

## **DESIGN AND OPERATION OF A LARGE VOLUME TWIN CELL HEAT-FLOW CALORIMETER FOR THE MEASUREMENT OF BOTH TRITIUM AND PLUTONIUM SAMPLES**

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### **ABSTRACT**

This paper describes the design and operation of a large volume twin cell heat-flow calorimeter intended for the measurement of samples containing either tritium or plutonium. In contrast to isothermal calorimeters, which are optimized for shorter measurement times with adequate measurement precision, this large volume twin cell heat-flow calorimeter is based on updating the design of the traditional twin cell heat-flow calorimeter, which is characterized by long measurement time constants. The advantage of the twin cell heat-flow method is that better measurement precision can be achieved for large volume measurement cells. Through the use of high output voltage thermopile differential temperature sensors and enhanced thermal insulation, a significant sensitivity improvement has been achieved for a large volume measurement cell (typically 50 litres in volume). The transportable calorimeter incorporates a removable electrical calibration heater and a method of controlling the calorimeter peripheral temperature environment without using a liquid coolant. Eliminating the liquid coolant reduces any potential criticality hazard and the possibility of tritium contamination of the coolant. The thermal performance of the calorimeter is considered and measured data for different sample powers is presented. Calorimeter measurement sensitivity is determined using electrical calibration data and results for measurement precision and accuracy are presented.

### **INTRODUCTION**

This paper reports on the development of an improved sensitivity twin cell heat-flow calorimeter for large samples. The instrument is intended for the measurement of plutonium or tritium bearing samples or the measurement of other radio-nuclides with an adequate heat-output. The design is described and preliminary performance results are presented and discussed in this paper.

In a sense the present work represents a return to the past. Over the last 25 years, much effort has been expended to improve the performance of isothermal calorimeters, which while delivering adequate measurement precision, are focused on reducing measurement times. In isothermal calorimeters the measurement time is not effected by the thermal time constants of the calorimeter measurement chamber as this is maintained at constant temperature.

In marked contrast, with heat-flow calorimeters the thermal time constants of the calorimeter measurement chamber can be the dominant factor controlling the measurement time. The advantage of the twin cell heat-flow method, and the reason

that it was selected in the present case is that better measurement precision can be achieved for large volume measurement cells. The improvement in precision has been bought at the cost of significantly longer measurement times. The advantage of thermally close coupling cylindrical samples to the walls of a cylindrical measurement chamber has also been removed.

The model 400HF-5300 twin cell calorimeter is shown in Figure 1. In the photograph the twin cell thermal element body, the plug unit extraction carriage, the instrument enclosure and a panel-mounted computer can be seen.



Figure 1. The model 400HF-5300 twin cell heat-flow calorimeter

## **MECHANICAL AND ELECTRICAL AND HEAT-TRANSFER DESIGN**

The calorimeter consists of a twin chamber (cell) heat-flow design thermal element and instrumentation enclosure with computer workstation mounted on a single chassis fitted with lockable wheels.

The thermal element contains two identical measurement chambers, also referred to as cells, into which canisters containing the samples to be measured are placed. System control and measurement electronics, including the control and analysis computers and amplifiers, are located in the instrumentation enclosure. The instrument enclosure is connected to the thermal element by shielded signal and control cables.

The calorimeter is designed to provide a stable exterior environment around each measurement cell so that the sample being measured and the reference cell are isolated from the effects of fluctuations in the ambient temperature of the laboratory.

Particular attention has been paid to the performance, including design and selection of the hardware components for the thermal element and measurement instrumentation in order to meet stability, sensitivity and limit of detection requirements. The instrument has been designed for a limit of detection of 0.0005 Watts and with an operating range from 0.001 to greater than 10 Watts.

