U.S. ATOMIC ENERGY COMMISSION

REGULATORY GUIDE

DIRECTORATE OF REGULATORY STANDARDS

September 1974

REGULATORY GUIDE 5.38

NONDESTRUCTIVE ASSAY OF HIGH-ENRICHMENT URANIUM FUEL PLATES BY GAMMA RAY SPECTROMETRY

A. INTRODUCTION

Part 70 of Title 10 of the Code of Federal Regulations requires each licensee authorized to possess more than 350 grams of contained U-235 to conduct a physical inventory of all special nuclear material in his possession at intervals not to exceed 12 months. Each licensee authorized to possess more than one effective kilogram of high-enrichment uranium is required to conduct measured physical inventories of his special nuclear materials at bimonthly intervals. Further, these licensees are required to conduct their nuclear material physical inventories in compliance with specific requirements set forth in Part 70. Inventory procedures acceptable to the Regulatory staff for complying with these provisions of Part 70 are detailed in Regulatory Guide 5.13, "Conduct of Nuclear Material Physical Inventories."

For certain nuclear reactors, the fuel consists of highly enriched uranium fabricated into flat or bowed plates. Typically, these plates are relatively thin so that a significant percentage of the U-235 gamma rays penetrate the fuel cladding. When the measurement conditions are properly controlled and corrections are made for variations in the attenuation of the gamma rays, a measurement of the U-235 gamma rays can be used as an acceptable measurement of the distribution and the total U-235 content of each fuel plate. In lieu of assaying the product fuel plates, fuel plate core compacts may be assayed through the procedures detailed in this guide, provided steps are taken to ensure the traceability and integrity of encapsulation of each assayed fuel plate core compact. This guide describes features of a gamma ray spectrometry system acceptable to the Regulatory staff for nondestructive assay of high-enrichment uranium fuel plates or fuel plate core compacts.

B. DISCUSSION

The number, energy, and intensity of gamma rays associated with the decay of U-235 provide the basis for nondestructive assay of high-enrichment fuel plates by gamma ray spectrometry (Ref. 1). The 185.7-keV gamma ray is the most useful U-235 gamma ray for this application; it is emitted at the rate of 4.25×10^4 gamma rays per second per gram of U-235. Lower-energy gamma rays emitted by U-235 are less penetrating and more sensitive to errors due to fluctuations in clad and core thickness. In general, more accurate fuel plate assays may be made by measuring only the activity attributable to the 185.7-keV U-235 gamma ray.

Assay measurements are made by integrating the response observed during the scanning of single fuel plates and comparing each response to a calibration based on the response to known calibration standards.

1. GAMMA RAY MEASUREMENT SYSTEM

1.1 GAMMA RAY DETECTION SYSTEM

1.1.1 Gamma Ray Detector

High-resolution gamma ray detectors, i.e., intrinsic or lithium-drifted germanium, provide resolution beyond that required for this assay application. While the performance of such detectors is more than adequate, their low intrinsic detection efficiency, extensive operational and maintenance requirements, and high cost make them unattractive for this application.

Most sodium iodide [NaI (Tl)] scintillation detectors are capable of sufficient energy resolution to be used for the measurement of the 185.7-keV gamma rays. The detector diameter is determined by the fuel plate width and the scanning method selected (see Section B.1.2 of this guide). The thickness of the NaI crystal is selected to avoid unnecessary sensitivity to gamma rays above the 185-keV region which produce a background in the 185-keV energy region as a result of Compton scattering.

For measurements to be reproducible, it is necessary to assure that the detection system is stabilized on the

intended portion of the gamma ray spectrum during measurements. Internally "seeded" NaI crystals which contain a radioactive source (typically Am-241) to produce a reference energy pulse are commercially available. The detection system is stabilized on the reference, and the amplifier gain is automatically corrected to assure that that energy and the rest of the spectrum remain fixed in position.

1.1.2 Gamma Ray Collimator

To ensure that the only gamma ray activity detected originates from a well-defined segment of the fuel plate, the detector is shielded from extraneous background radiations and collimated to define the area "seen" by the detector crystal. The collimator consists of a disk of appropriate shielding material. A slit is machined through the center of the disk which will allow only those gamma rays emitted within the slit opening to strike the detector. The disk thickness is a minimum of six mean free path lengths to effectively stop all gamma rays emitted from outside the view area. To prevent gamma rays from striking the crystal around the edges of the collimator disk, the disk diameter exceeds the crystal diameter by at least twice the crystal depth.

