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Author(s): J. K. Sprinkle, Jr., A. Christiansen, R. Cole, M. L. Collins, S. Hsue, P. L. Knepper, T. O. McKown, and R. Siebelist

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Low-Resolution Gamma-Ray Measurements Of Uranium Enrichment

J. K. SPRINKLE, JR.,¹ A. CHRISTIANSEN,¹ R. COLE,¹ M. L. COLLINS,¹ S.-T. HSUE,¹
P. KNEPPER,¹ T. MCKOWN,¹ and R. SIEBELIST¹

¹Los Alamos National Laboratory, Los Alamos, NM 87545 USA

1. Introduction

Facilities that process special nuclear material perform periodic inventories. In bulk facilities that process low-enriched uranium, these inventories and their audits are based primarily on weight and enrichment measurements. Enrichment measurements determine the ²³⁵U weight fraction of the uranium compound from the passive gamma-ray emissions of the sample (Reilly T. D., Walton R. B., and Parker J. L., 1970; Parker J. L. and Brooks M., 1988). Both international inspectors and facility operators rely on the capability to make in-field gamma-ray measurements of uranium enrichment. These users require rapid, portable measurement capability. Some in-field measurements have been biased, forcing the inspectors to resort to high-resolution measurements or mass spectrometry to accomplish their goals.

2. Measurement Physics

The fundamental relationship underlying this technique is: *The intensity of 186-keV gamma rays emitted from a fixed area of a thick sample of uranium is proportional to the ²³⁵U enrichment of the sample.* In this context, "thick" means several mean free paths at 186 keV. In practice, this thickness is generally quite small. Table 1 has some dimensions.

A schematic illustration of the geometry of enrichment measurement is given in Fig. 1. The detector views the uranium sample through a fixed solid angle provided by an appropriate

TABLE I. Linear Sample Dimensions Required for 99.9% Gamma Response

Uranium Compound	Mean Free Path (cm)	Mass Attenuation Coefficient (cm ² •g ⁻¹)	Sample Density (g•cm ⁻³)	Sample Thickness for 99% Response (cm)
U metal	0.036	1.47	18.7	0.25
UO ₂	0.76	1.31	1.0 (solution)	5.27
	0.38		2.0 (powdered)	2.67
	0.69		11.0 (sintered)	0.49
U ₃ O ₈	0.78	1.27	1.01 (solution)	5.44
	0.39		2.0 (powder)	2.72
			8.3 (highly packed powder)	0.66
UF ₆	0.095	1.03	1.0 (solution)	6.80
	0.97		4.7 (solid)	1.43
	0.21		1.0 (solution)	9.10
Uranyl nitrate	1.30	0.77	1.0 (solution)	9.10
UO ₂ (NO ₃) • 6H ₂ O	0.46		2.8	3.25

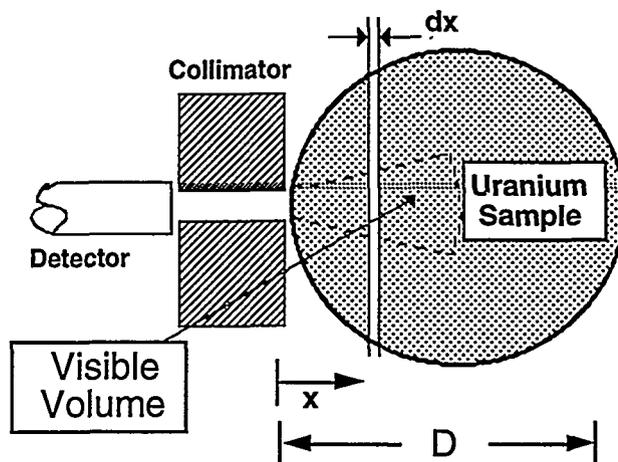


Fig. 1. Schematic illustration of uranium enrichment measurement.

collimator. The sample is infinitely thick for 186-keV gamma rays (transmission ≤ 0.002) and consists of a "uniform" mixture of uranium and matrix (everything else). The density of uranium is ρ_u , and matrix ρ_m . The mass attenuation coefficients at this energy are μ_u and μ_m . The count rate can be expressed by the integral relation

$$CR = \rho_{235} \Gamma T_c \varepsilon A \int_0^{\infty} \exp[-(\rho_u \mu_u + \rho_m \mu_m)x] dx, \quad (1)$$

where ρ_{235} = ^{235}U density,

ε = total counting efficiency,

Γ = 43 000 gamma/s \cdot g ^{235}U ,

T_c = container transmission

A = effective area of sample viewed, and

x = distance within sample to front edge of sample.