The calorimeter uses thermopiles embedded within an annular gap between the inner and outer walls of each of the measurement and reference cells to determine the thermal energy generated in the sample and the consequent heat-flow across the measurement chamber boundary. The number of thermopile sensors has been chosen in order to optimize calorimeter performance. Figure 2 is a plot of the calculated maximum calorimeter sensitivity as a function of the number of thermopiles, which

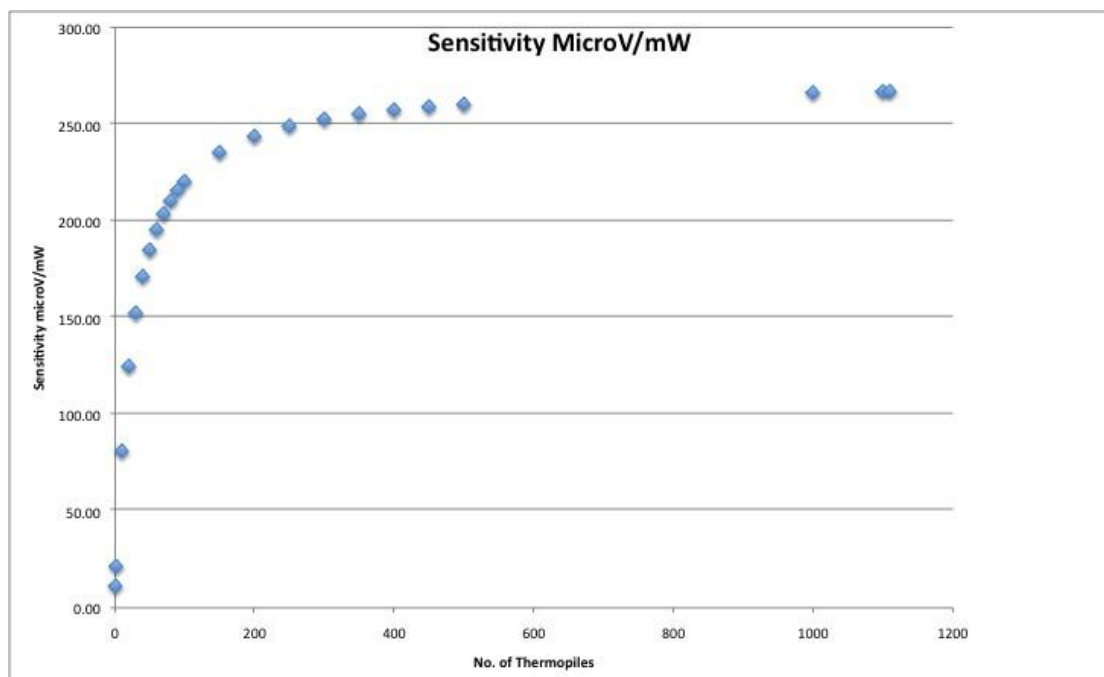


Figure 2. Calculated calorimeter sensitivity as a function of the number of thermopiles.

are deployed on all 6 internal surfaces of each cell. For the present design a theoretical maximum sensitivity of 0.225 V/W was used as the design basis. Initial measurements have confirmed an actual sensitivity value of 0.194 V/W. This confirms that the calorimeter has achieved one of its design objectives, which is to have greater sensitive than other equivalent large sample commercial calorimeters of the heat-flow type.

The measurement and reference cells can be seen prior to final assembly in Figure 3. The internal dimensions of each cell are 330 mm by 330 mm by 500 mm (length-width-height). The calorimeter incorporates a semiautomatic mechanism for removing and replacing the heavy measurement and reference chamber plug units. It is called the calorimeter plug unit extraction carriage. It is operated manually using a handle, which has some mechanical advantage. It is used to gain access to both cells of the calorimeter and ensures that the plug units are replaced in exactly the correct position. Once the plug unit has been removed, the operator opens the internal

calorimeter chamber lid manually. Samples may be loaded by hand or using an appropriate engine hoist or small crane.

The two cells are designed to have identical thermal characteristics. By making a differential measurement between the two cells both electrical noise and temperature variations that affect both cells are cancelled, improving both accuracy and measurement precision. The twin cells are imbedded in multiple layers of high performance ultra-low thermal conductivity insulation panels. These insulation panels are contained in an external aluminium box enclosure, which is mounted on the robust steel chassis fitted with lockable wheels.



Figure 3. Measurement and reference cells during assembly of the calorimeter

A design requirement of the calorimeter was that circulating liquid coolant (such as water) not be used as a means of providing temperature control for the external aluminium box enclosure. In order to meet this objective low voltage surface heating pads have been deployed on the external surface of the aluminium box enclosure. A computer based temperature controller regulates the temperature of the external heater pads in order to maintain a constant external surface temperature on the box enclosure.