The probability of detection for gamma rays emitted at the center of the collimator slit is greater than that for gamma rays emitted near the ends of the slit. This effect becomes increasingly important at small detector-to-plate spacing, especially when scanning near the edge of a plate. To minimize this detection nonuniformity and to minimize the sensitivity to jitter, the detector-to-plate distance can be made large, especially with respect to the dimensions of the slit opening. As an alternative means of reducing the detection nonuniformity across the slit, the slit opening can be divided into channels by inserting a honeycomb baffle into the slit or by fabricating the collimator by drilling holes through the disk in a pattern which ensures that each hole is surrounded by a minimum wall thickness of 0.2 mean free path length. A 7.0-cm-thick iron disk with holes less than 0.5 cm in diameter drilled in a pattern having 0.2 cm of wall between adjacent holes is one example of a collimator that would perform satisfactorily. A large number of small-diameter holes is preferable to a few large-diameter holes.

1.1.3 Multiple Detectors

Several detectors may be used to shorten the measurement time. The detectors can be positioned to simultaneously measure different segments of a single fuel plate or to simultaneously measure additional fuel plates. In some cases it may be useful to sum the response from two detectors positioned on opposite sides of a plate to increase counting efficiency. In such cases it is essential that the response of such detectors be balanced and checked at frequent intervals.

1.2 SCANNING TECHNIQUES

It is critical that the scanning apparatus for moving the plates relative to the detector provide a uniform, reproducible scan. The importance of a well-constructed, mechanically stable conveyor cannot be overemphasized. Either the detector can be moved and the plate held stationary, or the plate can be moved past a fixed detector. Care must be exercised to maintain the detector-to-plate spacing within close tolerances to minimize errors caused by the inverse-square dependence of detection on distance. This is especially important in the case of close spacing, which is sometimes desirable to maximize the count rate. Various commercial conveying systems have been used and found to be adequate. Such systems may significantly reduce the cost of designing and building new scanning mechanisms. High-precision tool equipment such as milling machines, lathes, and x-y scanning tables can be investigated. Numerically controlled units offer additional advantages when they can be incorporated into a scanning system. This is particularly true when an automated scanning system is being developed.

Fuel plate core compacts may be sufficiently small to permit total assay without scanning in a fixed-geometry counting system. The scanning techniques for fuel plates discussed in the following subsections can also be used for core compacts when total fixed compact counting is not possible.

1.2.1 Linear Total Scan

The detector collimation consists of a rectangular opening which extends across the width of the fuel plates beyond the edges of the uranium core contained within the plate cladding. Scanning the total plate is accomplished by starting the count sequence on the end of a plate and continuing to count until the entire length of plate has been scanned.

To ensure that gamma rays emitted anywhere across the face of the fuel plate have an equal probability of being detected, it is necessary that the diameter of the detector crystal exceed the plate width or that the detector be positioned away from the plate.

Use of the spot or circular collimator scan technique eliminates or reduces to insignificance most of these edge effects.

1.2.2 Sweeping Spot Scan (Ref. 2)

If the collimator channel width is smaller than the fuel plate width, the viewing area (spot) can be swept across the plate as the detector scans along the length of the plate. This scanning technique can be readily adapted to scanning bowed plates through the use of a cam which is designed to maintain the detector-to-plate distance constant over the entire geometry of the fuel plate. The collimator channel dimensions can be selected to provide compatible information on the uniformity of

the fuel plate which is frequently obtained by comparing fixed (static) spot counts at a variety of locations to reference counts.

1.2.3 Sampled Increment Assay

When used in conjunction with radiographic dimensional measurements performed on all fuel plates, the U-235 content of a fuel plate can be measured by scanning the ends of each fuel plate and sampling the balance of the plate. It is necessary to measure the dimensions of the fuel core loading radiographically, through gamma ray scanning along the length of the plate, or by spot scanning the fuel plate ends and measuring the distance between end spots where the fuel loading stops. The U-235 content of the plate is then determined by averaging the results of sample spot measurements of the U-235 content per unit area at a number of sites along the plate and multiplying this average value by the measured area of the fuel core. The radiograph of each plate is examined to ensure that the core filter is uniform.

