

# THE USE OF CALORIMETRY FOR SMALL SAMPLE PLUTONIUM MASS DETERMINATION

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## Abstract

Recent improvements in measurement precision and reductions in measurement times have made calorimetry attractive as an alternative to some Destructive Assay (DA) for plutonium measurement. These improvements include operating the calorimeter measurement chamber at room temperature in order to match the calorimeter to the sample (effectively preheating the calorimeter) and improving the equilibrium fit and the sample equilibrium power prediction procedures. A new feature used in measurement validation is an external electric sample. The sample power may be supplied from an external calibrated electrical power source. Alternatively, the calorimeter internal electrical calibration power supply and measurement circuitry may be employed. This paper illustrates the use of these new features and presents the results of measurements on small plutonium samples and calibrations using an external electrical sample.

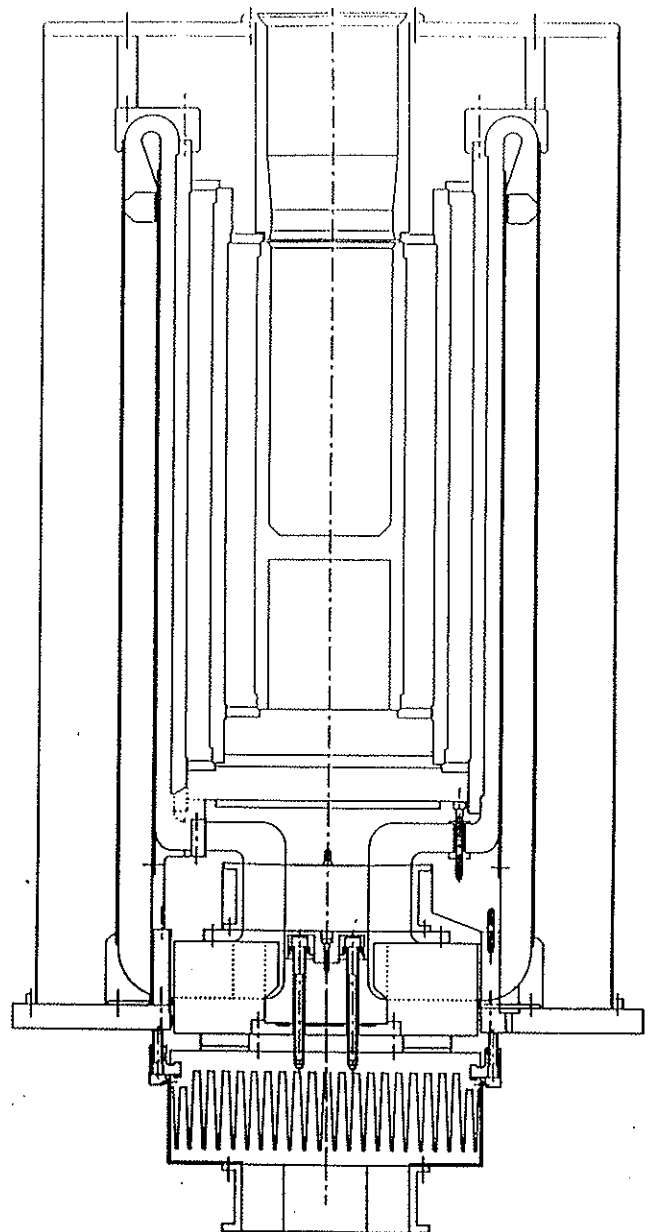
## 1. Introduction

The advantages of calorimetry as a non-destructive technique /1,2,3/ for the assay of plutonium bearing materials are well established. The insensitivity of calorimetry to sample inhomogeneity, neutron multiplication effects and the presence of moisture makes the technique more reliable than passive neutron coincidence counting over the range of possible sample materials.

A calorimeter measures the thermal energy or heat evolved from the radioactive decay by alpha particle emission of plutonium isotopes and americium. In order to obtain plutonium mass from the heat measurement it is necessary to obtain measured plutonium and americium isotopic ratio data and combine these data with well established plutonium and americium specific power data. With the reduction in calorimeter measurement times to between two and four hours and recent improvements in the measurement of isotopic ratio data by high resolution gamma-ray spectrometry, the potential applicability of calorimetry for both nuclear safeguards and materials assay has increased significantly.

The small sample prototype calorimeter has a number of novel features which are listed below:

- a. Traceable electrical calibration using internal electrical heater or external electrical calibration sample.
- b. Sample equilibrium power prediction procedure controlled by expert system software.
- c. Peltier thermo-electrical cooling enables room temperature operation of the measurement chamber.
- d. Sample simulation facility using the internal electrical calibration heater simulates a measurement. It may be used



Transportable Small Sample  
Calorimeter Thermal Element

Figure 1

as a comprehensive hardware and software test procedure and for operator training.

e. The instrument is transportable and consists of a thermal element mounted on a trolley frame and an electronic instrument rack on wheels.

