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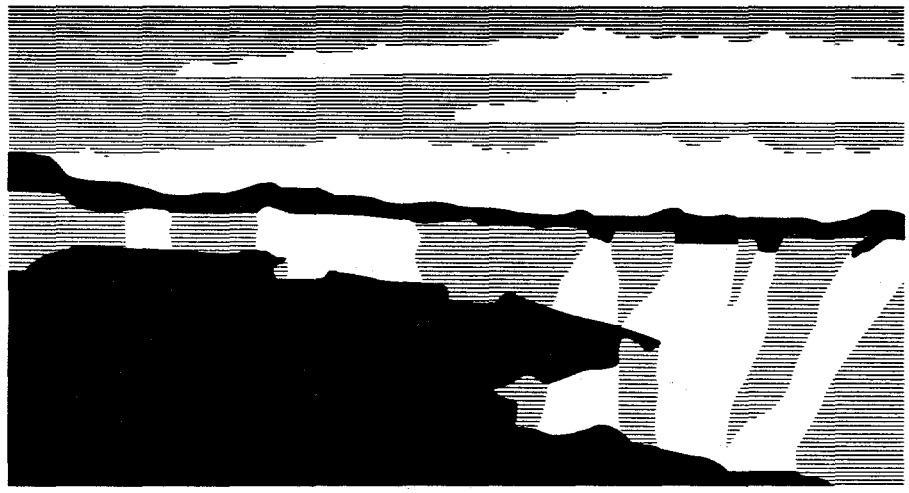

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CALORIMETRIC ASSAY OF MINOR ACTINIDES*

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ABSTRACT

This paper reviews the principles of calorimetric assay and evaluates its potential application to the minor actinides. We conclude that calorimetry and high-resolution gamma-ray isotopic analysis can be used for the assay of minor actinides by adapting existing methodologies for Pu/Am-241 mixtures. In some cases, mixtures of special nuclear materials and minor actinides may require the development of new methodologies that involve a combination of destructive and nondestructive assay techniques.

I. INTRODUCTION

Calorimetry is an important nondestructive assay (NDA) technique used for nuclear materials accountability and for accurate shipper/receiver measurements in most DOE plutonium and tritium facilities. Calorimeters are currently in use in DOE facilities such as Hanford, Pacific Northwest Labs, Lawrence Livermore, Sandia Livermore, Rocky Flats, Los Alamos, Savannah River, Mound, Idaho, and Pinellas. Also, calorimetry is now coming into use by the IAEA at the Westinghouse Hanford Site for bias-defects quality measurements of excess weapons materials. Calorimetry can determine sample power to about 0.2% for samples that emit more than 1 watt of thermal power. High-resolution gamma-ray spectroscopy or mass spectroscopy is used in conjunction with calorimetry to convert the measured sample power to total grams of plutonium. The combination of calorimetry and isotopic analysis is usually the most accurate NDA technique available for accountability measurements in a facility environment.

Most of the minor actinides that occur in the nuclear fuel cycle generate enough specific heat to make calorimetry a potentially viable technique for quantification. In some cases, the heat output is low enough to require very sensitive calorimeters and careful gamma-ray isotopic analysis. This paper reviews the principles of calorimetric assay and evaluates its potential application to the minor actinides.

II. PRINCIPLES OF CALORIMETRIC ASSAY

An isothermal calorimeter measures the constant heat output of nuclear material by allowing the sample to come to equilibrium in a constant temperature environment. Fine nickel wires in a Wheatstone bridge arrangement are used to measure the temperature rise through a calibrated thermal resistance (°C/Watt). Once the sample thermal power is determined, it is converted to mass using the sample specific power P_{eff}, as follows:

$$\text{MASS (grams)} = \frac{\text{POWER (Watts)}}{P_{eff}\ \text{(Watt/gram)}}$$

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The sample specific power is the sum of the specific powers of the isotopes that make up the sample as follows:

$$P_{eff}\,(\text{Watts/gram}) = \Sigma_i\ R_i P_i,$$

where R_i = mass fraction of isotope i,
P_i = specific power (Watts/gram) of isotope i.

Gamma-ray or mass-spectroscopy isotopic information is needed to determine the mass fractions of each isotope present in the sample.

Figure 1 shows a water-bath calorimeter developed by Mound Laboratories, and in common use throughout DOE facilities. The water bath is used to control the temperature of the calorimeter's environment. This water bath contains two twin 7-inch-diameter calorimeters of the type used by Westinghouse Hanford for nuclear materials accountability and for IAEA inspections of excess weapons materials.

