



U.S. ATOMIC ENERGY COMMISSION

JUNE 1974

REGULATORY GUIDE

DIRECTORATE OF REGULATORY STANDARDS

REGULATORY GUIDE 5.34

NONDESTRUCTIVE ASSAY FOR PLUTONIUM IN SCRAP MATERIAL

BY SPONTANEOUS FISSION DETECTION

A. INTRODUCTION

Section 70.51, "Material Balance, Inventory, and Records Requirements," of 10 CFR Part 70, "Special Nuclear Material," requires certain licensees authorized to possess at any one time more than one effective kilogram of plutonium to establish and maintain a system of control and accountability such that the limit of error (LE) associated with the material unaccounted for (MUF), ascertained as a result of a measured material balance, meets minimum standards.

Included in a typical material balance are containers of inhomogeneous scrap material that are not amenable to assay by the traditional method of sampling and chemical analysis. With proper controls, the nondestructive assay (NDA) technique of spontaneous fission detection (SFD) is an acceptable method for the assay of plutonium in containers of bulk scrap material. The use of SFD thus facilitates the preparation of a complete plant material balance whose LEMUF meets established requirements.

This guide describes procedures acceptable to the Regulatory staff for application of the technique of spontaneous fission detection for the nondestructive assay of plutonium in scrap.

B. DISCUSSION

Plutonium in scrap material can contribute significantly to the material unaccounted for (MUF) and to its associated limit of error (LEMUF). Unlike the major quantity of material flowing through a process, scrap is typically

USAEC REGULATORY GUIDES

Regulatory Guides are issued to describe and make available to the public methods acceptable to the AEC Regulatory staff of implementing specific parts of the Commission's regulations, to delineate techniques used by the staff in evaluating specific problems or postulated accidents, or to provide guidance to applicants. Regulatory Guides are not substitutes for regulations and compliance with them is not required. Methods and solutions different from those set out in the guides will be acceptable if they provide a basis for the findings requisite to the issuance or continuance of a permit or license by the Commission.

Published guides will be revised periodically, as appropriate, to accommodate comments and to reflect new information or experience.

Copies of published guides may be obtained by request indicating the divisions desired to the U.S. Atomic Energy Commission, Washington, D.C. 20545, Attention: Director of Regulatory Standards. Comments and suggestions for improvements in these guides are encouraged and should be sent to the Secretary of the Commission, U.S. Atomic Energy Commission, Washington, D.C. 20545, Attention: Chief, Public Proceedings Staff.

The guides are issued in the following ten broad divisions:

- | | |
|-----------------------------------|------------------------|
| 1. Power Reactors | 6. Products |
| 2. Research and Test Reactors | 7. Transportation |
| 3. Fuels and Materials Facilities | 8. Occupational Health |
| 4. Environmental and Siting | 9. Antitrust Review |
| 5. Materials and Plant Protection | 10. General |

inhomogeneous and difficult to sample. Therefore, a separate assay of the entire content of each container of scrap material is the only reliable method of scrap accountability. Nondestructive assay (NDA) is a method for assaying the entire content of every container of scrap.

The term "scrap" refers here to material that is generated incident to the main process stream due to the inefficiency of the process. Scrap material is generally economically recoverable. Scrap therefore consists of reject or contaminated process material such as pellet grinder sludge, sweepings from a glovebox, dried filter sludge, and reject powder and pellets. Scrap is distinguished from "waste" by the density or concentration of heavy elements in the two materials. The concentration of uranium and plutonium in scrap is approximately the same as it is in process material, i.e., 85-90% (U + Pu) by weight. Plutonium in fast reactor scrap material is 15-20% by weight and in thermal reactor recycle material 2-9% by weight. The main difference between scrap and process material is that scrap is contaminated and inhomogeneous. Waste, on the other hand, contains a low concentration of plutonium and uranium, i.e., a few percent or less (U + Pu) by weight. However, the recovery of combustible waste by incineration may produce ash that is high in uranium and plutonium concentrations. Such incinerator ash is also considered "scrap" in this guide.

