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ADVANCES IN ISOTHERMAL CALORIMETRY FOR PLUTONIUM ASSAY

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ABSTRACT

Calorimetry is a technique for measuring the thermal power of heat producing samples that is widely used in a variety of measurement fields. The technique has specific application in the area of nuclear materials accounting and safeguards. Isothermal calorimetry represents a further advance in the technique in that the relatively long thermal time constant associated with the heat capacity of the calorimeter measurement chamber is eliminated from the measurement process. This paper describes the isothermal (servo-controlled, power replacement) calorimeter method and recent improvements including expert system software to control the measurement data analysis procedure and enhanced instrumentation to allow software control of measurement chamber operating temperature and power. Results of recent measurements analyzed using the expert system software are reported.

INTRODUCTION

Calorimetric measurements of the thermal power of plutonium samples, when combined with a knowledge of the plutonium isotopic mass ratios, provide a convenient, accurate and non-destructive measure of the total plutonium mass of the samples[1,2]. The technique has advantages over other measurement methods such as passive neutron coincidence counting and destructive analysis. It does not suffer from neutron multiplication effects and it is not biased by inhomogeneity or the presence of moisture in the sample.

A calorimetry measurement is an absolute measurement of the heat evolved from the radioactive decay of a plutonium bearing material. As this process is mostly by means of alpha particle and beta particle emission by the different isotopes of plutonium and by americium, it is necessary to know the isotopic composition of the material.

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With the advent of efficient high resolution gamma-ray spectrometry systems and recent advances in the measurement of Pu and Am isotopic ratios by gamma-ray spectrometry, accurate isotopic ratio data is available in a timely manner and it can be readily combined with a calorimetric heat measurement to give Pu mass. An isotopic ratio data accuracy spanning from 0.5% to about 1% can be achieved for materials of low ^{242}Pu content and measurements with adequate counting statistics can be made in about one hour. The method has evolved so that improved accuracy is possible even with inhomogeneous samples by means of averaging the gamma-ray spectrum over the sample volume. Calorimetric heat measurement accuracy is often better than 0.5% so the overall accuracy in Pu mass determination is generally about 1% - 2% or better.

The operation of an isothermal plutonium calorimeter has been described in detail elsewhere[3]. Isothermal calorimetry, where the calorimeter measurement chamber is maintained at constant temperature, represents an improvement in the calorimetry technique in that the relatively long thermal time constant associated with the heat capacity of the calorimeter measurement chamber is eliminated from the measurement process. Improvements in measurement times can be achieved and acceptable levels of heat measurement accuracy are maintained, typically 0.5%. When this approach is combined with the ability to vary the calorimeter measurement chamber temperature and operating power, and hence match the calorimeter to the sample and container, a further measurement time saving is possible. Using this method sample preheating is not required.

POWER AND TEMPERATURE RANGE

Typically the calorimeter measurement chamber operating power may be varied over the range up to 100 Watts. The normal operating power is about 20 Watts. It is desirable to set the maximum operating power to a value about 10 % greater than the maximum sample power expected in any given campaign of measurements. The measurement chamber operating temperature is also variable. It is normally set, using software commands, to the value of the mean sample can surface temperature in order to eliminate the need for sample preheating and reduce measurement times[3]. Maximum measurement chamber operating temperature is normally 50 °C. The minimum temperature is determined by the requirement to dump heat to the ambient. The operating temperature range can be reduced significantly if a Peltier heat exchanger or other peripheral cooling system is employed.

Variable temperature operation is achieved by offsetting the resistance thermometer bridge balance point under software control for each of the three concentric cylindrical temperature controlled regions which constitute the calorimeter thermal element. Shifting the balance point for any cylinder changes the cylinder temperature,

the power applied to the cylinder and the overall calorimeter radial heat flow. By shifting the balance points of the cylinders in a coordinated way it is possible to vary both the measurement chamber temperature and applied electrical power over a wide range. This procedure is illustrated in figure 1 (which is reproduced from reference 3). In this case only the balancing bridge resistance for the measurement chamber (inner most concentric temperature controlled region) was varied. A greater temperature range can be achieved by varying more than one temperature controlled region at the same time.

