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CHARACTERISTICS AND PERFORMANCE OF A LARGE VOLUME TWIN CELL HEAT-FLOW CALORIMETER FOR PLUTONIUM AND TRITIUM MEASUREMENT

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ABSTRACT

In this paper the characteristics and performance of a large volume twin cell heat-flow calorimeter is described and reported. The transportable calorimeter incorporates a removable electrical calibration heater and an internal calibrated power supply for both calibration and performance checks. Provision is made for calibration using a calibrated external precision power supply or radioactive heat standards. The calorimeter is able to achieve a high degree of sensitivity and thermal stability through the use of high output voltage thermopile differential temperature sensors and enhanced thermal insulation. Using twin cell heat-flow measurement technology, a significant sensitivity improvement has been achieved for large volume measurement cells, each with a volume in excess of 50 litres. The present instrument achieves improved performance with an external heat sink made of a laminate of dry solid insulation combined with high thermal conductivity panels, which represents an improvement in technology over water bath calorimeters. The elimination of water as a heat sink medium reduces any potential criticality hazard and the possibility of tritium contamination. Measured precision and accuracy for different sample powers will be presented and minimum levels of detection are determined using the electrical calibration sample and zero power measurements. The stability of the calorimeter and its response to variations of ambient temperature has been studied. Measurements of both electrical samples and radioactive samples with varying thermal powers will be considered. Results will be compared for measurements at different thermal powers using both equilibrium end point power prediction and end point equilibrium power determination. The paper includes results of measurements of the spatial variation of sensitivity within a measurement chamber cell. Sensitive large sample volume calorimeters of this type have application to the measurement of a wide range of radioactive materials in variety of containers.

INTRODUCTION

In this paper the calibration and performance of a large twin cell heat-flow calorimeter is presented. The twin cell heat-flow device has two identical 53-litre measurement chambers and is intended for use in measuring a wide range of heat producing samples including plutonium and tritium or other radio-nuclides with an adequate heat-output. Data is presented from measurements of both electric test samples and tritium samples. The initial development of the ANTECH Model 400HF-

5300 calorimeter was described in an earlier paper [1]. In contrast with isothermal calorimeters, where measurement time is optimized to achieve adequate measurement precision [2, 3], heat-flow calorimeters achieve better measurement precision but at a cost in measurement time. In this paper work is described which is directed towards improving the effectiveness of end point sample power prediction algorithms and towards improving measurement times by providing effective thermal coupling between the calorimeter sample and the measurement chamber.

The twin cell heat-flow calorimeter is shown in Figure 1. In the photograph the twin cells of the thermal element body make up most of the volume of the instrument. The instrument electronic enclosure, with a panel-mounted computer, is located at the right hand end of the photograph.

Figure 1. The model 400HF-5300 twin cell heat-flow calorimeter consists of two 53-litre measurement chambers.

HEAT-FLOW CALORIMETER DESIGN

The calorimeter has been designed with a measurement power operating range from 0.001 to greater than 10 Watts and with a limit of detection of better than 0.0005 Watts. Measurements on the prototype instrument [1] suggested a short-term (48 to 72 hours) measurement chamber power variation in the absence of external disturbances of less than 150 µWatts (0.000150 Watts). All of the instruments of this type have similar calibration factors of typically 0.193µVolts/mWatt.

The two large measurement chambers or cells have dimensions of 330 mm by 330 mm by 500 mm (L x W x H). Although they are identical one is designated the Measurement Cell and the other the Reference Cell. Each of the two cells is surrounded on all sides by thermopile heat-flow sensors, which effectively measure the temperature difference (magnitude of heat-flow), which arises as a result of the

presence of an heat producing sample in the measurement chamber. When thermal equilibrium is reached by the sample, the constant temperature difference or heat-flow rate across the walls of the measurement cell is used with the calibration constant to determine the heat generation rate or thermal power of the sample. The Measurement and Reference cells are measured in series so that thermal or electrical disturbances that affect both cells are effectively cancelled, increasing the sensitivity of the measurement process. The cells are surrounded by a region of significant thermal insulation to reduce the effect of changes in the ambient temperature. This region also has a large heat capacity so that it maintains a reasonably constant temperature during measurements.

An important aspect of the design is that the measurement chamber geometry remains constant. When samples are loaded and unloaded it is essential that the chamber lid or plug unit is precisely re-positioned. This is achieved by the Plug Unit Extraction Carriage, which is shown in Figure 2.

