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Reference Materials for Nondestructive Assay of Special Nuclear Material

Volume 2: Thin Metal Foils of Highly Enriched Uranium

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Volume 2: Thin Metal Foils of Highly Enriched Uranium

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REFERENCE MATERIALS FOR NONDESTRUCTIVE ASSAY OF SPECIAL NUCLEAR MATERIAL

Volume 2: Thin Metal Foils of Highly Enriched Uranium

by

J. K. Sprinkle, R. N. Likes, and H. A. Smith

ABSTRACT

This manual describes the fabrication of reference materials for use in gamma-ray-based nondestructive assay of small high-density uranium samples. The sample containers are small Petri dishes. The reference materials consist of thin circular discs of highly enriched uranium metal foil. The ^{235}U content ranges from 0.2 to 10 g. The manual also describes the assay procedure used with low-resolution detectors.

PART 1

PRODUCTION OF HIGHLY ENRICHED URANIUM REFERENCE MATERIALS FROM THIN METAL FOILS

I. INTRODUCTION

A. Role of Reference Materials in Nondestructive Assay

Reference materials provide two functions in nondestructive assay (NDA). They are used to calibrate NDA instruments and to verify the constancy of the calibrations. Traceability is important only for the calibration aspect. Constancy of the calibration can be verified with any material that provides a stable, consistent signal. Clearly, a calibration is misleading if the reference material loadings are not well known. However, the loading values need not be known to much greater accuracy than that achievable by the assay. If a reference material is known to one-fourth the expected measurement uncertainty, it contributes little to the overall uncertainty of the measurement. (The investigation of systematic effects has more rigorous requirements on the accuracy requirements for the reference materials.)

The reference materials should represent the samples in aspects to which the particular measurement technique is sensitive. However, contrary to popular thought, exact representation is not necessary if the measurement physics is well understood and properly applied. For example, for transmission corrections, the reference materials must have transmissions for which a correction can be made accurately, but they do not need precisely the same transmission as each sample.

The second function (that of verifying the constancy of the instrument's calibration) merely requires a stable reference material. Because the technique is nondestructive, the same sample can be used repeatedly to verify the same instrument response. It is much easier to make a stable reference material than to ensure that multiple reference materials are accurate and appropriate in all necessary ways.

B. Measurement Applications

The measurements addressed in this manual concern the bulk assay of uranium samples using transmission-corrected gamma-ray assay techniques. The emphasis is on small samples of high-density material with uniform uranium dispersion. The transmission of a 185.7-keV gamma ray through the sample must be finite and measurable. The low-resolution technique described in Part 2 becomes difficult to use at transmissions below approximately 5%. Changing to a high-resolution detector and a different transmission source allows this technique to be extended to transmissions below 1%.

As the sample transmission approaches zero, the 185.7-keV assay changes character significantly and a quantitative assay becomes impossible. The operator may be able to use an enrichment measurement plus a sample weight and an assumption of uniformity to yield a ^{235}U assay. The high-energy gamma rays from the ^{238}U daughter, ^{234m}Pa, may be used if (1) the material is more than a hundred days old (that is, if more than a hundred days have elapsed since the protactinium was separated), (2) the count rate is sufficient, and (3) the sample composition is sufficiently uniform. But the transmission-corrected 185.7-keV assay is meaningless for those samples that have a transmission of zero at 185.7 keV.

C. General Character of the Selected Reference Materials

The uranium loadings were chosen to be typical of those expected to undergo assay, allowing for accurate transmission measurement and, in turn, accurate transmission correction. The nominal ^{235}U loadings (0.2, 0.5, 1, 2, 5, 10 g) cover the range of expected sample loadings. Therefore, the reference materials have sufficient material to allow for good counting precision, and reasonable transmissions are obtained (20 to 90%). All of the samples will be in 4-cm-diameter, 1-cm-high Petri dishes; consequently, those dimensions were chosen for the reference material container. Other containers may require different corrections.

