IMPROVED END-POINT PREDICTION IN ISOTHERMAL CALORIMETRY

J.A. Mason, on secondment from Imperial College, London, U.K.
N. Bainbridge, A.N. Technology Ltd., Ascot, Berkshire, U.K.
S. Guardini, CEC, JRC, Ispra

Abstract

An "Expert System" software procedure has been developed for use with isothermal calorimeters in order to improve the sample end-point power prediction process. The procedure examines the measurement data and determines the onset of the exponential end-point prediction procedure. It also determines when sample thermal equilibrium conditions have been reached. The procedure has been tested using archived data from two different calorimeters and sample power predictions are obtained in a total measurement time of about 2 hours with sample power errors of less than 0.5%.

1. Introduction

Recent measurements with isothermal calorimeters/1,2,3/have demonstrated both the accuracy in the technique and reductions in measurement times compared to conventional heat-flow calorimeters. The present work has been directed towards further reductions in measurement times. This is motivated, in part, by the needs of safeguards inspectors for short measurement times.

2. Measurements

Table 1 contains results from isothermal calorimeter measurements performed using PERLA plutonium oxide standards. Although the results are generally satisfactory, typically a period of about 4 hours was required for each measurement to achieve a satisfactory predicted sample end power. The reason for the long time period to reach an adequate prediction value can be seen in Fig. 1. The figure is a plot of the measured sample power P (Po-IC) and the predicted sample power P (Pred) as a function of time. From the figure it is clear that the predicted sample power reaches an asymptotic value after a period of about 3 hours and only after a large number of power measurement data points are included in the exponential fitting algorithm. It would appear that the prediction process is being biased by early data points which do not fit the assumed single exponential form of the prediction equation.

3. Analysis

On the basis of measurement data from two calorimeters an analysis of the behaviour of the prediction procedure was undertaken. In particular the behaviour of the coefficient of the exponential was examined. It was found that for most measurements this exponential coefficient reached an asymptotic value in a range of time from about 20 minutes to 60 minutes from the start of the measurement. It was further observed that if the onset of prediction was delayed until the exponential coefficient achieved an asymptotic value a better prediction was obtained at an earlier time. This is in contrast to the previous prediction procedures where the prediction was started after a fixed delay period (usually 15 minutes) following the start of the measurement (sample insertion into the calorimeter measurement chamber).

A software procedure has now been devised to control the prediction process. A rolling value of the exponential coefficient of the fitting function is calculated using 18 averaged sample power data points and the achievement of an asymptotic value is tested using a rolling linear least squares fit to the last six values of the calculated exponential coefficient. The test criteria involve a number of parameters including the slope and standard deviation in slope of the coefficient itself. Once the criteria are satisfied the single exponential fitting/prediction procedure is initiated and similar tests and criteria are used to determine the earliest acceptable value for the predicted sample power.

4. Results and Conclusions

The new procedure has been tested on data from two calorimeters including measurements of both plutonium metal and oxide samples. The results are displayed in Table 2. Sample numbers with a suffix of P1 or P2 have been preheated for one or two hours respectively.

The results are very satisfactory and show that the new procedure works well. At the time of the "first guess", which corresponds to the first predicted value, the error (when compared to equilibrium or certified values) is less than 5% and this is achieved in times ranging from about one hour to about 90 minutes for samples with no preheating. The prediction achieves an acceptable value (error less than 0.5%) in about two hours. These results represent a significant improvement in measurement times when compared to the results from the conventional prediction procedures. It is anticipated that additional refinements to the prediction procedure will further improve the results.

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5. References

/1/ J.A. Mason, R.W. Wilde, J.C. Vickery, B.W. Hooton and G.M. Wells, "Development and Evaluation of a Plutonium Assay Calorimeter Test Bed", Proceedings of the 29th INMM Annual Meeting, Las Vegas, 26-29 June 1988 699-704.

/2/ J.A. Mason, R.W. Wilde, J.C. Vickery, B.W. Hooton, G.M. Wells, M. Cuypers and S. Guardini, "Plutonium Calorimetry", Proceedings of the 11th Annual ESARDA Symposium on Safeguards and Nuclear Material Management", Luxembourg 30 May - 1 June 1989, 519-524.

/3/ J.N. Lowe and J.A. Mason, "Development and Application of a Low Power Transportable Calorimeter for Plutonium Assay", Proceedings of the 31st INMM Annual Meeting, Los Angeles 15-18 July 1990, 496-500.

