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PLUTONIUM ASSAY CALORIMETERS

by

R. B. Perry

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The work on calorimeters that we originally started under the NRC program was not completed because of the decision by the government to delay the introduction of plutonium into the commercial fuel cycle. That work has now developed into a task for the International Safeguards Project Office involving three instruments for the IAEA, because the rest of the world is still contemplating recycle and breeders, so I can discuss the progress that has been made since the NRC program was terminated.

Air-chamber calorimetry is the name that we have coined for these instruments, but some of the larger calorimeters have other material than air between the cylinders. The idea behind the calorimeter design at Argonne was to try to eliminate the large heat sinks, thus making the calorimeters more portable, and to design them so that they were more applicable to long rods. That was what we initially had started out to do.

We are developing three instruments for the IAEA which are similar in concept to those we were developing for the NRC. One is a small-sample calorimeter for making measurements on a few grams of plutonium oxide, mixed oxide, or pellets. The design parameters are shown in Table I. The idea was to have a very small instrument that could be hand-carried by inspectors in the field. The second instrument is a calorimeter for up to 2 kg of plutonium in cans and is capable of measuring up to about 25 watts. And the third instrument is for four-meter long light water reactor plutonium-recycle rods.

One small-sample calorimeter was completed and has been delivered to the IAEA. A similar instrument was completed for the NRC Office of Inspection

TABLE I. Design Parameters for ANL-Built Calorimeters

A. Small-Sample Calorimeter
   1. Physical Description: two (2) packages weighing $\leq$ 10 kg each
      a. Data Acquisition Module consisting of a microprocessor, printer, and system power supplies
      b. Measurement Module consisting of calorimeter and preheater
   2. Sample Size: up to 10-g (6-cc) plutonium oxide
   3. Measurement Precision: better than 0.5%
   4. Measurement Time: less than 20 min

B. Bulk Calorimeter
   1. Physical Description
      a. Sample measurement chamber in a 20-L drum
      b. Sample chamber
      c. Data Acquisition consisting of a microprocessor and printer
      d. Power supplies
   2. Sample Size: up to 2-kg Pu
   3. Measurement Precision: better than 0.5%
   4. Measurement Time: less than 60 min

C. Four-Meter Calorimeter (for LWR Pu-Recycle Fuel Rods)
   1. Physical Description
      a. Two demountable segments, each two meters long
      b. Data Acquisition Module consisting of a microprocessor and printer
      c. Two AEC bins containing temperature-control circuits and power supplies
   2. Sample Size: Fuel column up to 4.25 m (500 g Pu)
   3. Measurement Precision: better than 0.1%
   4. Measurement Time: less than 30 min

ANL SMALL-SAMPLE CALORIMETER

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 ± 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 polyethelyne 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: $s(P_0)/P_0 = 0.1\%$

TABLE II

and Enforcement for Region I. The data-acquisition module consists of an 8085 microprocessor. This is essentially an 8K computer in a suitcase and weighs about 5 kilograms. The measurement module weighs about 13 kilograms and is about the size of a woman's makeup case. Sample capacity is about 10 grams of plutonium oxide or 6 cm^3 of mixed oxide, and the measurement range is about 25 milliwatts total power. Other design parameters were to make it as insensitive as possible to line-voltage fluctuations and noise, and to operate in a temperature range of 10°C to 35°C. The temperature drift is less than 0.1% per °C. Measurement time is less than 20 minutes for preheated samples.

The performance capability of the IAEA instrument is given in Table II. Figure 1 briefly describes the principals of operation. The calorimeter is a constant-temperature instrument. When a sample is inserted, it disturbs the system which will then settle back, but the power now required to heat the instrument will be less because of the power supplied by the plutonium. We then take the difference between the power measurements to get the sample power.

Figure 2 shows the NRC instrument. The package on the right is the calorimeter; the attache case contains the control and readout system. In this instrument we used a microprocessor coupled to a Texas Instruments SR52 calculator and printer.

A similar instrument with improved performance was built for the IAEA and is shown in Fig. 3. For this instrument we went completely to a microprocessor system, eliminating the calculator. The system is much easier to operate because each of the subroutines in the program is initiated by pressing a single key that is labeled appropriately, and the calorimeter is somewhat modified to improve its temperature stability.

Figure 4 is a blowup of the inside of the instrument and shows two chambers: one for the calorimeter, and the other for the preheater. The sample is transferred from the preheater to the calorimeter, and another sample can be preheated while measurements are being made on the one that is in the calorimeter.