- f. Water is not used as a heat-transfer medium reducing the criticality hazard.
- g. The measurement chamber operating power and temperature is user software adjustable.
- h. The instrument is fitted with a redundant and fail-safe sample overheating protection system.
- j. The thermal element is highly instrumented with direct temperature indication using linear thermistors and heater power measurement for all temperature controlled surfaces.
- k. Comprehensive hardware and software diagnostic and test features are included in the system.

## 2. Calorimeter Operation

The sample thermal power is determined by measuring the change in the electrical power applied to the measurement chamber once a sample has been inserted. Precise measurements of the difference in the applied electrical power are obtained through measurements of electrical current and voltage and these are based on electrical calibration.

Sample preheating has been used in the past to reduce the time a sample needs to spend in the calorimeter measurement chamber during a measurement. This procedure is avoided by operating the calorimeter measurement chamber at the sample can surface temperature. Typically for small samples this is just above room temperature. This form of operation is made possible using a Peltier air-to-air heat exchange cooling system.

A cross section diagram of a prototype small sample calorimeter is displayed in Figure 1. The Peltier thermo-electric air-to-air heat exchanger or heat pump can be seen at the bottom of the figure. The outer cylinder is maintained at a low temperature, typically 12 °C, by air which is circulated in a closed loop passing through the cold side of the Peltier cooling unit.

As a result of maintaining the outer cylinder, which acts as the heat sink, at 12 °C it is possible to operate the inner cylinder at around room temperature, typically 25 °C. The benefits of calorimeter operation at room temperature are further enhanced with the use of improved equilibrium power fit and the sample equilibrium power prediction procedures.

## 3. Measurement and Calibration Results

Measurements were performed with a series of calibrated small plutonium samples at the PERLA Laboratory at JRC Ispra. The results of these measurements are displayed in Table 1. For short measurement times, 1 - 2 hours, the results were obtained with sample equilibrium power prediction. Other results correspond to the sample reaching thermal equilibrium in the calorimeter.

Precise electrical calibration provides a means by which the performance of the instrument may be regularly verified. Electrical measurements of voltage and current are based on a NAMAS (UK National Measurement Accreditation Service) certified calibration which is traceable to standards at the National Physical Laboratory in the United Kingdom and at NIST in the United States.

Plutonium Sample No.	Measurement Time [hr]	Plutonium Sample [mW]	Measured Power [mW]	Error [%]	Power Error [mW]
31	2.3	5.5	9.0	63.64	3.5
31	1.6	5.5	6.4	16.36	0.9
31	2.3	5.5	7.9	43.64	2.4
31	2.8	5.5	6.7	21.82	1.2
21	2.7	9.8	10.2	4.08	0.4
21	1.8	9.8	12.5	27.56	2.7
21	1.9	9.8	12.6	28.57	3.8
21	4.1	9.8	12.3	25.51	2.5
21+31	3.0	15.3	16.3	6.54	1.0
21+31	3.4	15.3	16.1	5.23	0.8
21+31	2.9	15.3	15.8	3.27	0.5
21+22+31+32	5.1	30.6	35.2	15.03	4.6
6	36.3	130.9	132.7	1.38	1.8
6	2.1	130.9	133.1	1.68	2.2
6	3.0	130.9	129.3	-1.12	-1.6
6	1.2	130.9	132.9	1.53	2.0
6	1.2	130.9	132.4	1.15	1.5
9	1.7	131.2	131.7	0.38	0.5
8+9	1.3	261.2	265.4	1.61	4.2
6+7	2.3	263.0	263.9	0.34	0.9
6+8	2.5	392.9	392.5	-0.10	-0.4
7+9	1.0	393.2	396.2	0.76	3.0
6+9	1.9	524.1	525.2	0.21	1.1
6+9	2.3	524.1	521.3	-0.53	-2.8
6+9	2.1	524.1	523.5	0.11	-0.6

**Table 1:** Measurements of Small Plutonium Standard Samples

Run Number	Electrical Calibration Power (mW)	Predicted Power (mW)	Error (mW)	Time (m)	Equilibrium Power (mW)	Error (mW)	Time (m)
0809201	1222.1	1212	-10	56	1223	1.1	314
1009201	87.8	91	3.2	46	85	-2.8	420
1409202	93.6	97.1	3.5	74	94.3	0.7	188
1509201	92.4	90.7*	-1.7	86	94.7	2.3	108
1409201	93.8	118.7	24.1	47	98.7	4.9	230
1309202	93.9	86	-7.9	96	88.1	-4.8	226
1209201	1055.6	-	-	-	1052+	3.6	18