Nondestructive assay for plutonium can be accomplished primarily by the passive methods of gamma ray spectrometry, calorimetry, and spontaneous fission detection. Regulatory Guide 5.11¹ provides a framework for the utilization of these NDA methods.

Gamma ray spectrometry of scrap consisting of dense materials can be unreliable because of the attenuation of gamma rays. Gamma ray spectrometry is most applicable to waste assay.

Calorimetry is an accurate method of plutonium assay when there is an accurate knowledge of the relative abundance of each plutonium isotope and americium-241. Scrap may contain a mixture of materials of different radionuclidic compositions, especially different americium-241 concentrations, necessitating the measurement of the average radionuclidic composition. The average radionuclidic abundances can only be accurately measured when the scrap

is reasonably homogeneous. When the radionuclidic abundances can be accurately measured or controlled, calorimetry can be applied to scrap assay.² However, calorimetry is time-consuming for heterogeneous materials of high heat capacity and may not be a practical method for the routine assay of large numbers of containers.

Spontaneous fission detection (SFD) is the most practicable and generally applicable NDA technique for the assay of plutonium in scrap material. Spontaneous fission radiations are sufficiently penetrating to provide a representative signal from all the plutonium within a container. The plutonium isotopic composition must be known for SFD assay, but the accuracy of SFD is not as dependent on the accuracy of analysis for the minor plutonium isotopes as is that of calorimetry. Nor is SFD sensitive to americium-241 ingrowth. The quantity of scrap material on inventory when a material balance is computed can be reduced through good management, and the scrap remaining on inventory can be assayed by SFD to meet the overall plant MUF and LEMUF constraints required by paragraph (e)(5) of Section 70.51 of 10 CFR Part 70.

This guide gives recommendations useful for the SFD assay of containers, each containing a few liters of scrap and having contents ranging from a few grams to a few hundred grams of plutonium or approximately 50 grams of effective plutonium-240.* Containers with a larger plutonium content, i.e., on the order of 500 grams of plutonium or more, give a spontaneous fission response that is difficult to interpret due to high counting rates and possible neutron multiplication. A large quantity of plutonium can be assayed by SFD by subdividing the scrap into smaller amounts, or the items may be more amenable to nondestructive assay by calorimetry.

*The effective plutonium-240 mass is a weighted average of the mass of each of the plutonium isotopes. The weighting is equal to the spontaneous fission neutron yield of each isotope relative to that of Pu-240. Since only the even-numbered isotopes have significant spontaneous fission rates, the effective Pu-240 mass is given approximately by:

$$M(240)_{\text{eff}} = M(240) + 1.64M(242) + 2.66M(238)$$

where M is the mass of the isotope indicated in parentheses. The coefficients in this equation are only known to approximately $\pm 5\%$.

C. REGULATORY POSITION

The method of spontaneous fission detection (SFD) for the nondestructive assay for plutonium in bulk inhomogeneous scrap material should include: (1) discrimination of spontaneous fission radiations from random background by coincidence techniques and (2) measurement of the relative plutonium isotopic composition of the scrap by an independent measurement technique. An acceptable SFD method of plutonium assay is described below:

1. Spontaneous Fission Detection System

a. Detectors. Instruments based on moderated thermal neutron detectors, i.e., neutron well coincidence counters,^{4,5} are recommended for applications in which the gross neutron detection rate does not exceed 2×10^4 neutrons/sec. The dead time inherent in these slow coincidence systems can be reduced by employing a shift-register coincidence circuit.⁶ If the gross neutron detection rate is due primarily to random background and exceeds 2×10^4 neutrons/sec, then a fast neutron detection, single coincidence system can be used, provided that adequate corrections can be made for matrix effects. Matrix effects are more severe in fast neutron detection systems, as shown in Table I.

