ANL-77-8

NUREG-0182

(Distribution Category NRC-13)

PLUTONIUM CALORIMETRY AND SNM HOLDUP MEASUREMENTS

Progress Report for the Period March 1976–August 1976

ARGONNE NATIONAL LABORATORY

for

U. S. Nuclear Regulatory Commission

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by

S. B. Brumbach, A. M. Finkbeiner,
R. N. Lewis,* and R. B. Perry

Nondestructive Assay Section
Special Materials Division

February 1977

ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

Prepared for the U. S. Nuclear Regulatory Commission
under
U. S. Energy Research and Development Administration Contract W-31-109-ENG-38

*Electronics Division

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PLUTONIUM CALORIMETRY AND SNM HOLDUP MEASUREMENTS

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ABSTRACT

Part I of this report discusses the calorimetric instrumentation developed at Argonne National Laboratory (ANL) for making nondestructive measurements of the plutonium content of fuel rods. Measurements with these instruments are relatively fast (i.e., 15 to 20 minutes) when compared to the several hours usually required with more conventional calorimeters and for this reason are called "fast-response." Most of the discussion concerns the One-Meter and the Four-Meter Fuel-Rod Calorimeters and the Analytical Small-Sample Calorimeter. However, to provide some background and continuity where needed, a small amount of discussion is devoted to the three earlier calorimeters which have been described previously in the literature.

In Part II, a brief review is presented of the literature on plutonium holdup measurements. The use of gamma-ray techniques for holdup measurements is discussed and results are given for the determination of sample thickness using the ratio of intensities of high- and low-energy gamma rays. The measurements cover the plutonium metal thickness range from 0.001 to 0.120 inches. The design of a gamma-ray collimator with 37 parallel holes is also discussed. Neutron-counting experiments using BF_3 proportional counters embedded in two polyethylene slabs are described. This detector configuration is characterized for its sensitivity to sample and background plutonium, counting both coincidence (fission) and total neutrons. In addition, the use of infrared imaging devices to measure small temperature differences is considered for locating large amounts of plutonium holdup and also for performing fast attribute checks for fabricated fuel elements.

PART I. PLUTONIUM CALORIMETRY

I. INTRODUCTION

A. General

Calorimetry has been shown to be a relatively precise method of determining plutonium content in many different types and sizes of fuel rods. Each of the ANL calorimeters possesses a measurement precision that is currently much better than that of other nondestructive assay techniques and destructive chemical analyses. Models I and II are designed to accommodate 6-inch ZPPR rods, and they have a measurement uncertainty of about 0.2% relative. Models III and IV accommodate one-meter fuel columns, and they have a measurement uncertainty of less than 0.1% relative. The four-meter calorimeter is under construction and further efforts are being made to optimize temperature control and data treatment. An analytical calorimeter is being constructed to accommodate small-volume samples of plutonium solids encountered in plutonium recycle fuel fabrication. Investigations into bulk and kinetic calorimetry are also in progress.

B. Nature of a Dry, Fast-Response Calorimetric Measurement

Plutonium assay with these calorimeters is done by making electrical power (i.e., wattage) measurements. Measurements are made of the number of watts that are needed to heat the measurement chamber and keep it at a constant temperature. When the measurement chamber contains a plutonium-loaded fuel rod, less electrical heating is needed to maintain the chamber temperature because some heat is supplied by the radioactive decay of the fuel in the rod. This radioactive heating comes primarily from the plutonium isotopes and from a daughter product, americium-241, and is directly proportional to the total abundance contained in the rod. Any contribution from the uranium portion of a mixed-oxide loaded rod is too small to have measurable significance. Therefore, the difference in the wattage required to heat an empty measurement chamber (a chamber containing a rod with no plutonium in it) and the wattage required when it contains a

plutonium-loaded rod is directly proportional to the amount of plutonium and americium-241 in the fuel rod. To convert this difference measurement from watts to grams of each of the plutonium isotopes and of the americium-241, the isotopic abundance ratios (i.e., weight percent) and the specific power constants (i.e., watts/gram) are used. The specific power constants are reliably known and usually the isotopic abundance ratios have been well characterized for the plutonium blend being calorimetrically assayed. The conversion has been described previously by Beyer, et al.,[1,2] and by Nutter, O'Hara and Rodenburg.[3,4] In some cases, this conversion to grams of material is required. However, it is not necessary if transfers of plutonium fuel rods are made by accepting the agreement between heat output determined by a receiver and that predicted by a shipper. Nor is it necessary if accountability measurements made after receipt are made on this basis. In this situation, wattage measurements would be compared and the excellent precision and accuracy (currently ±0.1% or less for a single measurement) that is possible with these instruments could be applied to the safeguarding of plutonium fuel.