II. PREPARATION

A. Desired Characteristics

The reference materials do not need to resemble the sample closely,[1] but the calibration will be easier if they do. Reliable corrections must be applied to account for the sensitivity of the measurement technique to certain characteristics. The important parameters for a passive measurement technique are the distance that the gamma radiation must travel to the detector, the gamma-ray energy, and the attenuation it suffers. Therefore, computation of a correction for the gamma-ray attenuation requires that the reference material (and sample) be homogeneous; lumps of gamma emitters or absorbers can decrease the amount of gamma radiation emitted from the reference material with no indication to the user from the bulk transmission measurement. In addition, the reference material must be stable with respect to settling and migration of the special nuclear material.

If the reference materials and samples are the same size and packaged in similar containers, the same correction factor (CF) expression applies to both. Otherwise, a different CF is applicable even when the transmissions are identical. The CF explicitly corrects for different transmissions and different sample geometries. The CF can be a significant source of uncertainty in the assay if the transmission is very low. Consequently, a lower limit applies for permissible transmissions.

It is more important that the reference materials verify the calibration and operation of the instrument than that they span the range of representative loadings. Similar loadings for the sample and reference materials may be used, but are not required. In many cases, adequate measurement precision obtained from high loadings is more important than the use of similar loadings.

B. Production Using Thin Metal Foils of Highly Enriched Uranium

Thin metal foils exhibit the desired characteristics of stability and homogeneity that reference materials require, without sacrificing the capability of measuring a transmission at 185.7 keV. Metal foils of highly enriched uranium can be produced in the desired thicknesses from cast billets by standard hot, warm, and cold rolling techniques. After the appropriate thickness is obtained, circular disks are cut from the metal foil. The rough cut can be cleaned up to obtain a uniform circle with smooth edges by sandwiching the uranium foil between acrylic sheets and turning the sandwich on a lathe. Then the foil alone is etched to remove the oxide, weighed, and coated with Krylon Crystal Clear.* The outer coating is very important for the thinnest foils, which tend to flake apart during oxidation.

C. Preparation Results

Twelve new reference materials and two transmission sources are shown in Fig. 1. Two reference materials with different masses were cut from each foil thickness, one with a diameter of 3 cm and one with a diameter of 4 cm.

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*Trademark for clear acrylic spray.