b. Detection Chamber. The chamber should permit reproducible positioning of standard-sized containers in the location of maximum spatial response uniformity.

c. Fission Source. A spontaneous fission source with a neutron intensity comparable to the intensity of the largest plutonium mass to be assayed should be used for making matrix corrections, using the source addition technique.⁷ A nanogram of Cf-252 is approximately equivalent to a gram of effective Pu-240.

d. Readout. Readout should allow computation of the accidental to real coincidence ratio in addition to the net real coincidence rate. Live time readout or a means of computing the dead time should also be provided.

e. Performance Specifications. The performance of a SFD instrument should be evaluated according its stability, uniformity of spatial response, and insensitivity to matrix effects. Therefore, information should be obtained regarding:

(i) The precision of the coincidence response as a function of the real coincidence counting rate and the accidental to real coincidence ratio. Extremes in the background or accidental coincidence rate can be simulated by using a source of random neutrons (nonfission).

TABLE I

MATRIX MATERIAL EFFECTS ON NEUTRON ASSAY

5.34-5

Matrix Material (in ~4 liter can)	Mass (kg)	Neutron Detection Efficiency ⁸			Coincidence Efficiency, ³ He Detector, Thermal	Corrected ^{a,7} Coincidence Efficiency ³ He Detector, Thermal
		³ He Detector, Thermal	⁴ He Detector, Fast	ZnS Detector, Fast		
Empty Can	--	1.00	1.00	1.00	1.00	1.00
Carbon Pellets	1.89	1.03	--	--	1.05	0.97
Metal	3.60	1.04	0.83	0.75	1.09	1.02
Slag-Crucible	1.80	1.03	0.94	0.91	1.08	1.01
Concrete	3.24	1.05	0.84	0.79	1.10	1.02
String Filters	0.60	1.07	0.95	0.86	1.17	1.05
CH ₂ (ρ=0.65 g/cc)	0.27	1.06	0.96	0.92	1.11	1.00
CH ₂ (ρ=0.12 g/cc)	0.43	1.09	0.92	0.90	1.19	0.98
CH ₂ (ρ=0.27 g/cc)	0.97	1.19	0.71	0.67	1.36	1.04
H ₂ O (ρ=1.00 g/cc)	3.62	0.98	0.36	0.35	0.98	0.96

^aCorrected using the source addition technique.

(ii) Uniformity of spatial response. Graphs should be obtained on the relative coincidence response from a point source of fission radiation as a function of position in the counting chamber.

(iii) Sensitivity of matrix interference. A table of the relative coincidence response from a point source of fission radiation as a function of the composition of the matrix material surrounding the point source should be obtained. Included in the matrix should be materials considered representative of common scrap materials. Table I is an example of such a tabulation of the relative response for a wide range of materials.

This information should be used for evaluating the expected instrument performance and estimating errors. The above performance information can be requested from the instrument suppliers during instrument selection and should be acquired during preoperational instrument testing.

2. Analyst

A highly trained individual should oversee SFD assay for plutonium and should have primary responsibility for instrument specification, preoperational instrument testing, standards and calibration, writing an operation manual, measurement control, and error analysis. Experience or training equivalent to a bachelors degree in science or engineering from an accredited college or university and a laboratory course in radiation measurement should be the minimum qualifications of the SFD analyst. The SFD analyst should review SFD operation at least weekly and should authorize all changes in SFD operation.

3. Containers and Packaging

A single type of container should be used for packaging all scrap in each category, as discussed below. A recommended uniform container that would facilitate accurate measurement and would standardize this segment of instrument design is a thin-walled metal (steel) can with an inside diameter of approximately 10 cm or less.