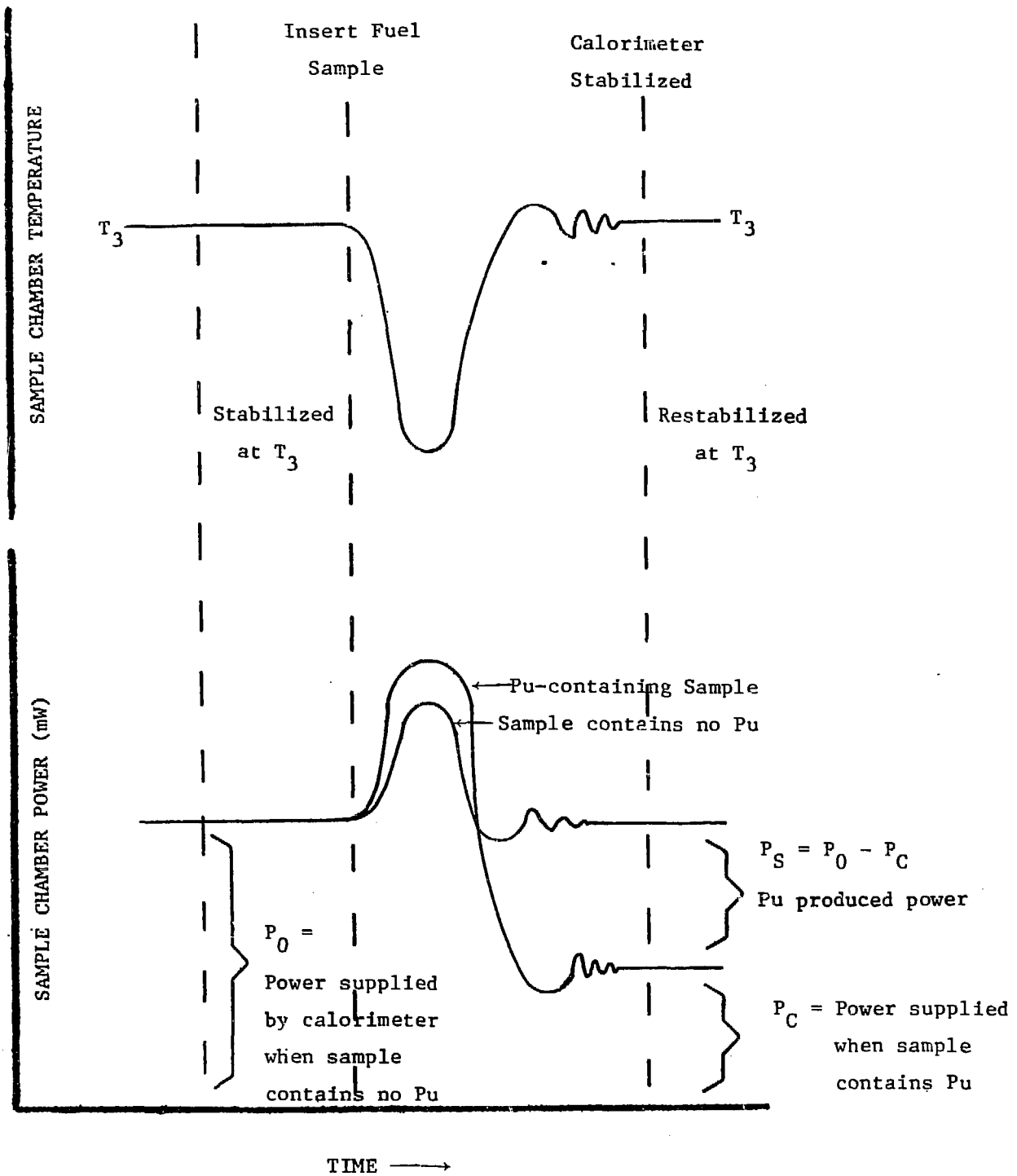

Insert Fuel Sample
Calorimeter Stabilized
SAMPLE CHAMBER TEMPERATURE
T_3
T_3
Stabilized at T_3
Restabilized at T_3
SAMPLE CHAMBER POWER (mW)
Pu-containing Sample
Sample contains no Pu
$P_S = P_0 - P_C$
Pu produced power
P_0 = Power supplied by calorimeter when sample contains no Pu
P_C = Power supplied when sample contains Pu
TIME