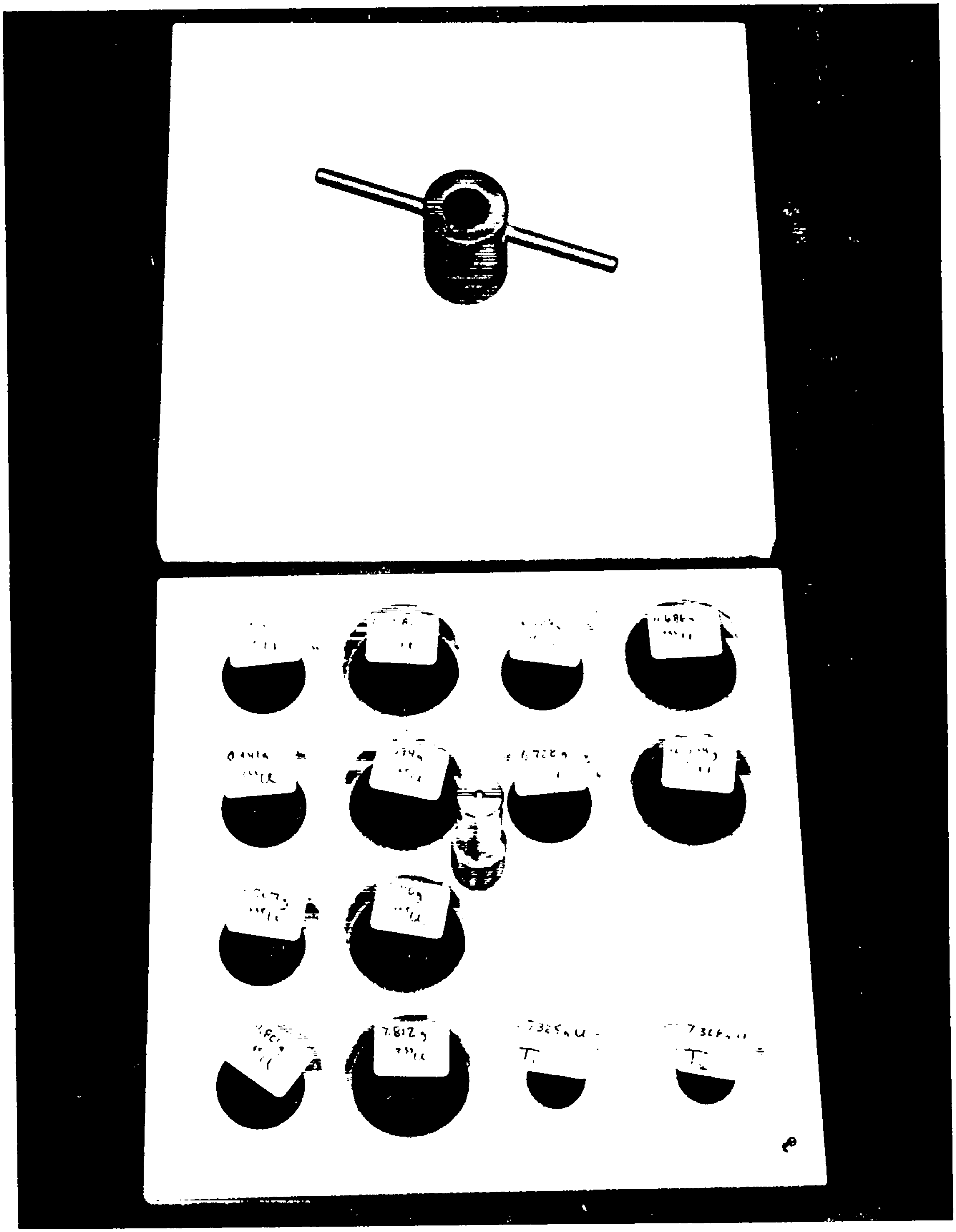

Fig. 1. Twelve foil reference materials and two transmission sources.

The verification data indicate that the detector responses are independent of the reference material diameters. The two transmission sources have a smaller diameter (2 cm), to preclude the possibility of their shining around a sample directly into the detector. Rubber cement was used to bond the foils to the Petri dishes. The Petri dishes can be handled extensively, however severe mishandling could break the rubber cement. Any movement of the foil inside the Petri dish could bias the assay. The Petri dishes should not be opened, and care should be taken to avoid external contamination.

The transmissions through the foils at 185.7 keV range from 0.20 to 0.95. The isotopic analysis for ^{235}U varies from 93.15 to 93.40% for the different thicknesses. Table I summarizes the uranium loadings for each Petri dish. The uncertainty in the uranium mass is 0.001 g. The fourth column lists the nominal thicknesses of the foils before they were etched to remove the oxide.

III. VERIFICATION

A. Introduction

Because the traceability of these reference materials is straightforward and relies on very few assumptions, only two assay procedures were used to verify the uranium content of the new reference materials. It is quite reasonable to assume that the metal foil is homogeneous and that its mass is due to uranium alone. The high-resolution procedure provides assays with less than 1% uncertainty; the low-resolution procedure provides assays with a few percent uncertainty. Reference 2 provides a detailed explanation of gamma-ray assay techniques.

B. Calibration Reference Material

The foil used to calibrate the assay systems has been compared to several solutions.[1] It has an enrichment of 93.15% and a uranium mass of 2.51 g; consequently, its ^{235}U mass is 2.34 g. The foil is 0.001 in. thick.

