

**A PORTABLE CALORIMETER SYSTEM
FOR NONDESTRUCTIVE ASSAY
OF MIXED-OXIDE FUELS**

by

**C. T. Roche, R. B. Perry, R. N. Lewis,
E. A. Jung, and J. R. Haumann**

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Nondestructive Assay Section
Special Materials Division

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ABSTRACT

Calorimetric assay provides a precise, nondestructive method to determine sample Pu content based on the heat emitted by decaying radionuclides. This measurement, in combination with a gamma-spectrometer analysis of sample isotopic content, yields the total sample Pu mass. The technique is applicable to sealed containers and is essentially independent of sample matrix configuration and elemental composition. Conventional calorimeter designs employ large water-bath heat sinks and lack the portability needed by inspection personnel. The ANL air-chamber isothermal calorimeters are low-thermal-capacitance devices which eliminate the need for large constant-temperature heat sinks. These instruments are designed to use a feedback system that applies power to maintain the sample chamber at a constant electrical resistance and, therefore, at a constant temperature. The applied-power difference between a Pu-containing sample and a blank determines the radioactive-decay power. The operating characteristics of a calorimeter designed for assaying mixed-oxide powders, fuel pellets, and Pu-containing solutions are discussed. This device consists of the calorimeter, sample preheater, and a microprocessor-controlled data-acquisition system. The small-sample device weighs 18 kg and has a measurement cycle of 20 min, with a precision of 0.1% at 10 mW. A 100-min gamma-ray measurement gives the specific power with a precision of better than 1% for samples containing 1-2 g of plutonium.

A. Introduction

The use of calorimeters to perform a nondestructive determination of the plutonium content of nuclear materials has been well documented.^{1,2,3} The technique is both highly sensitive and precise. Calorimeters are capable of detecting plutonium concentrations in the 20-ppm range. This corresponds to $0.1 \mu\text{W}/\text{cm}^2$, or approximately $0.1 \text{ g } ^{239}\text{Pu}/\text{liter}$. Precision better than 0.1% is obtainable on the measurement of plutonium-produced power.⁴ Among the advantages of calorimetry, when compared with other nondestructive-analysis techniques, are its insensitivity to the chemical form of the plutonium and its independence of measurement-bias problems due to sample geometric configuration and sample matrix composition. However, the lack of portability of the instrumentation and the relatively long sample assay time, when compared to neutron and gamma-ray assay procedures, have prevented calorimetry from being used as an in-field analytic technique. In designing ANL air-chamber isothermal calorimeters, we have constructed low-thermal capacitance devices which eliminate the necessity for the large water-bath heat sinks used by classical heat-flow calorimeters. The first of a set of instruments designed to assay the types of plutonium-containing materials encountered by International Atomic Energy Agency (IAEA) inspection personnel has been completed and tested. This device, the "Small-Sample Calorimeter," has been designed to measure the plutonium content of fuel pellets, powders, and solutions. The reduction in instrument size and mass, coupled with the improvement in measurement time should enable inspectors to employ this technique for in-field verification of nuclear safeguards systems.

B. Radioactive Decay and Sample Specific Power

Calorimetry is a technique which can be used to measure the thermal power produced by decaying radionuclides. This power is related to the nuclide mass through the total decay energy and the specific activity of the disintegration. The nuclear constants for the isotopes present in mixed-oxide fuel which are of importance to calorimetric assay are listed in Table I.⁴ These include the plutonium isotopes ($A = 238-242$) and ^{241}Am . The uranium isotopes present in MOX fuel are not included since the power produced by these nuclides is insignificant when compared to plutonium.

Table I. Nuclear constants for the important heat-producing nuclides in plutonium-bearing materials.

Nuclide	Q Total Disintegration Energy (MeV)	Principle Decay Mode	%Q Emitted as γ	$T_{1/2}$ (γ)	λ (days^{-1})	Specific Power mW/g
^{241}Am	5.6402 \pm 0.00016	α	0.38	434.1 \pm 0.06	4.372×10^{-6}	114.23 \pm 0.16
^{238}Pu	5.5921 \pm 0.0016	α	0.0005	87.79 \pm 0.08	2.1617×10^{-5}	567.16 \pm 0.57
^{239}Pu	5.2428 \pm 0.0016	α	0.0003	24082. \pm 46	7.880×10^{-8}	1.9293 \pm 0.0053
^{240}Pu	5.2551 \pm 0.0016	α	0.0005	6537. \pm 10	2.903×10^{-7}	7.098 \pm 0.015
^{241}Pu	0.00553 \pm 0.00001	β	0.0045	14.35 \pm 0.02	7.322×10^{-4}	3.39 \pm 0.002
^{242}Pu	4.985 \pm 0.01	α	0.0	370399. \pm 2900 ²	5.01×10^{-9}	1.18 \pm 0.01 ²

¹ ANSI N15-22-1975

² J. M. Meadows, ANL/MDM-38

With the exception of ^{241}Pu , the principal decay mode of the isotopes in Table I is alpha-decay, with a total disintegration energy between 5-6 MeV. Alpha particles in this energy interval are very short-ranged (< 50 mg/cm² of Al). The other major radiations produced by these isotopes, a 5.5 keV beta from ^{241}Pu and a 59.5 keV gamma from ^{241}Am , are also short-ranged and, like the alpha's, will deposit their energy within the sample or the measurement-chamber walls. Sources of energy which will not be measured by the calorimeter include high energy gamma-rays, $\bar{\nu}$, and neutrons; however, these account for less than 0.01% of the total decay energy. Consequently, calorimetry is a technique for which the theoretical detection efficiency approaches 100%.

In order to convert the calorimetrically measured wattage into plutonium content, it is necessary to determine independently the power emitted per gram of sample within a particular isotopic composition. The power emitted by a sample composed of a single isotope is:

$$P_i = 1.1167 \times 10^3 \times Q \times \lambda \times M_i/A$$

where

P_i	-	specific power of the isotopic i (mW/g)
Q	-	total disintegration energy (MeV)
λ	-	decay constant (day ⁻¹)
A	-	gram atomic weight of isotope i (g)
M_i	-	mass of isotope i (g)

Most material encountered by IAEA inspection personnel during an assay contains a mixture of the isotopes in Table I. Thus, the sample power would be:

$$P_S = \sum^n M_i P_i = M_T \sum^n R_i P_i = M_T P_{\text{eff}}$$

where

P_S	-	sample power (mW)
M_T	-	total sample mass of Pu (g)
R_i	-	mass fraction of isotope i
P_{eff}	-	effective specific power (mW/g)

Typical isotopic composition of nuclear fuels which may be encountered are given in Table II.^{5,6} The principal constituent is ^{239}Pu which varies approximately 30% over the fuels listed. The variation in the other principal heat-producing nuclides is larger. Plutonium-240 ranges over a factor of 4, and ^{238}Pu a factor of 10. Americium-241 is the decay product of ^{241}Pu , and, consequently, the americium content of the sample is dependent upon the length of time since the fuel was reprocessed. As a result of the ingrowth of ^{241}Am , the effective specific power of a fuel sample will increase with time. The effective specific power in mW/g of Pu is also given in Table II.