FIG 1

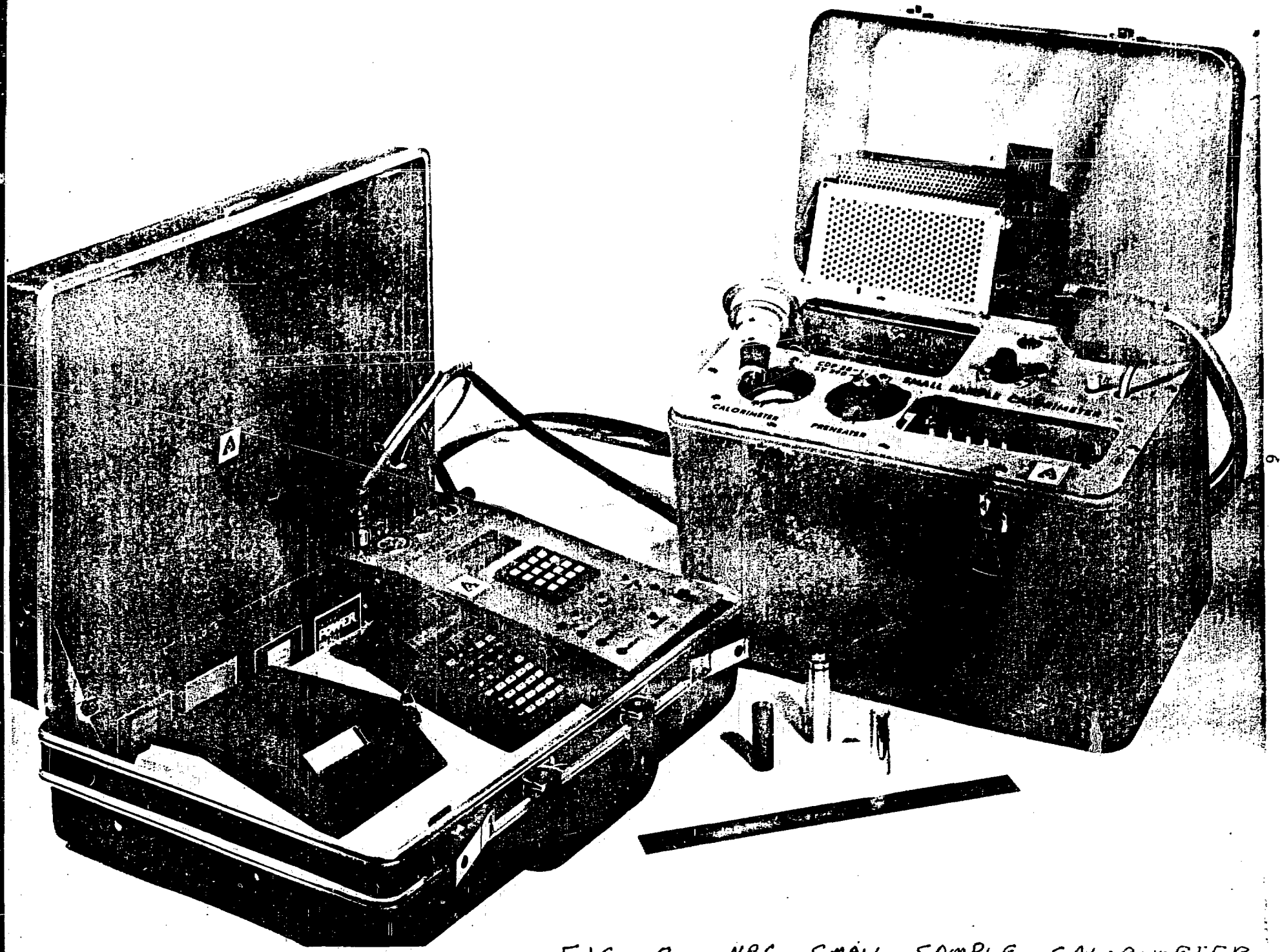


FIG. 2 NRC SMALL SAMPLE CALORIMETER