The precision of a single power measurement averaged over 1 minute is less than 5 mW. The calorimeter is capable of an accuracy of heat measurement of better than 0.5% over most of the sample power range. At the bottom end of the range samples with a thermal power output of less than 0.25 Watts can be measured with a precision of about 1% or better. In most practical applications the errors on plutonium isotopic data are the limiting factor in plutonium mass determination.

PLUTONIUM ASSAY CALORIMETER SOFTWARE

The calorimeter system is driven by programs written in the "C" programming language and operating in a DOS and Windows environment. The software is menu driven with function keys defined on many screens for ease of use. During a measurement the user-interface screens display the state of the measurement and the operating conditions of the instrument. In addition to the display on the computer CRT monitor, the measurement results are recorded on the printer and on the system hard disk as the measurement proceeds. The data associated with each measurement, including sample description, plutonium isotopic data (input by the user), predicted and measured results and measurement errors are recorded as an archive file on the system hard disk. Extensive software driven test and diagnostic facilities are incorporated into the system as is the software controlled electrical calibration procedure. Some of the main feature of the software are summarized below:

a. The automated measurement procedure is controlled by an 'expert system' software module incorporating an end point prediction routine which indicates to the operator when a satisfactory result has been obtained, providing both a measurement result and its uncertainty.

b. A radioactive decay correction algorithm is provided to decay correct the plutonium and americium isotopic ratio data to the date of the measurement.

c. The software includes an automatic electrical calibration procedure. The results of calibrations and any variations in the baseline measurement chamber

electrical power are recorded on a 'measurement quality assurance' data file.

MEASUREMENT RESULTS

Measurements have been performed with two calorimeters[4,5] measuring both oxide and metal samples and operating at different baseline powers. The results have been analysed using the end point prediction routine of the software 'expert system' analysis module. The prediction analysis is based on fitting a single exponential function to successive measurements of the measurement chamber applied electrical power. Preliminary results are displayed in Table 1 for preheated (sample No. contains a 'P') and un-preheated samples. The 'first guess result' or first predicted value, corresponds to the first point at which the measurement data is appropriate for a single exponential function fit. The 'predicted result' corresponds to the point at which the algorithm criteria (target accuracy of 0.5%) is satisfied.

It can be seen that the 'first guess result' is generally less than 5% and this is achieved after about 60 to 90 minutes of measurement time. In most cases the 'predicted result' achieves an error of less than 0.5% in about 120 minutes for un-preheated samples and less time if preheating has been performed. It can be seen, however, that there is no advantage to sample preheating if the preheating time is considered as part of the measurement time.

CONCLUSION

Two significant enhancements have been incorporated into isothermal calorimetry which improve measurement performance. First software controlled variable temperature and power operation has been implemented and this facility aids the process of matching the calorimeter and sample temperatures and powers. Matching the calorimeter measurement chamber and plutonium sample can temperatures should significantly reduce the requirement for sample preheating.

The second feature is the implementation of an expert system software procedure to direct the measurement and data analysis process. Initial tests of this analysis procedure indicate significantly reduced measurement times. Preliminary measurements suggest that results for preheated samples are obtained in less than one hour and often less than 30 minutes. Sample power measurements on un-preheated samples suggest measurement times of about 2 hours with a heat measurement accuracy of about 0.5 % or better for sample powers above 1 Watt.