JRC Ispra Plutonium Assay Calorimeter - Measurements with PERLA PuO_2 Standards

Sample No.	Data File	Calculated Power (Watts)	Measured Power (after 3 Hrs) (Watts)	Predicted Power (after 3 Hrs) (Watts)	Error ΔP (%)	Final Predicted Power (after 5 or 6 Hrs) (Watts)	Error ΔP (%)	Sample Treatment
Pu113		15.44	14.866	15.4971	+ 0.32	15.4425	+ 0.02	no preheating (V1.5)
Pu113	30019001	15.511	14.06	15.574	+ 0.38	15.60	+ 0.58	1 hr preheat (V1.7)
Pu113	30019003	15.512	14.93	15.585	+ 0.44	15.565	+ 0.34	1 hr preheat, Al, (V1.7)
Pu113	01029001	15.513	14.79	15.473	- 0.25	15.512	0.00	no preheating, Al (V1.7)
Pu113	02029001	15.514	14.73	15.559	+ 0.29	15.523	+ 0.06	0.5 hr preheat
Pu113	05029001	15.516	14.77	15.582	+ 0.42	15.548	+ 0.20	1 hr preheat
Pu113	06029001	15.517	14.85	15.478	- 0.26	15.587	+ 0.45	2 hr preheat
Pu113	07029001	15.518	15.10	15.460	- 0.37	15.528	+ 0.06	3 hr preheat
Pu113	08029001	15.519	14.80	15.506	- 0.08	15.528	+ 0.06	0.25 hr preheat
Pu106	I23039001	1.566	1.512	1.578	+ 0.78	1.5615	- 0.28	1 hr preheat, canister top
Pu106	I22039001	1.566	1.458	1.571	+ 0.32	1.5706	+ 0.29	1 hr preheat, canister centre
Pu106	I21039001	1.566	1.496	1.554	- 0.77	1.5555	- 0.70	1 hr preheat, canister bottom
Pu111	31019002	5.872	4.556	5.753	- 2.0	5.859	- 0.22	1 hr preheat, brass packing
Pu111	I301101	5.845	5.20	5.863	+ 0.3	5.8398	- 0.08	
Pu115	I20039001	22.440	21.245	22.5307	+ 0.43	22.466	+ 0.11	Po without canister
Pu115	I19039001	22.438	21.07	22.466	+ 0.10	22.436	- 0.00	Po with canister
Pu120	ISP281102	37.96	36.48	38.04	+ 0.21	37.918	- 0.12	no preheating

$$\text{Error } \Delta P\ (\%) = \frac{(\text{Predicted Power}) - (\text{Calculated Power})}{(\text{Calculated Power})} \times 100$$

Table 1

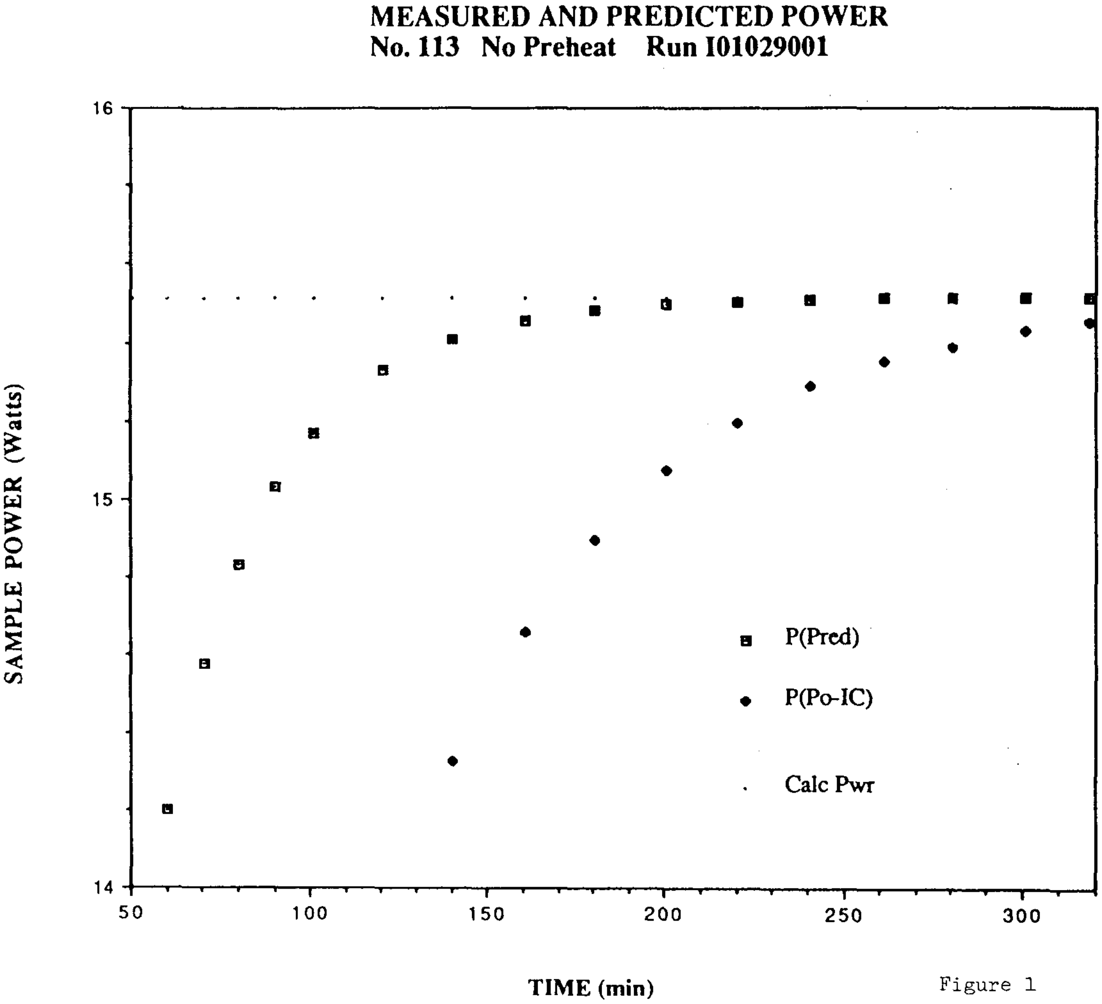

Figure 1

MEASUREMENT RESULTS USING EXPERT SYSTEM SOFTWARE

SAMPLE N°	END POWER (W)	FIRST TIME (min)	GUESS % ERR.	PRED. TIME (min)	RESULT % ERR.
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	15.715	69	2.0	121	0.2
G	15.726	71	3.6	96	0.9
HP2	53.653	46	0.6	82	0.1
IP1	57.542	44	0.8	83	0.2
JP1	59.880	36	3.4	81	0.3
KP2	58.481	40	3.7	73	0.4
L	39.010	46	0.3	94	0.3

TABLE 2