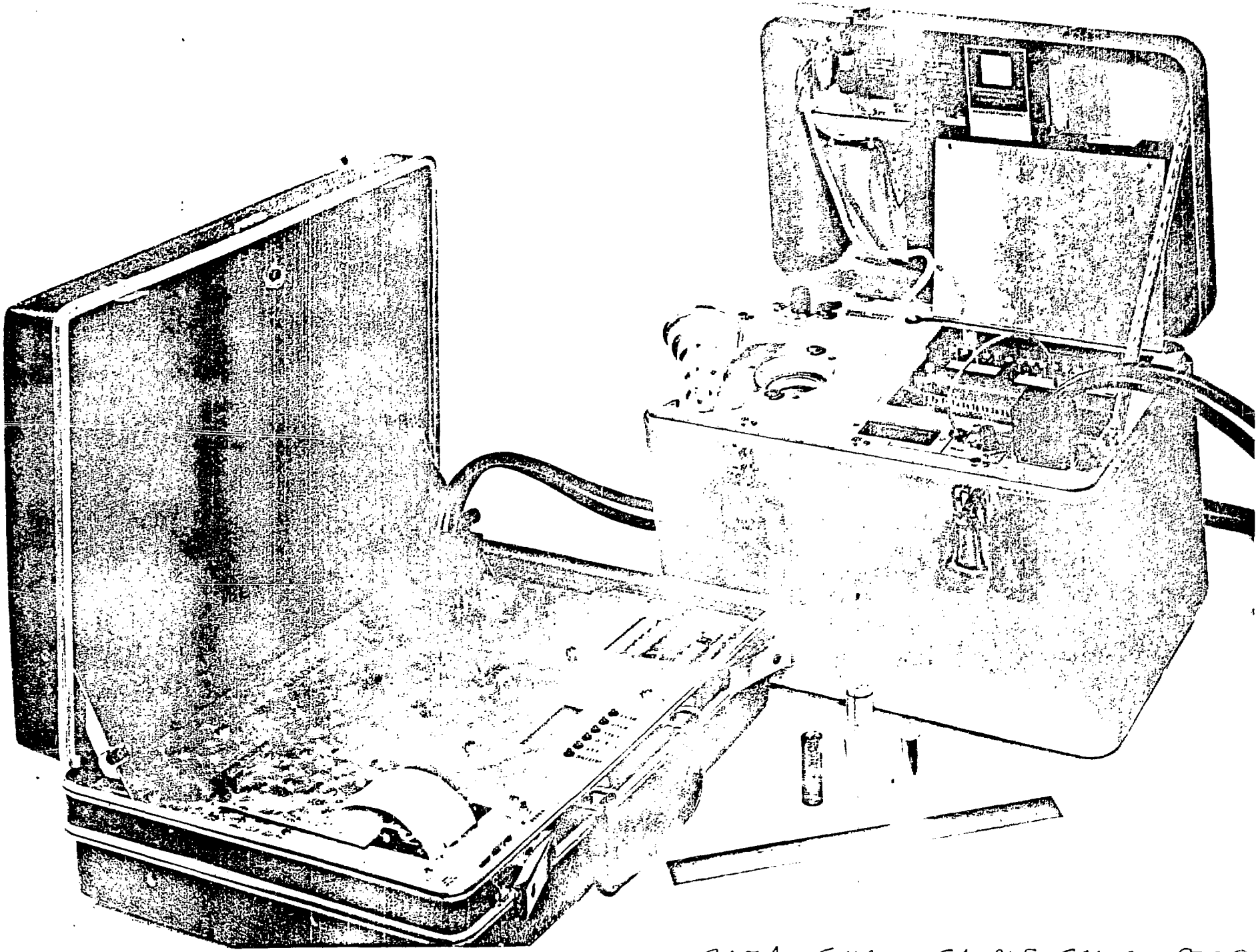
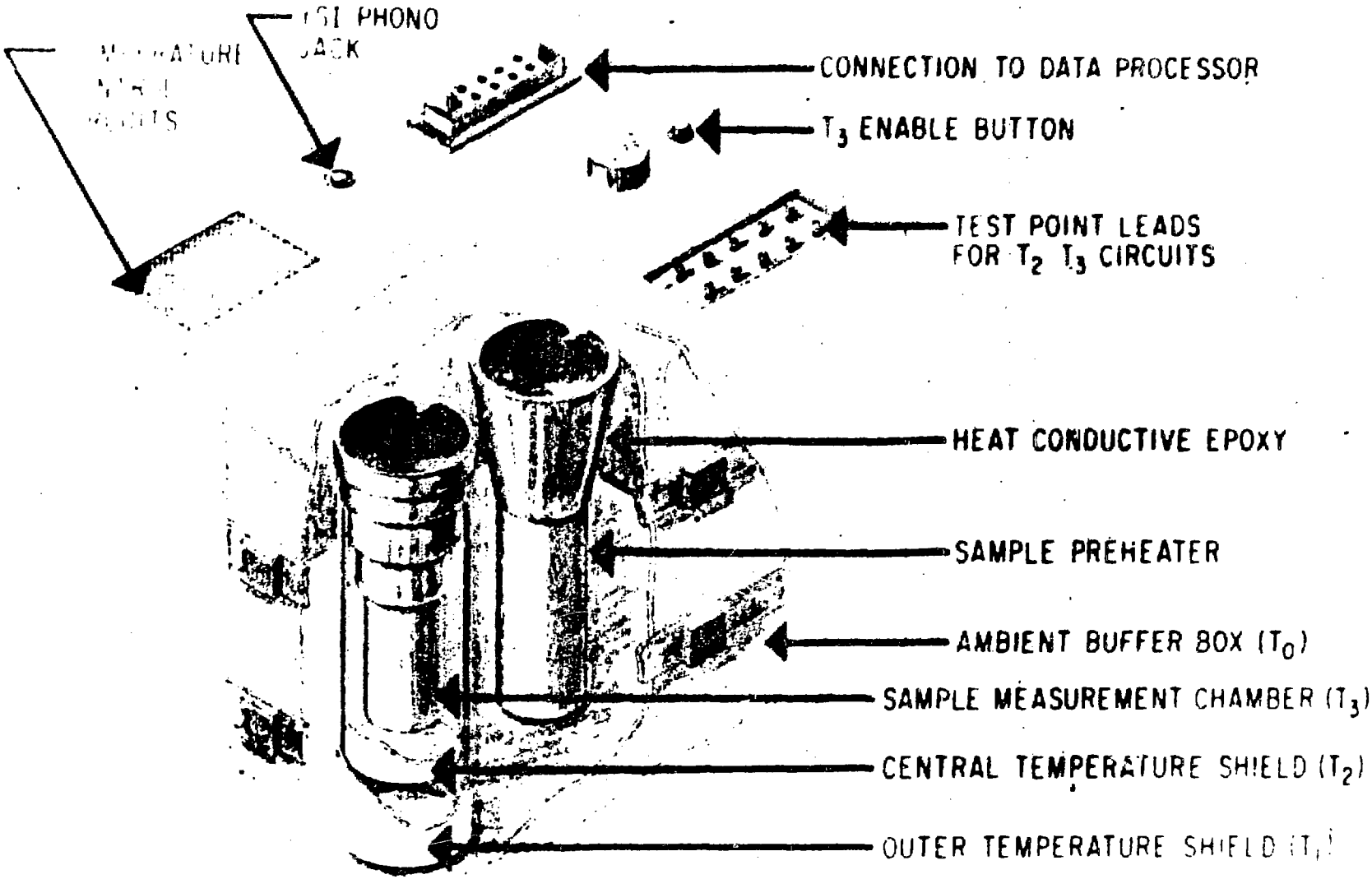