C. Low-Resolution Spectroscopy

Gamma-ray assays with low energy resolution usually involve high-efficiency detectors (such as NaI detectors) and hence provide high count rates and

TABLE I

TOTAL URANIUM AND URANIUM-235 MASSES OF THE REFERENCE MATERIALS

ID[a]	Uranium (g)	^{235}U (g)	Nominal Thickness (in.)
8778-A	0.208	0.194	0.001
8778-B	0.384	0.358	0.001
11404-A	1.010	0.941	0.003
11404-B	1.710	1.594	0.003
1412-1-A	3.440	3.207	0.010
1412-1-B	6.125	5.710	0.010
1455-4-A	5.154	4.801	0.0135
1455-4-B	8.387	7.812	0.0135
1456-4-A	6.990	6.529	0.022
1456-4-B	12.512	11.686	0.022
1456-2-A	7.203	6.728	0.024
1456-2-B	13.436	12.549	0.024
Trans-A	7.325	6.823	0.050
Trans-B	7.308	6.807	0.050

---

[a] The A and B designations following the ID numbers denote the 3-cm-diameter and 4-cm-diameter foils, respectively.

consequently good statistical precision. However, assays based on low-resolution detectors are not as reliable as those based on high-resolution detectors. Small interferences from gamma-rays with energies close to the gamma-ray of interest can be impossible to resolve and can bias an assay significantly. It is also a nontrivial matter to determine the Compton continuum under a peak.

Although far-field assays are relatively simple to execute, their use is not appropriate for very small samples. Unfortunately the low count rates from less than 1 g of ^{235}U in some of the foil samples require the samples to be much closer to the detector than the far-field procedure allows. The small sample-to-detector distance makes the assay more sensitive to sample positioning and sample nonuniformities. It is therefore important that the sample be thin and have a uniform thickness.

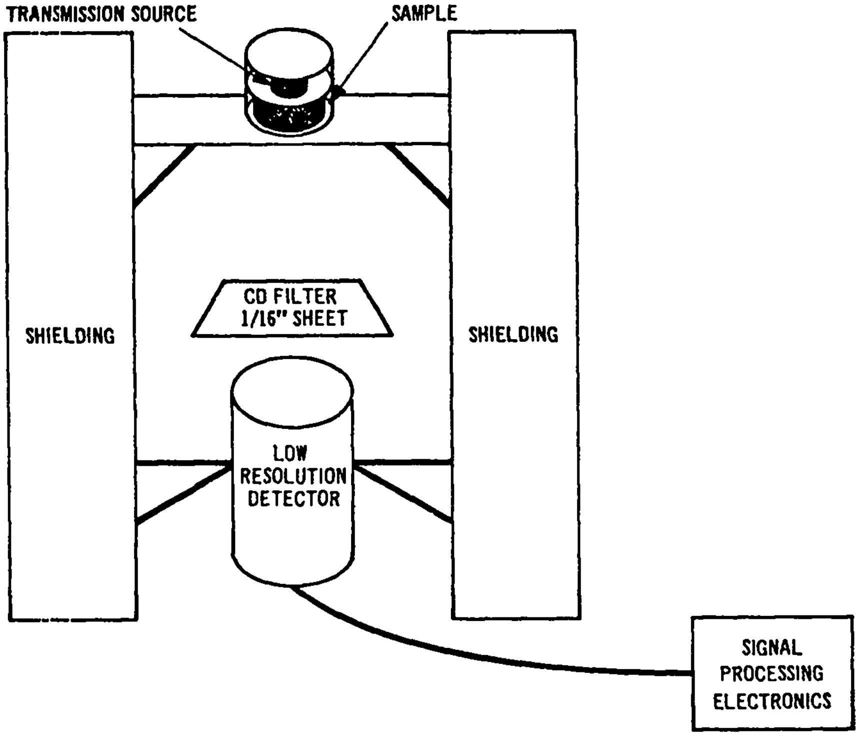

Fig. 2. Low-resolution assay configuration.

