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ATTRIBUTES MEASUREMENTS BY CALORIMETRY IN 15 TO 30 MINUTES*

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ABSTRACT

An analysis of the early portion of the power-history data collected with both of the IAEA's air-cooled bulk calorimeters has demonstrated that such calorimeters can measure the power from preheated containers of plutonium oxide with an accuracy of 2-5% in 15 to 30 minutes. Material accountancy at plutonium facilities has a need for such a capability for measurement of Pu scrap. Also, the IAEA could use just two calorimeters and a gamma-ray assay system for reliable variables and attributes measurements of plutonium mass during a two-day physical-inventory verification (PIV) at a mixed-oxide(MOX) fuel-fabrication facility. The assay results would be free of the concerns about sample moisture, impurities, and geometry that previously have limited the accuracy of assays based on neutron measurements.

INTRODUCTION

Calorimetry yields a highly accurate measure of the power from a container of plutonium-bearing material independent of the container's moisture, impurities, and geometry, as well as other uncontrolled parameters, in contrast to the accuracy problems encountered with neutron detection of the same container. However, high accuracy takes time, i.e., the time for the container's temperature profile to equilibrate. The time for a measurement of the container's power to an accuracy of 0.2% using a bulk calorimeter is typically 2-6 hours. What has been overlooked, or at least not stressed, is that one can measure the power from a well-preheated container in 15 to 30 minutes with an accuracy of 2 to 5% (see data below) and still be independent of moisture, impurities, etc. Such a fast calorimetric method has two applications. First, it would be appropriate for process control and material control and accountability (MC&A) at plutonium facilities where large amounts of impurity-laden plutonium scrap are generated. This scrap is difficult to measure accurately using neutron detection. Secondly, a coarse but reliable "attributes" measurement would be useful to the IAEA during a physical-inventory verification (PIV) at a plutonium-reprocessing and/or MOX-fuel fabrication facility. For a moderately sized facility, the calculated number of attributes measurements (15-30) and highly accurate "variables" measurements (3-4) of containers of PuO_2 could be carried out using two calorimeters to do all heat measurements in just two days[1].

PREHEATING OF SAMPLES

The prerequisite for measuring power with an accuracy of 2-5% in 15 to 30 minutes using the Agency's air-cooled 20- and 40-Watt bulk calorimeters[2] is a long preheating period of 3 hours or more. For the application to MC&A and process control, several containers could be preheated overnight in several inexpensive preheaters, and then measured the next day. For the IAEA, the preheating could be done during the morning of the two days of PIV measurements. Meanwhile, the calorimeters could be used for the highly accurate variables measurements. The preheaters could be kept in the vault to avoid violating operator limitations on plutonium mass in a single room.

MEASUREMENT TIMES

The raw data in a heat measurement of a plutonium-bearing container is the power history of the container in the calorimeter, i.e., the electrical power supplied by the calorimeter to its concentric chambers to keep these chamber temperatures constant. Typically, the calorimeter power is sampled 64 times per minute and an average value printed out and/or stored in the computer memory. After a Pu container is placed in the calorimeter, the power drops sharply with time as the container's own heat replaces the calorimeter's power. A computer algorithm (a *double exponential* function with a constant term) is used to fit these power histories, $P_c(t)$;

$$P_c(t) = A*e^{Bt} + C*e^{Dt} + E. \qquad (1)$$

The more sharply dropping the power, the less important are the exponential terms at 15 minutes, and the more accurate is the end-point power prediction (E), the constant term. From E, the power of the container's contents (P_s) can be determined;

$$P_s(t) = P_{BL} - E, \qquad (2)$$

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where P_{BL} is the baseline power of an empty calorimeter.

Several variables affect how sharply the power drops, such as preheat time, heat resistance of the container's contents, transfer time to the calorimeter, etc. For those containers whose power falls off sharply, the time per measurement would be about 15 minutes; for other containers which have not been preheated long enough, 30 minutes may be required before the power prediction is within 5% of the correct value. To avoid premature prediction, the computer algorithm could be written to predict the end-point power after each measurement point and to test the time constants of the fitting function against expected values before accepting the predicted value. Thus, the time per measurement for this calorimetric attributes-mode measurement could be a variable to adjust for the differences in the container's preheat history or temperature profile. However, the simplest solution is always to preheat the container long enough to guarantee a sharply dropping power history.