FIG. 3 IAEA SMALL SAMPLE CALORIMETER



Internal Construction of ANL Small Sample Calorimeter

FIG 3 4

A flowchart of the program that is in the data-acquisition system is shown in Fig. 5. The program provides automatic electrical calibration. The inspector can initiate the calibration routine before lunch, and the calibration data will be all printed out when he returns. The program also allows him to put in isotopic data. The instrument will then perform the correction for decay and give him the current isotopic and the specific power. After the system has come to equilibrium and is stable, it measures the power of the unknown source and calculates the plutonium content from the specific power that has been calculated from the input data.

The conceptual design for the Bulk-Sample Calorimeter for NRC, shown in Fig. 6, was completed, but the construction was never started. The instrument was then built with funding from the Argonne Safeguards and Security Improvement Project. The electronics and readout package has been miniaturized for the IAEA instrument. The control package is just slightly larger than one NIM-BIN, and the data-acquisition system is identical to the one used for the Small-Sample Calorimeter.

The uncompleted four-meter instrument that was started for NRC is shown in Fig. 7. The calorimeter portion of it was completed, but the electronics package was never completed. It could not be adapted for the IAEA because of its length—16 feet or so, with a preheater of equal length—and because it was not intended to be a transportable instrument. The one we are building for the Agency is being made in two halves that can be coupled together, and it will have an instrument package and readout system similar to that in the bulk calorimeter.

Table III gives an example of some data obtained from the small-sample calorimeter that was supplied to the IAEA. This data was arrived at by doing a gamma-ray isotopic analysis of the sources and using that isotopic value to calculate the specific power. Then the calorimeter power measurement was used to get the total plutonium. The uncertainties, therefore, include the gamma-ray assay and the power measurement of the calorimeter. As you can see, we have very good agreement with the book values. The samples that are used in the zero-power reactors are called coin sources. They are about the size of a dime and are plutonium foils that are sealed up in stainless steel cans. By putting several of the coin sources in a can, we can get larger samples.