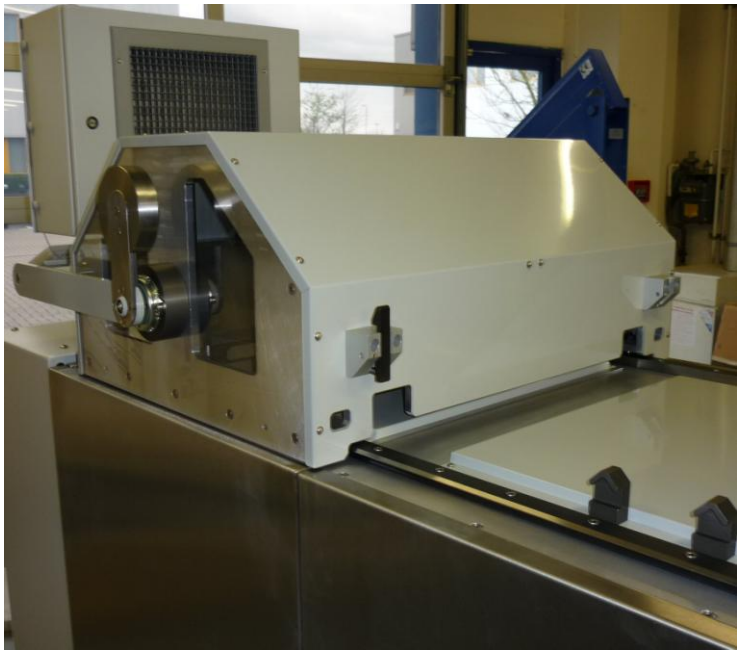

Figure 2. Calorimeter Plug Unit Extraction Carriage which is designed to precisely remove and replace the plug units of either the Measurement or Reference Cells.

ELECTRICAL CALIBRATION

Calibration can be performed using either calibrated heat standards or calibrated electrical standards. While radioactive heat standards have traditionally been preferred, they are more difficult to manage and calibrate and they also have associated security issues. In marked contrast, electrical standards are easier to manage, are traceable to national standards and do not have the security requirements associated with the handling and management of nuclear material**.**

In order to facilitate electrical calibration, a precise digitally controlled power supply, an high precision digital voltmeter (DVM) and a precision calibrated electrical resistance are built into the calorimeter internal calibration system**.** The system is

capable of delivering precisely controlled rates of electrical power, which is converted by an electrical calibration sample into equivalent rates of thermal power. In this manner, an heat or thermal energy generating sample, for example a tritium sample, is precisely simulated. The system is very robust and requires only annual calibration of the DVM and precision resistance, which are traceable to national standards**.**

Set Power (mW)	Measured Voltage (mV)	Measured Power (FAT Cal)	Accuracy (mW)	Accuracy (%)
0.5	0.092879257	0.481263158		
0.5	0.101006192	0.523373684	0.044804338	8.96%
0.5	0.085719814	0.444165789		
5	0.947755418	4.910889474		
5	0.951818885	4.931944737		
50	9.566563467	49.57010526		
50	9.525154799	49.35554211		
500	95.96942724	497.2751842		
500	96.04682663	497.6762368		
500	95.71594427	495.9617368	4.388675228	0.88%
500	95.70394737	495.8995737		
5000	967.2987616	5012.155263		
5000	964.2027864	4996.113158	10.25659203	0.21%
5000	963.622291	4993.105263		

Table 1. Calibration data obtained during Factory Acceptance Testing using an electrical heat standard.

