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Servo-Controlled Calorimeter System*

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EVALUATION OF THE ARGONNE NATIONAL LABORATORY SERVO-CONTROLLED CALORIMETER SYSTEM

by

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ABSTRACT

The control system of a replacement mode, twin-bridge, water-bath calorimeter originally built by Mound EG&G Applied Technologies was modified by Argonne National Laboratory. The calorimeter was upgraded with a PC-based computer control and data acquisition system. The system was redesigned to operate in a servo-control mode, and a preheater was constructed to allow pre-equilibration of samples. The instrument was sent to the Plutonium Facility at Los Alamos National Laboratory for testing and evaluation of its performance in the field using heat source standards and plutonium process materials. The important parameters for calorimeter operation necessary to satisfy the nuclear materials control and accountability requirements of the Plutonium Facility were evaluated over a period of several months. These parameters include calorimeter stability, measurement precision and accuracy, and average measurement time.

The observed measurement precision and accuracy were found to be acceptable for most accountability measurements, although they were slightly larger than the values for calorimeters in routine use at the Plutonium Facility. Average measurement times were significantly shorter than measurement times for identical items in the Plutonium Facility calorimeters. Unexplained shifts in the baseline measurements were observed on numerous occasions. These shifts could lead to substantial measurement errors if they are not very carefully monitored by the operating facility. Detailed results of the experimental evaluation are presented in this report.

INTRODUCTION

A modified Mound EG&G Applied Technologies twin-bridge, water-bath calorimeter was shipped from Argonne National Laboratory to Los Alamos National Laboratory for field testing and independent evaluation. The new system was tested for long-term and short-term baseline stability, and for precision and accuracy of routine assays of heat source standards and process materials in the operational environment of the Los Alamos Plutonium Facility.

The original Digital Equipment Corporation VAX computer system and the stand-alone digital volt meters required for data collection were replaced with a new PC-based data acquisition system utilizing an internal analog-to-digital conversion (ADC) card for data collection. This conversion to a PC-based system replaces an outdated VAX computer system with reliable, inexpensive, and readily available off-the-shelf components.

The calorimeter was originally designed to operate as an adiabatic calorimeter in replacement mode. In this mode of operation, the sample is placed in the sample chamber, and the power is inferred from the temperature rise in the sample chamber after the temperature has reached equilibrium or by using a prediction algorithm to predict the end-point temperature. A Wheatstone bridge is used to sense the temperature difference between the reference and sample chambers. With the new Bulk Plutonium Assay Calorimeter (BPAC) control system, the calorimeter operates in a servo-control mode, also known as an isothermal mode. In the servo-control mode, a constant power is supplied to both the sample and reference chambers of the calorimeter using internal heater coils which are wrapped around the outside of each chamber. The power to the sample chamber is controlled so that the Wheatstone bridge is always in balance. When a heat producing sample is inserted into the sample chamber, the power supplied to the sample chamber is reduced until the Wheatstone bridge is brought back into balance. The power produced by the sample is then simply the difference between the power supplied to the sample chamber with the sample in place—the assay power—and the power supplied to the sample chamber without the sample—the baseline power.

SYSTEM DESCRIPTION

The calorimeter is designed to measure the thermal power emitted by plutonium-bearing materials. The nominal measurement range is from 0.1 to 10 watts. Maximum sample dimensions are approximately 4.5 inches in diameter and 8 inches tall. The calorimeter consists of a two-unit EG&G Mound Applied Technologies twin-bridge, water-bath calorimeter, of which only one twin-bridge unit is currently functional. Each calorimeter unit consists of two identical calorimeter chambers which are immersed in a large-volume water bath. The water bath serves as a stable, constant-temperature environment around the calorimeter chambers. One chamber is the reference well, which remains empty at all times; the other is the sample well. The system also includes an Argonne-designed calorimeter control unit, a sample preheater, a Hart Scientific water-bath temperature controller, and the data acquisition/control computer, which is a 66 MHz, 80486 IBM compatible computer.

All data acquisition and control functions are handled through a Strawberry Tree ACPC 16/16 ADC and Digital I/O card. The internal ADC card is used for all voltage measurements on the reference and sample chambers. This ADC card provides voltage measurement precision equivalent to a 5 1/2-digit digital volt meter. The 16 bit digital I/O function provides communication with the Argonne Twin-Bridge Calorimeter Controller which supplies power to the heater coils on the reference and sample chambers.

The BPAC software consists of three separate tasks that run under the Quarterdeck DESQview 386 multitasking environment. The three tasks are: the Data-Acquisition-System (DAS) task, which handles all user interface operations and data manipulation; the I/O task, which handles all analog and digital I/O operations; and the Control task, which performs the PID control of the power supplied to the reference and sample chambers.