4. Reduction of Error Due to Material Variability

The SFD response variation due to material variability in scrap should be reduced by: (1) segregation of scrap into categories that are independently calibrated, (2) correcting for matrix effects using the source addition technique,⁷ or (3) applying both categorization and the source addition technique. Categorization should be used if the SFD method is more sensitive to the material variability from scrap type to scrap type than to the material variability within a scrap

type. Application of the source addition technique reduces the sensitivity to material variability and may allow the majority of scrap types to be assayed under a single calibration. Material characteristics that should be considered in selecting categories include:

- a. Plutonium Isotopic Composition
- b. Uranium/Plutonium Ratio
- c. Containerization and Packaging
- d. Abundance of High-Yield alpha-neutron Material, i.e., low-atomic-number impurities
- e. Plutonium Content
- f. Density (both average density and local density extremes should be considered)
- g. Matrix Composition

5. Calibration

A guide to calibration for nondestructive assay is presently under development by Task Force 8.3 of the N15 committee of the American National Standards Institute* and will include details on calibration standards, calibration procedures, curve fitting, and error analysis. Guidelines relevant to SFD are given below.

a. A minimum of four calibration standards of the same isotopic composition as the unknowns should be used for calibration. If practicable, a calibration curve should be generated for each isotopic blend of plutonium. When plutonium of different isotopic composition is assayed using a single calibration, the effect on the SFD response of isotopic composition should be determined over the operating ranges by measuring standards of differing plutonium isotopic compositions. The use of the effective Pu-240 concept can lead to error because of the uncertainty in the spontaneous fission half-lives, as shown in Table II, and the variation in response with isotopic composition.

b. Calibration standards should be fabricated from material having a plutonium content determined by a technique traceable to or calibrated with National Bureau of Standards standard reference material. Well-characterized homogeneous material similar to the process material from which the scrap is generated can be used to obtain calibration standards.

c. Fabrication of calibration standards that are truly representative of the unknowns is difficult for scrap assay. To measure the reliability of the calibration based on the fabricated standards discussed above, and to improve this calibration, unknowns that have been assayed by SFD should periodically be

*When copies become available, they may be obtained from the American National Standards Institute, Inc., 1430 Broadway, New York, New York 10018.

TABLE II
EFFECTIVE PLUTONIUM-240 ABUNDANCE AND UNCERTAINTY
CORRESPONDING TO DIFFERENT BURNUP CATEGORIES^a

BURNUP (MWd/t)	Approximate Abundance (%)					
	Pu-238	Pu-239	Pu-240	Pu-241	Pu-242	Pu-240 _{eff}
8,000- 10,000	0.10	87	10	2.5	0.3	10.75±0.03(0.3%)
16,000- 18,000	0.25	75	18	4.5	1.0	20.30±0.08(0.4%)
25,000- 27,000	1.0	58	25	9.0	7.0	39.14±0.50(1.3%)
38,000- 40,000	2.0	45	27	15.0	12.0	52.00±0.87(1.7%)

^aComputed using the following coefficients for Pu-238 and Pu-242 in the equation for Pu-240 effective:

$$M(240)_{\text{eff}} = M(240) + 1.64 \pm 0.07 M(242) + 2.66 \pm 0.19 M(238)$$

The uncertainties in the coefficients and in the effective Pu-240 abundances in the table are from the reported standard deviations in the most reliable data available.³

selected for assay by an independent more accurate technique. Calorimetry² can be used to assay a random selection of scrap in containers and provide reliable data that should be fed back into the calibration fitting procedure to improve SFD calibration. The original calibration standards should be retained as working standards.

6. Measurement Control

For proper measurement control, a "dummy" item should be assayed on each day of scrap assay as a background measurement. Also, control (or working) standards should be assayed each day scrap is assayed for normalization and to assure reliable operation.

The source addition technique⁷ is recommended for correcting the SFD response for each assay. If not used routinely, the source addition technique should be applied to a random selection of items but in no case should be used less frequently than daily. The results of random applications of the source addition technique can be used in two ways:

- a. As an average correction factor to be applied to a group of items, and
- b. As a check on the item being assayed to verify that it is similar to the standards used in calibration and that no additional matrix effects are present, i.e., purely as a qualitative assurance that the calibration is valid.