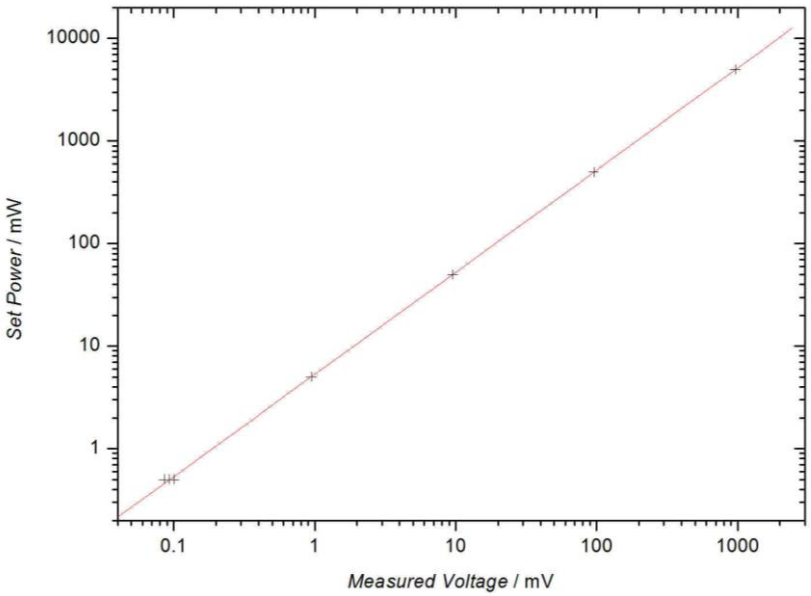

Figure 3. Electrical calibration curve is linear over 4 decades of sample power.

An electrical heat standard calibration was performed during the Factory Acceptance Test of the calorimeter and the data is presented in Table 1. Electrical calibration data is linear over 4 decades of power and this can be seen in Figure 3, where the data is plotted in log – log format.

MEASUREMENT CHARACTERISTICS AND PERFORMANCE

The two plots in Figures 4 and 5 illustrate the range of measurement power. They show electrical sample measurements at powers of 0.5 mW (500 µW) and 500 mW respectively. A variety of measurements have been made both with electrical and tritium samples. Performance is assessed on the basis of electrical measurements performed at Factory and Site Acceptance Tests, as the sample power is easier to establish accurately from electrical sample measurements. Electrical sample measurements covering three decades of power are tabulated in Table 2.

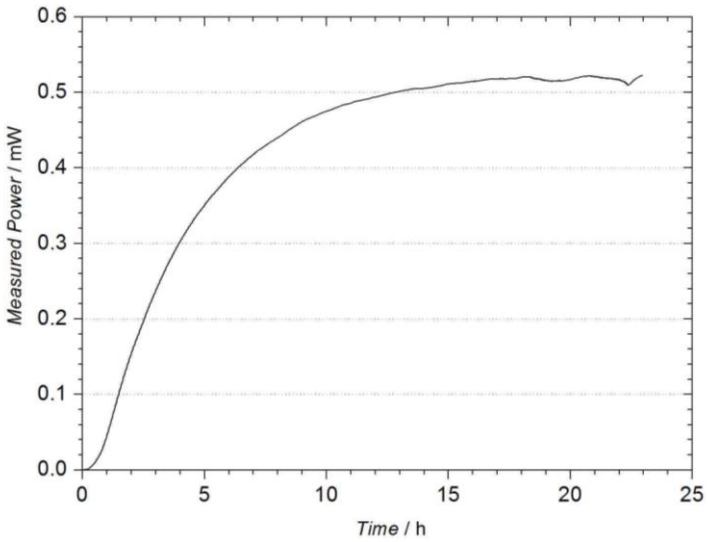

Figure 4. Measurement of an electric sample with a set power of 500 µW

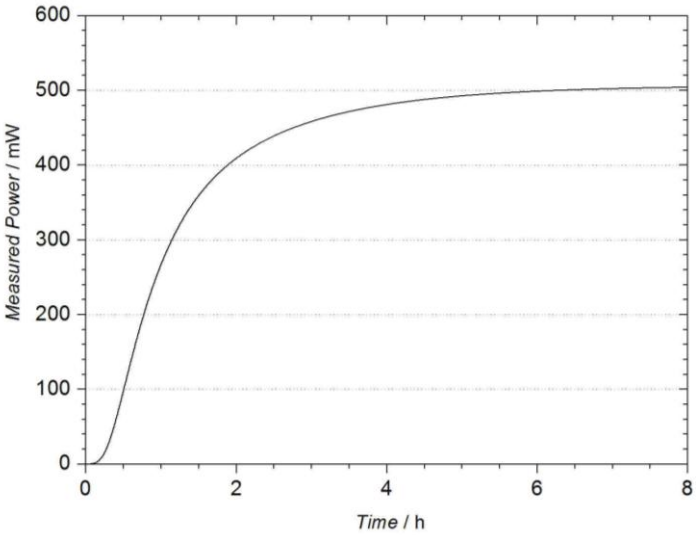

Figure 5. Measurement of an electric sample with a set power of 500 mW.