## **DATA ACQUISITION AND ANALYSIS**

A Keithley Instruments model 2701 digital voltmeter is used to measure the measurement and reference cell output voltages. The unit is fitted with a model 7702 multiplexer and also measures the output of a variety of additional temperature sensors distributed over the calorimeter. The output of these additional sensors is used for diagnostic purposes and to determine the state of health of the calorimeter. A Keithley Instruments model 2601A SourceMeter is used to power the electric calibration and confirmation sample.

The software operates under Microsoft Windows XP™ and the calorimeter may be monitored and controlled over a network. The software includes both data acquisition and data analysis functions. Sample power, end point predictions and equilibrium fitting routines are included. Measurement data is archived and may be analysed off-line.

The calorimeter employs sample thermal equilibrium determination as well as end point prediction. Thermal equilibrium determination provides an indication to the operator when the sample has reached thermal equilibrium and the measurement is finished. End point prediction provides a means of shortening the measurement time by predicting the thermal equilibrium or end point sample power.

## **OPERATION AND MEASUREMENT RESULTS**

Figure 4 provides a view of the measurement cell with electric calibration and confirmation sample inserted. Samples are loaded into the measurement cell after the calorimeter plug has been removed using the plug unit extraction carriage.

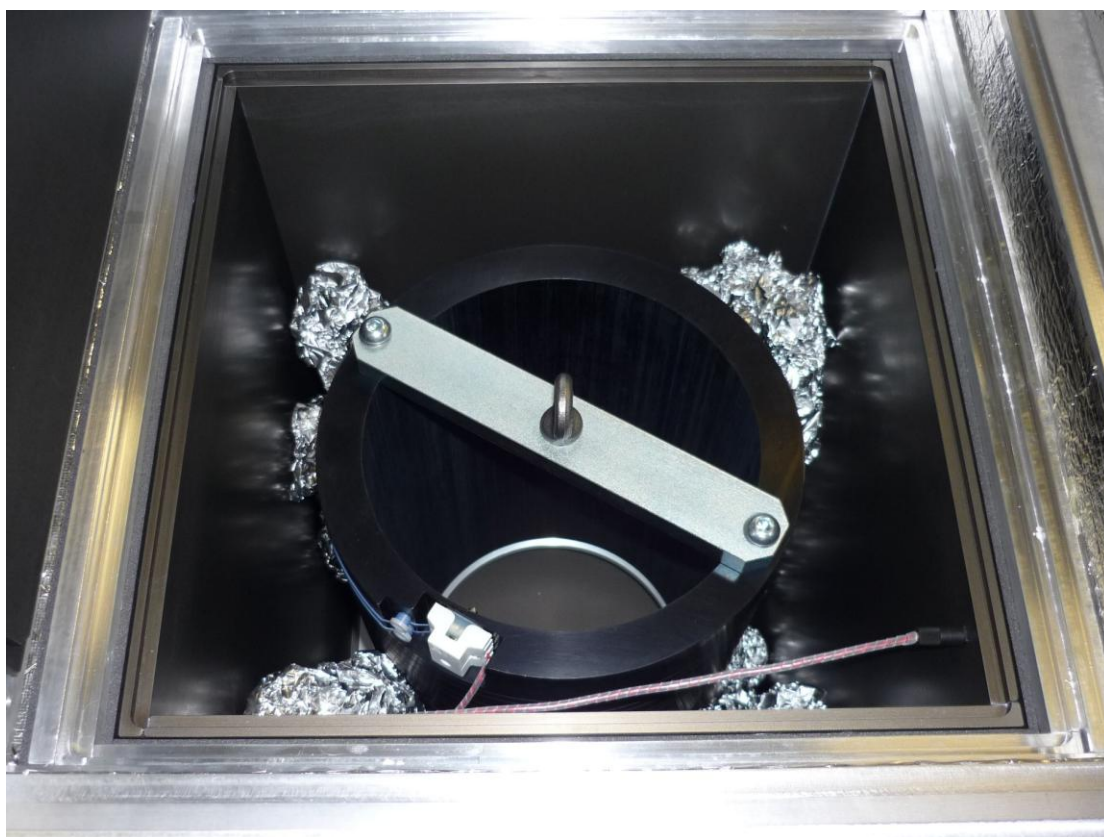


Figure 4. This is a view of the measurement cell with electric calibration and confirmation sample inserted. Note the use of aluminum foil as a heat-transfer medium to increase heat conduction from the sample to the measurement cell walls.