The sample preheater is a crucial component for operation of the calorimeter with optimum throughput. Assay times can be greatly reduced if the samples are preheated to match the inner temperature of the sample chamber prior to the assay. The preheater has a self-contained, AC temperature controller; a digital temperature meter; and a top-panel, operator-adjustable temperature control dial. Samples should be placed in the preheater for 30 to 240 minutes before the assay, depending on the thermal conductivity of the sample. Since the thermal conductivity is generally unknown, a minimum preheat time of 240 minutes is recommended.

RESULTS

The first step in the experimental evaluation was to adjust the temperature of the preheater to match the temperature of the inner well of the calorimeter sample chamber. The calorimeter temperature is sensitive to the water-bath temperature and the baseline power. Unfortunately, the inner-well temperature cannot be measured directly as the calorimeter is currently configured.

An iterative procedure was used to set the temperature of the preheater. An inert sample was preheated to different temperatures, and the equilibrium temperature of the calorimeter well was inferred from the equilibration time for an assay. For these measurements, a block of aluminum weighing approximately 2 kg was used. The block was placed in the sample chamber and assayed to equilibrium for each preheater temperature. The sample was first assayed from room temperature. Assays were then performed after preheating the sample for three or more hours at varying preheater temperatures. The equilibration time was determined for each assay. In addition, the power supplied to the sample chamber was written to disk in one minute intervals. These data are presented in Figure 1.

Figure 1. Power supplied to calorimeter sample chamber as a function of assay time for different preheater temperatures.

The baseline power in the sample chamber, which was measured before each assay, was approximately 9.5 watts, meaning that the Wheatstone bridge was in balance at this power level. Since the sample is not providing any additional power, when the system is returned to equilibrium, the power should be the same as the baseline power. For the assay from room temperature, approximately 23.5 °C, the equilibration time is 269 minutes. In this case, the initial sample temperature was well below the inside temperature of the calorimeter. The supplied power saturates at the maximum power output level for approximately 40 minutes while attempting to bring the sample to equilibrium. The power then approaches the correct equilibrium value over a period of approximately two hours.

The assay from 35.5 °C is the highest preheat temperature shown in Figure 1. In this case, the power immediately drops off when the sample is inserted. This indicates that the initial temperature of the sample is higher than that of the calorimeter well. As the excess heat dissipates from the sample, the power slowly returns to the baseline value. The next two cases, at 33.4 and 33.0 °C, show the assay power as the temperature approaches the correct preheat temperature. For a preheat temperature of 33.0 °C, the initial temperature is very closely matched to that of the calorimeter, and there is very little fluctuation in the calorimeter power when the sample is inserted. A preheater temperature of 33.0 °C was used for all subsequent measurements discussed in this report.

The equilibration time for an assay is very sensitive to the choice of preheater temperature. If the sample is placed in the calorimeter at a temperature that is not equal to the calorimeter temperature, the equilibration time is determined by the rate at which the calorimeter can supply or dissipate thermal energy, as well as by the thermal conductivity and heat capacity of the sample. In the Al block example demonstrated in Figure 1, the equilibration time was reduced from 269 minutes for the room temperature assay to 64 minutes for the assay at 33.0 °C.

The baseline stability was determined through repeated baseline measurements over the course of the evaluation period. The stability of the baseline is important for accurate assays of nuclear material because the sample power is determined from the difference between the baseline power and the power measured with the sample in place. Any changes in the baseline power between the time of the baseline measurement and the time of the assay will directly affect the assay results. The stability of the baseline power was shown to be acceptable over short time intervals. However, relatively large and discrete shifts in the baseline were observed on numerous occasions throughout the experiment. These shifts could not be correlated with any external influence on the calorimeter.

All of the baseline measurements discussed here were made after a disturbance to the calorimeter, i.e., either a sample was removed from the calorimeter or an empty canister was inserted from the preheater. The results of the baseline measurements taken throughout the evaluation fell into closely matched groups rather than a random distribution of values. The baseline results are shown in Table I. The results in Table I show the averages of the groups, the one sigma standard deviation of the data set within the group, the number of baseline runs that fell into that group, and the average measurement time for the baselines within the group. It is worth noting that a particular set of baseline measurements in a group was not necessarily collected in a chronological sequence. A given set of sequential baseline measurements might have as few as one or as many as ten measurements belonging to a particular group before a major shift occurred. This most likely indicates that there is a problem in the servo-control algorithm which causes the calorimeter to stabilize at these discrete baseline power levels.