Set Power (mW)	Measured Voltage (mV)	Measured Power (FAT Cal)	Deviation (%)	Standard Error 2 Sigma (%)
5	0.93742	4.857335472		
5	0.93518	4.845728688	-2.895	0.2573
5	0.93948	4.868009568		
50	9.5236	49.34748576		
50	9.56588	49.56656381	-1.005	0.2995
50	9.56795	49.57728972		
500	96.3251	499.1181382		
500	96.6253	500.6736545	-0.0638	0.0019
500	96.3506	499.250269		
5000	964.2007	4996.102347		
5000	966.7347	5009.232522	0.0804	0.0016
5000	966.2533	5006.738099		

Table 2. Repeated measurements with an electrical sample to determine calorimeter performance.

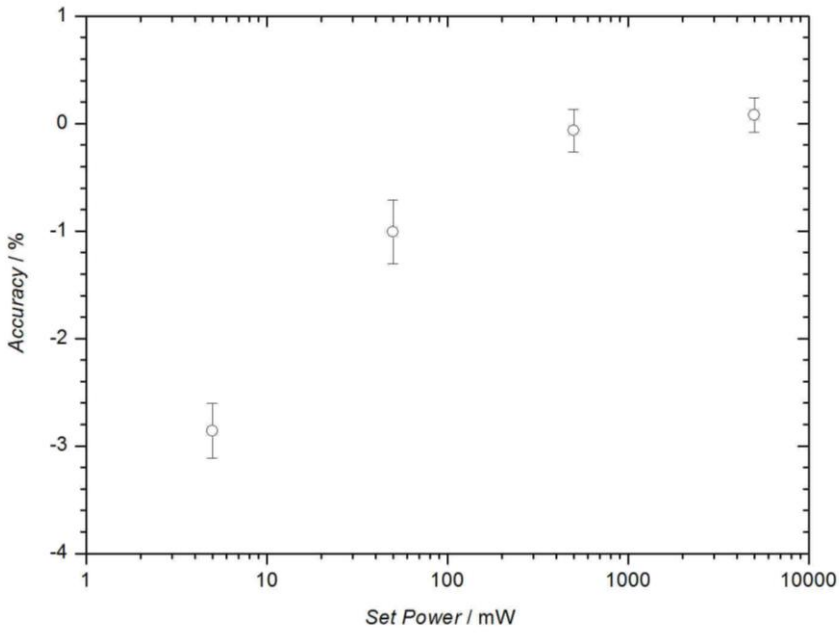

Figure 6. Deviation (accuracy) is presented as a percentage of measured power. Error bars are the standard error (2 standard deviations).

It can be seen from the data that the deviations are relatively small and that the measurements have very good precision over the power range. The data was obtained once the calorimeter had been installed in a facility where ambient temperature was controlled. The performance data has less deviation and greater precision largely due to a better-controlled environment during the Site Acceptance Testing.

The prediction algorithm employs a single exponential fit to the data based on rolling averages. A preliminary algorithm using a three-point fit determines when the full single exponential fitting process can be started. Parameters are set to determine acceptance criteria at different stages of the entire fitting process. If the criteria are set too "tight" then the fit is required to achieve smaller errors and this process will take a longer period of time but produce better results. If the criteria are set too "loose" then the fit will tolerate larger errors and this process will declare a result, with poorer precision and accuracy in a shorter period of time.

Run	Actual power (W)	Original fit parameters		Revised fit parameters	
		Prediction power (W)	Time to prediction or time run ended (min)	Prediction power (W)	Time to prediction or time run ended (min)
Sample 1	0.230941	0.230945	1125	0.231096	385
Sample 2	0.230678	0.230873	927	0.230993	387
Sample 3	0.063769	0.0642	1808	0.05975	346
Sample 4	0.00388	0.003947	1470	0.004846	260
		Average	1332.5		344.5

Table 3. Improved prediction results for several tritium samples.

Table 3 shows the improvement that can be achieved by careful selection and adjustment of the fitting criteria for different sample powers. Note that for a slight degradation in the measurement result, a substantial reduction of approximately a factor of four is achieved in the time required to obtain a prediction result. As expected the errors that result are larger for smaller sample powers.

As part of the process of assessing the characteristics and performance of the calorimeter, measurements were made with a small electric sample in order to assess the spatial variation of the calorimeter response across the measurement chamber. The results are displayed in Table 4 and the measurement positions are described below.

Position		Power (mW)
1	Corner	9.841
2	Edge	9.76
3	Centre Bottom	9.796
4	Centre Chamber	9.86

Average (mW)	9.81425
SD (mW)	0.045036097
SD (%)	0.46%

Table 4. Measurements of spatial variation of sample power.

