U.S. ATOMIC ENERGY COMMISSION

REGULATORY GUIDE

DIRECTORATE OF REGULATORY STANDARDS

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REGULATORY GUIDE 5.35

CALORIMETRIC ASSAY OF PLUTONIUM

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) of the material unaccounted for (MUF), ascertained as a result of a measured material balance, meets established minimum standards. The selection and proper application of an adequate measurement method for each material form in the fuel cycle is essential in meeting these requirements.

Calorimetry is a precise nondestructive method for plutonium assay that is applicable primarily for material of high plutonium concentration such as oxide powder and fuel pellets. Calorimetry, with proper calibration, is capable of an accuracy sufficient to meet LEMUF requirements established by 10 CFR Part 70 and is not dependent on representative sampling. Since calorimetry can also be applied to sealed containers, it is an accurate and timely method for verifying receipt quantities of plutonium. Calorimetry is also useful as a method for improving the calibration of other NDA techniques, as suggested in two Regulatory Guides,[1,2] and thus improving the control and accountability of plutonium.

This guide describes procedures acceptable to the Regulatory staff for operating and calibrating heat flow calorimeters in measuring the power associated with plutonium decay.

B. DISCUSSION

Calorimetry is the measurement of heat or the rate of heat emission (power). Calorimetry can therefore be applied as a nondestructive method for determining the quantity of plutonium in material in bulk form by measuring the thermal power generated as a result of radionuclide decay. As a plutonium assay technique, calorimetry must be accompanied by supplemental measurements to determine the proper relationship for converting the thermal power to the quantity of plutonium. Although the actual power measurement is accurate on a wide range of materials, it is the ability to accurately determine the conversion relationship that limits the application of calorimetric plutonium assay.

The conversion of a power measurement to a mass of plutonium is sometimes accomplished by considering calorimetry an absolute plutonium assay method, i.e., an assay technique that does not depend on representative calibration standards for accurate assay. Theoretically, a measurement of the thermal power from plutonium can be converted directly to plutonium mass if the relative abundance of each plutonium isotope and americium-241 is known. However, practically, this general method for converting calorimetric measurements of power to plutonium mass has two limitations: (1) the nuclear decay constants of the plutonium isotopes used in the conversion are known only to within approximately $\pm$0.5% and (2) the present state of the art in measuring the relative abundance of plutonium isotopes limits the accuracy of this approach. Even with these limitations, the use of calorimetry as an absolute measurement whose interpretation is based on fundamental nuclear constants is adequate in many situations.

Calorimetry is capable of higher accuracy and is applicable to more materials when the assay results are interpreted through a calibration based on representative standards rather than conversion of the power measurement on the basis of radionuclide abundances and decay constants. A representative standard for calorimetry need not have all the same physical characteristics of the unknowns as it does for other NDA methods. For calorimetry, the standard need only be similar to the unknowns in a few characteristics, the most important of which is the relative abundance of each radionuclide. If material is processed in batches and the batches are characterized by a single plutonium isotopic composition, a single standard from each batch is usually sufficient to normalize the calorimeter calibration to allow accurate interpretation of the power measurement. Such a

standard can be obtained from the batch at a point where the batch material is reasonably pure and homogeneous. Calibration based on this standard is then applicable, with corrections for decay of the plutonium isotopes, for the life of the material in the process. Nondestructive gamma ray spectrometry can be used to verify the plutonium isotopic composition and to assure that the correction of the calibration relationship for decay is valid.*

Under the sponsorship of the Institute of Nuclear Materials Management (INMM), Subcommittee N15.8 of the American National Standards Institute (ANSI) Standards Committee N15, Methods of Nuclear Control, has developed American National Standard N15.22, "Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Material Control." This standard identifies procedures for the calibration and operation of calorimeters to be applied for measuring the thermal power generated from the radioactive decay of plutonium and its daughters. The standard also describes, in detail, methods for interpreting the power measurement in terms of plutonium mass, either through a calculation using the measured radionuclidic abundances and their decay constants, or through a representative calibration. Methods for correcting the calibration or relative radionuclidic abundances for Pu-241 decay and Am-241 ingrowth are also discussed in this standard, as well as procedures for computing an estimate of the total error in the plutonium assay to be used in calculating the limit of error (LE) in the material unaccounted for (MUF).

Calorimetry can be used not only for routine plutonium assay but also to improve other NDA methods for plutonium assay and to ease the task of estimating the error in other NDA methods. Radiometric plutonium assay methods of spontaneous fission detection and quantitative gamma ray spectrometry are susceptible to interferences resulting from variation in the composition of the material being assayed and rely on highly representative standards for accurate assay. Calorimetry is not susceptible to most of these interferences. Therefore, if the relative abundance of the plutonium isotopes is constant for each batch, errors can be detected and calibrations corrected by comparison of assay results from these NDA methods with calorimetric results.

*A regulatory guide is under development that identifies acceptable methods for verifying radionuclidic abundances. Reference 3 provides information for the selection of gamma rays to be used for verification.

C. REGULATORY POSITION

The procedures on operation and calibration of heat flow calorimeters specified in the June 1974, draft of American National Standard N15.22-1974,* "Calibration Techniques for Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Material Control," should be used for measurements of the power associated with plutonium decay. The conversion of the power measurement to plutonium mass should be made according to one of the methods discussed in ANSI N15.22-1974, i.e., either by a calculation using the measured abundances of the plutonium isotopes and americium-241 and the nuclear decay constants of these radionuclides given in N15.22-1974, or by reference to a calibration based on representative standards.

When material is processed in identifiable batches characterized by a single blend of plutonium isotopes, the calibration or radionuclidic composition used to convert power measurements to plutonium mass is the same for all material in the batch. If calorimetry is applied for plutonium assay using a batch calibration or batch radionuclidic composition, then the following procedures should be used to assure that calorimetric assay is accurate and reliable:

1. Measurement of the effective specific power** of the plutonium should be made on a sufficient number of samples to generate a reliable estimate of the sampling error. Preferably, samples should be taken at a point in the process where the blend originates.

2. The rate of increase of the effective specific power due to ingrowth of Am-241 should be used to determine the current value of the effective specific power at the time of each measurement.

3. The process should be studied to determine if separation of americium from plutonium could occur in process steps between the point of sampling for measuring the effective specific power and the point of measurement of the thermal power. If separation behavior is unpredictable, a redetermination of the effective specific power at later steps of the process is required for proper calorimetric plutonium assay.

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*Copies may be obtained from the Institute of Nuclear Materials Management, 505 King Ave., Columbus, Ohio 43201, ATTN: Mr. H. C. Toy.

**Effective specific power is defined in ANSI N15.22-1974 and is the quantity used to convert power to mass of plutonium.

4. If mixing of plutonium isotopic blends is suspected, either the effective specific power should be redetermined or the radionuclidic abundances should be monitored to detect such mixing and/or to allow the average effective specific power to be computed.

5. For each container of material assayed, the ratio of the abundances of two of the plutonium isotopes and americium-241 should be measured by gamma-ray spectrometry to verify the plutonium isotopic composition[3] and to verify the predicted americium-241 abundance.

REFERENCES

1. Regulatory Guide 5.11, "Nondestructive Assay of Special Nuclear Material Contained in Scrap and Waste."

2. Regulatory Guide 5.34, "Nondestructive Assay of Plutonium in Scrap by Spontaneous Fission Detection."

3. R. Gunnink and R. J. Morrow, "Gamma Ray Energies and Absolute Branching Intensities for ^{238}, ^{239}, ^{240}, ^{241}Pu and ^{241}Am," UCRL-51087, July 1971.