Once the cell lid and plug have been replaced the measurement can begin. The calorimeter corrects for any temporary offset in the power data due to installing the sample. As the measurement proceeds data is displayed in graphical and tabular form on the panel PC, which controls the analysis of measurement data. The equilibrium determination and prediction algorithms begin to operate once sufficient data is available.

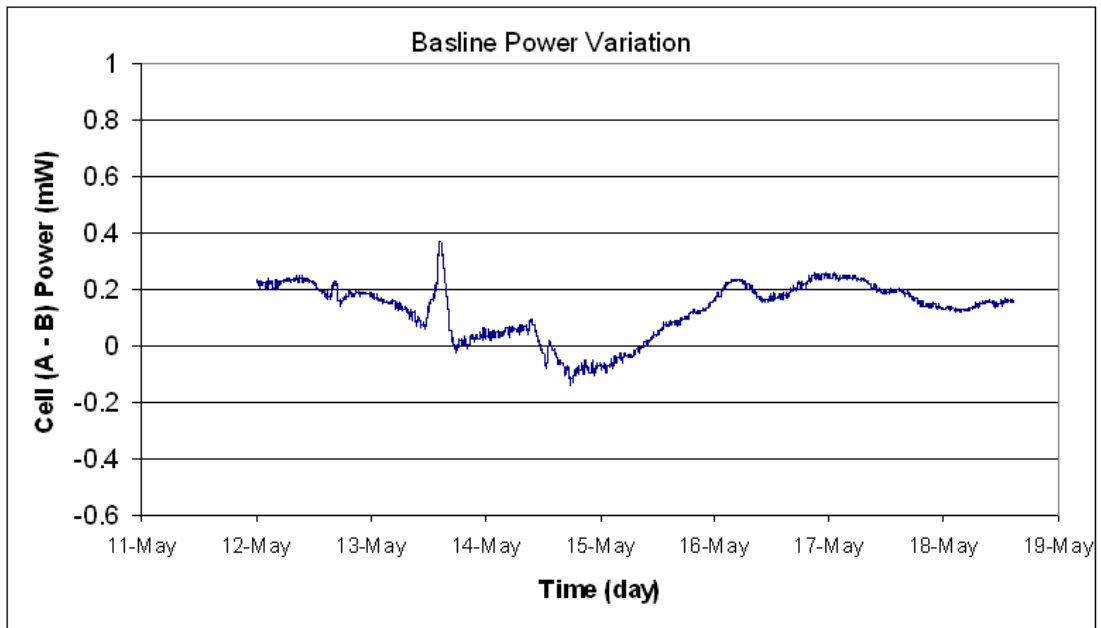


Figure 5. This graph shows the drift of the calorimeter differential power over a period of several days. Aside from an externally induced excursion the variation is within an envelope of 400 microwatts.

