities of the peaks in the two spectra, to confirm the validity of the item in question[3]. A similar procedure has been employed in Russia for some time, where a low-resolution scintillation spectrum and the gross neutron count for a given item are obtained and used as a "passport" to confirm the authenticity of an item in later confirmatory checks.

Transmission Measurements

In addition to the information provided by the intrinsic gamma rays emitted by an item, additional information can be obtained by measurements of the transmission through the item of the gamma rays emitted by an external source. The isotopes Co-60, emitting gamma rays with energies of 1173 and 1332 keV, and Th-228, which emits a rich spectrum of gamma rays with energies up to 2614 keV, are preferred for this application. They are readily available, have reasonably long half-lives, and the gamma rays cited have energies high enough that their absorption occurs largely by Compton scattering, so that the attenuation in a given item will depend largely on the density and thickness (strictly, the electron density) and be relatively independent of the atomic number of the nuclear material in the item. This information, combined with the information from the intrinsic spectra, which verify the integrity and homogeneity of the material in an item, is sufficient to characterize the material completely.

Examples

1. Critical Facilities. Since many different core configurations must be modeled by critical facilities, these assemblies typically utilize thousands of small fuel elements, each containing from ~20 to several hundred grams of fissile material. The ZPPR assembly, at the Argonne West facility, for example, employs rectangular plates, 5 cm wide, 2.5 to 20 cm in length, and 0.16 to 0.63 cm thick, or 1 cm diameter fuel pins, 7.5 to 15 cm long. The BFS assemblies, at the Institute for Physics and Power Engineering (IPPE), Obninsk, Russia, utilize disks, ~ 5 cm diameter, and ~ 2 to ~6 mm thick, containing from ~20 to ~ 200 g of fissile material. As a specific example, we consider a BFS disk, 4.69 cm diameter, .57 cm thick, containing 157 grams of HEU enriched to 90% in U-235. Three separate measurements are proposed:

Collimated gamma rays. In addition to the 185.7 keV gamma ray of U-235 and those of U-238, the gamma rays emitted from the U-232 chain (strong lines at 238, 583, 727, and 2614 keV) can be measured. The mean free paths of the 185.7 and 238 keV gamma rays are much less than the disk thickness, that of the 583 keV gamma ray is comparable, and the disk will be relatively transparent to the 2614 keV radiation. Thus the ratios of the high and low energy lines will be

uniquely related to the disk thickness and the enrichment can be obtained by the "enrichment meter" principle, and corroborated by the ratio of the 185.7 line to those at 766 and 1001 keV from U-238.

Detector Viewing Entire Disk. Since the lowest energy gamma rays are emitted from a thin surface layer, the ratio of the counting rates for these lines for the collimated and uncollimated measurements provides a check on the stated area of the nuclear material in the disk. The entire spectrum provides a unique "signature" uniquely related to the mass of the material and the geometry of the item. For this simple case, this can be calculated from known gamma-ray absorption coefficients, or it can be measured for a particular item, archived as a "passport", and utilized later for comparison with a confirmatory measurement. In this case the template comparison methods described earlier are useful.

Transmission Measurements. A measurement of the attenuation of the 1173 and 1332 keV gamma rays of Co-60 traversing the disk will provide a precise value for the surface density (g/cm^2) of the material. This method is useful for disks or other items up to several cm in thickness; for thinner items the transmission can be measured at other than normal incidence, or a lower energy gamma ray utilized, for example, the 661 keV gamma ray of Cs-137.

These measurements provide a mutually reinforcing set of data which determine the mass and enrichment of the material and provide strong evidence for the integrity and identity of the item. For items which have already been reliably characterized, it may not be necessary to perform the complete set of measurements on all such items.

2. Canister for plutonium or uranium oxide. In the Russian Federation, oxides of plutonium and uranium are stored or shipped, typically, in double-walled stainless steel canisters. The plutonium canister has an inside diameter of 12.0 cm and a height to the shoulder of 164 cm. The uranium canister is slightly larger, with a diameter of 14.8 cm. We consider the example of a canister containing 3.5 kg of plutonium oxide. If the oxide powder fills the canister to the shoulder, its density will be fairly low, ~$2 g/cm^3$. In this case, the material is fairly transparent to the highest energy gamma ray of Pu-239 with a useful intensity, that at 769 keV. For gamma rays of this energy emitted from the center of the canister, about 35% will reach the surface; for those within 2 cm of the surface, which represent a large fraction of the total mass, most will penetrate to the surface. Thus, the measurement of this gamma ray will sample the entire volume of the material. As for the previous example, the following three measurements are proposed (For all of these measurements, the canister should be situated on a rotating turntable, to average out any non-uniformities and to sample the entire volume uniformly.):

Collimated Gamma Rays. For the fixed geometry of the canister, the measured spectrum of plutonium gamma rays will be directly related to the density of plutonium therein, and since it samples the entire volume, will verify the integrity of the material. The isotopic composition of the plutonium can be determined by standard procedures, and a vertical scan of the container will determine the height of the contents.

Detector Viewing Entire Canister. As in the previous example, the gamma-ray signature of the entire canister verifies the integrity of the material, and can be compared with a previously obtain template or "passport". A further check on the height of the contents can be obtained by comparing the intensity of the 129 keV gamma ray for the collimated and uncollimated spectra, since the intensity of this low energy gamma ray , which is emitted from a thin layer of material at the surface, will, in the latter measurement, be proportional to the surface area viewed by the detector.

Transmission Measurements. A measurement of the transmission of the 2614 keV gamma rays from an external Th-228 source will, as in the previous example, give a direct measurement of the path integral of the density along a diameter of the canister, and hence, in combination with the volume, the mass of material therein. A vertical scan will provide a second determination of the height of the material in the canister, and, if necessary, other transmission measurements can be used to verify the uniformity and integrity of the material.

CONCLUSIONS

For items containing fissile materials which possess a known and reproducible geometry, high resolution gamma-ray spectrometry provides a complete and reliable means for determining the fissile mass and for verifying the integrity of the item. For such items, this can be accomplished in a cost-effective way with existing techniques of gamma-ray spectrometry and template matching algorithms, without the use of neutron coincidence measurements or techniques such as computer assisted tomography, which are required for more complicated problems, such as determining the character and quantity of randomly distributed nuclear materials inside closed containers. This methodology has the advantage that all measurements necessary to characterize or verify the material in an item, including both the isotopic composition and fissile mass, can be performed with one measurement system.

References

1. Anthony J. Peurrung, "Predicting 232U Content in Uranium", Pacific Northwest National Laboratory Report PNNL-12075, Dec. 1998.

2. James Lemley, Walter Kane, Peter Vanier, and Leon Forman, "High-Resolution Gamma-Ray Measurements of Sapphire Material", Proc. 41st Annual Meeting of the INMM, New Orleans, July 16-20, 2000.

3. Peter B. Zuhoski, Jr., Joseph P. Indusi, and Peter E. Vanier, "Building A Dedicated Information Barrier System for Warhead and Sensitive Item Verification", Proc. 40th Annual Meeting of the INMM, Phoenix, July 25-29, 1999.