DATA ANALYSIS FLOWCHART

Electrical Calibration

1. Apply reference voltage (P_A)
2. Monitor calorimeter output (P_M)
3. Calculate A, B (LLSQ)

   $\bar{P}_M = A + B\bar{P}_A$

Plutonium Calibration

1. Assay calibrated standard (P_K)
2. Calculate $F = P_{STND}/P_K$

System Stability

1. Measure P_C for sample containing no Pu (P_0)
2. t-test comparison of P_0 with A

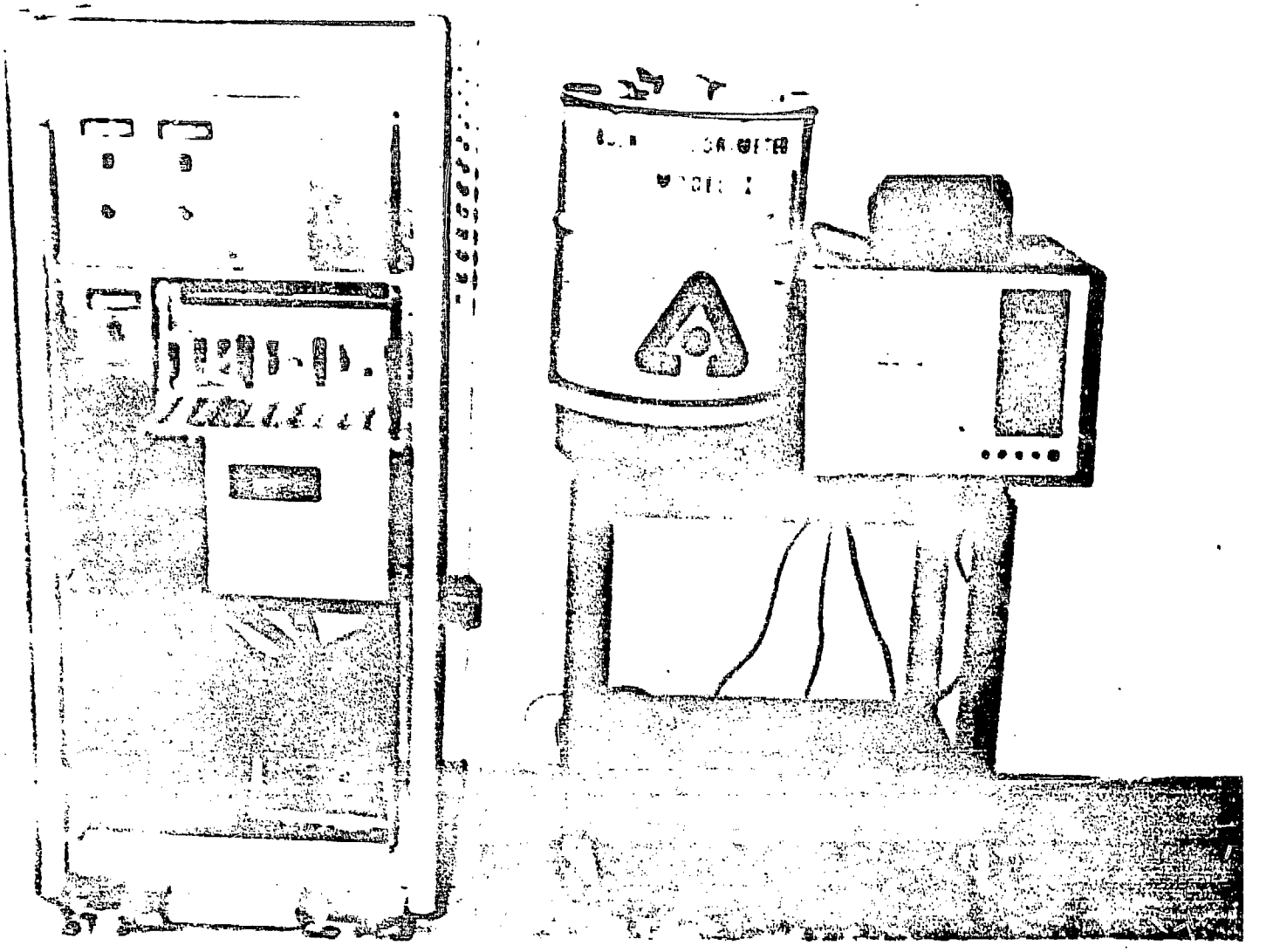
Sample Assay

1. Measure P_C for unknown source
2. Calculate $\bar{P}_S = F(\bar{P}_C - A)/B$
3. Calculate

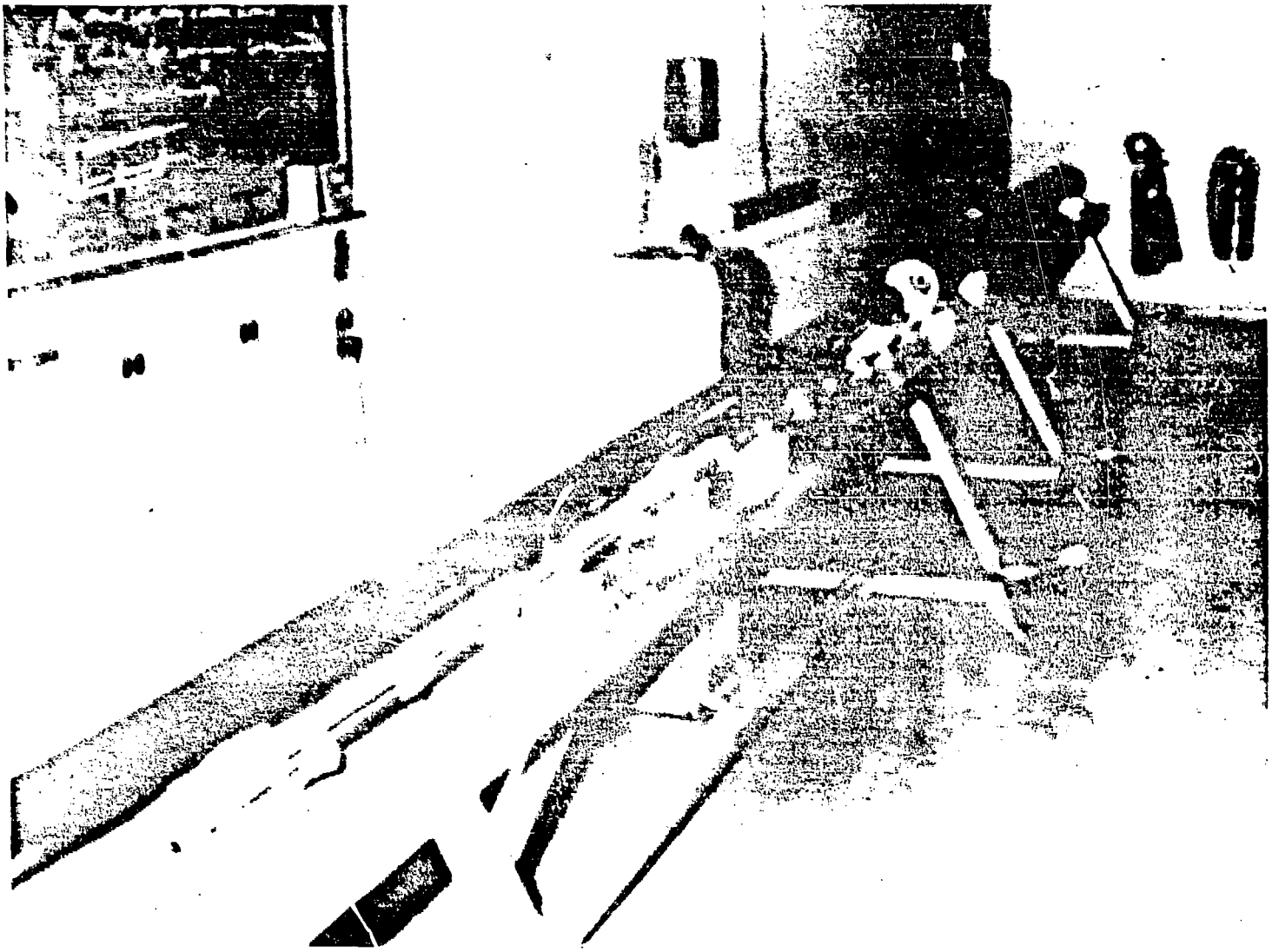
   $s^2(\bar{P}_S) = s^2(HDE) + s^2(TEMP) + s^2(REP) + s^2(CALIB) + s^2(\bar{P}_C)$
4. Calculate $M_S = \bar{P}_S/P_{eff}$
5. Calculate

   $s\ (M) = s\ (\bar{P}_S) + s\ (P_{eff})$

FIG 5



2. The Device under Construction for IAEA Will Have an Electronics Package Contained in a Single NIM Bin and Will Use a Data Acquisition Module Similar to That for the Small Sample Calorimeter.



. Calorimeter for Mixed-oxide Fuel Rods up to Four (4) Meters in Length Built by ANL for U. S. Nuclear Regulatory Commission. The foreground of this photograph shows an in-line sample preheater and its inner sensing and heating element. The device under construction for the IAEA consists of two demountable sections with internal preheating capability.

