Design and Performance of a Sensitive Multi-Mode Calorimeter for Single Cell Isothermal or Single and Twin Cell Heat-Flow Measurements of Plutonium or Tritium-14003

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ABSTRACT

In this paper the design and performance testing of a sensitive multi-mode calorimeter intended for single cell isothermal or both single and twin cell heat-flow measurements of plutonium or tritium or other heat producing radioactive substances is presented. The calorimeter measurement chamber and thermal element consists of three concentric cylinders. High sensitivity and thermal stability are achieved through the use of high output voltage differential thermopile heat-flux sensors positioned between the inner and middle cylinders. Conventional electrical resistance sensors with a high temperature coefficient of resistivity are used to determine the average surface temperature of the middle and outer cylinders of the thermal element. In the present application the calorimeter is operated in single cell isothermal mode for the measurement of plutonium in 3013 containers, which have a diameter of 125 mm and a height of 254 mm. The thermal element is insulated from ambient temperature fluctuations and employs a peltier cooling unit to remove heat from samples during measurements. Three calorimeters of this design form part of an automated assay system for use at the Mixed Oxide Fuel (MOX) facility being constructed at the Savannah River Site. They are designed for automatic operation with the plant controlled through a Process Computer and with loading and unloading conducted by a gantry robot crane. Alternatively, they can be operated as stand-alone units. The calorimeters incorporate a removable electrical calibration sample with electrical heater and an internal calibrated power supply for both calibration and performance checks. Calibration can also be performed using traceable radioactive heat standards. Measured precision and accuracy for different sample powers is presented and minimum levels of detection are determined using the electrical calibration sample and zero power measurements. Results from electric sample measurements of different thermal powers are presented. These measurement end point results are “declared” when the calorimeter equilibrium fit criteria are met by the data-fitting algorithm. Although designed specifically to accept automatically loaded 3013 containers, the sensitive and flexible multi-mode calorimeter concept is applicable to the measurement of a wide range of radioactive materials in a variety of sample containers.

INTRODUCTION

Radiometric calorimeters tend to be of two distinctly different types: Isothermal or Heat-flow, with different operating characteristics [1]. Isothermal calorimeters [2, 3], (single cell) employ servo-control and use the power replacement mode of operation. Measured electrical power (called the Base Power) is applied to the measurement chamber (cell) in order to maintain a constant temperature profile on the inner cylinder and between the cylinders. Following the insertion of a heat-producing sample, the servo control mechanism reduces the applied electrical power necessary to maintain the constant temperature profile, in such a way that the thermal power of the sample replaces the applied electrical power. At thermal equilibrium, the difference in the applied electrical power is a measure of the thermal power of the sample – hence the description "power replacement mode". Calibration, using a heat or electrical standard involves confirming a linear response (slope equal to 1) with only a small bias. Typically, isothermal calorimeters have shorter measurement times but with less accuracy and precision than heat-flow calorimeters.

Higher sensitivity and longer measurement times but with improved precision and accuracy are the characteristics of heat-flow calorimeters [4]. They usually employ twin cells – both a measurement chamber or cell and a reference chamber for compensation. Calibration involves establishing the linear relationship between the output signal of the sensors of the measurement chamber and the thermal power of the measured sample. In the present case we are correlating the voltage output of thermopile sensors of the measurement chamber with sample thermal power and determining the calorimeter sensitivity, typically expressed in units of microvolts per miliwatt (µV/mW). Improved precision is achieved in the twin cell configuration where the output signal of the reference cell is subtracted from the output signal of the measurement cell. Common mode effects such as disturbances caused by variations in the ambient environmental temperature are thus eliminated.

A novel feature of the present design is that the thermal element can be configured as a single cell isothermal calorimeter operated in power replacement mode. This is how the calorimeters are configured and will be operated at the Shaw AREVA MOX plant at Savannah River, where they are to be installed as model CD285-3013 Isothermal Calorimeters. Alternatively, the thermal element can be configured as a single or (with another identical thermal element) a twin cell calorimeter operated in heat-flow mode. The choice to deploy the calorimeters in the isothermal configuration was base on meeting the requirements for measurement time (8 to 12 hours) with adequate precision and at the same time dissipating heat from samples producing more than 15 watts.