Figure 2 is a sketch of the assay configuration. The transmission source is uranium and is slightly smaller than the sample to prevent it from shining around the sample into the detector. The source-to-detector distance is 8 cm. The detector is a 3 in. by 3 in. NaI, and the amplifier is gain-stabilized. The Compton continuum under the peak is estimated by using a background region above the peak. Foil positioning hardware is important to ensure reproducible counting geometry.

D. High-Resolution Spectroscopy

The addition of a high-resolution detector significantly improves the quality of a gamma-ray-based assay. The drawback is a loss in detection efficiency. However, the increased ability to resolve interferences and to accurately determine peak areas often offsets the loss in efficiency.

This setup had a sample-to-detector distance of 20 cm. The detector was a large coaxial Ge(Li). Because the transmission at 185.7 keV is larger than 0.20, the uranium source provided adequate precision for the transmission measurement.

E. Verification Results

The low-resolution assay of ^{235}U is generally believed to have an accuracy of a few percent, if it is done carefully and the sample attributes are reasonable; 5% uncertainty is expected for most instances. A conservative estimate of the accuracy of a high-resolution assay, subject to the same restrictions, is 1%. Table II lists the tag ^{235}U mass values and the verification assay results.

Because these reference materials came from six foils with different origins, consistency among them is a partial verification of their validity. Figure 3, a plot of the response (corrected counts per gram) versus the mass (g ^{235}U) for these reference materials, shows consistency among the different isotopic analyses and mass determinations.

TABLE II

VERIFICATION ASSAY RESULTS FOR ^{235}U

ID[a]	Tag (g)	Low Resolution (g)	High Resolution (g)
8778-A	0.194	0.195	0.192
8778-B	0.358	0.359	0.363
11404-A	0.941	0.942	0.934
11404-B	1.594	1.58	1.596
1412-1-A	3.207	3.21	3.169
1412-1-B	5.710	5.75	5.712
1455-4-A	4.801	4.82	4.792
1455-4-B	7.812	7.85	7.758
1456-4-A	6.529	---	6.483
1456-4-B	11.686	11.59	11.623
1456-2-A	6.728	6.71	6.780
1456-2-B	12.549	12.56	12.408

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[a] The A and B designations following the ID numbers denote the 3-cm-diameter and 4-cm-diameter foils, respectively.

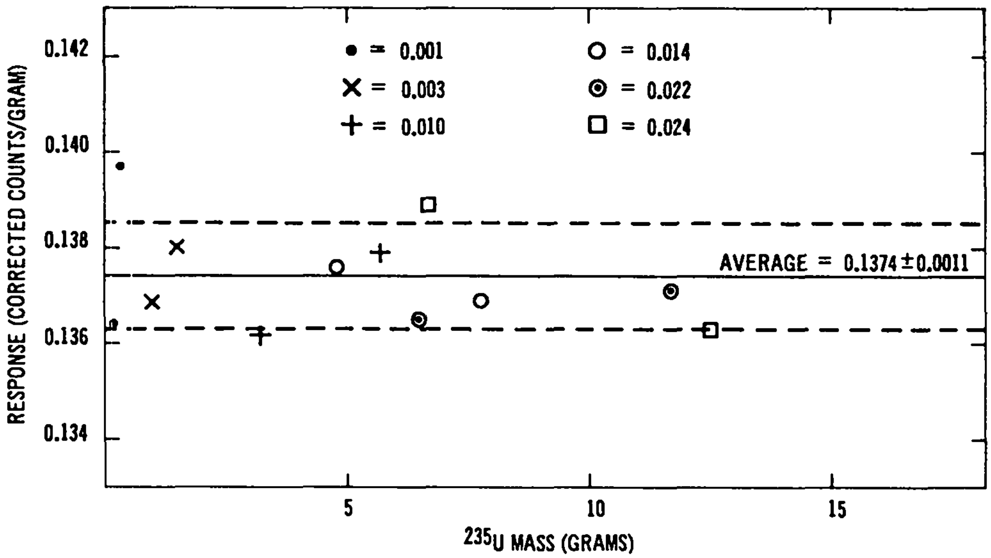

Fig. 3. The response (corrected counts per gram) for each of the foil reference materials as a function of ^{235}U mass.