In positions 1, 2 and 3 separate measurements were made using a small cylindrical electric sample in an aluminum can (approximately 8 cm in diameter and 10 cm high). The sample was placed in direct physical contact with the bottom of the measurement chamber in three positions: in the corner, half way along one side and in the centre. A fourth measurement was made with the same sample in the centre of the volume of the cell, not in direct contact with the walls and supported by aluminum foil "balls". It can be seen from the data contained in Table 4 that the spatial variation of the response across the calorimeter measurement chamber s less than 0.5%.

A final aspect of the investigation into calorimeter performance involved studying the effects of thermal coupling on equilibration time. An important factor in determining the time for a sample to reach equilibrium is the degree to which it is thermally coupled to the calorimeter measurement chamber internal surfaces. Coupling materials with low heat capacity, and high thermal conductivity (good thermal diffusivity) are most effective. Efficient contact with both the surfaces of the sample and calorimeter internal surfaces is also important. Aluminum foil "balls" have been found to be a relatively poor coupling material due to inefficient surface contact, despite satisfying the other criteria.

Figure 7 shows a view of the calorimeter measurement chamber (cell) with a canister, which contains an electric calibration sample. The sample canister is coupled to the walls of the measurement chamber by a thermal coupling mechanism (thermal insert). Measurements have been performed with the electric sample in this configuration (small sample), with the sample and canister directly coupled to the cell floor and on an insulating pedestal (large sample). Note that power is delivered to the electric calibration sample by means of an electrical connector fitted to the inside of the cell. Care has been taken in the design of the instrument to bring the electric sample power wiring through the cell wall in such a way that heat leaks from or to the cell are not introduced and at the same time, the electrical power liberated into the cell is correctly measured.

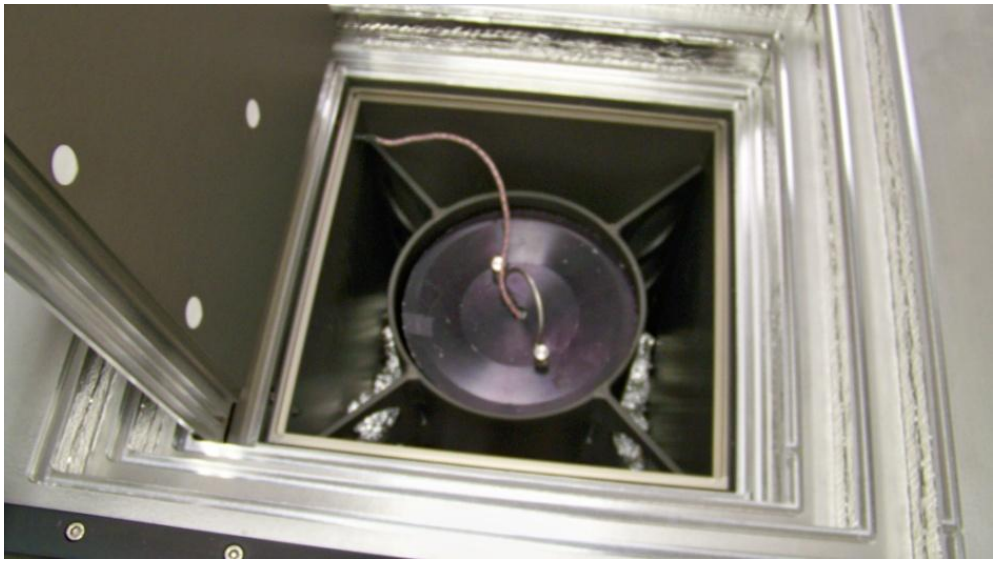

Figure 7. View of the calorimeter measurement chamber with the canister (containing an electric calibration sample) coupled to a thermal coupling mechanism.

The results of measurements with different sample thermal coupling configurations can be seen in Figure 8 with a supplied electric power of 300 mW. The initial shape the sample power curve (falling or rising) is determined by the relative magnitude of the power measurement, which took place before the measurement shown in the plot. In the case of falling curves, the previous measurement was of an electric sample with a higher power. The rising curves correspond either to a previous measurement of a smaller powered sample or a period where the calorimeter has been at thermal equilibrium with no electric sample power supplied.