The collimator shape and dimensions can be selected to provide compatible information on the uniformity of the fuel plate.

1.3 COMPUTER CONTROL

The reproducibility of measurements can be improved and the measurement time per fuel plate can be reduced by using a computer to control the fuel plate scanning operation. The computer can be used to control data acquisition by accumulating counts according to a predetermined scheme. Also, the computer can be used for data analysis, including background corrections and intermachine normalization, calibration, error analysis, and diagnostic test measurements and analyses. Report preparation and data recording for subsequent analysis are also readily accomplished through an appropriately designed computer-controlled system.

2. INTERPRETATION OF MEASUREMENT DATA

The three factors discussed below may give rise to significant errors in interpreting measurement data.

2.1 ENRICHMENT VARIATIONS

Licensees authorized to possess highly enriched uranium are required to account for element and isotope as prescribed in §70.51. Under the conditions detailed in this guide, the U-235 content of individual plates is measured. To determine the total uranium content of each plate, the U-235 enrichment must be known from separate measurements.

Enrichment variations may alter the radiation background in the gamma ray energy region of interest. Uranium-238 decays by alpha-particle emission to Th-234. Thorium-234 then decays by beta-particle emission with a half-life of 24.1 days to Pa-234 which, in turn, decays by beta-particle emission to U-234. Approximately 1% of the Pa-234 decays are followed by high-energy (e.g., 1001 keV, 766 keV) gamma rays. These gamma rays frequently lose energy through Compton scattering and may appear in the 185-keV spectral region. It is important to note that activity from Pa-234 may be altered by disturbing the equilibrium between U-235 and Th-234, as frequently occurs in uranium chemical conversion processes. The interference due to variations in U-238 daughter activity becomes less important as the enrichment of U-235 increases. At enrichment levels above 90%, this problem can essentially be ignored.

2.2 RADIATION ATTENUATION

The number of U-235 gamma rays which escape from the fuel plate (and are thus available for detection) without losing energy depends on the characteristics of the fuel plate core and cladding. Gamma rays from U-235 are attenuated in the uranium, in the cladding, and in the inert material that may be added with the uranium to form the core of the fuel plate. Through well-controlled product tolerance limits, each of these potential sources of signal variability can be controlled to permit accurate accountability assays.

2.2.1 Self-Attenuation

The uranium photon attenuation coefficient for gamma ray energies corresponding to U-235 emissions is quite large (Ref. 3). Small changes in uranium density resulting from increased fuel loading or from variations in the manufacturing process can significantly change the number of gamma rays which escape from the fuel plate.

2.2.2 Cladding Attentuation

Small variations in cladding thickness may cause significant attenuation variations. Variations in cladding attenuation can be measured by a simple gamma ray absorption test using thin sheets of cladding material as absorbers and varying the clad thickness over the range of thicknesses to be encountered in normal product variability.

2.2.3 Core Filler Attenuation

Radiation intensity measurements may be made of plates fabricated with different ratios of uranium to filler to show the effects of this type of attenuation. If significant effects are noted, plates can be categorized by core composition characteristics and the assay system can be independently calibrated for each category of fuel plates.

2.2.4 Attenuation Corrections

When the thickness of the core and cladding of each plate is known, an attenuation correction can be applied to improve the accuracy of the assay. Ultrasonic gauging may provide such a measure, provided the metallographic zones within the plate are sufficiently defined to provide a detectable interface.

The alternative attenuation correction is based on a micrometer measurement of the total thickness of each plate. The clad thickness of a plate is estimated by subtracting the mean core thickness of the product plates, which is determined by periodically sampling product plates and cutting a cross section to permit visual measurement of clad and core thickness.

2.3 INTERFERING RADIATIONS

As noted in Section B.2.1 of this guide, an internal background variation may arise from changes in the amount of U-238 present in a fuel plate or from changes in the ratio of Th-234 to U-238 resulting from fuel manufacturing processes. Fluctuations in the internal background cause the response of the unknown items to be different from the calibration standards, thereby creating a measurement bias. Such interferences can be compensated by measuring additional regions of the gamma ray spectrum.