A second important improvement involves the mounting of rectangular thermopile sensors in a cylindrical geometry configuration. This feature is particularly relevant to the measurement of cylindrical 3013 containers where, as a result of the use of a cylindrical measurement chamber, close thermal coupling is achieved between the measurement chamber and the sample, reducing measurement time. Many measurements reached equilibrium in less than 6 hours and all in less than 10 hours.

In this paper the design and performance testing of a series of sensitive multi-mode calorimeters designed for single cell isothermal or both single and twin cell heat-flow measurements of plutonium or tritium or other heat producing radioactive substances is described. In the present case, the Isothermal Calorimeters are part of an automated plutonium handling facility including the three calorimeters, two gamma spectrometers for plutonium isotopic ratio determination and an automated gantry robot for automatic loading and unloading of 3013 containers.

DESIGN

In Fig. 1, which shows the model CD285-3013 Isothermal Calorimeter, the thermal element is on the left and the control electronics are on the right. The calorimeter plug unit (top left) has a lifting feature (as found on the 3013 sample containers) and the feature on the top right is a storage well where the plug unit is positioned by the gantry robot during sample loading and unloading.

Fig. 1. Model CD285-3013 Isothermal Calorimeter.

Fig. 2 shows a cross section view of the top of the calorimeter thermal element. It consists of three concentric cylinders manufactured from aluminium alloy. Copper heater and nickel sense coils are wound around the outer surface of the middle and outer cylinders for electrical heating and temperature control. A copper heater coil is also included on the inner cylinder. Thermopile sensors for heat flow measurement are located in positions in the gap between the inner and middle cylinders.

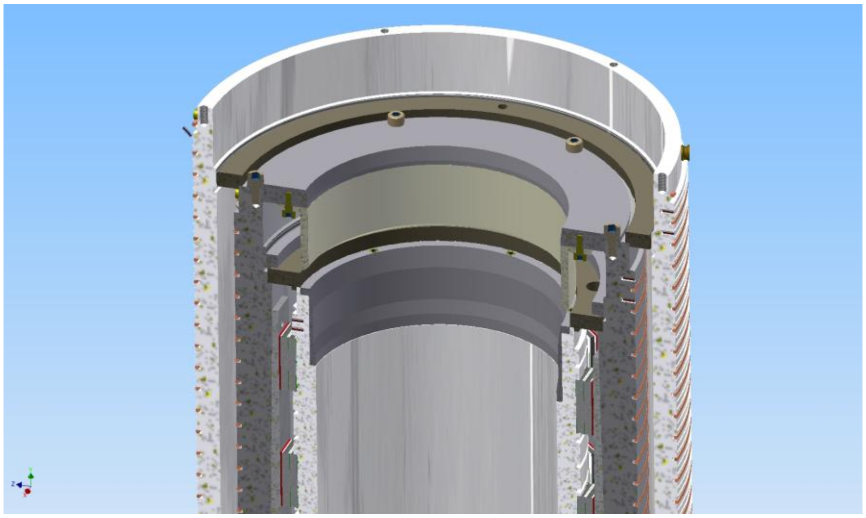

Fig. 2. A cross section view of the top of the calorimeter thermal element.

Care has been taken in this novel design to ensure good thermal contact between the double thermopile assemblies and the walls of the inner and middle cylinders between which they are sandwiched. At the same time an air gap of almost 10 mm has been maintained between the cylinders where the thermopiles are not fitted. The thermopile sensor assemblies occupy less than 20% of the volume of the gap between the cylinders.

The calorimeter sensitivity as a function of the number of double thermopile sensor assemblies embedded within the annular gap between the inner and middle-cylinders has been calculated using a simplified heat-transfer model of the calorimeter and the results are displayed in Fig. 3.