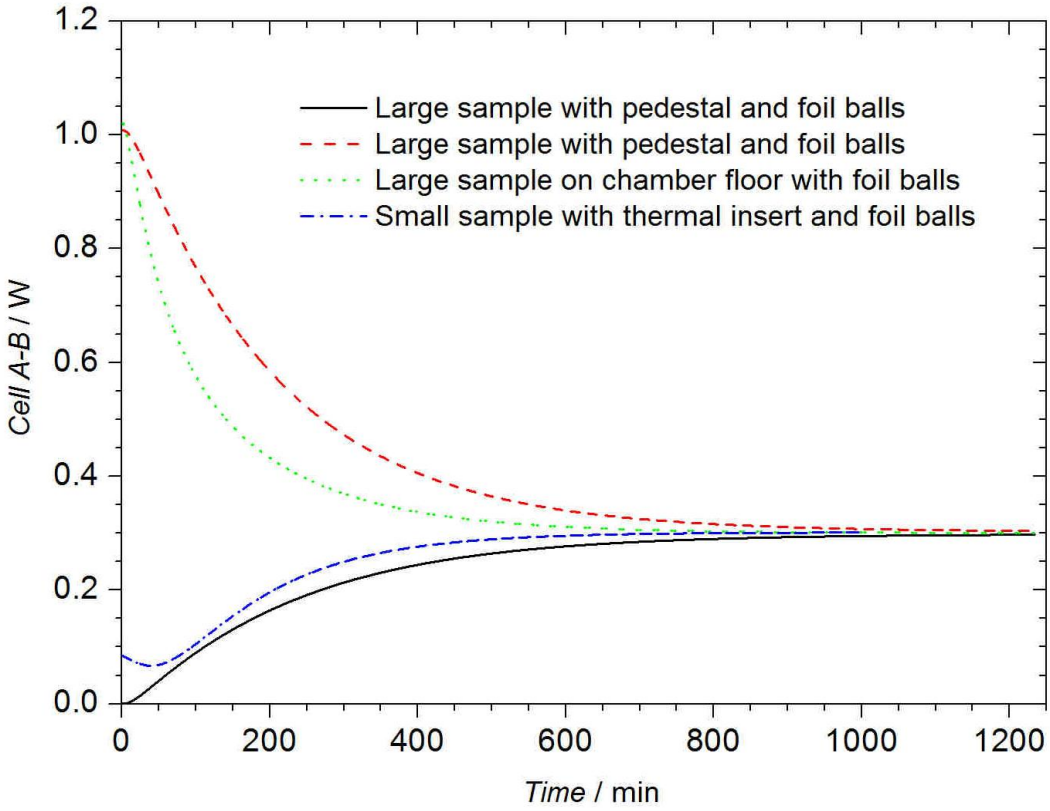

Figure 8. Measured sample power for electric samples of 300 mW but with different thermal coupling.

It can be seen from Figure 8 that calorimeter samples that are more effectively thermally coupled to the calorimeter measurement chamber reach thermal equilibrium with the measurement chamber more quickly than those with poor thermal coupling. Work continues to optimize the thermal coupling process in order to further reduce measurement times.

CONCLUSIONS

The performance characteristics of the ANTECH heat-flow calorimeter model 400HF-5300 have been demonstrated in this paper. The calorimeter achieves a high level of sensitivity for a large volume instrument. Measurements have shown that the measurement chamber is relatively insensitive to spatial variations in the distribution of heat sources. The calorimeter has excellent performance for low powered samples and achieves expected high precision for higher-powered samples. Initial work to shorten the time required to achieve a prediction of the sample end point power has produced results that show a factor of 4 improvement on average while maintaining

adequate precision and accuracy.

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REFERENCES

[1] J. A. Mason, K. J. Burke, N. J. Challacombe, A. R. Packman and A. C. N. Towner, *Design and Operation of a Large Volume Twin Cell Heat-flow Calorimeter for the Measurement of both Tritium and Plutonium Samples*, Proceedings of the 50th Annual Meeting of the Institute of Nuclear Materials Management, Tucson, Arizona, USA, July 2009.

[2] J. A. Mason, *Advances in Isothermal Calorimety for Plutonium Assay*, Proceedings of the 4th International Conference on Facility Operations - Safeguards Interface, Albuquerque, 29th September - 4th Oct. 1991, p251-254.

[3] M. I. Thornton, G. Vassallo, J. Miller, J. A. Mason, Design and Performance Testing of a Tritium Calorimeter, Nuclear Instruments and Methods in Physics Research, A.363 (1995) 598-603.