The simplest way of describing how measurements are made with the ANL fast-response fuel-rod calorimeters is to consider the instruments as constant-temperature ovens. They are operated by electronic-feedback control circuits which are capable of very precise maintenance of the temperatures of each chamber and of very precise measurement of the electrical power required to maintain the innermost chamber temperature.

II. EVALUATION OF THE PERFORMANCE CAPABILITIES OF THE ONE-METER FUEL-ROD CALORIMETER

A. Major Attributes of the Model IV Calorimeter

The precision of the measurements that can be made with the Model IV (one-meter) calorimeter is considerably better (i.e., ±0.1-0.2% relative) than can be attained with other nondestructive

assay (NDA) instrumentation. It is also possible to calibrate with a higher degree of dependability and accuracy because electrically heated calibration rods can be used which may be standardized against well-established and highly accurate electrical standards. These desirable attributes, coupled with a nondestructive measurement of reasonable duration, make these calorimeters attractive for safeguards measurements.

Covered in this section of the report are six general areas: (1) the nature of the Model IV calorimetric measurement; (2) design features of the Model IV calorimeter; (3) measurement reliability of previous ANL calorimeters; (4) calibration linearity; (5) performance under simulated field conditions; and (6) data control and precision after stabilization.

B. The Nature of the Model IV Calorimeter

The basic components of the ANL fast-response fuel-rod calorimeters are shown in Fig. 1, which is a photograph of the Model IV calorimeter. The calorimeter measurement box (on the far left) contains the measurement chamber, thermal guards, temperature sensors, and a few circuit boards. Attached to this box is the preheater, which is used to warm fuel rods before measuring them. The control console contains the main electronic feedback control circuits and potentiometers which allow adjustment of the temperature settings. The HP 45 Controller provides a means for precise data control and automatic statistical analysis. The controller comprises a combination of a commercially available portable electronic calculator, a number of electronic logical elements, and a small printer which provides a hard-copy printout of power measurements and calculations. Figure 2 is a closeup photograph of the control console and the 45 Controller.