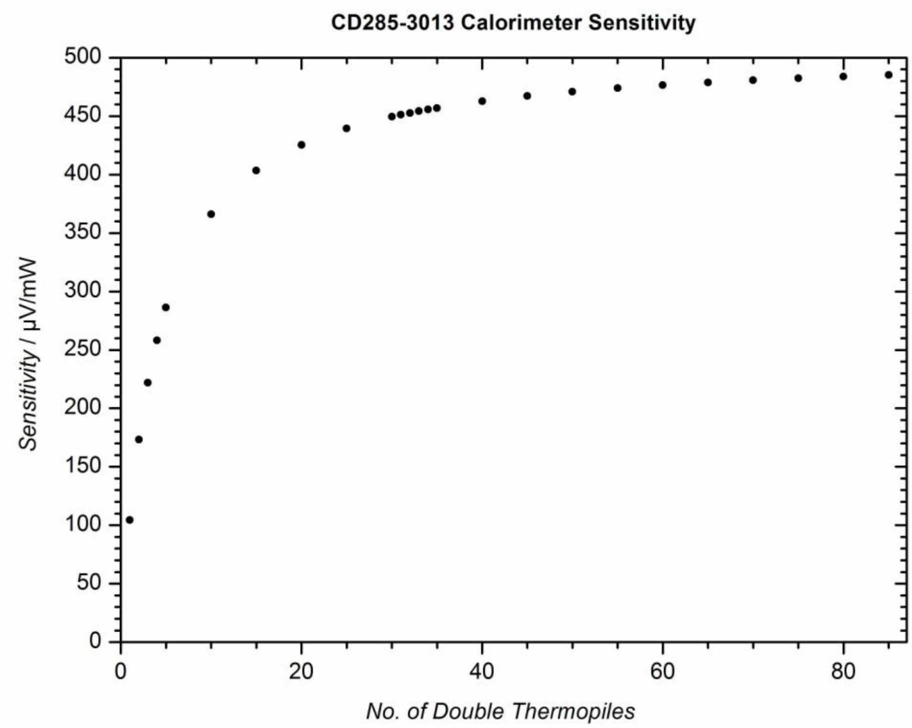

Fig. 3. Using a heat-transfer model, the calculated calorimeter sensitivity is plotted as a function of the number of double (two fixed together) differential thermopile heat-flow sensor assemblies that are positioned in the gap between the inner and middle cylinders.

A total of 32 sensor assemblies were selected and installed in the thermal element, resulting in a very high estimated calorimeter sensitivity of 452.7 µV/mW, when operated in single cell heat-flow mode. The errors in the calculation are greater than the errors for a similar calculation performed for the large volume calorimeter model CHF400-5300 [4], which had rectangular cells with a constant gap between the inner and outer walls of each cell. In the present case the model used an approximation to the cylindrical geometry where the inter-cylinder gap was not constant, resulting in larger errors in the sensitivity calculation.

In isothermal operation the middle cylinder, with significant heat capacity and hence thermal inertia, is regulated at a fixed average temperature by a nickel sense coil wound around the cylinder surface that controls the electrical power supplied to the middle cylinder heater winding. The inner cylinder average temperature is controlled in relation to the middle cylinder temperature by a similar heater winding but in this case control is achieved using the heat-flow thermopile sensor assemblies. The thermopile sensor assemblies are more sensitive than conventional nickel sense windings and have a significant voltage output resulting in a higher signal to noise ratio than can be achieved with a nickel sense winding. This fact explains the greater sensitivity of this multimode calorimeter technology.

An outer cylinder surrounds the middle cylinder and the gap between the two cylinders is filled with a silicon compound with a relatively low but constant thermal conductivity. The outer cylinder is controlled using a nickel temperature sense winding and a heater coil wound around the outer cylinder surface and the thermal resistance of the middle – outer cylinder gap creates a temperature gradient between the cylinders.

The resulting constant temperature gradient from the inner to the outer cylinder produces a continuous heat flow, whether a heat-producing sample is present in the measurement chamber or not. A peltier cooling unit removes heat from the outer cylinder as part of the process of maintaining the calorimeter constant temperature gradient.