PLUTONIUM MASS

The plutonium mass in the container measured by the calorimeter is the ratio of the power in Watts P_S to the effective specific power P_{EFF} in Watts/kg. P_{EFF} for most plutonium-bearing material measured in a calorimeter is known with good accuracy from chemical assay of samples drawn from the contents of the container, or from earlier samples taken from the batch of material that was processed to form the container's contents. In these cases, the uncertainty in the Pu mass is essentially that of the power measurement.

On the other hand, IAEA inspectors want their measurements to be independent of the operator. To do so, the effective specific power should be calculated from the isotopic fractions obtained from an analysis of a gamma-ray spectrum. For medium- to high-burnup samples, and for short (15 min) measurement times, the overall error in P_{EFF} is normally 2-3%. Therefore, the total sample power error is the combined error in P_S and P_{EFF}, both of comparable size.

RESULTS

The early power histories of several plutonium samples measured in the 20-Watt calorimeter and several heat standards measured in the 40-Watt calorimeter have been fit by means of regression analysis using the double-exponential plus a constant term[3]. The sample power P_S for each measurement has been compared to either the equilibrium or predicted power normally produced by the calorimeter after a few hours. Tables 1a,b show the results from five PuO_2 samples measured with the 20-Watt calorimeter and six heat standards measured with the 40-Watt calorimeter. The %Diff column(s) lists the percent difference between the equilibrium (or predicted) sample-power value measured normally by the calorimeter and either the 15-minute or 30-minute prediction.

20-Watt Calorimeter

For the 20-Watt calorimeter, containers of plutonium oxide were measured. The percent difference (%Diff) in power for these containers (Table 1a) ranged from 1% to 9%. It should be noted that the higher %Diff values for this calorimeter correspond to Pu containers which have undergone shorter preheating periods. There are a limited number of data sets because the power histories were not recorded for most of the calorimeter runs. Of those runs for which records were made, most included preheating for less than one hour.

40-Watt Calorimeter

For the 40-Watt calorimeter, heat standards of several sizes were measured. The seven power-history runs of 1.7 to 40 Watts are shown in Figures 1a,b. When all the data points for each run were used for the algorithm, the P_s results were essentially the same as the P_s values obtained using the calorimeter's own algorithm, i.e., a one-exponential fit to the tail end of the data. Then more and more of the tails of these data were eliminated and the double-exponential fit repeated. This continued until either too few data points were left or the results deviated too far from the known answer. The %Difference was calculated and some of the results shown in Figure 2. Also shown are the two exponentials contributing to the fit.

Notice that one exponential has a much shorter time constant and a larger amplitude than the other. It seems likely that the short time constant (≈2min) is associated with the calorimeter electronic-control circuits, while the exponential with the longer time constant is associated with the heat flow from the container. Ideally, if the container is preheated to exactly the same temperature as the calorimeter and if the transfer to the calorimeter is fast, the amplitude of the longer-time-constant exponential should approach zero.

When only a few data points are analyzed, a single poor point could cause a large error in the end-point prediction. The first few data points correspond to a quickly dropping calorimeter power and the average value printed out has a large uncertainty. Unfortunately, the algorithm available[3] did not have the capability to weight the data points with their errors, which would have reduced their impact on the fit. Instead, the first one or two points were dropped, allowing the end-point power prediction for a couple of the runs to greatly improve. In the future, data errors will be included in the algorithm.

Only the approximately 15-minute and 30-minute analyses are shown in Table 1b for 6 of the 8 heat standards measured. Of the other two standards, one (Cal-5) did not have its data recorded and one (Cal-1), the lowest-power heat standard, exhibited unusually noisy data. The %Diff values shown in the Table range from 3-4% for 30 minutes and 2-8% for 30 minutes. As seen in Figure 2, the predicted values do not follow any pattern; sometimes the end-point prediction improves as more points are dropped, sometimes not.

BIAS

There is a clear bias in the %Diff values listed in Tables 1a,b. The major source of this bias is the slight mismatch between the power-history curve that the early data follow and the double-exponential function chosen to fit these data. As more such data become available, a calibration curve could be generated to eliminate most of the 3-4% bias, leaving only a 2-3% random-error component.

CONCLUSIONS

Until now calorimetry has been relegated to the NDA-measurement domain of high accuracy and long measurement time. In contrast, neutron- and gamma-NDA measurements have accuracies and measurement times that reflect the properties of the material being measured and the accuracy needed. This paper focuses on the extended use of calorimetry for shorter measurement times and reduced accuracy, i.e., attributes measurements. All that is required is the ability to preheat the samples for 3 hours or more.