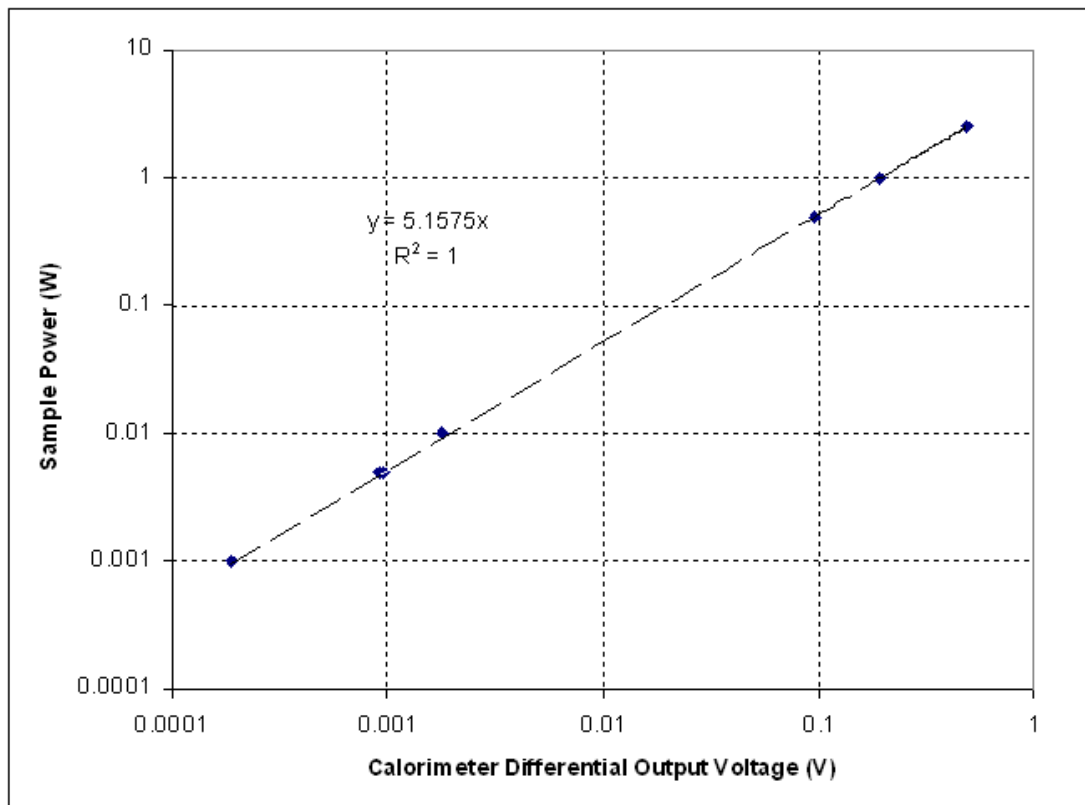


Figure 6. This graph is a log-log plot of the calorimeter calibration ranging from 0.001 to 2.5 watts. From this graph the calorimeter sensitivity is determined to be 0.194 V/W.

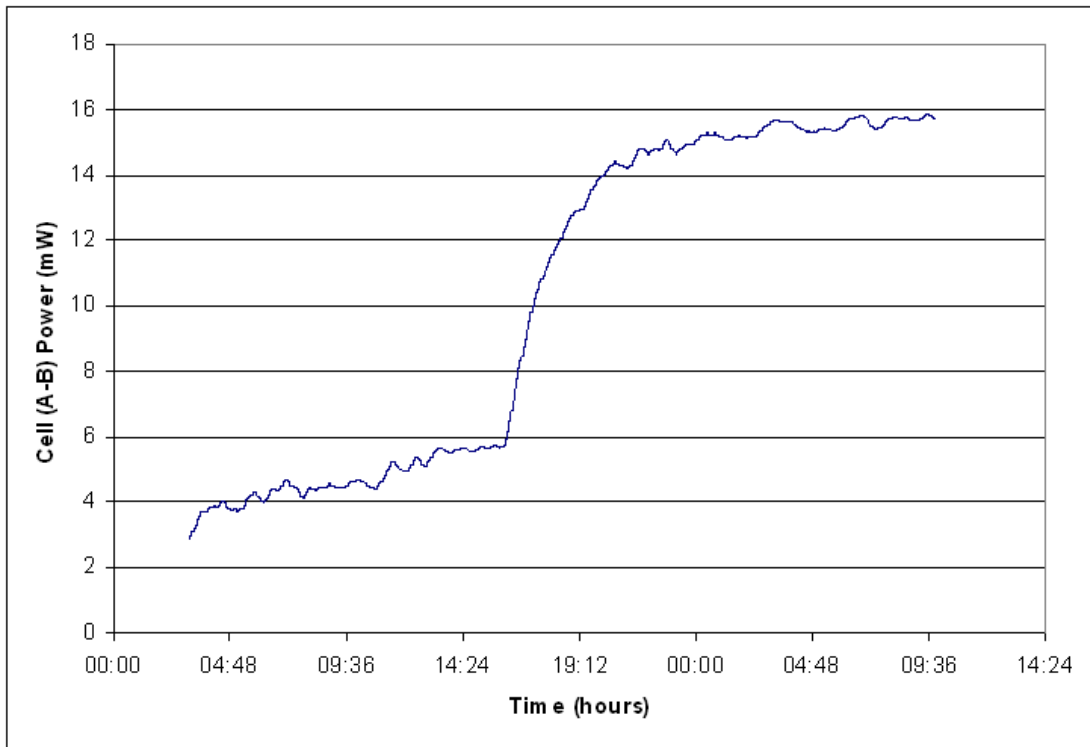


Figure 7. This graph shows the differential power variation (measurement cell – reference cell) for the measurement of a 10 milliwatt electric sample.