7. Error Analysis

The sources of error in SFD are discussed in Regulatory Guide 5.11.¹ Analysis of the error in the calibration is discussed in the literature^{4,9} and in the ANSI guide on calibration now under development. In addition to the calibration error there are errors due to the measurement process and due to variability in material composition.

The error due to the measurement process, i.e., the measurement-to-measurement error, accounts for most of the random error in NDA. At least fifteen unknowns selected at random should be repeatedly assayed to estimate the random error. Repeated measurements should be made under as many different conditions as are experienced in normal operation, e.g., different times of day, different operators, different ambient conditions. The standard deviation in the distribution of differences in replicate results should be used in constructing a 95% confidence interval. The mean difference in replicate results has an expected value of zero. Corrections for significant drift in the instrument performance should be made based on data from daily assay of control standards, i.e., the measurement control program.

The error due to material variability, i.e., the item-to-item error, is the major source of bias and systematic error in NDA. If proper calibration standards and a proper calibration relationship are used, the calibrating error should be a reliable estimate of the systematic error. To test these assumptions, and to determine the bias, SFD assay results on a random selection of unknowns should be compared with assays on the same items by an independent more accurate technique, as discussed in 4(c). Calorimetry is not sensitive to the majority of interferences that cause error due to material variability in SFD and is practical for this application because it is nondestructive. An alternative method for verifying SFD assay is to sample the scrap extensively and to perform chemical analyses for the plutonium concentrations in these samples.

The mean difference in comparative assays should be used as the bias for correcting SFD assay results. The bias correction should be made if the mean difference is greater than 0.1 times the standard deviation in the mean difference. The standard deviation in the bias (mean difference) is a systematic error that should be used in constructing a 95% confidence interval. (There will always be a potential bias and systematic error in the technique used to verify SFD. The systematic error should be known and should be insignificant compared to systematic error in SFD for the technique to be viable for verifying SFD assay results.)

Comparisons of SFD with a more accurate assay method should be made on at least two unknowns a week to determine bias and systematic error. Data may be pooled and used to improve the calibration although no data should be older than one year.

REFERENCES

1. Regulatory Guide 5.11, "Nondestructive Assay of Special Nuclear Material Contained in Scrap and Waste."
2. Regulatory Guide 5.35, "Calorimetric Assay of Plutonium."
3. C. Weitkamp, "Nuclear Data for Safeguards: Can Better Data Improve Present Techniques?" Symposium on Practical Applications of R&D in the Field of Safeguards, Rome, March 1974, and J. D. Hastings and W. W. Strohm, J. Inorg. Nucl. Chem., Vol. 34, pp. 25-28, 1972.
4. R. Sher, "Operating Characteristics of Neutron Well Coincidence Counters," BNL- 50332, January 1972.
5. J. E. Foley, "Neutron Coincidence Counters in Nuclear Applications," IEEE Transactions Vol. NS-19, No. 3, pp. 453-456, June 1972.

6. K. Böhnel, "Neutron Coincidence Counting with Overlapping Cycles," October 1972, Gesellschaft für Kernforschung mbH. 75 Karlsruhe, P. O. Box 3640, Germany, or L. V. East and J. E. Foley, "An Improved Thermal-Neutron Coincidence Technique," LA-5197, 1972.
7. H. O. Menlove and R. B. Walton, " 4π Coincidence Unit for One-Gallon Cans and Smaller Samples," LA-4457-MS, 1970.
8. H. O. Menlove, "Matrix Material Effects on Fission-Neutron Counting Using Thermal- and Fast-Neutron Detectors," LA-4994-PR, p. 4, 1972.
9. J. Jaech, "Statistical Methods in Nuclear Material Control," TID-26298, Section 3.3.8, 1974.