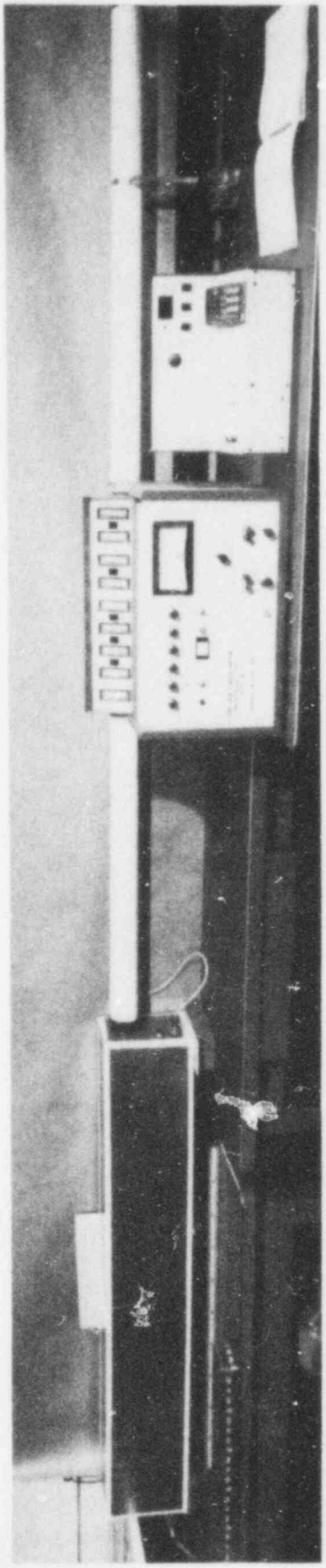

Fig. 1. Model IV Calorimeter

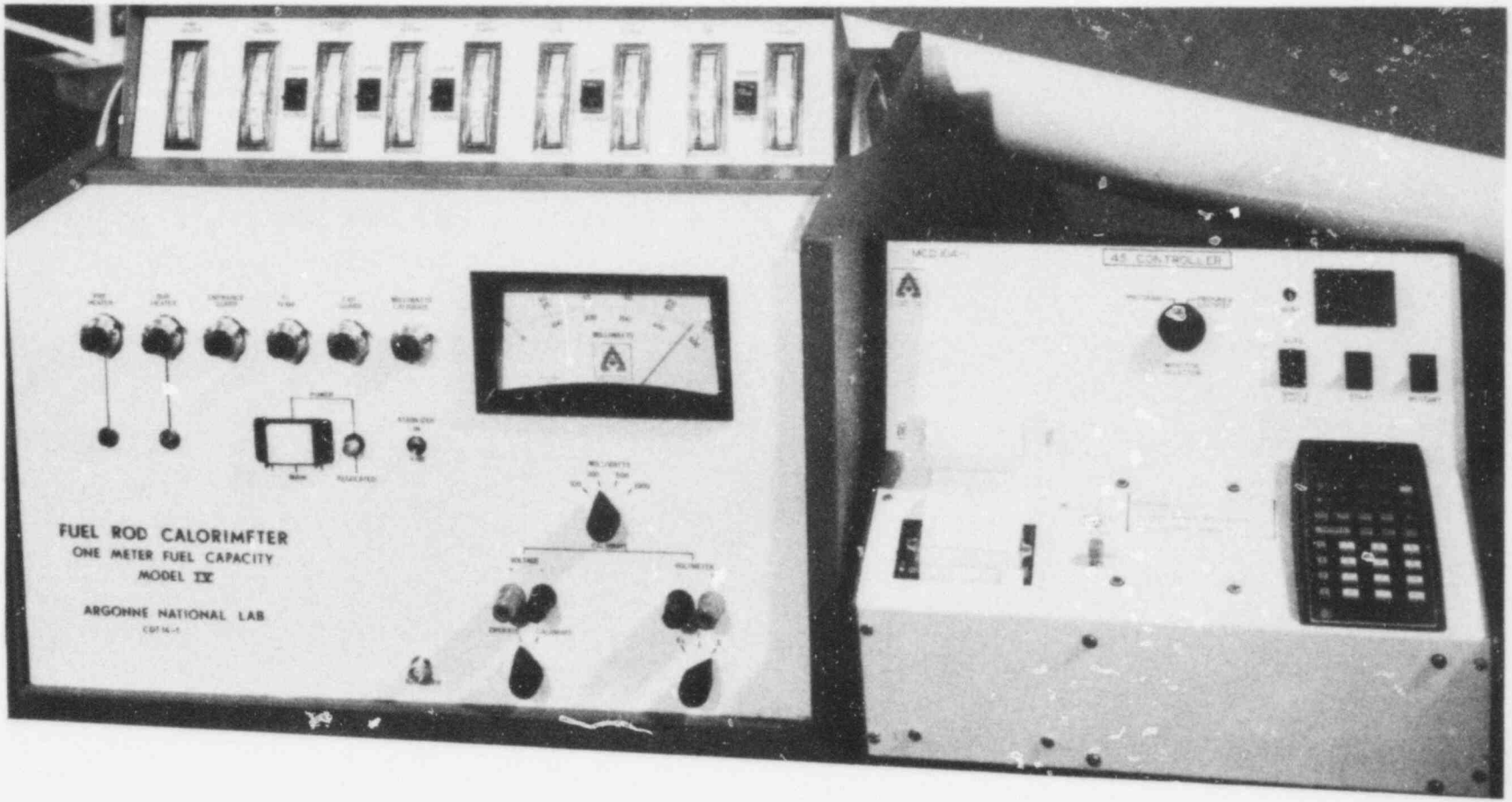

Fig. 2. Main Control Console and Readout/Controller for the Model IV Calorimeter

The steps in making a measurement with these four basic components are as follows: 1) A fuel rod is inserted in the preheater. After the rod heats for 10 minutes, a meter on the control console indicates it is within ±0.0001°C of the temperature of the measurement chamber and is ready to be measured. 2) Another fuel rod is now inserted into the preheater, pushing the preheated rod ahead of it into the measurement chamber. 3) The circuits which control the heating of the measurement chamber automatically start to operate. Within fifteen minutes the measurement chamber temperature has stabilized to within ±20 microdegrees C of the measurement temperature. 4) The HP 45 Readout/Controller automatically senses this temperature stabilization, indicates the electrical power (milliwatts) required to sustain it, converts this to grams of plutonium, and calculates the measurement precision.

