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Uranium Enrichment Measurements without Calibration
Using Gamma Rays Above 100 keV*

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Abstract

The verification of UF_6 shipping cylinders is an important activity in routine safeguards inspections. Current measurement methods using either sodium-iodide or high-purity germanium detectors require calibrations that are not always appropriate for field measurements, because of changes in geometry or container wall thickness. The introduction of the MGAU code demonstrated the usefulness of intrinsically calibrated measurements for inspections. MGAU uses the 100-keV region of the uranium gamma-ray spectrum. The thick walls of UF_6 shipping cylinders and the low-energy analysis preclude the routine use of MGAU for these measurements.

We have developed a uranium enrichment measurement method for measurements using high-purity germanium detectors, which do not require calibration, and uranium gamma rays above 100 keV. The method uses seven gamma rays from ^{235}U and ^{238}U to determine their relative detection efficiency intrinsically and with an additional gamma ray from ^{234}U, the relative abundance of these three uranium isotopes. The method uses a function that describes the basic physical processes that predominately determine the relative detection efficiency curve. These are the detector efficiency, the absorption by the cylinder wall, and the self-absorption by the uranium contents. We will describe this model and initial testing on various uranium materials and detector types.

1. INTRODUCTION

The traditional uranium enrichment measurement method is based on the measurement of the 186-keV peak and is commonly referred to as the "enrichment meter" method [1]. This method can be used on UF_6 shipping cylinders with either germanium or NaI detector based systems, but is limited in accuracy and requires calibration for each type of sample material or container type. The measurements must be made using the calibration geometry and on a sample with a thickness of several mean paths at 186 keV.

To overcome these limitations, the MGAU code was developed [2]. This code eliminated the need for calibration and demonstrated the usefulness of an intrinsically calibrated uranium enrichment measurement method in an inspection regime. The MGAU code uses the 100-keV region with gamma- and x-ray peaks that are close in energy and reasonably intense for most types of measurements.

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UF_6 shipping cylinders are made of steel and have walls that are approximately 8 to 16 mm thick. A 16mm steel wall reduces the intensity of a 100-keV gamma-ray line by a factor of 250. Measurements on these types of containers have shown that MGAU has limited applicability, when the container wall thickness is greater than 10 mm [3].

An alternative energy region for measuring the uranium enrichment is the 186- to 1001-keV region. Although the 186- and 1001-keV peaks are easy to measure, it is difficult to determine the relative detection efficiency of these two gamma rays because of the large difference in their energies [4]. However, our experience in developing the MGA code [5] has taught us how to approach the determination of the relative detection efficiency based on the physical processes involved.

2. DESCRIPTION OF THE METHOD

The basic method for determining the relative abundance of the uranium isotopes is to measure the intensity of two more peaks from gamma rays of similar energy, but arising from different isotopes as is done in MGA and MGAU. Because the gamma-ray emission probabilities and half-lives are known, the atom ratios can be calculated if the relative detection efficiencies for the peaks can be estimated. Usually a curve is used to describe the relative efficiency over an energy range that is determined by fitting a function to the observed peak intensities divided by their respective emission probabilities. The functions that are commonly used are adequate to describe the relative efficiency of peaks that are closely spaced in energy. However, it is difficult for these functions to accurately relate the relative efficiency of the 186- and 1001-keV peaks, because the energy spacing is so large.

To overcome this, we use a functional form, which was developed for MGA that describes three major components involved in the detection process. These are 1) the detector efficiency, 2) absorption by the steel container walls, and 3) self-attenuation by the uranium in the sample. The advantage of basing the functional form on known physical processes is that its shape is determined not only by the data but also by the physical-interaction processes. Because the variables involved in the three processes are not usually known or controlled, they must be determined from the gamma-ray spectrum itself. The following equation is used to describe these processes as a function of energy:

$$A_j \equiv \sum_{k=1}^{3}(p_{j,k}\times X_k)\times\exp(-\mu_{Fe}(E_j)\times FE)\times((1-\exp(-\mu_U(E_j)\times U))/(\mu_U\times U))\times\varepsilon_j^0\times(1+bE_j+cE_j^2)$$

Where A_j are the areas of eight peaks in the spectrum due to ^{234}U, ^{235}U, and ^{238}U and X_k are the unknown amounts of these isotopes. The gamma-ray emission probabilities are represented by the terms $p_{j,k}$ for each peak j belonging to uranium isotope k. The absorption coefficients, μ_j, at each peak energy are known for steel and the uranium composition, UF_6, but the thickness of the container wall, FE, and the uranium sample thickness, U, are treated as unknown variables. The final term in the equation, ε^0, is the estimated relative detector efficiency for peak j where b and c are unknown variables in a quadratic function used to account for small variations in the efficiency. The term, ε^0, may be calculated using the dimensions of the detector. This equation is very nonlinear in form and therefore the variables must be solved by an iterative least-squares method. The results from this analysis are the relative amounts of ^{234}U, ^{235}U, and ^{238}U and a relative efficiency curve.
Thus the enrichment is determined independent of calibration and standards. The contributions to the relative efficiency curve are shown graphically in Fig. 1.