PART 2

USE OF HIGHLY ENRICHED URANIUM REFERENCE MATERIALS CONSISTING OF THIN METAL FOILS

IV. INTRODUCTION

The highly enriched uranium reference materials consisting of thin metal foils were developed for use in high- or low-resolution assay procedures. Part 2 of this manual emphasizes the low-resolution technique; however, where appropriate, the manual recommends future implementation of improvements that can be achieved with more sophisticated procedures and equipment.

Transmission-corrected low-resolution assay can be used with high-density uranium samples, subject to several restrictions. The samples (and reference materials) should have similar container sizes and configurations. They should be homogeneous. They must have measurable transmissions at 185.7 keV (typically >5%). If the samples do not fulfill these requirements, incorrect assays usually result. The amount of bias can be related (but not simply) to how poorly the samples fulfill these requirements. The more advanced techniques are less susceptible to some or all of the biases.

An understanding of the measurement physics is essential. Good reference materials do not guarantee good assays. The assay technique must be properly applied to appropriate samples.

V. LOW-RESOLUTION ASSAY PROCEDURE

A. Assay Configuration

In the assay configuration shown in Fig. 2, the detector should be shielded from all sources of radiation except the sample and the transmission source. A thickness of 1.3 cm of lead is sufficient shielding for 185.7-keV gamma rays. Gamma rays of higher energy can usually be reduced to acceptable intensities by 5.1 cm of lead, however 1-MeV gamma rays may require 20 cm of lead shielding. Note that the detector is sensitive to radiation from all directions. A 0.16-cm-thick cadmium filter (placed between the detector and sample) is helpful in reducing low-energy background, such as uranium or lead x rays.

The detector axis, the sample center, and the center of the transmission source should all lie on the same axis. The sample-to-detector distance should be 8 cm or larger for the small Petri-dish samples. If the sample consists of discrete particles, the sample thickness must equal at least 10 particle diameters to obtain a valid transmission.

Figure 4 shows details of the sample and the transmission source. The low-resolution system essentially requires the use of the same energy gamma ray for both the assay peak and the transmission source. A good candidate for this type of transmission source is a thick foil whose diameter is approximately one-half to two-thirds that of the sample. The transmission source must have a uniform distribution of uranium, and it should not be allowed to shine around the sample into the detector. The capability of repositioning the transmission source to give the same intensity is extremely important.

Reliable assays can be accomplished with single channel analyzers, but multichannel analyzers allow more flexibility, give a better indication of whether problems are arising, and are easier to set up. Stabilization of the system is required, despite the fact that broken stabilizers could well be the most difficult problem to troubleshoot. Photomultipliers are not sufficiently stable without assistance, and small drifts in the peak location can bias the peak area determination significantly. Low-resolution systems can use short amplifier time constants; consequently, they are less sensitive to pileup and deadtime effects. However, it is still good practice to limit the count rate to 10 000 counts/s or less. High-resolution systems should use a rate loss correction source,[2] enabling them to tolerate count rates up to 30 000 counts/s or more.