The effect of changing isotopic composition on the relative heat contribution of the isotopes in MOX-LWR (light water reactor) fuel is shown in Fig. 1.⁷ As fuel burn-up proceeds, the relative amounts of the nuclides with large specific powers increase. Consequently, the P_{eff} for a particular fuel may vary significantly depending upon its residence time in the reactor.

The effective specific power may be determined in two ways. The first method, referred to as the empirical technique, requires that a set of representative samples be collected. These samples are measured calorimetrically, and a subsequent chemical assay is performed to determine their plutonium content. A time spread in the analysis of the sample is necessary to determine the change in specific power due to the ingrowth of ^{241}Am . In general, this technique is not suitable for inspection personnel because of the time delay required to account for the americium. The second method, referred to as the computational technique, requires that an isotopic analysis be performed and that the mass ratios be used to calculate a weighted-average specific power. The isotopic analysis may be performed either by mass-spectrometric or NDA techniques. The total assay may be performed nondes-

Table II. The isotopic abundances of the heat-producing nuclides in typical nuclear fuels. (The mass ratios are relative to plutonium.)

Nuclide	wt %					
	Fuel Type					
	LWR	FBR (ZPPR)		British	Recycle	WR
		F & G	H			
²⁴¹ Am	0.7	0.15	--	0.1	1.5	0.05
²³⁸ Pu	0.2	0.05	0.09	0.1	0.5	0.01
²³⁹ Pu	75.7	86.6	68.5	90.2	65.0	93.5
²⁴⁰ Pu	18.4	11.5	25.6	8.5	24.0	6.0
²⁴¹ Pu	4.6	1.7	4.53	1.0	8.0	0.5
²⁴² Pu	1.1	0.2	1.4	0.2	2.5	0.05
P _{eff} (mW/g)	5.19	2.92	3.80	3.05	7.85	2.35

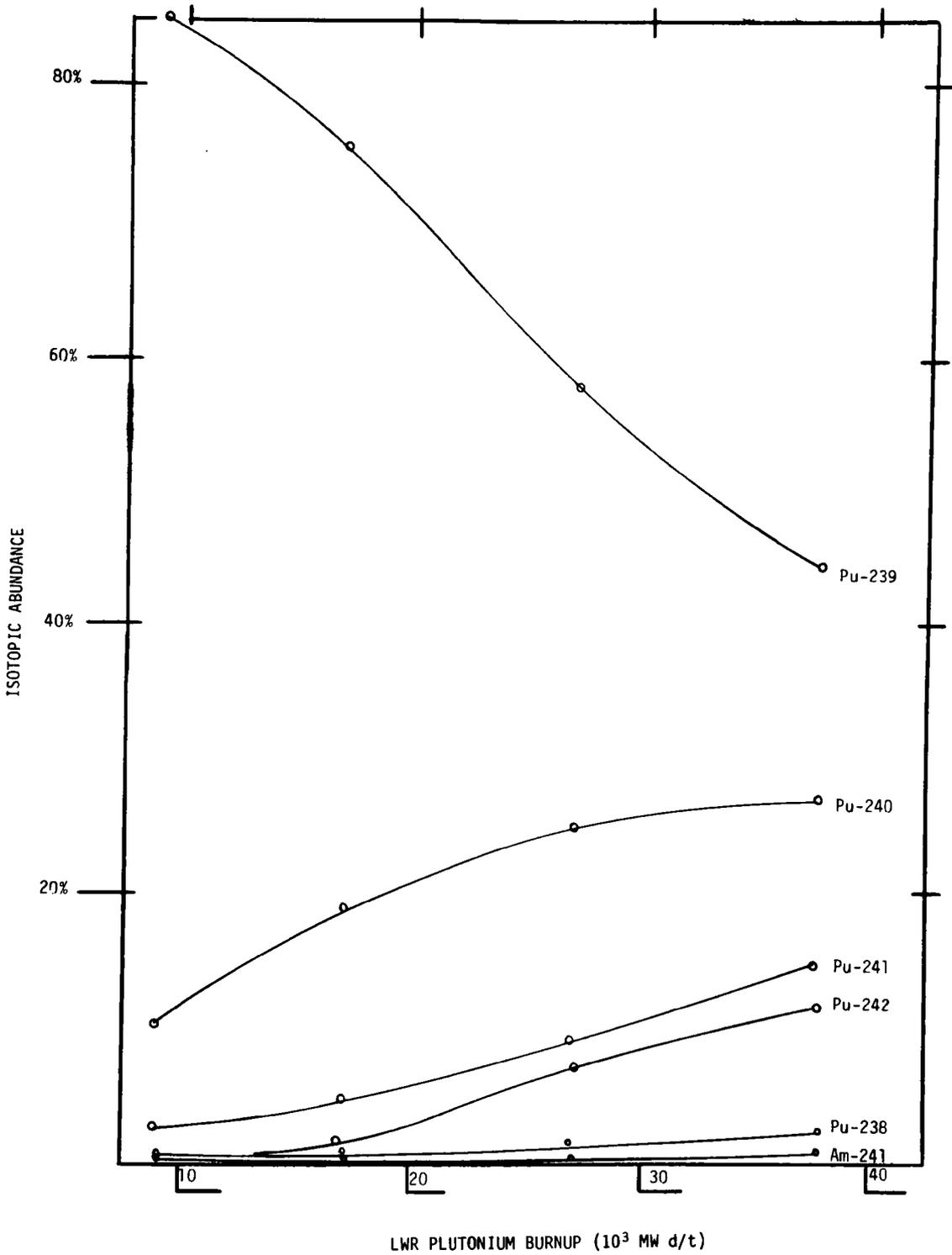


Fig. 1. The isotopic abundances of a mixed-oxide light-water reactor (MOX-LWR) fuel as a function of irradiation history.