C. Design Features of the Model IV Calorimeter

The basic hardware of the ANL instruments consists of concentric cylinders that are wrapped with heater-wire coils and sensor-wire coils. The cylinders are surrounded by air. No water bath is used with the ANL rod calorimeters, called "dry" calorimeters. The earlier models (Models I and II) which are described in Ref. 1, and the latest instrument (Model IV) use three cylinders. Model III, described in Ref. 2, has only two cylinders. The cylinders are housed in a measurement box. For Models III and IV, temperature of the interior of the box is controlled by a box heater. The basic features of the Model IV are represented in Fig. 3, which is a functional block diagram of the calorimeter. The measurement chamber is one meter in length and can be used to measure fuel columns up to approximately 98 centimeters in length.

The measurement chamber box of the Model IV contains six temperature-controlled regions: three concentric and thermally gradated cylinders; entrance and exit guards; and the box itself. All six thermal regions are maintained at temperatures that are a few degrees above ambient temperature. The temperature gradation from T_3 (the highest temperature

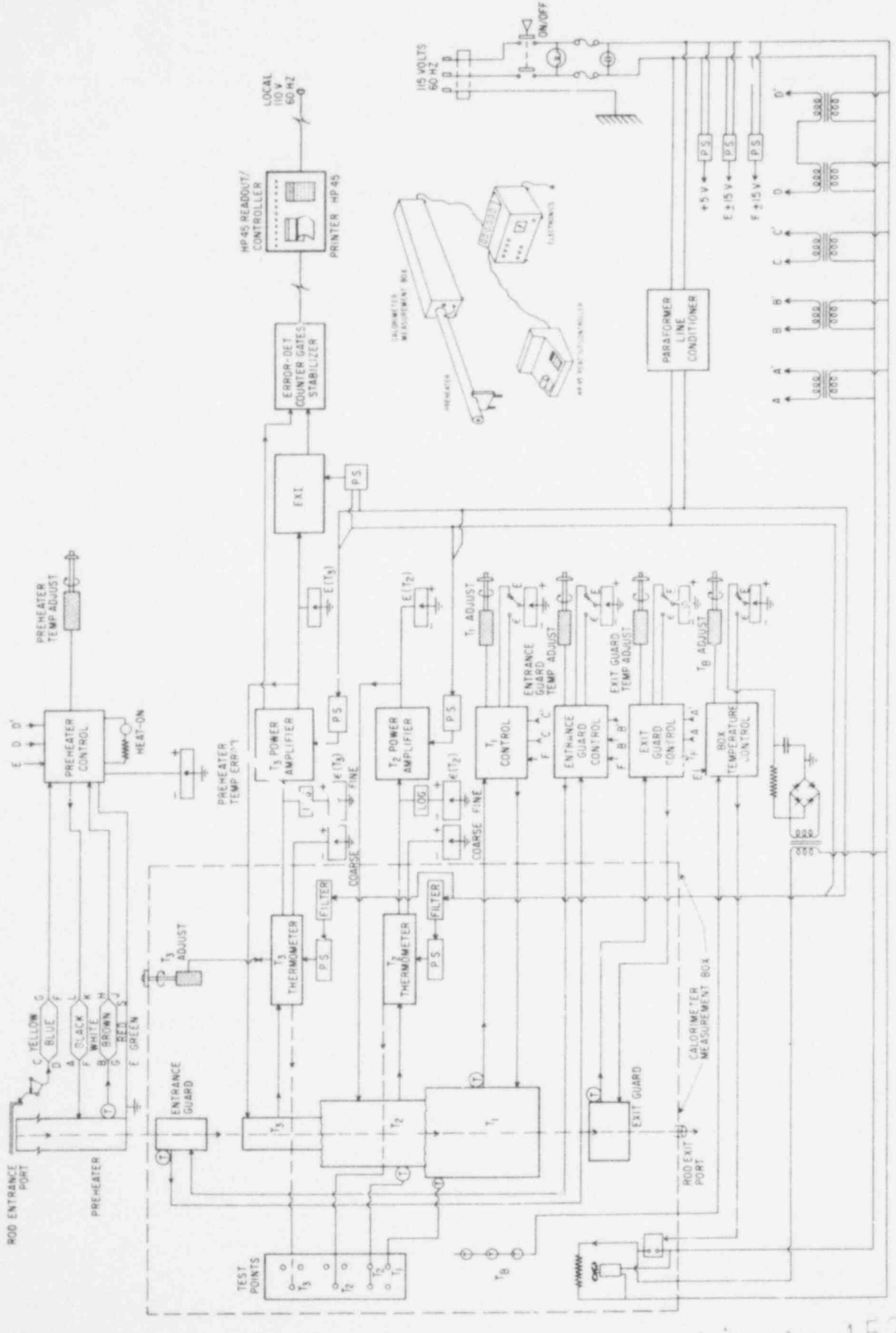

Fig. 3. Model IV Calorimeter Block Diagram

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and that of the innermost cylinder) to T_2 to T_1 to T_{box} to room temperature requires a constant flow of heat outward. The power to the T_3 heater that is required to maintain the temperature difference $\Delta T = T_3 - T_2$ is constant under steady state conditions and determines the maximum sample power that can be measured by the calorimeter. The precision of control varies considerably among the different temperatures, with T_3 and T_2 controlled with the greatest refinement. The measurement chamber temperature, T_3, is maintained to within $\pm$ 20 microdegrees C. The ends of the T_3 cylinder are thermally protected by cylindrical entrance and exit guards of the same diameter and maintained at T_3. The guards help to maintain a zero thermal gradient at the entrance and exit to the measurement chamber. They are required because fuel rods often extend beyond the ends of their fuel column and, in this case, they provide protection against heat losses (or maintain the losses very constant) out of the ends of the measurement chamber. The box also contains circuit boards for the T_2 and T_3 regions and a heating element to create a constant temperature environment.