The 14 data runs on well-preheated plutonium samples and heat standards analyzed with the double-exponential algorithm indicate that sample powers could be measured with an accuracy of <5% in about 15 minutes. Field-test measurements of plutonium scrap are soon to be performed in order to confirm this conclusion and to add to the store of data for this calorimetry application.

Table 1a. Predicted Sample Power

20-Watt Calorimeter - Plutonium Oxide Samples

Sample #	Preheat time	Prediction Algorithm 30 min	Prediction Algorithm Equilibrium	%Diff
1	unknown	3.31 watts	3.44 watts	4%
2	unknown	2.02 watts	2.04 watts	1%
3	3 hrs **	4.24 watts	4.38 watts	3%
4	2 hrs **	4.06 watts	4.36 watts	7%
5	1.7 hrs **	3.97 watts	4.35 watts	9%

** We would like to thank Gordon Wells(Harwell) for supplying these calorimetry data.

Table 1b. Predicted Sample Power

40-Watt Calorimeter - Heat Standards

Standard #	Preheat time	Prediction Algorithm 15 min	Prediction Algorithm 30 min	Prediction Algorithm Equilibrium	%Diff (15)	%Diff (30)
1	>3 hrs	1.646	1.661	1.712	4.1%	2.9%
2	>3 hrs	5.152	5.140	5.373	4.1%	4.3%
3	>3 hrs	7.800	7.909	8.207	3.0%	4.0%
4	>3 hrs	25.597	25.467	26.329	2.8%	3.3%
5	>3 hrs	31.033	32.981	35.026	7.9%	3.9%
6	>3 hrs	41.757	39.307	39.918	1.3%	1.3%
				ave=	3.9%	3.2%

DISCLAIMER

ACKNOWLEDGEMENTS

R.P. wishes to thank John Lemming of Mound Laboratories for permitting the use of their facilities and heat standards.

REFERENCES:

[1] "Material Accountancy Verification Strategies for PuO_2 and MOX Powders and Pellets", S. Fiarman, W. Higinbotham, S. Mughabghab and J. Sanborn, Brookhaven National Laboratory report, ISPO-263, TSO-86-16, (November 1986).

[2] "The Bulk Assay Calorimeter", R.B. Perry et al., Argonne National Laboratory report ISPO-14, ANL-NDA-9 (January 1982).

[3] Hewlett Packard "STATPK" statistical package (copyright 1982) used on the HP 9816 computer.