Consider the measurement of a sample of thickness D , where the sample-to-detector distance is large compared to the depth of the visible volume. This permits one to neglect $1/r^2$ effects in integrating over the visible volume of the sample.

The viewed area of the sample, A , is a function of the collimator opening and depth. The integral is easily evaluated and yields the expression

$$CR = \frac{\rho_{235} \Gamma T_c \varepsilon A}{\rho_u \mu_u} \frac{1}{\left(1 + \frac{\rho_m \mu_m}{\rho_u \mu_u}\right)}, \quad (2)$$

or

$$CR = \frac{EK}{1 + \frac{\rho_m \mu_m}{\rho_u \mu_u}}, \quad (3)$$

where $E = \frac{\rho_{235}}{\rho_u}$ is the enrichment, and

$$K = \frac{\Gamma T_c \varepsilon A}{\mu_u}$$

is a constant to be determined by calibration with a known standard.

Note that K includes the container transmission (Reilly T. D. et al., 1991, Sec. 7.3.2).

Equation (3) is the fundamental relation that contains all the basic information for measuring enrichment. There is a direct dependence between count rate and enrichment if the density and absorption characteristics of the uranium-bearing samples and containers are constant. Specifically, the $(\rho_m/\mu_m)/(\rho_u/\mu_u)$ ratio and T_c must be constant. This usually implies the same chemical form of the sample and identical sample containers. Equation (3) does not constrain the total density of the sample or the sample-to-detector distance.

If $\mu_m \rho_m / \mu_u \rho_u \leq 0.1$, the observed count rate CR is proportional to the enrichment, regardless of matrix, with an error of 10% or less. For matrix material with $Z \leq 30$, $\mu_m / \mu_u \sim 0.08$. Thus, $\mu_m \rho_m / \mu_u \rho_u \leq 0.1$ if $\rho_m / \rho_u \leq 1$. A half-and-half mixture of uranium and low- Z material would give less than 10% fewer counts than a slab of uranium metal having the same enrichment. A wide range of compounds and mixtures may be measured by comparison to a single standard with only small corrections for the variation in composition. If a standard and a series of unknown samples are known to be the same compound or mixture, the enrichments will be directly proportional to the observed rates. For example, if both standard and samples are pure UF_6 , the term $(1 + \mu_m \rho_m / \mu_u \rho_u)$ can be neglected. If the standard was UO_2 and the unknowns UF_6 , a small correction might be applied.

Below are a few values for $[1 + \mu_m \rho_m / \mu_u \rho_u]$:

Material	$[1 + \mu_m \rho_m / \mu_u \rho_u]$	$[1 + \mu_m \rho_m / \mu_u \rho_u]^{-1}$
U metal	1.000	1.000
UO_2	1.012	0.988
UF_6	1.041	0.961

Use of low-resolution detectors has historically included either hardware or software gain stabilization to compensate for energy calibration drifts caused by the photomultiplier's sensitivity to minor temperature changes. Newer detectors do not require this compensation inside of temperature-controlled laboratories, but in-field conditions still cause significant drifts when external temperatures vary widely.

Use of low-resolution detectors has historically relied on two Regions-of-Interest (ROIs) for the data reduction and analysis. A region from 161 to 211 keV is set around the 186-keV peak,

and a second region from 220 to 270 keV is used to estimate the Compton continuum underneath the desired peak. The calibration equation has often been simplified to

$$E = a S_1 - b S_2,$$

where $S_1 =$ the summed counts in region 1,
 $S_2 =$ the summed counts in region 2, and
 a and b are the calibration constants
determined from two or more well-
characterized standards.

3 . Measurement Equipment

We have used a portable multichannel analyzer (Halbig J. K. et al., 1994) available from Aquila Technologies Group, Inc, for this development. The M3CA contains all the necessary electronics support for typical gamma-ray detectors: the high- and low-voltage supplies, the amplifier, the ADC, and the MCA memory, all in a 10 x 20 x 9 cm unit that can run for six hours on a removable 12-V, 2.2-Ah battery. The M3CA has firmware that allows software gain drift compensation, and uses a serial interface. The serial interface was chosen because it is widely used and available on virtually all computers and potential controllers. The slowness of the serial interface is compensated for by increasing the data reduction that can be done in the M3CA, generally only a few numbers are transmitted for each measurement rather than the complete channel-by-channel spectrum.