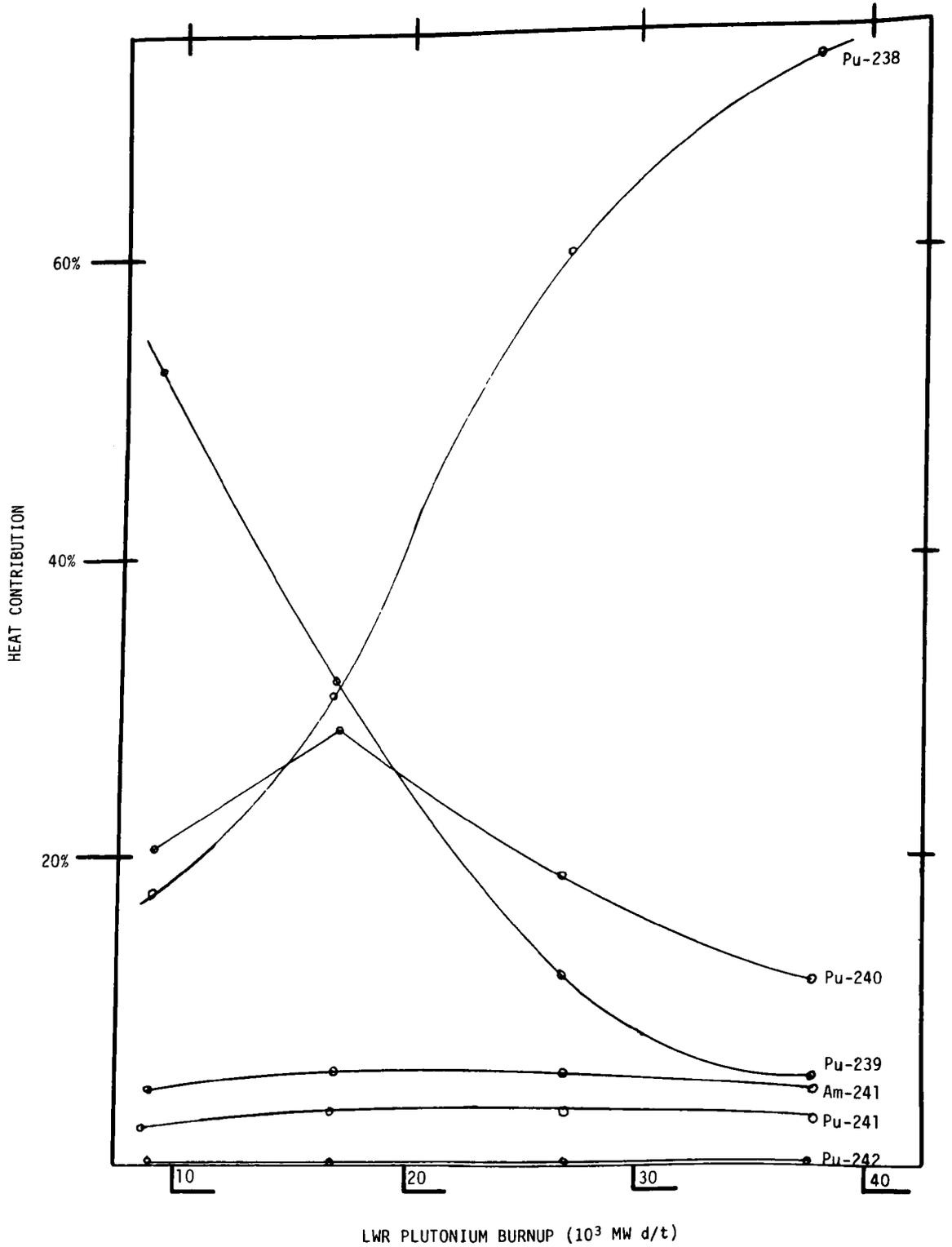


Fig. 2. The heat contribution of the isotopes in MOX-LWR fuel as a function of irradiation history.

tractively by combining a gamma-spectrometric analysis of the sample isotopic with a calorimetric measurement. A 100-min gamma analysis which uses a version of GAMANL, a gamma-stripping program designed for analysis of plutonium,⁸ gives the specific power with a precision of better than 1% for a sample containing 1-2 g of plutonium.

After determining the P_{eff} for the representative material, the mass of other samples may be determined from the relation

$$\bar{M}_S = \bar{P}_S / P_{\text{eff}}$$

The uncertainty in the sample mass is given by

$$(s(\bar{M}_S)/\bar{M})^2 = (s(\bar{P}_S)/\bar{P}_S)^2 + (s(P_{\text{eff}})/P_{\text{eff}})^2$$

C. Air-Chamber Calorimeters

The classical heat-flow calorimeter measures the temperature across a thermal resistance for a plutonium-containing chamber in contact with a large isothermal heat sink. The entire system is allowed to reach a steady-state condition in which the heat absorbed by the water-bath heat sink is equal to the heat produced by the decaying radionuclides. The plutonium-produced power is then proportional to the difference between the sample chamber temperature and the heat-sink temperature. The ANL air-chamber calorimeters are constructed from a series of concentric cylinders which act as a constant-temperature oven. The servo circuits measure the electrical power necessary to maintain this constant temperature. This design eliminates the necessity for a large isothermal heat sink. The temperature-power relations in the air-chamber calorimeter are shown in Fig. 3. At equilibrium an amount of calorimeter-supplied electrical power, P_0 , is necessary to maintain the measurement chamber at some constant temperature, T_3 . If a fuel sample at some temperature less than T_3 is inserted into the measurement chamber, the heaters will apply power to raise the combined sample-sample chamber system to T_3 . The total power necessary to reestablish T_3 is P_0 ; however, the amount of power supplied by the control circuits, P_C , depends

