

DEVELOPMENT AND APPLICATION OF A LOW POWER TRANSPORTABLE CALORIMETER FOR PLUTONIUM ASSAY

J N Lowe

Atomic Weapons Establishment (Aldermaston)
U K Ministry of Defence (Procurement Executive)
Aldermaston, Reading, RG7 4PR, England

J A Mason *

A. N. Technology Ltd.
Technology Transfer Centre
Silwood Park, Ascot, SL5 7PW, England

ABSTRACT

As a precise and non-destructive technique calorimetry is an appropriate method for the measurement of plutonium bearing samples. The sample thermal power measured by the calorimeter, when combined with isotopic ratio data obtained from mass or gamma-ray spectroscopy, provides a measurement result in terms of plutonium mass. The transportable calorimeter described in this paper is of improved design and it is intended for use in the assay of low power (1 to 10 Watts) plutonium bearing samples. The samples, which are in sealed containers, have compositions which range from metal to oxide and metal-salt mixtures. The thermal and electrical characteristics of the calorimeter have been optimized in order to enhance measurement performance. Measurement chamber temperature stability is improved over earlier calorimeters of a similar design and heat-transfer modelling has been employed in the design to reduce measurement times. The device may be used with a preheater and the normal measurement process includes equilibrium sample power prediction. A 'friendly' software-user interface is employed so the instrument may be operated by semi-skilled staff. Comprehensive diagnostic and test software is also implemented on the system. Preliminary measurements are reported on a series of plutonium metal billets and smaller plutonium metal samples.

INTRODUCTION

The advantages of calorimetry as a non-destructive technique for the assay of plutonium bearing materials^(1,2) 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 this 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.

* On leave of absence from Imperial College of the University of London.

The present paper describes an isothermal low power (1 to 10 Watts) transportable plutonium assay calorimeter for use in measuring samples in sealed containers. The calorimeter operating temperature and measurement chamber power may be varied in order to match the calorimeter to the expected characteristics of a series of samples. The instrument includes a preheater and two sample canisters. The preheater is used to precondition a sample in its canister by matching the sample surface temperature to that of the calorimeter measurement chamber temperature. This process can significantly reduce measurement times.

INSTRUMENT DESIGN

The design of the instrument is similar in principle to earlier instruments^(3,4,5) however the thermal element is of an improved design based on heat-transfer analysis and the operating experience of earlier devices. The electronic system design has been driven by the need for robustness and reliability and a 'friendly' software user interface has been implemented in order to simplify the operation of the instrument. The isothermal calorimeter system contains extensive hardware and software diagnostic and test facilities.

The calorimeter system consists of three main hardware components, an instrumentation rack, the thermal element and the preheater. The first two of these may be seen in Fig. 1 in which a sample canister is also shown. The preheater can be seen in Fig. 2.

The calorimeter thermal element consists of three concentric aluminium cylinders which are separated by annular regions filled with a heat-transfer medium. Helical electrical heating coils wound around each cylinder are used to regulate the cylinder temperatures in order to obtain uniform and constant heat-flow between the cylinders through the heat-transfer medium. The inner, middle and outer cylinders are maintained at nominal temperatures of 45, 35 and 33 °C respectively and this configuration results in a measurement chamber operating power of about 20 Watts. These temperatures can be modified and as a result the calorimeter operating power may also be varied.

The cylinder heating coils form part of a closed loop control system. The cylinder temperatures are measured using nickel resistance thermometers each of which forms one arm of a sensitive bridge circuit. The output signal from each bridge circuit indicates the deviation of the appropriate cylinder average surface temperature from a set point temperature. These bridge output signals are amplified and digitized and form the error input signal to a proportional control system. The proportional control algorithms for cylinder temperature control are implemented on the system industry standard PC-AT compatible computer. The control

algorithm output signal is used to drive the cylinder heater power amplifiers through 16 bit digital-to-analog converters thus closing the temperature control loop.

The electronic measurement system has been optimized and the bridge circuitry employed in cylinder temperature measurement has been incorporated into both the thermal element and the preheater. This close coupling of the bridge circuitry to the measurement chamber has eliminated temperature effects in the cables and stray resistance problems associated with connectors.

