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**PERFORMANCE EVALUATION OF A COMMERCIALY
AVAILABLE HEAT FLOW CALORIMETER AND
APPLICABILITY ASSESSMENT FOR SAFEGUARDING
SPECIAL NUCLEAR MATERIALS**

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SAFEGUARDING SPECIAL NUCLEAR MATERIALS**

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Performance Evaluation of a Commercially Available Heat Flow Calorimeter and Applicability Assessment for Safeguarding Special Nuclear Materials

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Abstract

The performance characteristics of a commercially available heat-flow calorimeter will be presented. The heat-flow sensors within the calorimeter are based on thermopile technology with a vendor-quoted sensitivity of $150 \mu\text{V/mW}$. The calorimeter is a full-twin design to compensate for ambient temperature fluctuations. The efficacy of temperature fluctuation compensations will also be detailed. Finally, an assessment of design applicability to special nuclear materials control and accountability and safeguarding will be presented.

INTRODUCTION

The use of commercially available systems and components for materials control and accounting (MC&A) has improved and reduced the costs of safeguarding special nuclear material (SNM) in the United States and abroad. The use and/or applicability of a system or component for a particular safeguarding task must be determined by a safeguards measurement expert. The design, performance, and safeguard applicability evaluation of a commercially produced heat-flow calorimeter will be presented.

The heat-flow calorimeter manufactured by International Thermal Instrument Company [1] uses heat flow sensors based on thermopile technology with a cost of approximately \$10,000 in 1997, this does not include any readout electronics or data acquisition software. A photograph of the calorimeter with the foam insulation removed from the top is presented in Fig. 1. The calorimeter is a full-twin design with a passive heat-transfer media. The system is portable and contains no water. Details of the system design can be found in the next section.

Fig. 1. Left: side view picture of temperature-compensated thermoelectric microcalorimeter fabricated by International Thermal Instrument Company. Right: top view picture of calorimeter with top insulation removed and measurement chambers empty.

CALORIMETER SPECIFICATIONS

The calorimeter body is made from a single block of aluminum. The aluminum block is surrounded on five sides by 1 in. of foam insulation which is covered by a thin stainless-steel shell. The last side of the aluminum block, the top, is insulated with a single piece of Styrofoam. The top surface of the Styrofoam is covered by a piece of G-10 fiberglass circuit-board material. Both the Styrofoam and the circuit-board material have two holes for access to the two measurement chambers. The exterior dimensions of the calorimeter are 14 in. long by 12 in. high by 8.5 in. deep. The total weight of the calorimeter is less than 100 lbs. Two identical holes are bored perpendicular to the top of the aluminum block for the heat flow sensors. The interior measurement chamber size is 2.75 in. in diameter by 2 in. deep.

The measurement chamber circumference and bottom are surrounded by thermoelectric heat-flow sensors. These sensors consist of thousands of copper/Constantan thermocouple junctions embedded in a polyimide/glass substrate. One set of junctions is in thermal contact with the aluminum block via the outer circumference of the heat flow sensor, and the other set is in contact with the inner circumference of the sensor. A voltage is generated by the thermocouple junctions that is proportional to the temperature difference between the inner and outer surface of the sensor. The manufacturer-specified sensitivity of the thermopiles is 150 $\mu\text{V}/\text{W}$.

Ambient room temperature fluctuations that change the aluminum block temperature are partially compensated by the use of a full-twin design. Compensation is achieved by taking the difference output of two matched sensors. Voltage output for the sensors is matched to one percent according to supplier specification. One sensor is used as a reference and the other to measure the sample heat output. The change in the A and B sensor outputs with heat flow into and out of the aluminum block is canceled by subtracting the reference sensor signal from the sample sensor signal.

The calorimeter is equipped with three voltage outputs, one each for the sample and reference thermopiles; the third output is the electrical difference between the A and B sensors.

DATA COLLECTION

Data collection was performed using a personal computer interfaced to a general purpose interface bus (GPIB). Signal measurements were made using digital multimeters (models Fluke 8840A, HP 34401A). Readings of the three calorimeter outputs (mV), room temperature (mV), and bath temperature (mV) were taken once every 2 s. Sixty readings were averaged together and this value was saved as a single data point for each of the signals giving a file with data points every 2 min. The meter resolutions for the calorimeter output measurements were 0.1 μV and 1.0 μV for the bath and room temperature readings.

All of the performance data, presented in the next section, was collected with the bottom 7 in. of the calorimeter submerged in a water bath controlled to $\pm 0.001^\circ\text{C}$. Although the full-twin design significantly reduces signal noise due to ambient room temperature fluctuations, placing the calorimeter in a controlled environment further reduced this noise component.

CALORIMETER PERFORMANCE

The sensitivity of side A and side B heat-flow sensors was determined using ^{238}Pu heat standards and electrical resistance heaters. The heat standards ranged from 0.7 mW up to 430 mW and heater measurements ranged from 1.0 mW to 250 mW. The majority of the heater measurements were made using a 99.3-k Ω resistance heater, although several measurements were made in side B using a 550- Ω resistance heater.

The sensitivity based on the A minus B output of the calorimeter was 0.1307 mV/mW for power input to side A and -0.1295 mV/mW for power input to side B. The sensitivity of side B is negative because with heat in side B the A minus B output becomes more negative with increasing heat. The sensitivity was determined by making a linear least-squares fit to the heat standards and heater data. The data and the fits are presented in Fig. 2. No biases were observed between heater measurements and heat standard measurements within the noise level of the system. The nonlinearity of the system was determined to be zero within the scatter of the data points. This was based on the coefficients of the second order fits to the data being at least four orders of magnitude smaller than the first order coefficients. The zero offsets of the sensitivity curves were essentially zero with values of 18 μV and 13 μV for side A and B, respectively.