We have used a compact shielded NaI(Tl) detector developed to minimize the weight the user must carry from measurement location to measurement location. The shielding covers all directions except the front of the detector, allowing measurements to be made successfully when the user is surrounded by uranium samples, and includes a 1-inch-diameter by 1-inch-deep collimator in front of the 1-inch diameter detector.

This data was obtained with IBM ThinkPads, but the serial port on the M3CA allows the use of a variety of controllers. We have developed software to utilize both WINDOWS-style platforms like the ThinkPad and DOS platforms like the Hewlett-Packard Palmtop computer.

4 . Software

While the basic gamma-ray measurement physics are well-understood and the measurement techniques evolve very slowly, computer hardware and software change very rapidly. Therefore, we did not include a controller in the M3CA. Instead, the user selects the appropriate controller for the application. We are currently planning on continuing to support both DOS and Windows applications for the M3CA, while others have chosen platforms such as the Intermecc datalogger.

The software is being used by personnel with a wide variety of experience and training. Consequently, it is desirable to have a simple, user-friendly interface. This is easier to achieve on a more sophisticated platform like WINDOWS than in a DOS environment. We have issued prototype software for both environments, we have some field experience with several users, and we are working on enhancements to the software. As this instrument is distributed further, we will continue to refine the user interface.

The software includes functions to control the MCA; to change parameters such as amplifier gain or detector high voltage; to archive data, results, and MCA parameters; and to calibrate the instrument. Since a wide variety of expertise is expected in the users, it is also important to build in simple diagnostics that inform the user as soon as possible if the instrument begins to malfunction.

5. In-Field Experience

Some facilities have sufficient quantities of minor uranium isotopes to bias the traditional low-resolution enrichment measurement. Generally, low-resolution measurements of "clean" uranium can be made to 1% precision over nearly the entire range of uranium enrichment. High-resolution measurements have been made to 0.1% (Parker J. L. and Brooks M., 1988). Measurements in both Europe and the Former Soviet Union have observed bias effects of 5-10% in low-resolution measurements, caused by minor isotopes of uranium. This level of bias has been deemed unacceptable, causing some inspectors to resort to more expensive, time-consuming alternatives like mass spectrometry or liquid-nitrogen-cooled high-resolution detectors. As the international safeguards community attempts to inspect more facilities with less resources, these alternatives become highly undesirable.

The minor isotopes come from the use of uranium recycled from reactors being used as feed in the enrichment plants. Daughters of ^{232}U or ^{236}U include the thorium decay chain which emits a 238.6-keV gamma ray from ^{212}Pb . This gamma ray falls in the ROI used to estimate the Compton continuum under the 186-keV gamma ray. In addition, the multitude of high-energy gamma rays from the daughters of ^{232}U change the shape of the Compton continuum that lies under the desired 186-keV peak. Just to make the entire issue more challenging, the level of this interference varies widely among the samples generally offered to the inspector, causing unpredictable, wide variations in the bias effects.

We are investigating two approaches to solving this minor isotope problem—medium resolution detectors and more sophisticated analyses of the spectrum. Cadmium-telluride detectors show a lot of promise in being able to resolve the 238.6- and 186-keV gamma rays. The improved resolution should also drastically reduce the effects of changes in the shape of the underlying Compton continuum. However, the emission rate for these measurements is low enough to require bigger detectors than have been yet fabricated if one wants to have reasonable count times like 100–1000 seconds. The alternative approach is to apply more sophisticated analysis methods to the low-resolution spectra. To date, we have completed a study that evaluated small changes in how the net peak area is determined. If the background ROI is moved to the energy range 240 to 290 keV, the effects are significantly reduced. Moving it farther reduces the effects from the 238.6-keV line, but increases the sensitivity to the changes in the Compton continuum. If a three-ROI analysis is applied, wherein a background region on both sides of the peak is used to draw a line under the peak to estimate the Compton continuum, it works well for higher enrichments. For natural and depleted uranium, the three-ROI analysis has a bias that depends on the magnitude and cause of the underlying Compton continuum. Different contaminant isotopes cause different shapes.

We are initiating a study using wide-energy-range response functions for analysis of these spectra. This study is not yet ready to report.

6. Summary

We have developed smaller, lighter hardware (MCA and shielded detectors) that has been successfully transferred to the commercial sector. We have developed software that has received limited testing, the next release should be sufficiently robust for wide release to the general public. We have had initial success with more sophisticated analysis methods. We are pursuing medium-resolution detectors and response function fitting in the expectation that both will cause significant improvements in this measurement technique.

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