CALIBRATION AND MEASUREMENT RESULTS – ISOTHERMAL OPERATION

The linear calibration of calorimeter No. 2 (for which more data was available than for the other calorimeters) is plotted in Fig. 4.

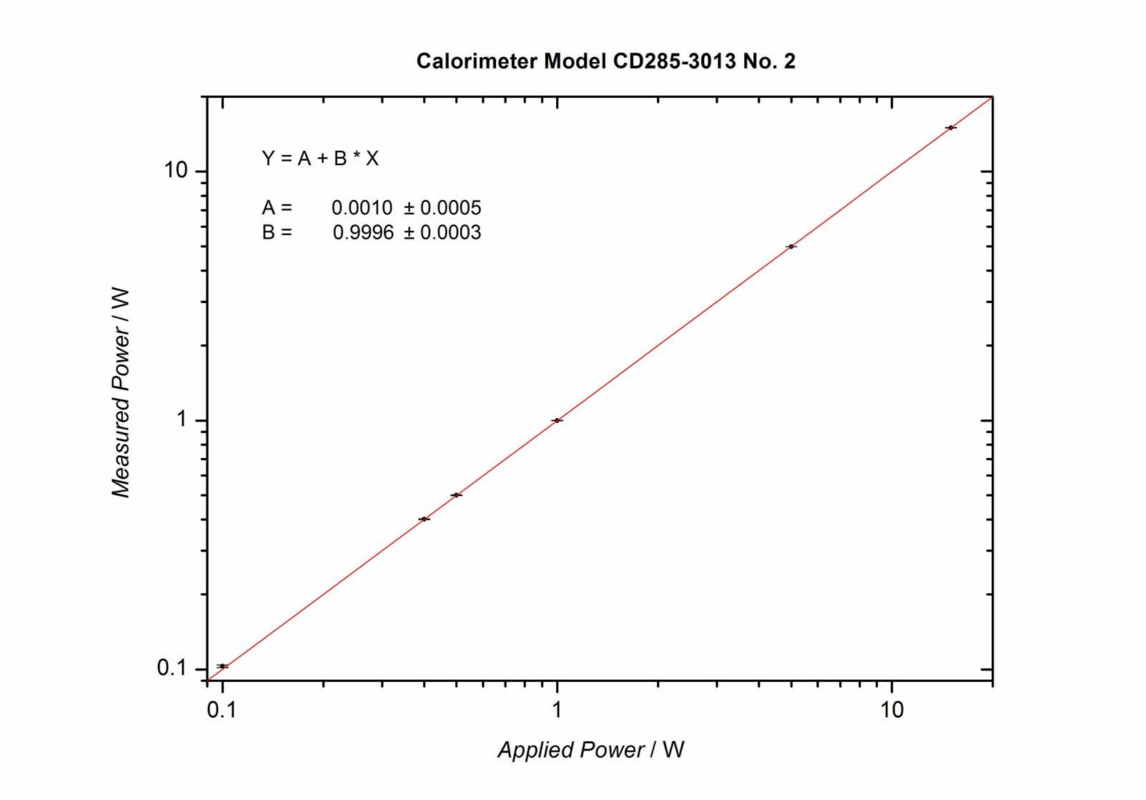

Fig. 4. Isothermal Calorimeter calibration data showing the measured calorimeter sample thermal power plotted as a linear function of applied sample electrical power provided by an electrical heat standard sample container, for calorimeter No. 2.

All three isothermal calorimeters have been calibrated using the same electrical heat standard, which consists of a suitably modified 3013 container with an internal electrical resistance-heating element. Electrical power is supplied to the heat standard through a thermally isolated electrical connector. Electrical power for calibration and performance check measurements is provided by a controlled precision power supply built into each calorimeter instrument rack.

A digital voltmeter and a precision resistor plate, which form part of the precision power supply, are calibrated (traceable to international electrical standards) on an annual basis. Fig. 4 shows, (for calorimeter No. 2) the measured calorimeter sample thermal power plotted as a linear function of applied sample electrical power. An electrical heat standard sample container, to which the calorimeter electrical calibration power supply is connected, provides the sample thermal power. Note that the slope of the plotted data is effectively 1.0 with a very small error.