The noise of the system can be quantified using baseline data which is collected when no heat is in either chamber of the calorimeter. The average of 25 baseline measurements was $-4.8 \mu\text{V}$ with a one sigma standard deviation of $9.1 \mu\text{V}$. The range of the measurements were from $-24.4 \mu\text{V}$ to $8.4 \mu\text{V}$. The baseline values collected over a two-month period are presented in Fig. 3. No drift on the baseline data is seen over the two-month period indicating good long-term stability of the heat-flow sensors. The scatter in the data is due to heat flowing in and out of the aluminum block due to changes in ambient room temperature. These fluctuations were partially damped out by submerging 7 in. of the calorimeter into a large temperature-controlled water bath. Using the standard deviation of the baseline, a zero point measurement uncertainty of $70 \mu\text{W}$ (28 mg of plutonium ($6\% \text{ }^{240}\text{Pu}$) equivalent) exists for this calorimeter. Three replicate measurements of the same 0.725-mW heat standard in side B gave a one sigma standard deviation of $64 \mu\text{W}$ and a negative bias of $221 \mu\text{W}$. These replicate heat measurements were made over a one-week period, interspersed between other heat standard measurements.

Fig. 2. Top panel: Sensitivity curve of calorimeter with power source in side A. Diamonds are ^{238}Pu heat standards data and squares are electrical resistance heater data. The solid line is a least-squares linear fit to the data points. Bottom panel: Sensitivity curve of calorimeter with power source in side B. Diamonds and squares are the same as top panel. Triangles are electrical resistance heater data collected with the calorimeter out of the temperature-controlled water bath. The solid line is a least-squares linear fit to the diamonds and squares only.

Fig. 3. A minus B calorimeter output in μV with no heat in either measurement chamber.

FULL-TWIN COMPENSATION

The calorimeter was run in room conditions where the temperature fluctuations had a range of 1.5°C . Over 50 h of data were collected after the calorimeter came to equilibrium with no heat in either A or B measurement chambers. The room temperature, A signal, B signal, and A minus B signal fluctuations are presented in Fig. 4, from top to bottom respectively. Figure 4 shows the A, B, and A minus B signals are roughly the inverse of the temperature fluctuations with the temperature fluctuation happening slightly earlier in time. The reason for the inversion of A and B relative to the room temperature is because when the room temperature increases, heat flows into the aluminum block through the outside of the heat flow sensors creating a more negative signal from the sensors. The structure found in the A and B individual signals is propagated to the A minus B signal output but reduced by a factor of more than 20. This graphically demonstrates the cancellation of noise due to ambient room temperature fluctuations using a full-twin design. Using the sensitivity determined for sensor B, the fluctuations with no compensation are equivalent to 31 mW or 12 g of plutonium (6% ^{240}Pu), but with compensation, using A minus B, the fluctuations are equivalent to 1.5 mW or 0.6 g of plutonium (6% ^{240}Pu).

Fig. 4. Room temperature, A and B, and A minus B signals are plotted as a function of time from top to bottom, respectively. The calorimeter was already at equilibrium at time zero.

SIGNAL CORRELATIONS

Examining the individual outputs from A and B revealed thermal cross talk between the heat-flow sensors. The voltage output of A and B as the calorimeter comes to equilibrium with 250 mW of power delivered to side A of the calorimeter is presented in the top panel of Fig. 5. The increase in voltage output from the side A sensor is what

would be expected with the addition of heat to the side A measurement chamber. There is also a negative deflection of the voltage output from side B which was not expected. This negative deflection of the B-side sensor voltage is displayed on an expanded scale in the bottom panel of Fig. 5. The curve has an exponential shape typical of all heat transfer processes. The magnitude of the voltage change is equivalent to -7.8 mW. The explanation for this negative signal is a fraction of the heat from side A is transversing the aluminum between the two sensors and creating a heat flow into the measurement chamber which gives a negative signal. This cross talk between measurement cells complicates the A minus B output, making it more difficult to predict equilibrium and creates a position sensitivity within the measurement chamber, both of which are detrimental effects for high-precision calorimetry measurements.

Fig. 5. Top panel: Calorimeter voltage output of A, positive curve, and B, negative curve, as a function of time with 250 mW of heater power delivered to side A. Bottom panel: B sensor output from top panel on expanded scale.

CONCLUSIONS

The full-twin calorimeter fabricated by International Thermal Instrument Company using thermopile heat-flow sensors is capable of measuring a 250-mW sample (100 g of plutonium (6% ^{240}Pu) equivalent) with less than 0.5% measurement bias. The full-twin compensation technique can decrease the effects of ambient temperature fluctuations, which is the major source of noise for this system, by a factor of greater than 20.

The calorimeter measurement capabilities nearly met manufacturer specification and performed well under laboratory measurement conditions. The ruggedness of the heat-

flow sensors and simplicity of design would be beneficial properties in a facility environment. The experimentally determined sensitivities in mV/mW for the A and B sensors were 0.1307 and -0.1295, respectively, slightly less than the specified 0.150 mV/mW. From these sensitivity values the mismatch of the heat-flow sensors is at the 1% level, in agreement with specifications. Improvements in the measurement precision and accuracy could be made by precisely controlling the aluminum block containing the heat-flow sensors and thermally isolating the individual sensors from one another.

The manufacturer offers internal heating and cooling of the aluminum block as an option on their heat-flow calorimeters. Measurement chamber sizes from 16 mL up to 50 L are available.

REFERENCES

1. International Thermal Instrument Company, P.O. Box 309, Del Mar, CA 92014.