CALORIMETRIC ASSAY OF ZPR-3 PLUTONIUM

Physical form - 1.6 cc stainless steel encapsulated sources

Chemical composition - 98.79% Pu, 1.17% Al

Typical isotopic composition - ^{238}Pu = 0.01%, ^{239}Pu = 95.2%, ^{240}Pu = 4.5%
^{241}Pu = 0.2%, ^{242}Pu < 0.2%, ^{241}Am = 0.2%

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

TABLE III

Any questions?

QUESTION: What type of software--are you using a machine language?

ANSWER (Mr. Perry): Yes.

QUESTION: And is it on EPROM or PROM?

ANSWER (Mr. Perry): Yes, it is on EPROMs. They can be erased and reprogrammed, but they don't lose the program when you turn the power off.

QUESTION: Has the IAEA had any experience with the small-sample calorimeter yet?

ANSWER (Mr. Perry): They haven't yet, no. The instrument was just shipped a few months ago. I understand it is in someone's office, and they turn it on every day and play with it. They don't have any plutonium at the Agency. If you are familiar with the Agency, you know that it is in downtown Vienna. So they made up some electrical sources using a battery and resistor, and they are playing with that.

They were talking about taking the calorimeter to Switzerland and trying it with the same little fuel rods that Steve Brumbach took autoradiographs of at the Proteus Reactor. I think they are also planning to take it to Belgium to Belgium Nuclear, but they haven't done anything with it yet.

QUESTION: One of the major problems with the instruments that are transportable is the beating they take. They arrive inoperable, dropped in handling. People just don't realize the problems.

They must be rugged.

ANSWER (Mr. Perry): Well, that certainly was a major consideration. I don't know how successful we have been. We didn't use any drop tests or anything on it.

But I hand-carried the one that was made for NRC to the agency last year,

and they had it on display for a conference. Then it was brought back to the Laboratory by someone else, and it survived the trip. That is probably the best I can tell you right now.

QUESTION (Dr. Shepard): I think I should make a statement since most of those projects were started for NRC and then stopped, and that kind of thing. But there were policy changes during the course of the work at Argonne, and I was sort of saddled with the responsibility of trying to redirect that program, which we were not successful in doing.

But I did understand that Region I was interested in some of those calorimeters at one time. Were you going to take the calorimeters that were produced for NRC and try to modify them for IAEA, or did you start from square one again?

ANSWER (Mr. Perry): No, we started from square one, as you put it, because we have gotten no indication that NRC doesn't want the instrument for Region I. We think that they are going to try to use the instrument. So, naturally, we couldn't give that instrument to the Agency.

The four-meter instrument in the configuration that we were building for NRC would not meet the IAEA requirements because it is too large to be shipped by commercial carrier. So we have to make the thing modular, and were unable to use the NRC instrument.

We have a one-meter instrument that was built for NRC that the IAEA is interested in. We reviewed that instrument carefully, and we do not think that it would meet IAEA requirements. We are about to start on a new instrument that provides them with the capability of measuring fast reactor fuel.

QUESTION: I have one other question. You know, we were supposed to try to get one of those calorimeters in place at New Brunswick.

ANSWER (Mr. Perry): Oh, yes, I forgot to mention that one. There was a sort of laboratory prototype instrument that was completed and delivered to New Brunswick, and I know they have made some attempts to use the instrument. I don't know how far they have gone with it. I know they helped us a little bit by encapsulating some plutonium for us so that we could use it to demon-

strate the instrument to the IAEA.

QUESTION: Has there been any experience with the NRC instrument in Region I?

ANSWER (Mr. Perry): No, because the delivery of it was delayed initially when NRC agreed to lend it to IAEA for a few months. When it came back, we were having some stability problems with it, and it sort of got put on the back burner because of the IAEA programs which were being pushed.

So now the instrument is ready for delivery. I talked to Walt Martin at the INMM meeting in Cincinnati. They may come up to Argonne and take the instrument, possibly, in September. It is hard for them to coordinate the people they wanted to come because of vacations and other schedules. So they actually haven't taken delivery yet.

QUESTION: Any more questions?

Thank you very much.

LIST OF ILLUSTRATIONS

Figure

1. Principals of operation of Small-Sample Calorimeter

2. NRC Small-Sample Calorimeter

3. IAEA Small-Sample Calorimeter

4. Internal construction of Samll-Sample Calorimeter

5. Data analysis flowchart

6. NRC Bulk-Sample Calorimeter

7. NRC Four-Meter Fuel-Rod Calorimeter

LIST OF TABLES

I. Design Parameters for ANL-Built Calorimeters

II. ANL Small-Sample Calorimeter Performance Capability

III. Calorimetric Assay of ZPR-3 Plutonium