Table 2: External Electrical Calibration Sample Data

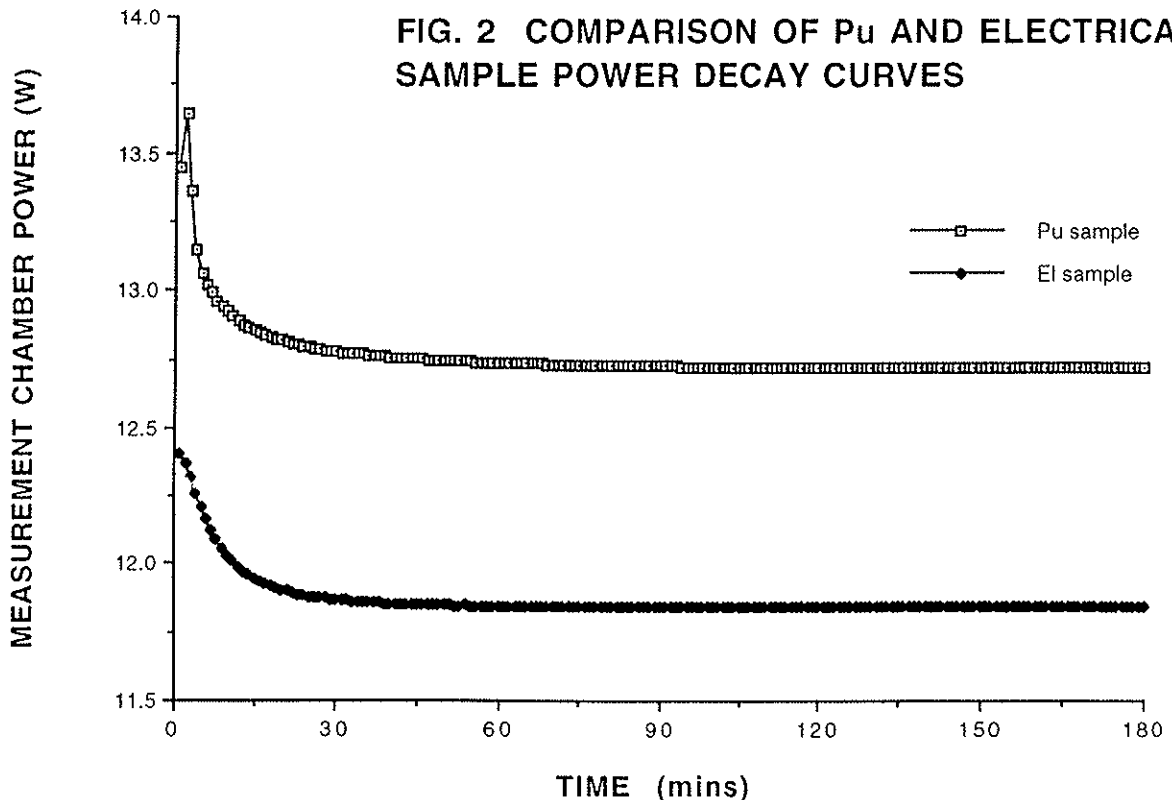
\* Equilibrium + Calibration Heater

As a further aspect of measurement validation the instrument is supplied with a calibrated external electric sample which may be connected to a calibrated external electrical power supply or to the calorimeter internal electrical calibration power supply and measurement circuitry. With corrections for heating in the connecting wires this external sample provides a very accurate method of calorimeter calibration and eliminates some of the need for the use of Pu heat standards. If independence of calibration is required, for example in international safeguards, an inspector may use the electric sample with an external calibrated power supply.

Data from the use of the external calibration sample are presented in Table 2. The predicted calibration power and the equilibrium calibration power are displayed in the table. The last result in the table used the internal electrical

heater and not the external calibration sample. For these measurements the uncertainty in the applied electrical calibration power is  $\pm 0.2$  milliwatts. A more precise electrical calibration is possible using the system.

The measurement and calibration results which are presented in Tables 1 and 2 were obtained using nickel wire resistance thermometry as a means of measuring the measurement chamber temperature. The instrument has also been fitted with high performance negative temperature coefficient thermistors as an alternative measurement chamber temperature sensor. Preliminary determinations of base power stability suggest that this new technology temperature sensor may lead to significantly improved power stability and hence power measurement precision. Further testing is in progress.



Typical measurement chamber electrical power decay curves for a plutonium sample and the electrical calibration sample are displayed in Figure 2. The absence of a sharp initial rise and subsequent fall in the measurement chamber power in the case of the electric sample is due the fact that the sample was present in the measurement chamber before electrical calibration power was applied.

#### 4. Conclusions

Calorimetry for small plutonium samples, when combined with gamma-ray isotopic data, provides a convenient, rapid and cost effective method for plutonium mass determination. Precise internal and external electrical calibration reduces the requirement for calibrated Pu heat standards.

The use of Peltier thermo-electric cooling allows measurement chamber operation at room temperature. The requirement for sample preheating is eliminated as the calorimeter is effectively 'preheated' to the sample temperature. Rapid measurements, with equilibrium sample power prediction in about 1.25 hours and final equilibrium in about 2.5 hours, are made possible. New technology in the form of thermistor temperature sensors shows promise as a means of significantly increasing measurement precision and reducing prediction times.

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