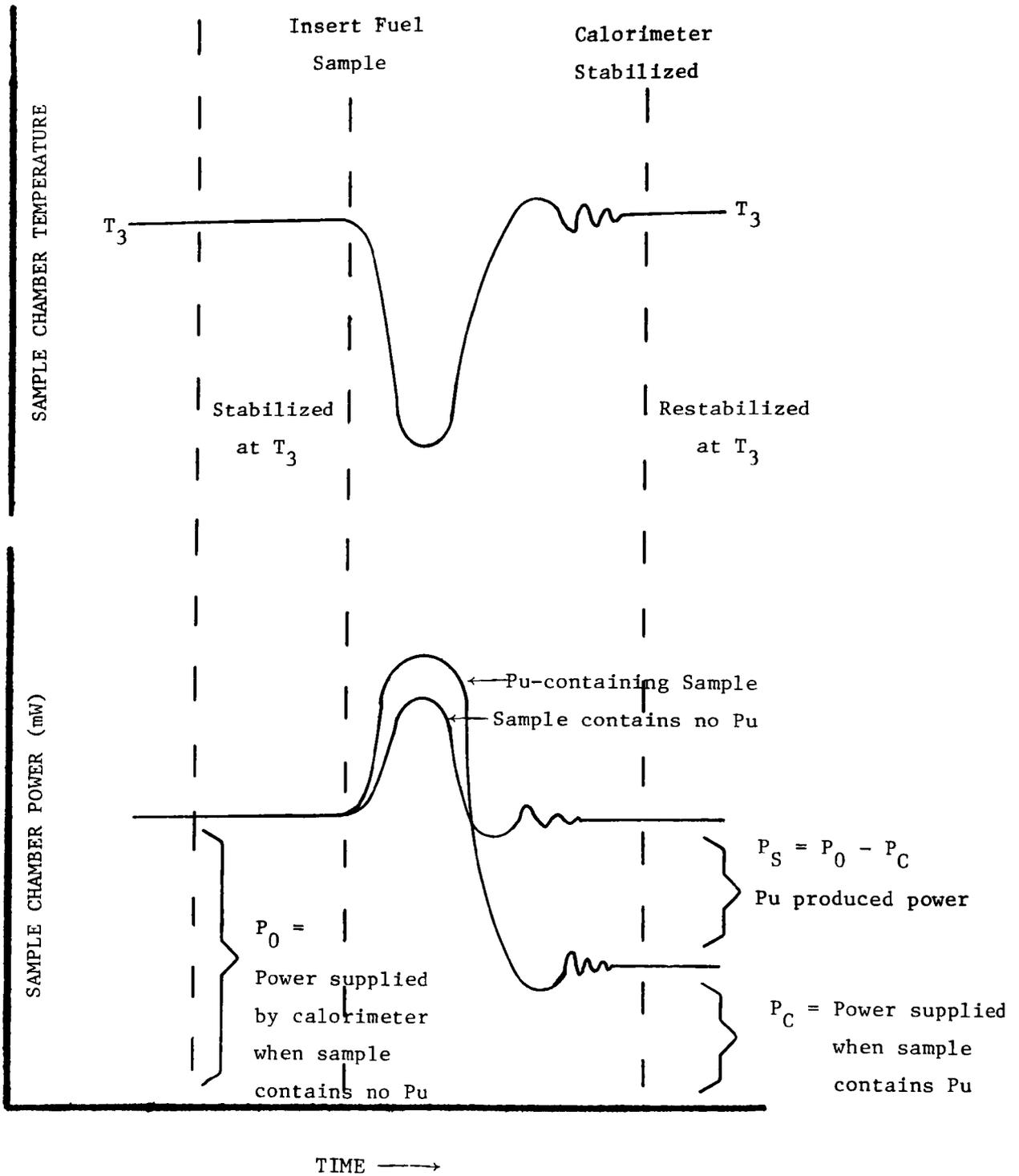


Fig. 3. The measurement-chamber temperature and power relations in ANL air-chamber calorimeters.

upon the sample composition. If the sample is producing heat due to radioactive decay, then P_C will be less than P_0 , and the sample power may be determined by the relation

$$P_S = P_0 - P_C$$

Table III shows the accuracy attainable with this technique. In these experiments, ZPPR fuel rods were assayed by neutron-coincidence, gamma-assay, and calorimetric techniques.^{9,10} The results were compared with a chemical analysis of representative fuel rods. The calorimeter used in these experiments was the ANL Model II, which had an assay time of 20 minutes. The calorimetric results agree with the chemical analysis within experimental uncertainty, while the neutron and gamma assays seem to display a measurement bias. This is a situation where the accuracy of the more rapid assay techniques can be improved by a concurrent calorimetric assay.

D. Small-Sample Calorimeter

The small-sample calorimeter is shown in Fig. 4. The system consists of two instrument packages: a measurement module and a data-acquisition module. The combined weight of both packages is 18 kg. The small-sample calorimeter is capable of measuring samples producing thermal power up to 32 mW. This is equivalent to approximately 10 g (6 cc) of plutonium oxide. This device has a measurement cycle of 20 min with a precision of 0.1%.

The data-acquisition system (DAS) is housed in a 47 cm × 35 cm × 16 cm attaché case and has a weight of 5 kg. It is a totally dedicated microprocessor-controlled device designed around the Intel 8085. The DAS obtains calorimetric power data through a 13-bit ADC. The system memory consists of 8-K bytes of erasable programmable read-only memory (EPROM) and 1-K bytes of random-access memory (RAM). The system program resides in the permanent memory (EPROM) and does not need to be reentered after the device is powered down. The upper 2-K bytes of this nonvolatile memory are located on an EPROM chip residing in the zero-force insertion socket on the face of the unit. The EPROM chip residing in this socket may be exchanged for other chips containing different programs. This permits the user to develop data-handling

Table III. A comparison of an ANL air-chamber isothermal calorimeter with other analytical techniques.

Physical composition: 0.95 cm × 15.25 cm stainless steel clad fuel rods
 Chemical composition: PuO₂, UO₂
 Nominal isotopic composition:

	<u>238Pu</u>	<u>239Pu</u>	<u>240Pu</u>	<u>241Pu</u>	<u>242Pu</u>
Rod type { F, G	0.05	86.55	11.5	1.7	0.2
H	0.09	68.45	25.56	4.53	1.38

<u>Technique</u>	<u>Plutonium wt %</u>		
	<u>F</u>	<u>G</u>	<u>H</u>
Neutron coincidence	13.35 ± 0.06	26.26 ± 0.07	NA
Gamma-ray assay	13.28 ± 0.03	26.42 ± 0.04	NA
Model II Calorimeter	13.20 ± 0.03	26.51 ± 0.03	15.73 ± 0.01
Chemistry	13.19 ± 0.02	26.48 ± 0.04	15.76 ± 0.06