The calibration data for all three calorimeters are very similar as expected and as illustrated in TABLE I. The calibration data for all three instruments have slopes of 1.0 with very small errors. This result is a conformation of both the quality of the components and the care taken, especially by one of the authors (BMS), during the calorimeter assembly process.

TABLE I. Calibration data for the three calorimeters

Cal No.	A		B		Regression		
	Value	Error	Value	Error	R	SD	N
1	-0.0001	0.0004	0.9997	0.0001	1	1.34118	6
2	0.0010	0.0005	0.9996	0.0003	1	1.35266	6
3	-0.0006	0.0006	1.0007	0.0004	1	0.84758	4

TABLE I displays calibration data for the three calorimeters showing the results of a linear fit of the function Y = A + B * X, where Y is the measured calorimeter sample thermal power and X is the applied electrical sample power. The linear regression parameters are included in the table and it should be noted that all three isothermal calorimeters have very similar performance. TABLE II and Figs. 5 and 6 display accuracy and precision data for isothermal calorimeter No. 2.

TABLE II. Calorimeter No. 2 measurement accuracy and precision data.

Applied Power (W)	Base Power (W)	Measured Power - Calorimeter No. 2			
		Accuracy (mW)	Precision (mW)	Accuracy (%)	Precision (%)
0.1	18.897	2.98	1.85	2.9772	1.8528
0.4	15.989	0.87	1.37	0.2181	0.3426
0.5	15.988	0.50	1.66	0.0998	0.3325
0.6	18.894	-0.11	0.90	-0.0180	0.1500
1.0	18.894	-0.17	1.55	-0.0168	0.1553
5.0	15.989	-1.87	0.90	-0.0374	0.0180
5.0	18.894	-4.44	2.70	-0.0888	0.0540
15.0	18.895	-0.07	8.14	-0.0005	0.0543

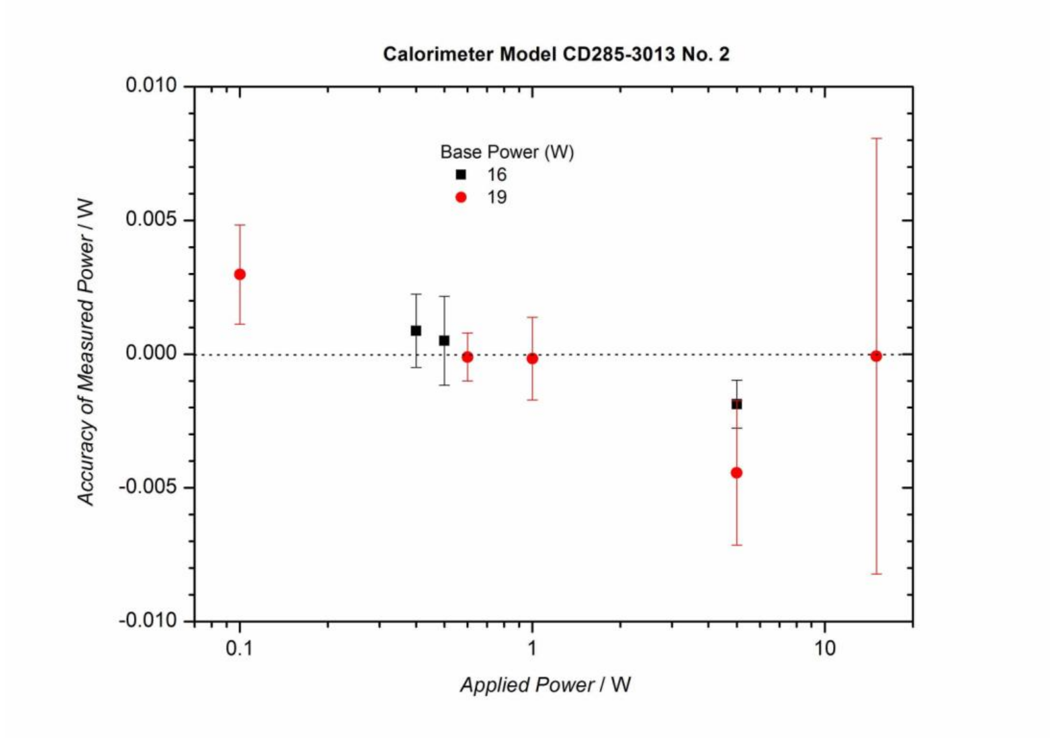

Fig. 5. The accuracy (deviation) of the measurement sample thermal power as a function of applied electrical power is plotted as absolute values.