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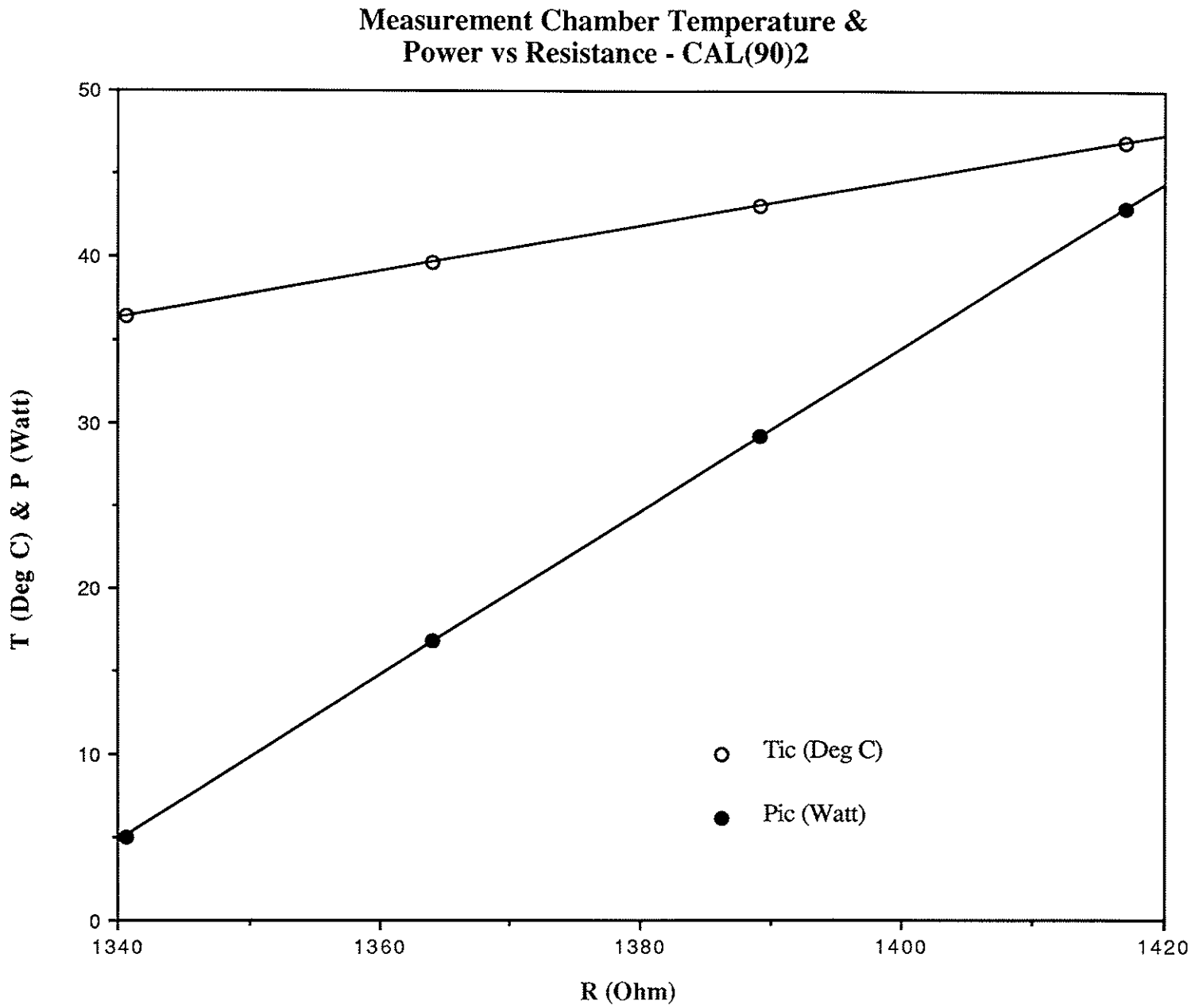
Measurement Chamber Temperature &
Power vs Resistance - CAL(90)2
T (Deg C) & P (Watt)
50
40
30
20
10
0
1340
1360
1380
1400
1420
R (Ohm)
Tic (Deg C)
Pic (Watt)

Table 1 Preliminary Measurement Results

Sample No.	End Power (W)	First Guess Result		Predicted Result	
		Time (min)	% Error	Time (min)	% Error
a	15.731	69	2.1	138	0.2
b	15.723	71	2.9	108	0.3
c	15.724	81	1.1	112	0.2
d	15.723	56	4.9	109	0.2
e	15.723	80	1.5	124	0.5
f	15.726	71	3.6	96	0.9
g P 2h	53.653	46	0.6	82	0.1
h P 1h	57.542	44	0.8	83	0.2
i P 1h	59.880	36	8.4	81	0.3
j P 2h	58.481	40	3.7	73	0.4