* N. S. Beyer, ANL-7906

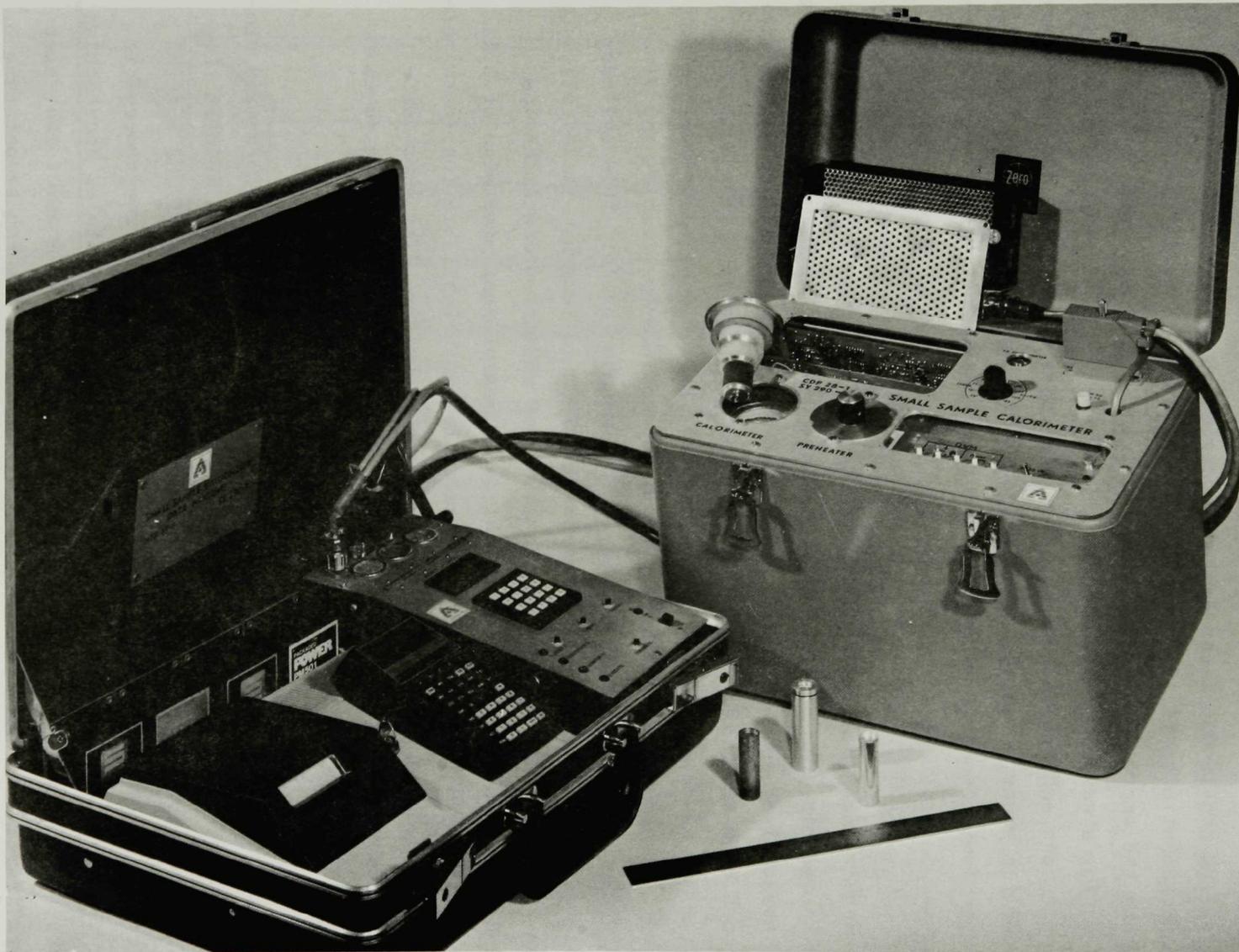


Fig. 4. The ANL small-sample calorimetric system.

and statistical programs tailored to his specific needs.

The software provided with the unit includes a number of data-acquisition and -handling routines. These include codes which calculate the average measurement-chamber electrical power (\bar{P}) and its standard deviation. The DAS will store the empty chamber baseline power for comparison with results obtained during a sample assay. Analysis programs are also included to obtain the sample mass and its uncertainty from the power measurement and the effective specific power. There is also a routine to calculate the P_{eff} from the sample isotopic data, as well as a routine to correct mass-fraction data for changes due to radioactive decay.

A double-encapsulation technique using metal sample containers was chosen to maximize the rate of heat transfer (see Fig. 4). The inner capsule is an inexpensive, commercially available drawn-aluminum cylinder with a diameter of 1.6 cm and a length of 5 cm. It would be sealed inside a glove box after the MOX powder or pellets were inserted and could be disposed of after the assay. The outer sample holder has been machined to minimize the air gap between the inner capsule and the sample-chamber walls. This cylinder has outer dimensions of 2 cm in diameter and 7 cm in length. It is filled with an "O" ring seal to minimize the chance of radiation contamination. By the use of this encapsulation technique, a measurement precision of 0.1% can be obtained in an equilibration time of 15 min. Equilibration tests were also conducted on sources double-bagged in polyethylene. The poor heat-conducting properties of the plastic slowed the equilibration time to about 30 min.

The measurement module contains the calorimeter chamber, the sample pre-heater, and the control circuits. Figure 5 shows the relation between the cylinders comprising the calorimetric unit and the measurement and control circuitry. The small-sample calorimeter is constructed of four concentric shells which are maintained at progressively higher temperatures approaching the center of the unit ($T_{\text{ROOM}} < T_0 < T_1 < T_2 < T_3$). This ensures that heat flow will remain constant in the outward direction across the temperature sensing coils. The outer shells, T_0 and T_1 , act as protective buffers for the inner measurement cylinders. They are controlled by a servo circuit which