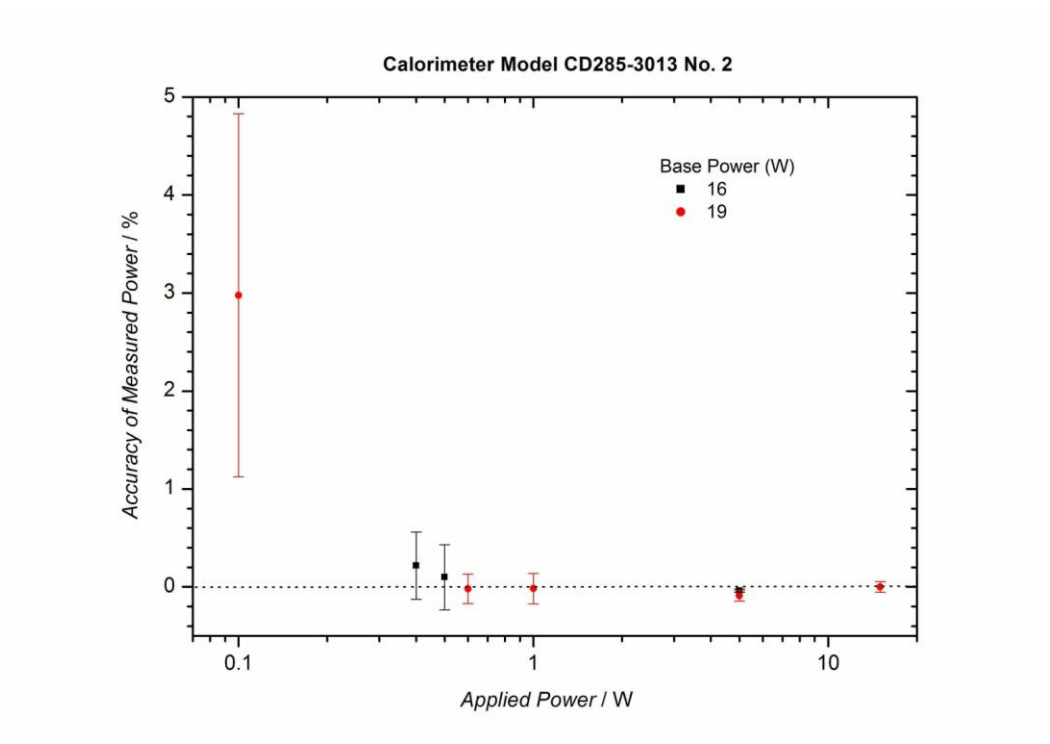

Fig. 6. The accuracy (deviation) of the measurement sample thermal power as a function of applied electrical power is plotted as percentage values.

In TABLE II, calorimeter measurement accuracy and precision data as a function of applied sample power are displayed in both absolute and percentage values. Repeated measurements have been performed at each applied power. Note that during the measurements the base power was adjusted from approximately 19 watts to 16 watts.

It is instructive to consider the accuracy of the calorimeter measurement by presenting the data both in terms of absolute values and as percentages and this is done in Figs. 5 and 6, respectively. The demonstrated accuracy of the data is high with a value of better than 0.2% in the power range from 0.6 to 15 watts and this has been observed across all three calorimeters. In both plots the error bars on the data are a measure of the measurement precision. The measured data points at 5 Watts have larger errors but still fall within the specification. Also, the expected trend of reduced accuracy and precision at lower sample powers is observed.