FIG. 1. The general effect of each of the three processes on the intensity of the peaks is shown superimposed on a typical uranium spectrum along with the total relative efficiency. Fe represents the transmission by the steel and U represents the transmission by the UF_6. *The plot of the residuals also shows the location of the peaks used in the analysis.*

The peak areas used in this analysis are the 120.9-keV gamma ray from ^{234}U, the 143.8-, 163.3-, 185.7-, and 205.3-keV gamma rays from ^{235}U, and the 258.3-, 766.6, and 1001-keV gamma rays from ^{238}U. The areas of all these peaks except the 185.7-keV peak are determined by simple window integration. The 185.7-keV peak is fit with a Gaussian and a low-energy exponential tail to account for the interference with the background from a peak at 182.7 keV.

3.0 MEASUREMENT RESULTS

Most of our initial testing has been performed on various uranium enrichment standards using steel absorbers with different thickness and various sizes of coaxial-type germanium detectors. Detector efficiency is an important parameter in these measurements, because of the use of the 1001-keV gamma ray. Energy resolution is not as important, because we use simple window integration methods for all the gamma-ray peaks except the 186-keV peak.

We measured two of the CRM 969 low-enriched uranium standards with enrichments of 1.94 and 4.46 weight percent using a coaxial-type detector with 20% efficiency. These standards consist of 200 g of U_3O_8 powder sealed in a cylindrical aluminum can of 70 mm inner diameter. We used a 15 mm steel absorber in these measurements with counting times that ranged from thirty minutes to eight hours. The long counting times were taken to reduce the effect of counting statistics on our study of the analysis methodology. Results from the thirty-minute measurements of these two standards are shown in Table I below.

Table I. Measurement results from thirty-minute measurements of two CRM969 standards with a 20% coaxial-type detector.

Declared Enrichment	Measured Enrichment ($\pm$ 1σ % relative)
1.94	1.96 ± 2.8 %
	2.15 ± 2.9 %
	2.00 ± 2.8 %
4.46	4.57 ± 2.8 %
	4.36 ± 2.8 %
	4.60 ± 2.9 %

We also measured the CRM 146 uranium isotopic standards with enrichments of 20.107, 52.488, and 93.1703 weight percent using a coaxial-type detector with 100% efficiency. These standards consist of approximately 230 g of U_3O_8 powder sealed in a cylindrical aluminum can of 70 mm inner diameter. We used a 12.7 mm steel absorber in these

measurements with thirty-minute counting times. We were unable to analyze the data from the 93% standard, because the 258.3-keV peak from ^{238}U was not visible even with counting times up to four hours. Results from the thirty-minute measurements of the other two standards are given in Table II below.

Table II. Measurement results from thirty-minute measurements of two CRM146 standards with a 100% coaxial-type detector.

Declared Enrichment	Measured Enrichment (± 1σ % relative)
20.107	20.29 ± 8.4 %
	20.89 ± 9.6 %
	19.82 ± 11.3 %
52.488	51.94 ± 7.6 %
	51.80 ± 8.2 %
	51.20 ± 7.0 %

We received two gamma-ray spectra of UF_6 cylinders that were taken during a cylinder cleaning project at the Portsmouth plant in Piketon, Ohio. Both of these cylinders were later emptied and mass spectrometry performed on the contents. One measurement is of a 30.5 cm cylinder, which contained 12.49% enriched material. The other measurement is of a 20.3 cm cylinder, which contained 37.59% enriched material. Both cylinders were measured using a coaxial-type detector with nominal 40% efficiency. Each measurement was for one hour with the detector placed close to the cylinder. Our analysis results gave 12.5 ± 2.1% and 38.1 ± 4.4%, respectively.

4.0 SUMMARY

We utilize a unique and accurate technique for delineating the relative detection efficiency that enables us to relate the intensities of widely separated peaks such as the 185- and 1001-keV peaks. As a result, we are able to determine without calibration the enrichment of uranium in containers where the 100-keV region is significantly attenuated. Our initial results demonstrate that our proposed analysis methodology may be utilized on uranium materials with enrichments up to 50%. Thirty-minute measurement times are possible, but a coaxial-type detector with an efficiency of at least 20% is required. The results indicate that measurement precision and accuracy better than ± 10% can be obtained with thirty-minute measurements.

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REFERENCES

[1] T.D. REILLY, R.B. WALTON, AND J.L. PARKER, Los Alamos National Laboratory, A-1 Progress Report LA-4605-MS (1970), p. 19.

[2] R. GUNNINK, W.D. RUHTER, P. MILLER, et. al., "MGAU: A New Analysis Code for Measuring U-235 Enrichments in Arbitrary Samples", UCRL-JC-114713, Livermore, CA 94550, prepared for the IAEA Symposium on International Safeguards, Vienna, Austria, March 14-19, 1994.

[3] PRIVATE COMMUNICATION from S. Abousahl, Institute for Reference Materials, Geel, Belgium, March 1995.

[4] R. LANIER, C. HAYEN, D. CLARK, AND W. PARKER, "Evaluations of the $^{235}U/^{238}U$ Isotope Ratio through thick-walled containers using the 185.7 keV and 1001.0 keV Gamma Rays," UCRL-ID-143390, Lawrence Livermore National Laboratory, Livermore, CA, 94550, ISPO Report 464.

[5] R. GUNNINK, "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances," UCRL-LR-103220, Vol. 1, Lawrence Livermore National Laboratory, Livermore, CA, 94550.