Table I. Baseline Stability Measurement Results.		
Baseline Power (watts)	Number of Measurements	Average Time (minutes)
8.1723 ± 0.0080	12	63
8.1570 ± 0.0006	2	85
8.6313 ± 0.0053	10	59
9.1191 ± 0.0050	4	67
9.5020 ± 0.0063	20	85
9.5611 ± 0.0088	11	67

The standard deviation of the baseline measurements in a given group varied from approximately 5 mW to 9 mW, with the exception of one group which had only two numbers. This standard variation is somewhat higher than the typical baseline observed in the calorimeters in operation in this facility. A recent study[1] showed a baseline variation of 3 mW over a period of several months for a Mound over/under water-bath calorimeter at the Los Alamos Plutonium Facility. The data averaged over all the groups for the Argonne servo-controlled calorimeter shows a variation of 0.064%, or approximately 6 mW. The average baseline measurement time for the Plutonium Facility calorimeter was 341 minutes, compared to an average measurement time of 71 minutes for the Argonne servo-controlled calorimeter. There is normally a tradeoff between measurement precision and assay time for any measurement process. The apparent discrepancy in the measurement precision and average assay time in the baseline results for these two calorimeters could be partly induced from the choice of parameters used to determine equilibrium conditions. In addition, it is expected that a servo-controlled calorimeter will have somewhat shorter measurement times as seen here.[2]

Plutonium heat source standards were used to test the precision and accuracy of the instrument for assays of heat-producing samples. All of the standards used were certified standards with well-known wattages. Seven different standards were assayed. Six were ²³⁸Pu heat standards and one, CALEX, was a certified, 400 g PuO₂ standard. Because of the unexpected shifts observed in the baseline power measurements, a baseline was run immediately before each assay, either the day of or the evening before the assay.

The results for the heat standard measurements are summarized in Table II. The table shows the certified wattage for each standard, the average ratio of the measured to certified wattage for that standard, the standard deviation of the data set for that standard, the number of assays in the data set, and the average assay time for each item. One statistical outlier with a measured to certified ratio R of 0.9463 was omitted from the CALEX data set. Also, two standards had only one measurement, and therefore the standard deviation is not applicable.

Table II. Measurement Results for Calorimeter Heat Standards.					
Standard ID	Cert. Watts	Measured/Certified	Std. Dev.	# of Assays	Time (min.)
CALEX	1.0053	0.9987	0.0039	6	184
MF13C	2.9363	0.9994	NA	1	85
UAS009T	4.0737	0.9984	NA	1	238
MF24	5.0257	0.9988	0.0004	4	96
MW7	0.6723	0.9975	0.0019	2	52
RW5	0.9020	0.9995	0.0016	4	82
UAS1385	3.6649	0.9990	0.0003	5	76
Ratio over All Standards = 0.9988 ± 0.0021			Average Assay Time = 121 min		

The results are presented graphically in Figure 2, where the ratio of the certified to measured wattage is plotted against the certified wattage. The trend in the data suggested by the average ratio in Table II of less than 1.0 is clearly illustrated in Figure 2. In fact, 18 of 23 measurements were below 1.0, indicating a real bias in the measurements. The average ratio over all the measurements is 0.9988. The standard deviation of the mean is 0.00045. Therefore, the average ratio of the entire data set is approximately 2.6 standard deviations below the expected result, suggesting that the apparent -0.12% bias is statistically significant. The assay precision over the entire data set was 0.21%. From Figure 2, it is evident that the precision is significantly different at the upper and lower portions of the

measurement range. Below approximately 1 watt, the standard deviation of the ratio was 0.29%. Above 1 watt, the standard deviation of the ratio was reduced to 0.036%. These results are reasonably consistent with an operational calorimeter in the facility that was recently demonstrated to have a precision of 0.15% at 3 watts and 0.075% at 7.3 watts.[1]

Figure 2. Ratio of the measured to certified heat value for the measurements of the standards shown in Table II.

In addition to the heat source standards, plutonium process items were measured to assess the performance of the calorimeter for weapons-grade plutonium materials. The measurement data are shown in Table III. The selection of items available for these measurements was very limited due to the size of the calorimeter. The inner diameter of the sample canister is just over 4.5 inches.