BLOCK DIAGRAM OF MAIN CONTROL COMPONENTS

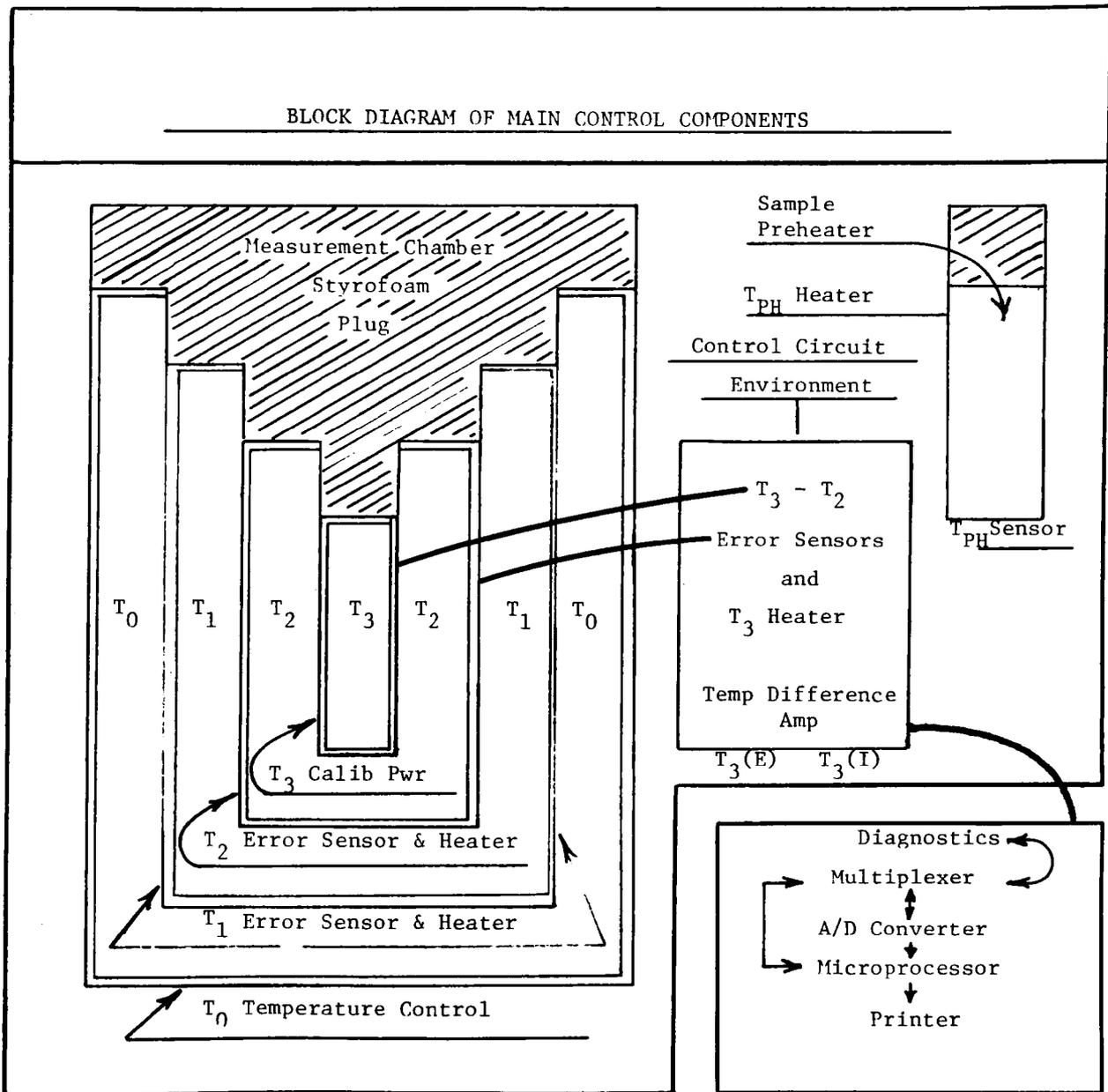


Fig. 5. A block diagram of the small-sample calorimeter with its measurement and control components.

has YSI thermistors as sensors and copper coils wound around the aluminum cylinders as heaters. The temperature difference between the inner measurement cylinders, T_2 and T_3 , must be controlled to the microdegree range. The N_i coils on these cylinders act as both temperature sensors and heaters by using the principles of resistance thermometry.

A simplified diagram presenting the control principles used in these circuits is shown in Fig. 6. The control circuits consist of two operational-amplifier (OP-AMP) networks. AMP•A senses the resistance imbalance between a precision resistor (1 ppm/°C) R_A and the N_i coil, R_3 , wound around the measurement chamber. The resistance of R_3 is dependent upon the temperature

$$R_3 = R_0[1 + \alpha T]$$

where

- R_0 - resistance at 20°C
 α - temperature coefficient of resistance (< 0)

AMP•B acts as a booster which determines the amount of power that will be applied to R_3 . This circuit forms a negative feedback system. If T_3 is too low, R_3 will be less than R_A . Consequently, the gain of AMP•A will be greater than 1.0. This will cause more power to be applied to R_3 , which will raise the temperature in the measurement chamber. The resistance of R_3 will increase until it equals R_A .

E. Data Analysis

The procedures followed during a typical assay are included in the flow-chart shown in Fig. 7. One of the more important features of the calorimetric technique is that the operator may calibrate the instrument using electrical heat standards. This is especially vital for in-field use of the device, since severe restrictions have been placed on the transportation of plutonium calibration sources. The DAS will automatically perform the calibration for a user-selected number of input calibration powers. The micro-processor calculates the proper input reference voltage to be applied across the calibration resistance coil. These powers simulate a set of plutonium