Other interfering radiations may come from external sources, from fuel plates awaiting assay, or from nearby radiation sources used for other measurements. This is not expected to be a major problem and can be controlled through (1) removing radiation sources, (2) shielding the detectors, and (3) monitoring the background at frequent intervals.

3. CALIBRATION AND VERIFICATION

3.1 INITIAL OPERATIONS

Calibration and the verification of assay predictions is an ongoing effort where performance is periodically monitored and the calibration relationship is modified to improve the accuracy of assay predictions. During initial operations, two means of basing preliminary calibrations are appropriate.

3.1.1 Foil Calibration Technique

Methods for calibrating scanning systems for high-enrichment uranium fuel plates through the assay of prepared uranium and clad foils are described in Reference 2. This method may be used in place of or in addition to the technique described in the following subsection.

3.1.2 Fabricated Calibration Plates

Calibration standard fuel plates can be fabricated using special precautions to ensure that the amounts of uranium, U-235, inert matrix, and cladding are accurately measured and that these parameters bracket the nominal range of product plates anticipated to fall within manufacturing tolerances.

3.2 ROUTINE OPERATIONS

The performance of the assay system is periodically monitored to ensure that the performance of the assay system has not shifted since its last calibration. Control limits for acceptable performance can be established for the response to an appropriate working standard. The control chart of the responses to the working standard can be checked for indications of short-term instrument drift or malfunction. The control chart can also be analyzed to detect long-term shifts within the measurement-to-measurement control limits that may be corrected by recalibrating the system. Severe changes in instrument performance are investigated promptly and their causes remedied.

To ensure that the calibration remains valid during normal operations and that accuracy estimates are rigorously justified, assay predictions are periodically compared with more accurate measurements of the content of typical fuel plates (see Section C.4 of this guide). Guidance on methods to relate this assay to the national measurement system and to reconcile verification measurements will be addressed in separate regulatory guides.*

C. REGULATORY POSITION

The content and distribution of U-235 in high-enrichment uranium plates can be measured through the gamma ray assay methods described in this guide. Combining this measurement with the results of an independent measurement of the U-235 enrichment enables the total uranium content of the fuel plates to be measured. The factors presented below should be taken into consideration for this assay method to be acceptable to the Regulatory staff.

1. MEASUREMENT SYSTEM

1.1 GAMMA RAY MEASUREMENT SYSTEM

1.1.1 Gamma Ray Detector

A thallium-activated sodium iodide scintillation detector or series of detectors is recommended for this assay application. When more than one detector is to be incorporated into the scan system, the performance characteristics of the detectors should be matched. The diameter of the crystal should be larger than the projected view onto the crystal face through the

*For example, regulatory guides related to measurement quality assurance and calibration of nondestructive assay systems are being developed.

collimator channel. The thickness of the crystal should be no more than one inch. The crystal should contain an internal cesium iodide seed which is doped with a suitable alpha-emitter for spectral stabilization. The seed should produce approximately 1,000 counts per second at the reference energy.

1.1.2 Collimator

A collimator should be fabricated of appropriate gamma ray shielding material such as iron, lead, or tungsten. The shielding should completely surround the detector and photomultiplier assembly and should be sufficiently thick to completely block extraneous radiations from the detector. The response variation from the center of the collimator opening to its edge should be less than 1%.

1.1.3 Electronic Apparatus

All electronic systems should be powered by filtered, highly regulated power supplies. The ambient temperature and humidity in the vicinity of the scanning system should be controlled so that permitted fluctuations do not significantly affect the assay measurements. All electronic circuitry in signal-processing components should feature temperature compensation. Residual sensitivity to fluctuations in the ambient environment should be tested and monitored periodically.

The capability for multichannel gamma ray pulse height analysis with cathode ray tube spectral display should be provided. Signal-processing electronics capable of stabilizing on the alpha radiations emitted within the doped cesium iodide seed should be provided to stabilize the energy spectrum.

1.2 SCANNING SYSTEM

A mechanically sound, highly reproducible scanning system should be employed. Scanning should be accomplished by one of the three techniques discussed in Section B.1.2 of this guide.