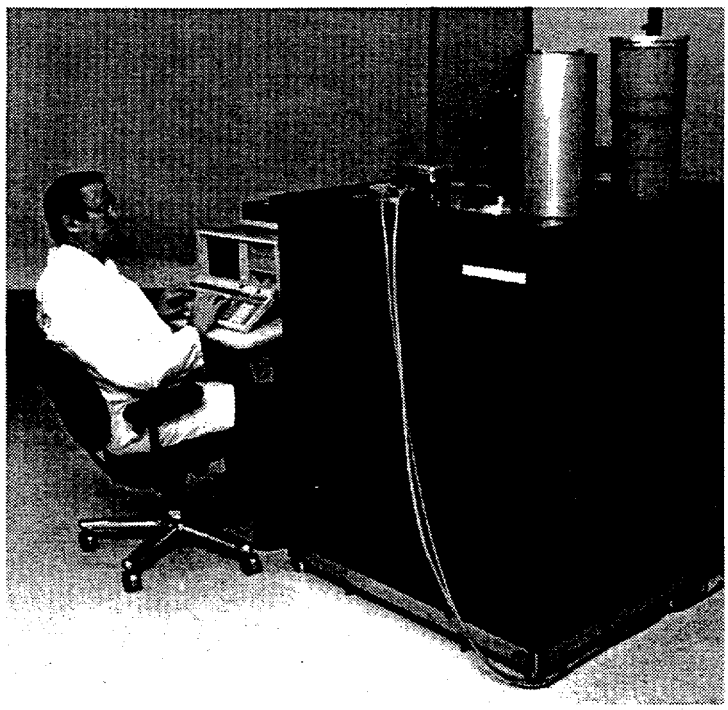

Fig. 1. A water-bath calorimeter developed by Mound Laboratories, and in common use throughout DOE facilities. The water bath contains a pair of twin calorimeters of the type used by Westinghouse Hanford for IAEA inspections of excess weapons materials.

Figure 2 shows a sensitive, transportable calorimeter developed recently by Mound Laboratories. This calorimeter is currently being used for measurements of highly enriched uranium (HEU), which emits only low amounts of heat, primarily from the U-234 that accompanies the U-235 in HEU (Ref. 1). A calorimeter of this type would be sufficiently sensitive and transportable to be suitable for IAEA measurements in a nuclear facility.

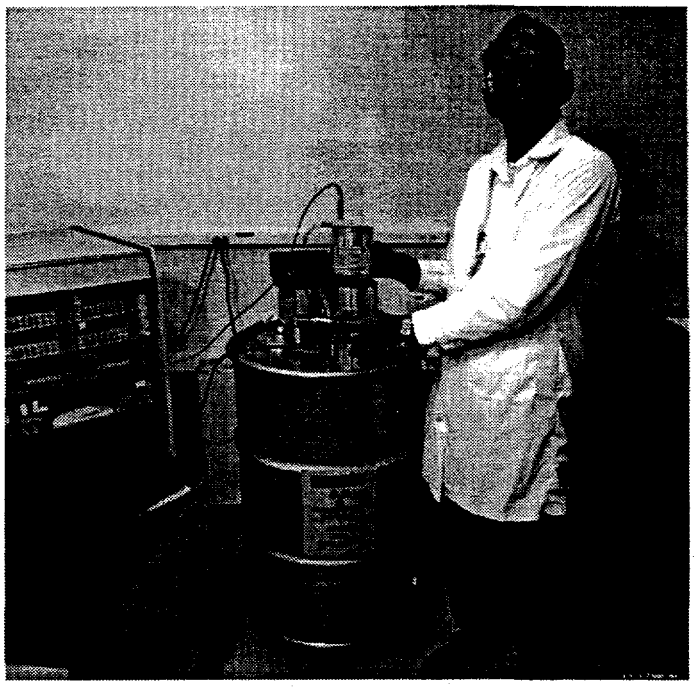

Fig. 2. A sensitive, transportable calorimeter developed at Mound Laboratories. This calorimeter is currently being used for measurements of HEU and would be suitable for IAEA measurements in a nuclear facility.

Important features of calorimetric assay include the following:

1. Sample power is routinely measured to an accuracy of 0.2% for high thermal power samples. Including the measurement error due to gamma-ray isotopic analysis, the accuracy of calorimetry/isotopics is typically 0.3 to 1.0%.
2. No physical standards are required. The calorimeter is calibrated relative to Pu-238 heat sources.
3. The measurement is matrix independent.
4. The measurement is traceable to the national measurement system through the Pu-238 heat source calibration and certification process.
5. American National Standards Institute and American Society for Testing and Materials measurement procedures are in place for Pu/Am assay using calorimetry and gamma-ray isotopics: (Refs. 2 and 3).
6. The calorimeter measurement is slow compared to neutron counting. Typical assay times are 4 to 8 hours for calorimetry, and 1 hour for the associated high-resolution gamma-ray spectroscopy analysis.

III. SPECIFIC POWERS FOR MINOR ACTINIDES

Table I summarizes the specific power in milliwatts per gram for some minor actinides and special nuclear materials. The same information is portrayed in Figure 3 as a graph of relative specific powers. The specific powers range from high values such as 324 milliwatts per gram for tritium to low values such as 0.0018 mW/g for HEU that contains 1% U-234. Actinides such as Am-241 and Am-243 have specific powers that make them easy to measure by calorimetry. For Np-237, the specific power is 0.0207 mW/g. A sample of Np-237 would have 100 times less the specific power of a Pu-239 sample with a similar mass. Thus the relative accuracy of a calorimetric assay of Np-237 would be less.