The function of the preheater is to raise the temperature of the fuel rod to a temperature close to that of T_3 in order to minimize the amount of time required to achieve thermal stability upon insertion of the rod into the measurement chamber.

A microswitch on the entrance door to the preheater causes the control circuit to go to standby/hold when the door is opened. This maintains a steady input power to the heater and prevents a sudden increase in applied power when a cooled rod replaces a preheated rod. Such an increase in power would overheat the rear portion of the preheated rod. For Models III and IV, the preheater is approximately 2.5 meters in length and therefore can be used for fuel rods up to that length.

The main control console houses the meters which monitor the temperature errors of the various temperature regions and the potentiometer knobs for the initial temperature adjustments. After setting appropriate levels for the temperatures of each thermal region, the operator does not need to adjust any controls when making a measurement.

The HP 45 Controller is one of the most interesting improvements that has been made in the ANL Model IV Calorimeter. The Hewlett-Packard 45 is a pocket calculator with callable memory registers and built-in programs for calculating means and standard deviations of input data. It has been interfaced so that keyboard access is electronically available, and printout of the display (X-register) can be accomplished. It is currently programmed to compute: a) the point at which thermal stabilization is sufficient to provide a desired measurement precision (usually 0.1% or less); b) the mean value and standard deviation of the electrical power of the T_3 circuit; and c) the mean value and standard deviation of the grams of plutonium, calculated by using the appropriate specific power constant (milliwatts/gram). The HP 45 may also be used off-line for other computations.

D. Measurement Reliability of Previous ANL Models

This section includes a discussion of our experience in evaluating measurement reliability of the ANL fuel-rod calorimeters. Although experience with Models I, II, and III has been reported previously in the literature,[1,2] a brief review is presented for the sake of providing background continuity.