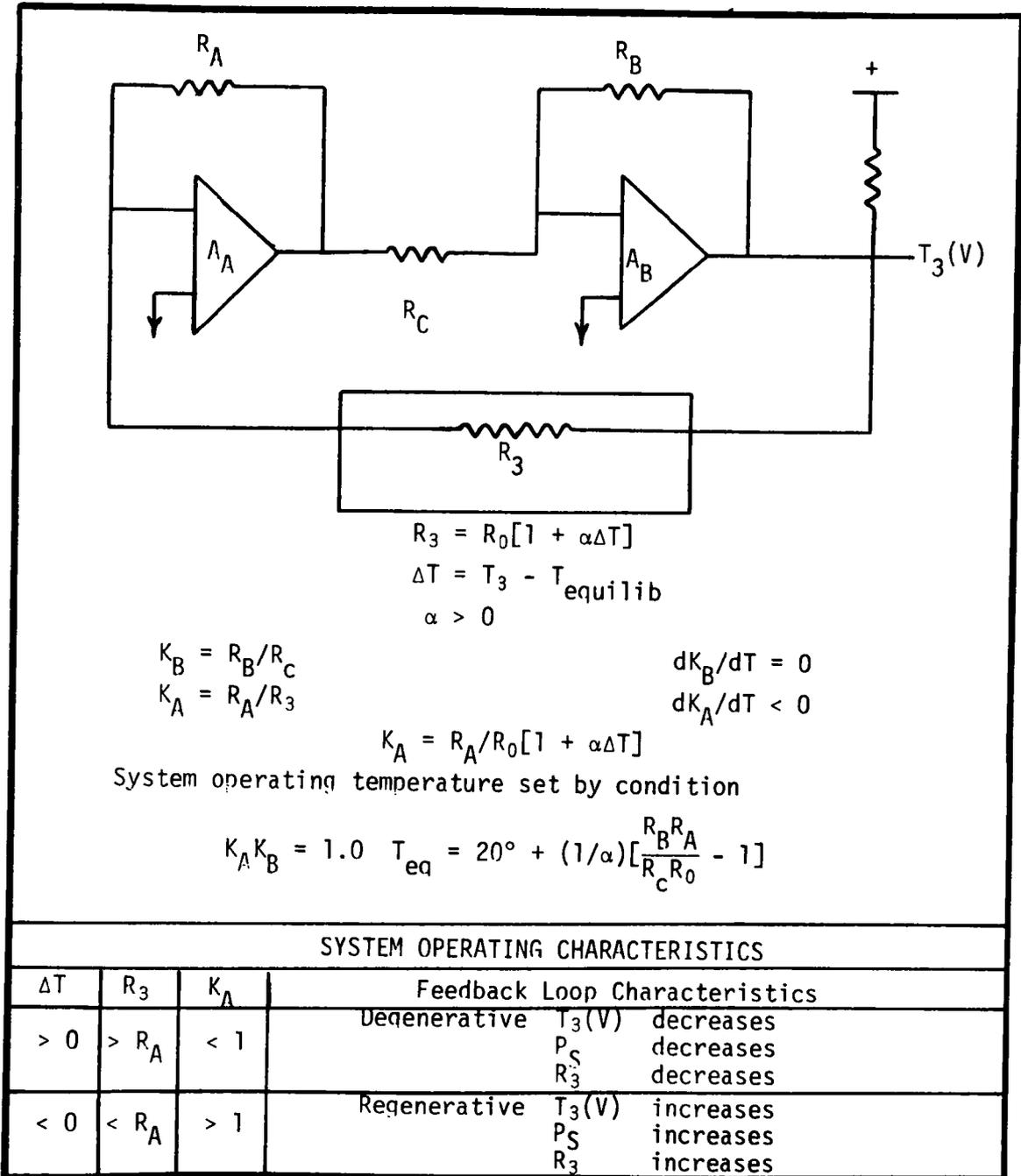


Fig. 6. Resistance thermometry and feedback control circuitry in ANL air-chamber calorimeters.

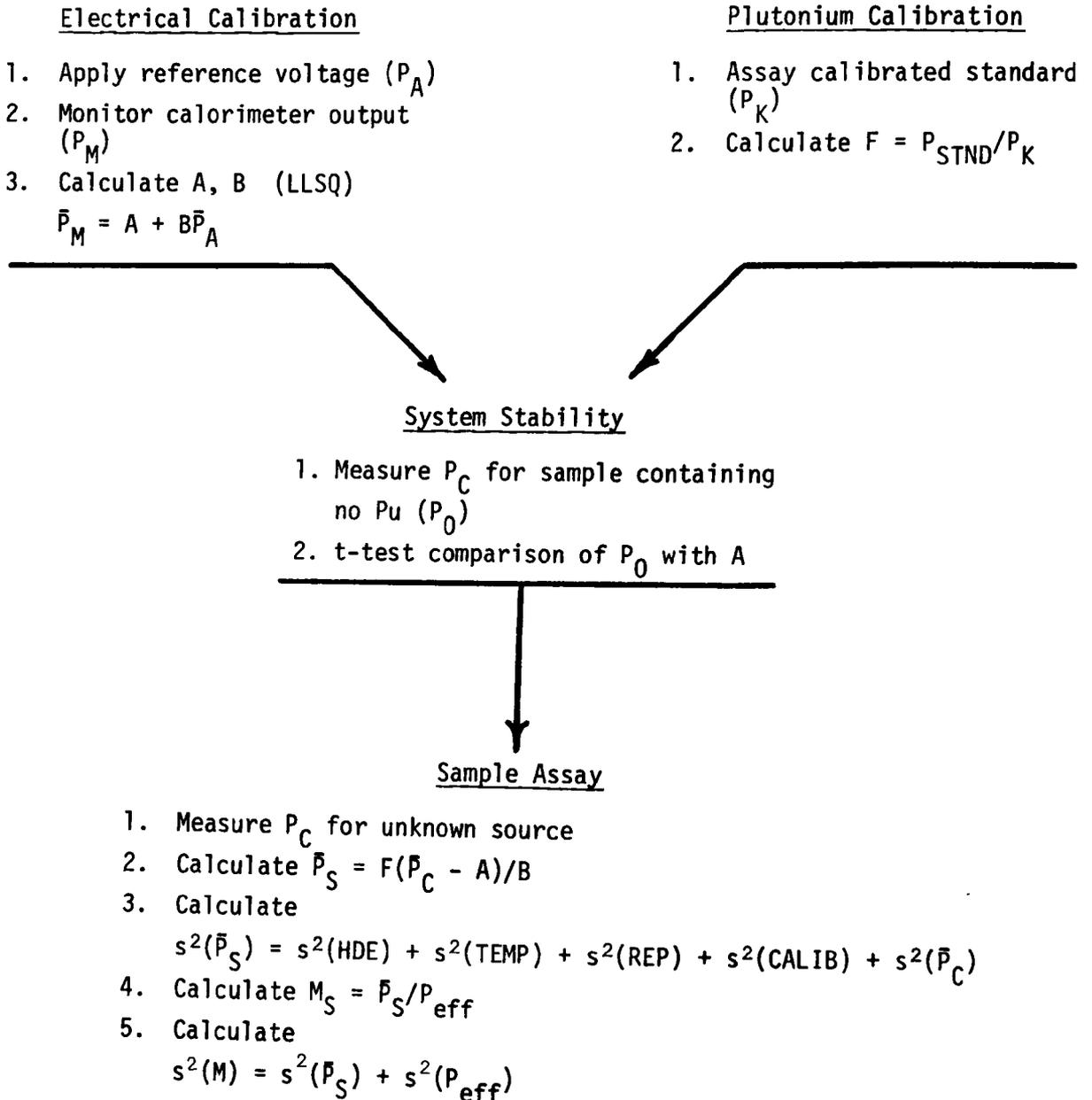


Fig. 7. Data analysis flowchart for the small-sample calorimeter.

standards over the measurement range of the instrument. The system then measures both the input power and the control-circuit power at each point. The results of this calibration are used in a linear least squares analysis to determine the zero-power intercept (A) and the power-measurement slope (B).