1.3 COMPUTER CONTROL

A dedicated minicomputer to control data acquisition, analysis, calibration, diagnostic testing, and report preparation should be employed for this assay application.

1.4 MULTIPLE SCANNING ASSAY SYSTEMS

When more than one scanning system is employed, assay response should be normalized so that each instrument provides consistent results. Verification data to establish the systematic assay error for each assay system should be obtained with the same plate.

2. MEASUREMENT INTERPRETATION

2.1 ENRICHMENT VARIATIONS

Procedures should be developed to ensure that the enrichment of the plates being scanned is known through separate measurements. Fuel plates generally satisfy the gamma ray penetrability criteria for quantitative U-235 assay; they do not satisfy the criteria for nondestructive enrichment measurement through gamma ray spectrometry.* Facilities processing more than one uranium enrichment should maintain strict isotopic control and characterize the enrichment through appropriate measurement methods.

2.2 ATTENUATION CORRECTIONS

Attenuation variations arising from plate-to-plate changes in core thickness, composition, and clad thickness should be determined over the range of product tolerance specifications. When such variations cause the assay error to exceed the error realized without the variations by 50% or more, procedures should be implemented to measure and apply a correction to the assay of each plate.

2.3 RADIATION INTERFERENCES

A clear plastic template which shows an acceptable spectrum display should be prepared. When radioactive interference may be encountered, the assay spectrum should be compared at appropriate intervals to the reference spectrum for indications of interference. Background radiation should be measured periodically during each operating shift.

3. MEASUREMENT CALIBRATION

During initial operations, the assay system should be calibrated either by the foil calibration method or with specially prepared sample fuel plates as described in Section B.3.1 of this guide.

4. RANDOM AND SYSTEMATIC ASSAY ERRORS

4.1 RANDOM ERROR ESTIMATION

A replicate assay program should be established to generate data for the evaluation of random assay errors during each material balance period. During each bimonthly interval, a minimum of fifteen plates should be selected for replicate assay. The second assay of each plate selected for replicate assay should be made at least four hours after the first assay. Replicate assay differences should be collected and analyzed at the end of the

*Criteria for uranium gamma ray enrichment measurements are given in Regulatory Guide 5.21, "Nondestructive Uranium-235 Enrichment Assay by Gamma Ray Spectrometry."

material balance period. The single-measurement standard deviation of the relative replicate assay differences should be computed as described in Reference 4.

4.2 SYSTEMATIC ERROR ESTIMATION

The systematic error associated with the assay of all fuel plates fabricated during a material balance period should be determined through one of the procedures* presented below.

4.2.1 Propagation through the Calibration Function

To estimate the systematic assay error through the calibration function, the calibration should be based on the regression analysis of an appropriate function to the calibration data. Uncertainties in the reference values of the calibration standards should be factored into the fit, and the errors propagated as demonstrated in Reference 5.

To ensure the validity of the predictions, the stable performance of the instrument should be monitored and normalized through the response to appropriate working standards which are assayed at frequent intervals. The frequency for assaying working standards should be determined through testing, but should not be lower than one test during each two-hour assay interval for spot response stability and one full scan test during each operating shift. Indications of shifting instrument performance should be investigated and remedied, and the instrument should be recalibrated to ensure the validity of subsequent measurements.

In order to ensure that the calibration standards continue to adequately represent the unknown fuel plates, key production parameters which affect the observed response should be monitored through separate tests. Data should be compiled and analyzed at the close of each material balance period. When a production parameter shifts from previously established values, the impact of the shift on the response of the assay instrument should be determined through an appropriate experiment or calculation (Ref. 6). A bias correction should be determined and applied to all items assayed from the point of the parameter change. The uncertainty in the bias should be combined with the systematic error predicted through the calibration function. When the bias exceeds 3% of the plate contents in a single material balance period, when a trend of 1.5% or more is observed in three consecutive material balance periods, or when the uncertainty in the observed bias is sufficient to increase the limit of error of the assay above 0.5%, new calibration standards should be obtained, and the scanning system should be recalibrated.

As a further check on the continued validity of the calibration standards, a program to periodically introduce new calibration standards should be implemented. A minimum of one new calibration standard fuel plate should be introduced during each six-month period.