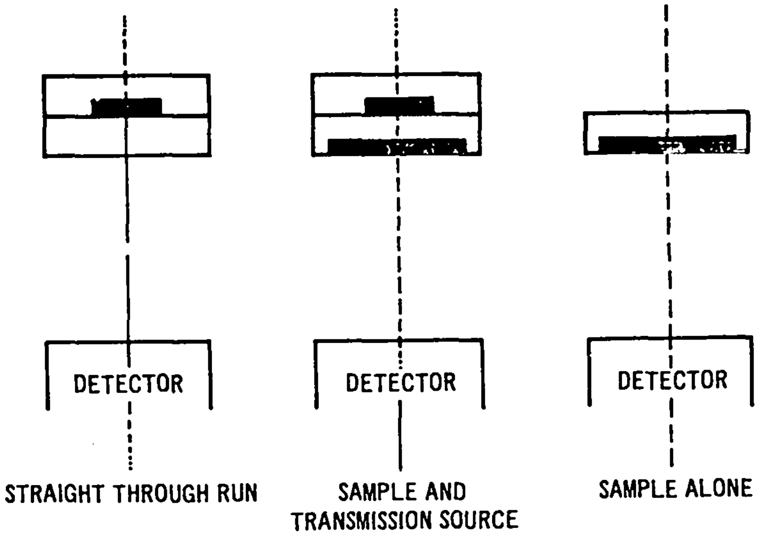

Fig. 4. Detail of the sample and transmission source.

B. Data Analysis

The total count in the peak region, P, is subject to two background subtractions, B_1 and B_2, to obtain the net area, A.

$$A = P - B_1 - B_2 \quad . \tag{1a}$$

The statistical uncertainty in A is expressed by Eq. (1b).

$$\sigma(A) = \sqrt{P + B_1 + B_2} \quad . \tag{1b}$$

The first background, B_1, is the total count in the region just above the 185.7-keV peak region. The two regions, peak and background, should be the same width and are counted during the same data acquisition. When single channel analyzers are used, a reasonable window width for P is 160 to 210 keV; for B_1, a reasonable window width is 220 to 270 keV. If only one single channel analyzer is available, the two regions require two separate counts. This practice can lead to difficulties if the background changes between the two counts. Background subtraction B_1 corrects for the Compton continuum under the 185.7-keV peak, which is due to higher energy gamma rays. Clearly, this subtraction is invalid if the background region contains a gamma-ray peak. The second background, B_2, is obtained from $P - B_1$ with no sample and no transmission source. If this background is nonnegligible, the assay position and shielding should be altered until it becomes negligible, if at all possible. Background subtraction B_2 corrects for 185.7-keV gamma rays that originate from somewhere other than the sample or transmission source. If B_2 is nonnegligible, the operator must determine that it does not vary, or else expect erroneous assays. High-resolution detectors generate spectra that allow for easier, more reliable peak area determinations.

The gamma rays emitted from within the sample may suffer attenuation before they reach the detector. The attenuation effects outside the sample container should be the same for all samples and reference materials; consequently, they can be ignored. The attenuation caused by the (empty) sample container is negligible and can be ignored in these assays. If different types of containers are used, the container transmission could well be different. If the matrix is homogeneous and the ^{235}U is spread uniformly throughout the sample, the average self-attenuation can be related to the transmission through the sample. Consequently, a correction factor based on the sample transmission can be calculated. The sample transmission, T, is given in Eq. (2a).

$$T = \frac{[A(S + T) - A(S)]}{A(T)} , \tag{2a}$$

where A(S + T) = the net area of the 185.7-keV peak determined with the sample and transmission source in place, A(S) = the net area with the sample in place and the transmission source removed, and A(T) = the net area with the transmission source placed over an empty container. The uncertainty in T is expressed in Eq. (2b).

$$\frac{\sigma(T)}{T} = \sqrt{\frac{\sigma^2[A(S + T)] + \sigma^2[A(S)]}{[A(S + T) - A(S)]^2} + \frac{\sigma^2[A(T)]}{A^2(T)}} \tag{2b}$$

Transmissions below 5% require a high-resolution system. At these low transmissions, the low-resolution technique requires very long count times and is susceptible to minor background fluctuations that may cause significant errors.