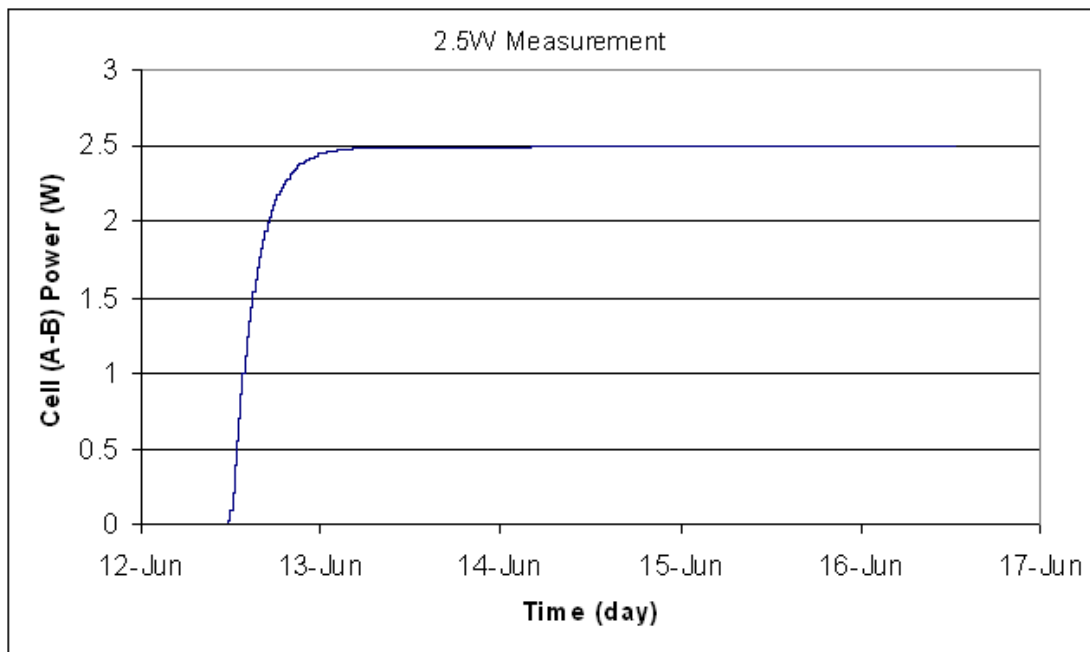


Figure 8. This graph shows the differential power variation (measurement cell – reference cell) for the measurement of a 2.5 watt electric sample.



Data is presented in figure 5, which gives a preliminary indication of the baseline drift of the differential measurement chamber power. Aside from an externally induced excursion the variation is within an envelope of 400 microwatts over a period of approximately one week.

Figure 6 provides a plot of measured data used to establish an initial calibration. The data is linear over the range 0.001 to 2.5 watts, although it is plotted on a log-log plot for convenience. This data also provides a measure of the sensitivity of the calorimeter of 0.194 V/W.

The results of two electrical sample measurements are presented in figures 7 and 8 for electrical powers of 10 mW and 2.5 W. It can be seen that the times to equilibrium are quite long compared to equivalent measurements performed with an isothermal calorimeter. Heat-flow calorimeter measurement precision is better, however than that which can be achieved using the isothermal calorimeter method. Typically measurement precision is 3% at 5 mW and better than 0.01% above 500 mW. Accuracy for measurement of power greater than 1 watt is typically 0.1%. Further testing of the instrument is currently underway.

## **CONCLUSIONS**

The ANTECH heat-flow calorimeter model 400HF-5300 has exceeded its design requirements and achieved a high level of sensitivity, close to the design target. It has excellent performance for low powered samples and achieves expected high precision for higher-powered samples. The low power performance will be useful in measuring containers that are considered to be empty and confirming that they contain very little radioactive material.