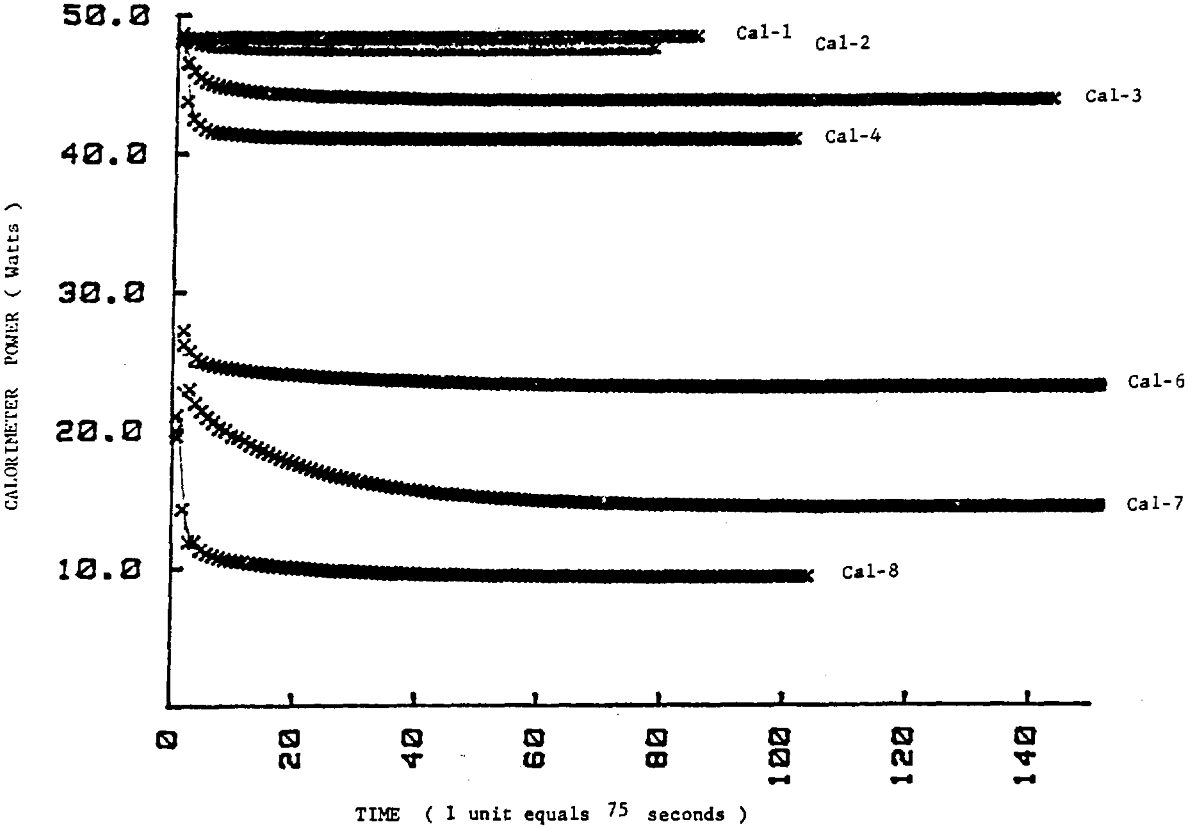


Figure 1a. HIGH-POWER ARGONNE CALORIMETER POWER HISTORIES FOR 7 HEAT STANDARDS

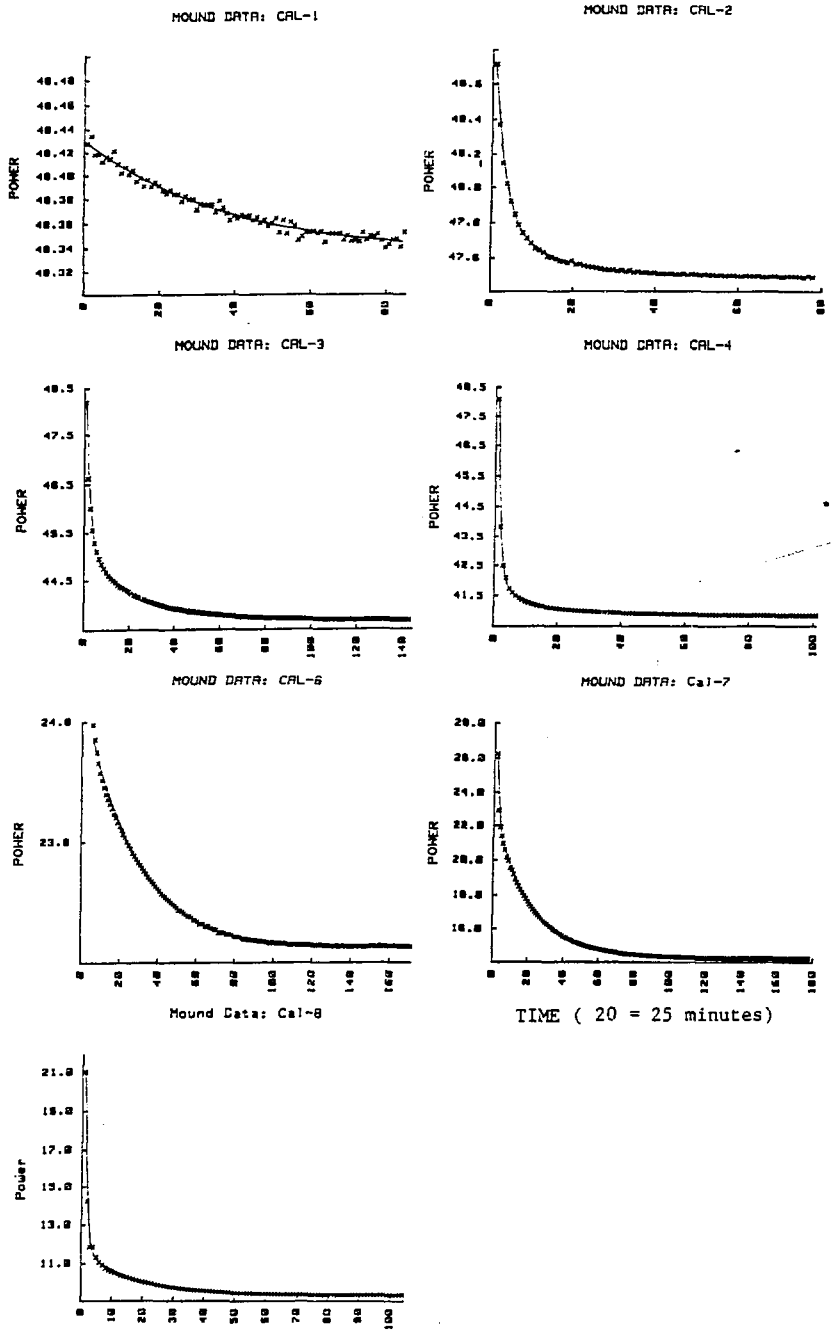
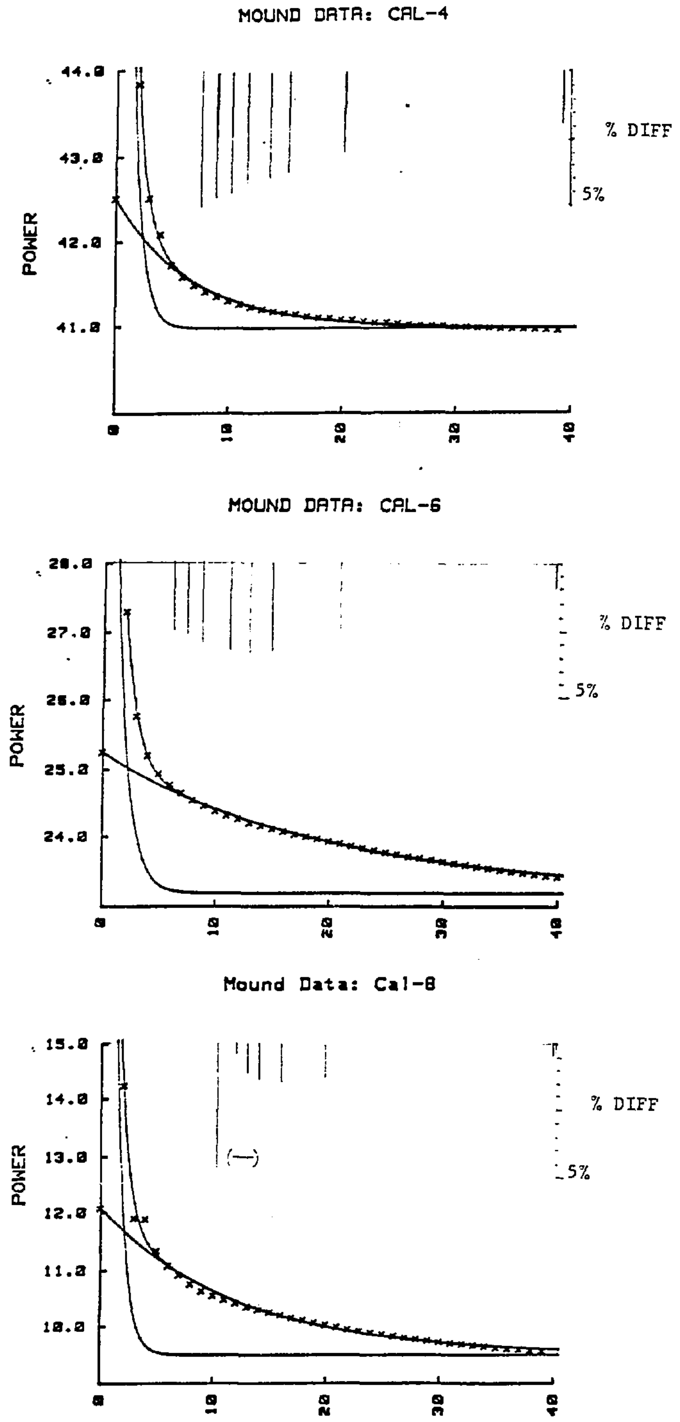


Figure 1b. EXPANDED SCALE FOR THE 7 POWER HISTORIES



MOUND DATA: CAL-4

MOUND DATA: CAL-6

Mound Data: Cal-8

Figure 2. POWER HISTORIES FOR THREE HEAT STANDARDS. THE INDIVIDUAL EXPONENTIAL TERMS ARE SHOWN. THE VERTICAL LINES ARE THE %DIFF VALUES USING THE INDICATED POINTS.