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 calibrations of the measuring instrumentation which are traceable to national standards for voltage and resistance. An advantage of the measurement system is that it does not rely on a calibration using heat standards, which is a requirement with many other types of plutonium calorimeter.

Two methods are employed to reduce measurement times. The first of these is equilibrium sample power prediction. The algorithm which is used employs a single exponential function and this is fitted to an average of the measurement chamber power data as it is obtained. The sample power is determined, once stable values of predicted power are computed, using criteria that are provided as a guide to the operator. The second method involves the use of a preheater which preconditions samples to the thermal element measurement chamber temperature. At the time of writing the preheater has yet to be employed in measurements.

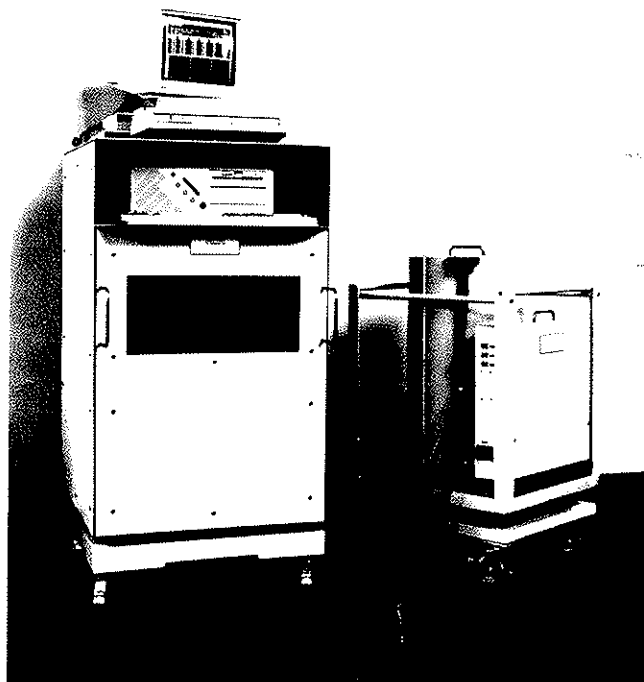


FIG. 1

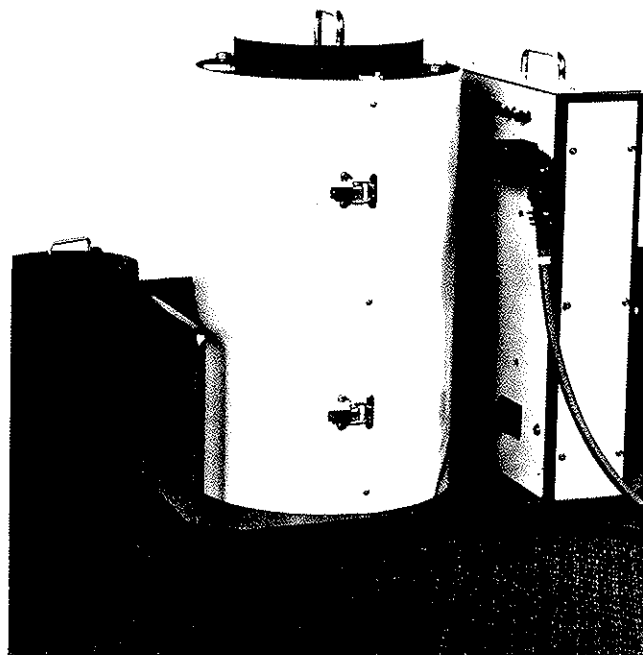


FIG. 2

In order to verify the correct functioning of the electronic measurement system an automatic electrical calibration procedure has been implemented under software control. Several fixed values of electrical power are delivered to the measurement chamber in sequence by means of a calibration heater winding. Both the applied calibration and the measurement chamber power are measured by the measurement system. At the conclusion of the procedure the ratios of measured to applied electrical power are determined and a least squares fit is performed and the results are reported to the operator.

'User friendly' software written in the 'C' language provides an interface to the operator and controls all of the functions of the instrument. The software is menu driven and the operator interacts with the calorimeter through a series of display screens. The screens are subdivided and some information is displayed as text, some in a numerical tabular form and some in graphical form. The screens contain descriptive information on the process currently in operation, on the state of the instrument and the results of the process when they are available. In addition to the screen displays information is provided to the operator by means of a printer which reproduces in hard copy form some of the data from the screens.