An assay is performed with a Pu standard to determine if a measurement bias exists between the electrically produced power and the radioactive-decay produced power. The results are incorporated in the normalization constant F.

If a number of samples are to be assayed, the operator will periodically check the system stability by measuring the control-circuit supplied power for samples which do not contain a heat-producing source (P_0). A t-test comparison may then be performed to assure that there has been no significant electronic drift from the calibration-determined empty-chamber power (A).

The power produced by an unknown source is then determined by the use of the above-mentioned parameters

$$\bar{P}_S = F(\bar{P}_C - A)/B$$

The uncertainty associated with this power measurement includes the following sources of error:

1. $s(\text{HDE})$ - the heat-distribution error contains the uncertainty in the measurement precision due to effects of geometric position of the heat source in the sample chamber.
2. $S(\text{Temp})$ - the ambient-temperature error contains the uncertainty due to fluctuations in the room temperature during the assay.
3. $s(\text{Rep})$ - the sampling-reproducibility error is determined by repeated measurements of the power from a single source.
4. $s(\text{Calif})$ - the calibration error includes the uncertainties in the electrical and radioactive calibrations.
5. $s(\bar{P}_C)$ - the statistical uncertainty is defined as the standard deviation of the power measurement $\div \sqrt{n}$.

The unknown plutonium content is then obtained by

$$M_S = \bar{P}_S / P_{\text{eff}}$$

and the power-measurement uncertainty is combined with the error in P_{eff} to give the uncertainty in M_S .

F. Results

The results of an experiment in which a set of plutonium-containing samples were assayed by the small-sample calorimeter are given in Table IV. These samples were constructed by placing encapsulated metal sources in sand to simulate the heat-conduction properties of PuO_2 powders. The sources were made from a PuAl alloy with a 98.79%-Pu composition. The isotopic composition of the sources was determined by a 30-min gamma assay which used a version of GAMANL specifically adapted for Pu analysis.⁷ P_{eff} was calculated by the computational method discussed in Section B. The sample power was determined in a 4-min measurement following a 15-min equilibration period. The sample Pu mass agrees well with the reported book value over the range of sample powers tested ($\sim 50\%$ of full scale). A mass-determination uncertainty better than 1% was obtained in almost all cases, with the major part of this error resulting from the determination of P_{eff} . Various device parameters, as well as the magnitude of the error contributions, are discussed in Fig. 8.

Table IV. An analysis of ZPR-3 plutonium samples with the ANL small-sample calorimeter.

Physical form	-	1.6 cc stainless steel encapsulated sources
Chemical composition	-	98.79% Pu, 1.17% Al
Typical isotopic composition	-	$^{238}\text{Pu} = 0.01\%$, $^{239}\text{Pu} = 95.2\%$, $^{240}\text{Pu} = 4.5\%$ $^{241}\text{Pu} = 0.2\%$, $^{242}\text{Pu} < 0.2\%$, $^{241}\text{Am} = 0.2\%$

Sample #	P_{eff} (mW/G) ¹	Sample Power (mW) ²	Sample Mass (g) ³	Reported Book Value
1	2.48 \pm 0.01	4.261 \pm 0.005	1.72 \pm 0.02	1.72
2	2.49 \pm 0.01	3.733 \pm 0.004	1.50 \pm 0.01	1.50
3	2.48 \pm 0.01	4.409 \pm 0.005	1.78 \pm 0.01	1.76
4	2.51 \pm 0.02	4.263 \pm 0.005	1.70 \pm 0.02	1.71
5	2.48 \pm 0.02	8.124 \pm 0.008	3.28 \pm 0.02	3.26
6	2.49 \pm 0.02	8.519 \pm 0.009	3.42 \pm 0.03	3.43
7	2.48 \pm 0.02	8.021 \pm 0.008	3.23 \pm 0.02	3.22
8	2.50 \pm 0.02	8.687 \pm 0.009	3.47 \pm 0.03	3.47
9	2.48 \pm 0.02	8.670 \pm 0.009	3.50 \pm 0.03	3.48
10	2.50 \pm 0.02	8.020 \pm 0.008	3.21 \pm 0.02	3.21
11	2.49 \pm 0.03	12.412 \pm 0.012	4.98 \pm 0.03	4.97
12	2.49 \pm 0.03	16.634 \pm 0.017	6.68 \pm 0.03	6.69

¹ Determined by gamma-assay

uncertainty contributions - counting statistics, ^{242}Pu bias (< 0.2%)

² Determined by 20 min calorimetric analysis

uncertainty contributions - counting statistics, sample heat distrib. (0.04%)

system temperature stability (0.09%/°C)

system reproducibility (0.02%)

³ Includes 0.1% uncertainty in radioactive standard calibration

I. Physical Description

- A. Data acquisition module - consisting of 8085 Microprocessor, 8K-byte nonvolatile memory, printer, and keyboard
 - weight - 5 kg
 - size - 47 cm × 35 cm × 16 cm
- B. Measurement module - consisting of calorimeter, sample preheater, and power supplies
 - weight - 13 kg
 - size - 30 cm × 41 cm × 26 cm

II. Sample Size - up to 10 g (6 cc) of plutonium oxide

III. Environmental Parameters

- A. Line power: 110 VAC (60 Hz) or 220 VAC (50 Hz)
 - Sensitivity to line noise - stable with ± 8 V spikes
 - Sensitivity to voltage fluctuations - stable at 110 V $\pm 20\%$
- B. Room temperature operating range - 10.5°C-35.0°C
 - Temperature drift < 0.09%/C°

IV. System Equilibration Time (Pre-heated Sample)

- A. Metal encapsulation - 15 min
- B. Double polyethylene bag encapsulation - 30 min

V. System Power Measurement Precision

- A. Sample heat distribution uncertainty - 0.04%
- B. Sampling reproducibility uncertainty - 0.02%
- C. System temperature stability - 0.09%/C°
- D. Typical statistical uncertainty
(4 min assay) - 0.01%

Combined precision: $s(P_0)/P_0 = 0.1\%$

Fig. 8. Data sheet for the ANL small-sample calorimeter.

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