Models I and II were designed to specifically handle the small (i.e., 6 inches long by 3/8 inch diameter) fuel rods used in the Zero Power Plutonium Reactor (ZPPR). These small, portable instruments were used to verify, on a sampling basis, the plutonium content of the ZPPR fuel rods. The experience gained in using these instruments to measure the ZPPR rods was very useful because it was possible to compare the calorimetric measurement results with other extensive analyses made by both nondestructive and destructive techniques (i.e., gamma-ray counting, neutron coincidence, and chemical measurement). A thorough comparison of results has been presented in the literature,[1,4,5] so only one set of results is presented here. The results were as follows:

MEASUREMENT TECHNIQUE	MEAN VALUE wt% TOTAL PLUTONIUM
Calorimetry	26.51
Chemical (Receiver)	26.48
Chemical (Shipper)	26.60
Gamma Ray	26.42
Neutron Coincidence	26.26

The uncertainty of the mean value for all cases was approximately 0.2% relative, or less. The measurement response time for Model I, which did not have a preheater, was 90 minutes. Model II used preheating and responded in 20 to 30 minutes.

The Model III instrument[2] was built to apply the design concepts of Models I and II to an instrument with a longer measurement chamber and to improve upon the measurement precision and response time. The instrument was able to handle a 92-centimeter fuel column, had a measurement response time of 13 to 23 minutes, and the single measurement precision for thermal power measurements was approximately 0.2% relative. The long-term stability or precision, as indicated by the uncertainty in the mean value of the thermal power, was less than 0.1% relative. Initially, this instrument was evaluated by measuring groups of ZPPR pins placed end-to-end to simulate longer fuel columns. Later it was modified to handle fuel rods for the Fast Flux Test Facility (FFTF) which are about 237 centimeters long, but contain a fuel column of about 91 centimeters. Studies with this modified instrument led to the development and construction of the Model IV calorimeter.

E. Calibration Linearity of the Model IV

The calibration of Models III and IV was done by comparative measurements of: 1) fuel rods which had been well characterized by chemical and mass spectrometric measurements; 2) rods (i.e., ZPPR rods) that had been assayed by the large, high-precision, water-bath calorimeters of Mound Laboratory; and 3) electrically simulated fuel rods built

for this purpose. The fact that it is possible to construct and use electrically simulated fuel rods with the ANL fuel-rod calorimeters is a fortunate circumstance, since the determination of the "real" value of the power supplied by the fuel is, in this case, independent of the uncertainties associated with mass spectrometric and/or chemical analyses which usually form a basis for determining the accuracy of other nondestructive assay instruments. The electrical measurements with an electrical fuel rod can be traced to the highly precise electrical standards available through organizations like the U. S. National Bureau of Standards. Figure 4 shows a recent calibration of the Model IV instrument, made with an electrically simulated FFTF fuel rod. A linear least-squares fit to the measurement data indicated a slope of -0.99085 $\pm$0.00089 and intercept of 274.66164 milliwatts with an uncertainty of $\pm$0.18004 or less than 0.1%. This linearity was shown to be unaffected by changes in room temperature between 21°C and 26°C.

Repetitive measurements made with the Model IV instrument using four FFTF fuel rods revealed that the precision for a single measurement of the thermal power is of the order of 0.1% or less and can be obtained in about 15 minutes. The overall measurement precision (i.e., including uncertainties in the conversion factors, specific power constants, and mass spectrometric analyses) obtained for a determination of the grams of plutonium in FFTF type rods was $\pm$0.2% relative at the one-sigma confidence level. The stability over a 12-hour period was on the order of $\pm$0.02% relative.

A complete discussion of calorimeter measurements and calibration techniques is given in ANSI N15.22, *Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Controls*,[6] and MLM-1798, *Calorimetry for Safeguards Purposes*, by F. A. O'Hara, et al.[4]

F. Performance under Simulated Field Conditions

In an attempt to predict actual use dependability, field conditions were simulated in the laboratory, and the T_3 power dependence on