CALIBRATION AND SENSITIVITY MEASUREMENT – HEAT-FLOW OPERATION

Limited time for testing prevented more extensive investigation of heat-flow operation, and measurements were only possible for the single cell configuration. Calibration data for single cell operation of calorimeter No. 2 are presented in Fig. 7. It is expected that twin cell operation will result in improved performance in terms of better measurement precision due to the cancelling effect when, for example, a temperature disturbance effects both cells.

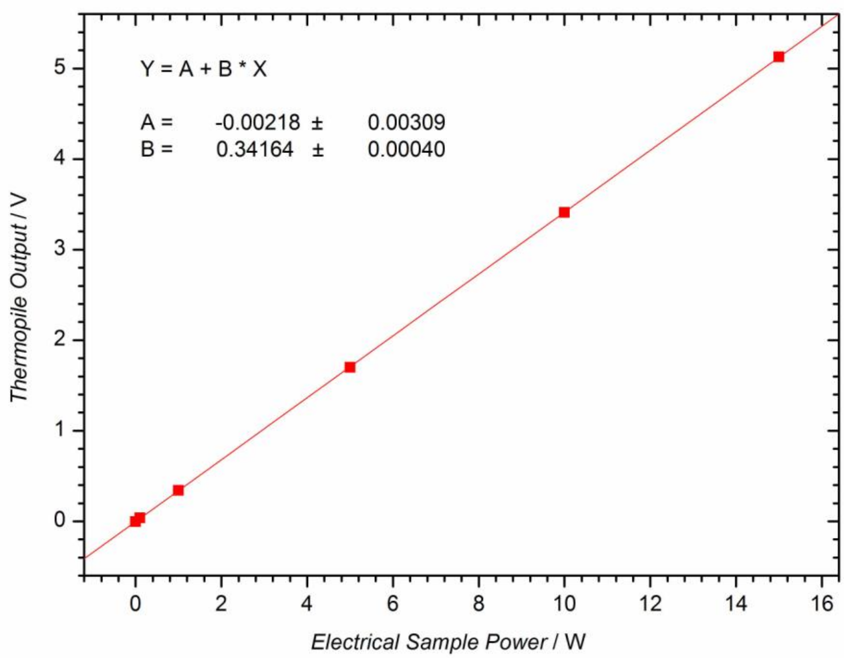

Fig. 7. Thermopile heat-flow sensor output voltage is plotted as a function of applied electrical sample power.

In Fig. 7, thermopile heat-flow sensor output voltage is plotted as a function of applied electrical sample power, for calorimeter No. 2. As expected, due to the linear response of the heat-flow sensors, the resulting graph is very linear, with only a small error in the slope of the line. The value of the slope of the linear regression line is the measured sensitivity of the calorimeter, with a very high value of 341.6 µV/mW. This value compares favourably with the value determined in the design calculation of 452.7 µV/mW. The difference corresponds to an under-estimation of calorimeter sensitivity of about 25%. This level of error is reasonable due to the simplicity of the model, which ignored both cylindrical geometric effects and the variations of the inter-cylinder gap due to the complicated internal geometry.

CONCLUSIONS

This paper describes the design and presents performance results for a new multimode calorimeter design that can operate as a single cell isothermal or both single and twin cell heat-flow calorimeter for measurements of plutonium or tritium or other heat producing radioactive substances. The design incorporates three new features including multimode operation, a novel implementation of doubled rectangular thermopile heat-flow sensors in a measurement chamber (cell) of cylindrical geometry and finally a high sensitivity in heat-flow mode of 341.6 µV/mW. The design is scalable and these features result in greater flexibility in operation, improved (reduced) measurement times, and a lower minimum detectable thermal power (MDP) compared to heat-flow calorimeters with rectangular measurement chambers. In heat-flow mode, the MDP is less than 500 µW.

For the present application of plutonium measurement, the calorimeter meets the performance requirements for accuracy of 0.16% above 1 watt. Below 1 watt the calorimeters achieve the required 0.2% down to 0.6 watts. The measurement time specification required that equilibrium be reached in 8 to 12 hours. Many measurements reached equilibrium in less than 6 hours and all in less than 10 hours, exceeding the specification.

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