The commands (executed using function keys) which are available to the operator at any given time are displayed on the screens as are warning messages if operating conditions are outside the acceptable range or if operator intervention is required. Many of the commands require the operator to issue a confirmatory command before action is initiated. The software is supported by an uncomplicated but comprehensive User Guide.

The calorimeter determines a result in terms of sample power. If isotopic ratio data is provided a result is calculated in terms of plutonium mass. The isotopic data is decay corrected to the date of the measurement and decay correction calculations may also be performed 'off-line' while the calorimeter is in an idle state.

Measurement results are automatically archived on the system hard disk drive and isotopic ratio data files may also be stored on disk. The system maintains a Quality Assurance file on which measurement chamber baseline power (P_0) and the results of electrical calibrations are archived. These data may be examined subsequently in order to verify the correct operation of the calorimeter over a particular time period.

In addition to the calibration procedure a variety of diagnostic and test procedures are implemented on the calorimeter system under software control. All of the normal calorimeter functions and display screens are duplicated in what is called Diagnostic Mode. In this mode of operation, which repeats all of the normal functions of the instrument, additional and more detailed information is provided on the display screens and printout in order to assist in assessing the performance of the instrument and as an aid to fault diagnosis should this be required. As part of the diagnostic software subsystem an electrical sample simulation procedure has been implemented using the electrical calibration heater winding. Samples with different thermal powers and heat-transfer time constants can be simulated and the procedure is particularly useful in testing both the calorimeter power measurement system and the data analysis algorithms, including the equilibrium sample power prediction algorithm.

Two safety features have been incorporated into the calorimeter system to ensure that the plutonium sample cannot be overheated. The first is a relay power cut-out system. Thermistors on the three cylinders of the thermal element and on the preheater cylinder independently measure cylinder temperature. If a cylinder temperature exceeds a value of about 65 °C, the cut-out relays operate to remove the applied electrical power. The second safety system, employing 'watch-dog' timer circuitry, is also linked to the same power cut-out fail-safe relays. An electronic pulse derived from the temperature control algorithm running in the controlling computer is monitored by the watch-dog timer circuits which are located on the thermal element and the preheater. The pulse is produced while the cylinder temperature control algorithm is functioning. If the circuits do not detect the pulse after a delay of a few seconds, the cut-out relays operate to remove heater power from the thermal element and the preheater. In order to restore power to the system the operator must manually reset the watch-dog timer circuits and cut-out relays.

MEASUREMENTS AND RESULTS

A number of measurements have been performed and data has been obtained during the short time that the instrument has been in operation. Two aims of the heat-transfer design process were to ensure measurement chamber power stability and to minimise the thermal element time constant for heat-transfer. An indication of calorimeter power stability is evidenced in Fig. 3 which displays a plot of measurement chamber power over a period of 2000 minutes. The data represent 30 point averages of measurement chamber power and all of the data points fall within a band with a width of 15 mW.

An indication of the small thermal time constant of the calorimeter may be seen in Fig. 4 which shows the measurement chamber power as a function of time following switching the instrument on from a cold condition. Although complete measurement chamber power stability is not achieved until several hours after initial switch-on, the plot demonstrates that measurements at a lower level of precision may be obtained after a delay of the order of one hour.

A preliminary estimate of the measurement precision of the instrument was obtained by repeatedly measuring the same Pu metal sample. The sample, which was housed in a large stainless steel can, was placed in the canister and the peripheral gap was filled with aluminium shot. The measurements were performed without sample preheating and the measurement data is summarised in Table 1 below:

Table 1

P_0 (Watts)	P_s (Watts)	Run Time (min)
20.406	4.677	240
20.404	4.649	250
20.406	4.662	250
20.406	4.667	240
20.406	4.665	245
20.404	4.673	220

The mean power is 4.666 Watts with a standard deviation of 9 mW and a coefficient of variation of 0.2 %. Sample power equilibrium prediction was not used in these measurements and the samples were allowed to approach the equilibrium sample power value. The very poor thermal properties of the sample packaging (involving an additional outer stainless steel can) account in part for the long measurement times.