Table III. Measurement Data for Process Materials.				
Sample ID	Item Type	Pu Mass (g)	Measured/Accepted	Assay Time (min)
CMF2519	Metal	610.88	0.9915	157
GVB-16-102-#1	Oxide	633.71	0.9964	405
GVB-16-M-10	Oxide	97.07	0.9979	103
HUD6-102-85-114-1	Oxide	626.27	1.0675	85
HUD6-102-85-223	Oxide	802.41	1.0004	96
HUD6-102-83-365	Oxide	736.29	0.9999	161
HUD6-4709-012-023	Oxide	1413.41	0.9994	320
LZB216-BG-259	Metal	2177.27	0.9971	21
Average over All Measurements =			1.0052 $\pm$ 0.0182	169 min

The data presented in Table III are again shown in terms of a ratio of the measured power to an accepted value. In the case of the process items shown in Table III, there is not a true "accepted" value. For these items, the assay value from the servo-controlled calorimeter is compared to the assay value from one of the calorimeters in the Plutonium Facility's

Non-Destructive Assay Laboratory. These calorimeters are maintained under a rigorous measurement control program and were known to be "in control" at the time of the measurements. It is apparent from the data in Table III that there is also a bias in this data set. There is also one statistical outlier in this data set. The measurement of HUD6-102-85-114-1 is over 6% high. After throwing out this data point, the average ratio becomes 0.9974, and the standard deviation of the mean is 0.0012. A statistically significant bias is again indicated since the apparent bias is greater than two standard deviations from the expected ratio of 1.0. The bias observed for these process items is statistically indistinguishable from the bias shown previously for the heat standards. The measurement precision over the entire data set is 0.31%. This is consistent with the precision demonstrated for the heat standards.

The average assay time for the eight metal and oxide items in the Argonne calorimeter was 169 minutes. The average assay time for the same items in the Plutonium Facility calorimeters was 347 minutes. On average, the assay time in the Argonne calorimeter is approximately half that in the other calorimeters. The precision of the Plutonium Facility calorimeter cannot be estimated for these items since there is no reference value for comparison. Typical measurement precision for the Plutonium Facility calorimeters is 0.1 to 0.2%.

CONCLUSION

The performance of the Argonne servo-controlled calorimeter was positive in many respects. The measurement precision given by the standard deviation of the pooled measurement data was 0.21% for the heat standards and was 0.5% for the process items. Although these numbers are slightly larger than those of the calorimeters at the Plutonium Facility, which are typically <0.2% on heat standards, they would still be acceptable in most cases. The accuracy measurements revealed a $-0.12\% \pm 0.045\%$ bias for the heat standards and a $-0.26\% \pm 0.12\%$ bias for the process items. These biases are indistinguishable when the measurement statistics are considered. The observed bias was reasonably consistent over the range of heat standards assayed. This indicates a constant bias that could most likely be removed or greatly reduced with a proper calibration or bias correction.

The average measurement times for the calorimeter were very impressive. The average time for a baseline measurement after a disturbance to the calorimeter was 71 minutes, significantly shorter than typical baselines for the operating calorimeters in the Plutonium Facility. The measurement times for the heat standards were impressive at 121 minutes on average. Measurement times for the process items were somewhat longer at just under three hours (169 minutes) on average. The average measurement time for the same items in Plutonium Facility calorimeters was nearly six hours. As stated earlier, it is expected that the servo-controlled calorimeter with a preheater will exhibit shorter assay times as verified in this evaluation. It should also be noted that at least part of the decrease in measurement time may be due to a reduction in assay precision since the precision of the Argonne servo-controlled calorimeter is not as good as that of the Plutonium Facility calorimeters.

Some disturbing results were also observed. The first was the instability in the calorimeter baseline measurements. Shifts as large as one watt were observed in the baseline from one day to the next. An error in the measured wattage of this magnitude would be the equivalent of approximately 400 g of low-burnup plutonium. Even with a baseline measurement performed before each measurement, the result of one measurement on GVB-16-102-#1 was 1.2171 watts, approximately 50% of the expected value. When the measurement was corrected by using the baseline measurement performed immediately after the assay, the result was 2.6106 watts, which was within 0.5% of the expected value. For this one item,

the baseline shift apparently occurred between the initial baseline measurement and the assay. If this calorimeter is to be used for accountability measurements, great care will have to be taken to ensure that the calorimeter is stable between the baseline and the assay and during the assay.

Another problem observed was the occurrence of two outliers that were many sigma away from the average results. One measurement from the CALEX data was 5.4% low, and one of the process items, HUD6-102-85-114-1, was 6.75% high. These outliers could not be directly attributed to shifts in the baseline power or to any external perturbations to the calorimeter or its environment. Even errors of this magnitude may be acceptable for some low-grade residues or waste materials, but they are clearly not acceptable for high-quality metals or oxides. Additional measurements would need to be performed before this calorimeter is officially used for accountability measurements in order to quantify the true magnitude and frequency of this problem.

REFERENCES

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