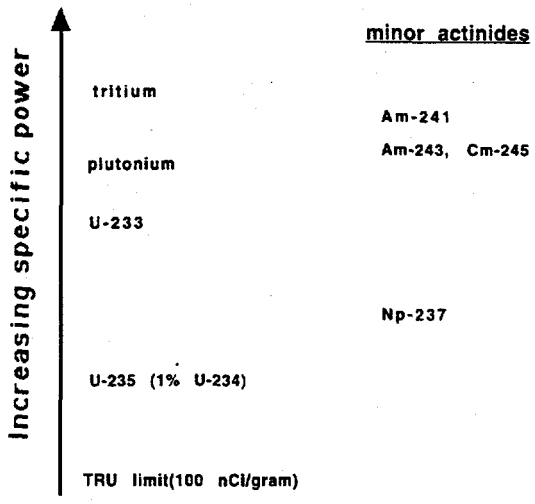

Fig. 3. Relative specific powers for some special nuclear materials and minor actinides.

TABLE I. The Specific Power, in Milliwatts Per Gram, for a Number of Minor Actinides and Special Nuclear Materials.

Minor Actinides		SNM	
Isotope	Specific Power (mW/g)	Isotope	Specific Power (mW/g)
^{232}U	689.7	tritium	324
^{232}U*	$5.3x10^3$	^{238}Pu	567.6
^{233}U	0.2808	^{239}Pu	1.929
^{234}U	0.1803	^{240}Pu	7.0824
^{241}Am	114.4	^{241}Pu	3.412
^{243}Am	6.436	^{242}Pu	0.1159
^{245}Cm	5.698	^{235}U	5.996E-05
^{237}Np	0.02071	^{238}U	8.51E-06
*including progeny at secular equilibrium			

IV. ISOTOPIC MIXTURES

There is considerable experience with the use of calorimetry and high-resolution gamma-ray spectroscopy to nondestructively determine the isotopic compositions and masses of mixtures of SNM. Mixtures that have been determined in this way include the following:

1. Am-241/Pu
2. Isotopically heterogeneous Am-241/Pu
3. U/Pu (mixed oxide)
4. (Pu/Am-241)/Np-239

However, for some mixtures of SNM and minor actinides, the use of calorimetry and high-resolution gamma-ray spectroscopy has not yet been studied, and further evaluation is needed. An example is given in Table II, below, which summarizes the heat output from a hypothetical sample containing 1 kg of Np-237 and 10 g of Pu of either weapons-grade or reactor-grade isotopic composition. In this example, the plutonium content is about 1% of the total nuclear material content. However, the plutonium may yield as much or more heat as the neptunium. Thus it is important to investigate the accuracy with which high-resolution gamma-ray spectroscopy can determine the Pu/Np ratio. At Los Alamos, we have begun to make calorimeter and gamma-ray isotopic measurements of Np-237/Np-239/Am-243 samples.

TABLE II. Heat Output Calculation for a Hypothetical Sample Containing 1 kg of Np-237 and 10 g of Plutonium of Either Weapons-Grade or Reactor-Grade Isotopic Composition.

Case 1:	^{237}Np and 1% by weight reactor grade Pu (25% ^{240}Pu)
	Total power for 1000 g ^{237}Np = 21 mW
	Total power for 10 g Pu = 135 mW
	% of power due to ^{237}Np = 13%
Case 2:	^{237}Np and 1% by weight weapons grade Pu (6% ^{240}Pu)
	Total power for 1000 g ^{237}Np = 21 mW
	Total power for 10 g Pu = 25 mW
	% of power due to ^{237}Np = 46%

V. CONCLUSIONS

Calorimetry and high-resolution gamma-ray isotopic analysis can be used for the assay of minor actinides by adapting the methodology developed for Pu/Am-241, as described in ANSI N15.22 and ASTM C 1030-89. We are beginning to carry out calorimetric measurements of minor actinides for test and evaluation of these methodologies. Mixtures of SNM and minor actinides will require further evaluation, and we are beginning to study the technical requirements for assaying such mixtures. In some cases we will develop new methodologies that involve a mixture of destructive analysis techniques and NDA techniques such as calorimetry, gamma-ray isotopics, active neutron counting, or passive neutron counting.

REFERENCES

1. C. R. Rudy, L. J. Satkowiak, W. W. Rodenburg, and J. A. McDaniel, "Calorimetric/Gamma-Ray Assay of High Enriched Uranium," *Nucl. Mater. Manage.* **XXII**, 1118-1123 (1993).

2. American National Standards Institute standard for nuclear materials ANSI 15.22 -1987, "Plutonium Bearing Solids - Calibration Techniques for Calorimetric Assay," approved January 1987.

3. American Society for Testing and Materials standard test method ASTM C 1030 - 1989, "Standard Test Method for Determination of Plutonium Isotopic Composition by Gamma-Ray Spectroscopy."