Similar measurements were made with the same sample but using aluminium foil as thermal coupling medium. A plot of the measured and predicted sample power is displayed as Fig. 5. It can be seen that the prediction routine produces an answer to within 0.2 % of the equilibrium value within two hours.

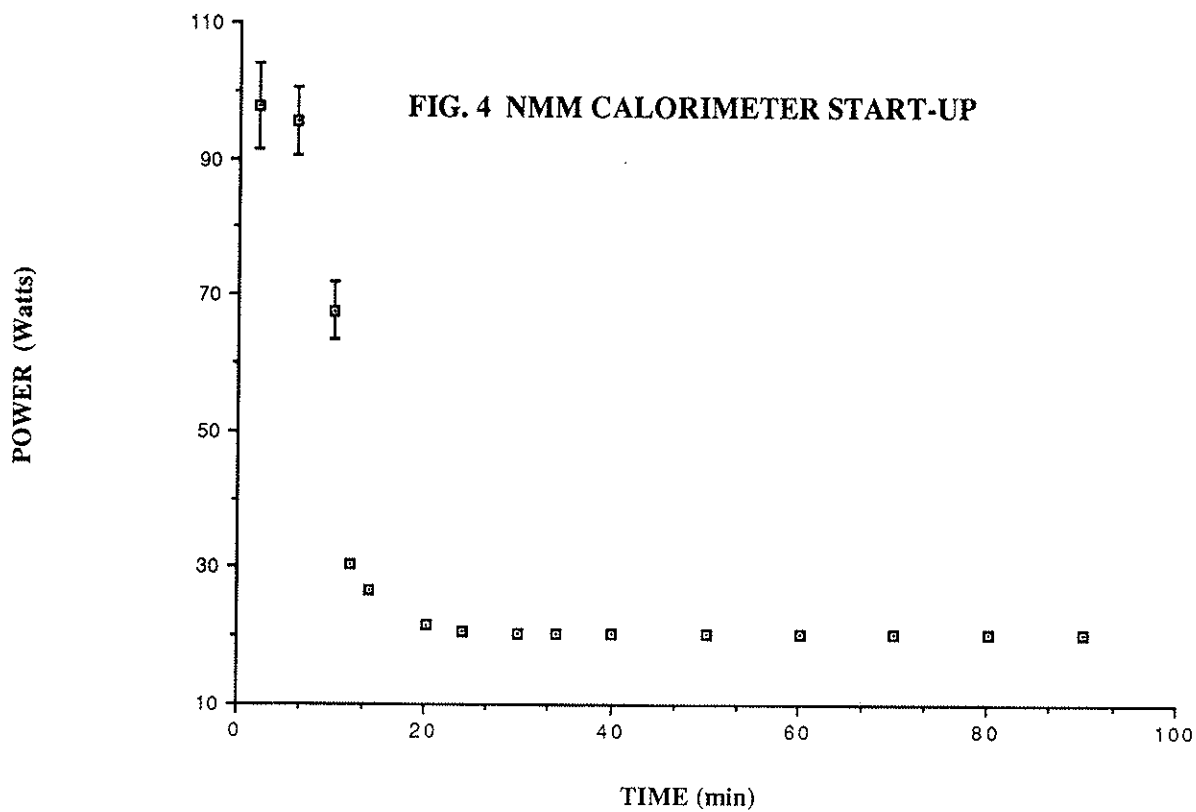
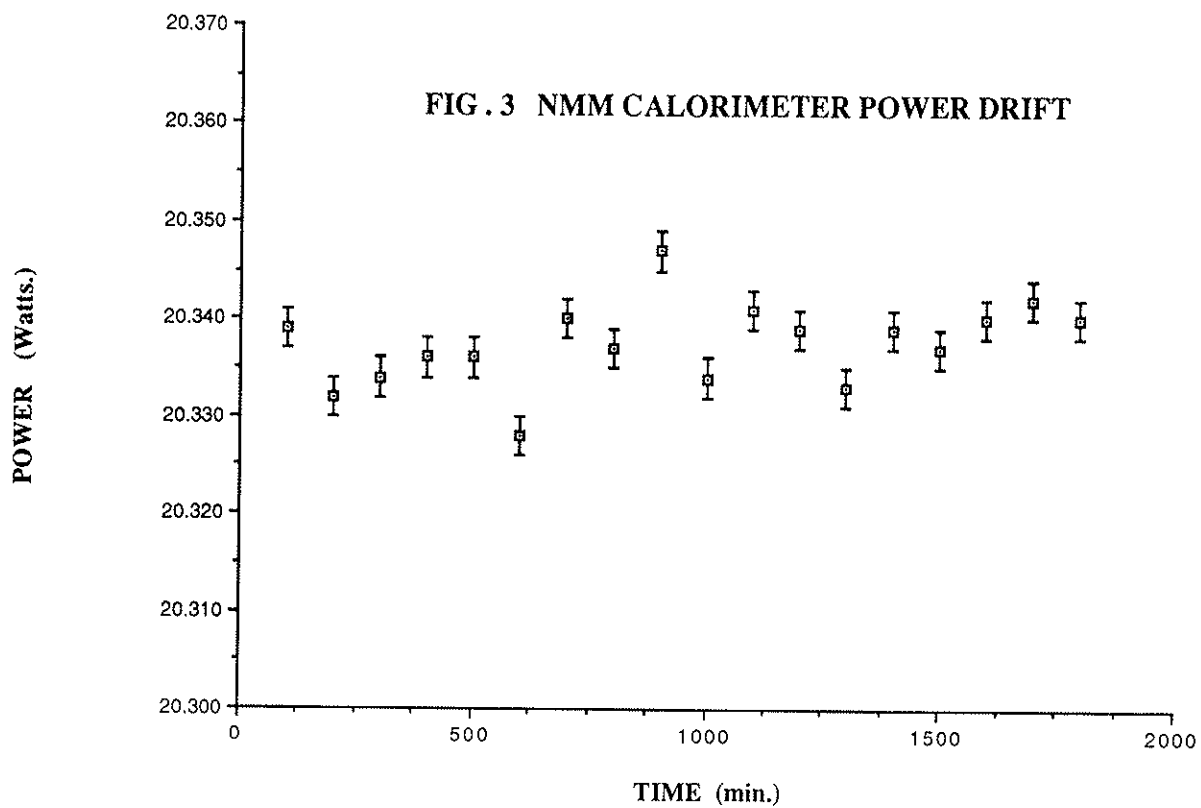
A limited number of measurements have been performed on small samples and the preliminary results are displayed in Tables 2 and 3. Equilibrium sample power prediction was employed to obtain the results in Table 2 for the 1.297 Watt sample and the short measurement times should be noted. Equilibrium sample power prediction was not employed to obtain the data presented in Table 3. In both cases the samples were contained in packaging which did not involve an additional stainless steel outer can and measurement times in the case of Table 3 are significantly reduced compared to those associated with the measurements presented in Table 1.