4.2.2 Comparative Evaluation

When two measurements are made on each of a series of items and the accuracy of one of the methods used is considerably greater than the other, the corresponding predictions can be compared to establish an estimate of bias between the measurement methods and to estimate the error associated with the less-accurate measurement method. To precisely determine the systematic error in the nondestructive assay, the fuel plates selected for comparative measurements should be randomly selected but should span the range of U-235 contents encountered in normal production. The selected fuel plates may be rejected from the process stream for failing to meet quality assurance requirements. Each plate should be repeatedly assayed to reduce the random assay error to less than 10% of the estimated or previously established systematic error. To determine its U-235 and total uranium content, the plate should be completely dissolved and the resulting solution should be analyzed by high-accuracy chemical and mass spectrometric procedures.

For one material balance period during the initial implementation of this guide, a product fuel plate should be randomly selected twice each week for an accuracy verification measurement. Following this initial implementation period, facilities manufacturing 100 or more fuel plates per week may reduce the verification frequency to one plate per week and pool the verification data for two consecutive material balance periods. Low-throughput facilities manufacturing less than 100 plates per week should verify at least 4 plates per material balance period through the procedures described above. At the close of each material balance period, data should be pooled to include only the 15 most current data points.

When the U-235 contents of the plates assayed using a common calibration relationship varies over a range of ±5% or more about the average of all plate loadings, the systematic error should be estimated as described in paragraph 1. below; when plate loadings are tightly clustered about a nominal value, the systematic error should be estimated as described in paragraph 2.

1. At the close of the reporting period, the assay value for each plate is plotted against the verified quantity. The verification data plot is examined for indications of nonlinearity or obvious outlier data. Anomalous indications should be investigated and remedied.

A linear regression analysis should be performed on the comparison data. The intercept should be tested against zero for an indication of a constant measurement bias. The slope should be tested against unity for an indication of a proportional bias. When bias is indicated, assays performed during the preceding operating period should be compensated. The systematic error should be

*These methods will be discussed in detail in a regulatory guide in preparation entitled "Calibration and Error Estimation Procedures for Nondestructive Assay."

estimated as the standard error associated with the verification line.

2. When all plates contain essentially the same U-235 content, the difference in the mean content values should be tested against zero as an indication of bias, and the systematic error associated with an inventory of plates should be quoted as the standard deviation of the *mean* difference. For individual plates, the systematic error should be quoted as the standard deviation of the difference distribution.

5. CORE COMPACT ASSAY

Final product assay in high-enrichment fuel plate manufacturing can also be accomplished through assaying each core compact following the procedures detailed in this guide and the following supplemental criteria:

1. Each core compact should carry a unique identification. Accountability records should be created for each compact. The fuel plate should carry an identification corresponding to the compact identification.

2. Each fuel plate should be radiographically examined to ensure that the entire compact has been encapsulated.

3. Each fuel plate should be checked with a gamma ray probe to qualitatively ensure that the plate core is uranium of the nominal product enrichment.

4. Calibration and error evaluation should follow the procedures for fuel plate assay.

REFERENCES

1. J.E. Cline, R.J. Gehrke, and L.D. McIsaac, "Gamma Rays Emitted by the Fissionable Nuclides and Associated Isotopes," ANCR-1029 (July 1972).
2. N.S. Beyer, "Assay of U-235 in Nuclear Reactor Fuel Elements by Gamma Ray Scintillation Spectrometry," Proc. 4th Intl. Conf. on Nondestructive Testing, London, 1963.
3. J.H. Hubbell, "Photon Cross Sections, Attenuation Coefficients, and Energy Absorption Coefficients from 10 keV to 100 GeV," Nat. Bur. Stds. NSRDS-NBS 29 (1969).
4. John L. Jaech, "Statistical Methods in Nuclear Materials Control," TID-26298 (1973).
5. American National Standard N15.20, "Guide to Calibrating Nondestructive Assay Systems," in preparation. Copies of the draft standard may be obtained from Institute of Nuclear Materials Management, 505 King Avenue, Columbus, Ohio 43201 (Attention: H.L. Toy).
6. See, for example, R.A. Forster, D.B. Smith, and H.O. Menlove, "Error Analysis of a Cf-252 Fuel Rod Assay System," LA-5317 (1974).