The correction factor, CF(T), is approximated in closed form for a slab geometry by

$$CF(T) = \frac{-\ell n(T)}{1 - T} . \tag{3a}$$

The uncertainty in CF(T) resulting from an uncertainty in T is shown in Eq. (3b).

$$\sigma(CF) = \frac{1}{T-1} \cdot \frac{\sigma(T)}{(T)} - \frac{\ell n(T)\,\sigma(T)}{(T-1)^2} \quad . \qquad (3b)$$

The low-resolution detectors require the use of the same energy gamma ray for both the assay peak and the transmission measurement. The intensity of the 185.7-keV gamma ray from a uranium sample is limited by its self-attenuation; consequently, no strong sources are available. This limitation restricts transmission measurements to transmissions above 5%. High-resolution detectors can take advantage of stronger sources with gamma rays at nearby energies and reliably measure transmissions below 1%.

The CF is used to generate the corrected counts, CC, from the net area, A.

$$CC = A \cdot CF(T) \quad . \qquad (4a)$$

The associated uncertainty in CC is shown in Eq. (4b).

$$\frac{\sigma(CC)}{CC} = \sqrt{\left(\frac{\sigma(A)}{A}\right)^2 + \left(\frac{\sigma[CF(T)]}{CF(T)}\right)^2} \quad . \qquad (4b)$$

C. Calibration and Measurement Control

The general procedure for gamma-ray assay is outlined in the assay equation:

$$M = \frac{CC}{K} \quad , \qquad (5a)$$

where M = the mass of ^{235}U and K = the calibration constant. The uncertainty is shown in Eq. (5b).

$$\left(\frac{\sigma(M)}{M}\right)^2 = \left(\frac{\sigma(CC)}{CC}\right)^2 + \left(\frac{\sigma(K)}{K}\right)^2 \quad . \tag{5b}$$

The calibration is determined with known reference materials and by inverting the assay equation, as shown in Eq. (6a).

$$K = \frac{CC}{M} \quad . \tag{6a}$$

The corresponding statistical uncertainty is shown in Eq. (6b).

$$\left(\frac{\sigma(K)}{K}\right)^2 = \left(\frac{\sigma(CC)}{CC}\right)^2 + \left(\frac{\sigma(M)}{M}\right)^2 \quad . \tag{6b}$$

The initial calibration should be based on repeated measurements of at least three reference materials. Because low-resolution systems typically have a calibration that varies with the transmission, more than three reference materials may be needed to determine the calibration curve. Two equations were investigated:

$$y = Ax + B \tag{7}$$

and

$$y = \frac{Ax}{1 + Bx} \quad , \tag{8}$$

where y = mass in g ^{235}U, x = CC, and A and B = the fit parameters. Equation (8) gave the best results.

When the calibration has been established, it should be verified with the assay of a reference material each day that the system is used to measure unknowns. At least one verification assay should be done both before and after the assays of unknowns. If there is any reason to suspect a system malfunction, more frequent verification is appropriate. The low-resolution gamma-ray assay technique does not assay nonhomogeneous materials correctly, nor can it distinguish between homogeneous and nonhomogeneous samples. If previous results with certain samples indicate that a bias may exist in the calibration constant, but the verification assays indicate that the calibration is valid, then the uncertainty associated with the assay should be increased unless an investigation of inhomogeneities or sample fill heights invalidates the assay results. Without detailed knowledge of the present sample, any attempt to use previous results to correct the calibration is unjustified. Because this detailed knowledge is rarely, if ever, available, the user is left with the choice of either guessing the calibration parameters or increasing the uncertainty to reflect the lack of information.

The continued validity of the uranium content of the reference materials can be assured by a comparison with fresh solution standards. A somewhat less rigorous assurance is obtained by demonstrating one or more of the following:

- the response does not change with time;
- the response is consistent with the responses of other known uranium samples;
- physical security is employed to prevent tampering; or
- visual inspection indicates no deterioration of the foils.

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