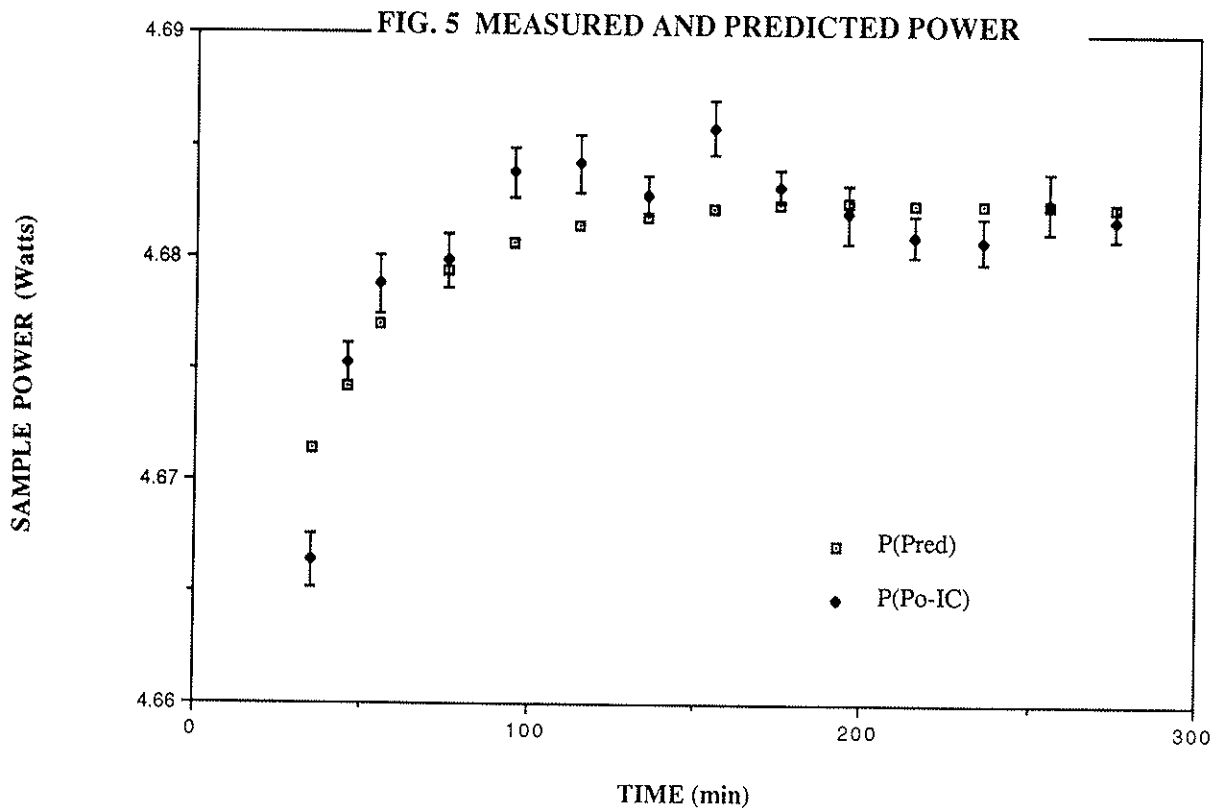
Table 2

P_0 (Watts)	P_s (Watts)	Run Time (min)
20.407	1.297	45
20.407	1.292	70
20.407	1.297	45

Table 3

P_0 (Watts)	P_s (Watts)	Run Time (min)
20.411	0.254	50
20.411	0.255	95
20.411	0.256	125





CONCLUSION

These preliminary measurement results confirm that the calorimeter has met its measurement precision design target of 0.5 %. At the time of writing the calorimeter has been operating reliably for a period of approximately three months and it is being used routinely to perform one or two measurements per day. In the case of the 0.255 Watt sample four repeat measurements were performed in one day. Although preheating was not used, the measurement procedure involving repeated measurements of the same sample was equivalent to the use of preheating.

The calorimeter used in these measurements was manufactured by A. N. Technology Ltd. of Ascot, England and the instrument is distributed by that company and by Jomar Systems Inc.

The authors wish to acknowledge the contribution to this work made by Mr J Preston, Mr L Benjamin and Mr T Sargent of AWE(A).

REFERENCES

1. J.A. Mason, The Use of Calorimetry for Plutonium Assay, Report SRDP-R100, Safeguards R and D Project, United Kingdom Atomic Energy Authority, 1982.
2. American National Standards for Nuclear Materials, Plutonium-Bearing Solids Calibration Techniques for Calorimetric Assay, Report ANSI N 15.22-1987, American National Standards Institute, Inc., 1987.

3. J.A. Mason, R.W. Wilde, J.C. Vickery, B.W. Hooton and G.M. Wells, Development and Evaluation of a Plutonium Assay Calorimeter Test-Bed, Proc. of the 29th Annual Meeting of the Institute of Nuclear Materials Management, Las Vegas, Nevada, USA, June 1988.

4. J.A. Mason, R.W. Wilde, J.C. Vickery, B.W. Hooton, G.M. Wells, M. Cuypers and S. Guardini, Plutonium Calorimetry, Proc. of the 11th ESARDA Symposium on Safeguards and Nuclear Materials Management, Luxembourg, June 1989.

5. J.A. Mason, B.W. Hooton, G.M. Wells, and B. Metcalf, Development and Evaluation of a Transportable Large Sample Plutonium Assay Calorimeter, Proc. of the 30th Annual Meeting of the Institute of Nuclear Materials Management, Orlando, Florida, USA, July 1989.