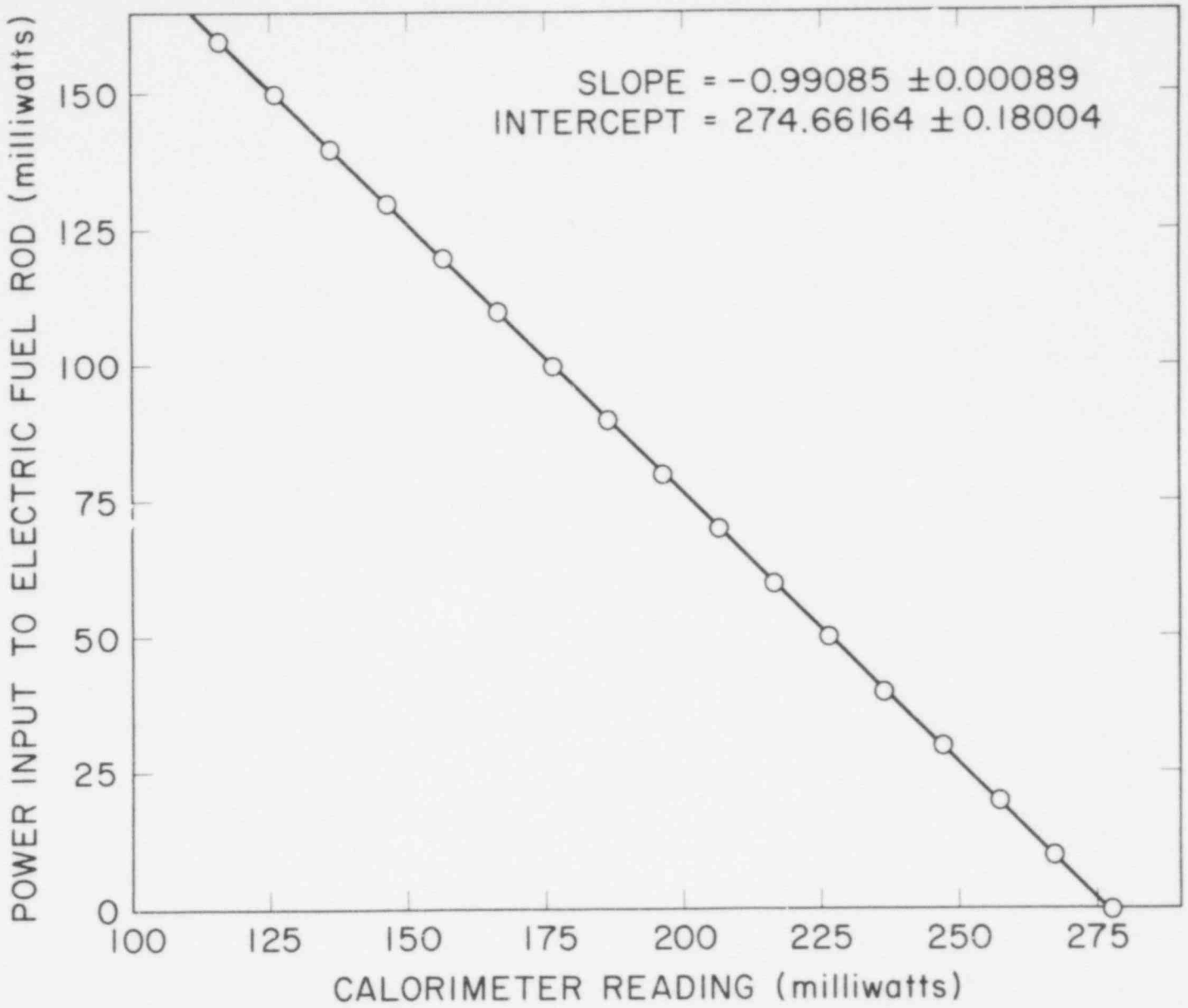


Fig. 4. Calibration of the Model IV Calorimeter

electrical and environmental extremes was examined extensively. These evaluation measurements were made, for the most part, with a dummy rod containing depleted uranium dioxide which simulated the thermal conductivity and geometry of a fuel rod. On the basis of the results, a set of operating conditions has been developed, which ensures maintenance of the usual performance precision of 0.1%. Although these condition requirements are broad, and adherence to them should present no difficulty to the operator, the 45 Controller has been equipped with the means to correct for any drift in the T_3 power which might or might not be caused by violation of these condition requirements.

Because of the inherent properties of electronic elements, the temperature control circuits were found to be minutely affected by a number of environmental and electrical conditions, most notably room temperature. These external conditions cannot be controlled to the precision required to reproduce any given set; that is, the state of the calorimeter at any given moment cannot be reproduced by gross control of ambience. This became especially clear when the T_3 power measurement of a depleted rod (i.e., the zero-power intercept) assumed two different values on two different days of similar conditions. This drift in the zero-power intercept occurred only when the set of conditions was violated (e.g., at night when the temperature dropped). As discussed above, the value to be converted to grams of plutonium is $\bar{x}_{alpha} = \bar{x}_o - \bar{x}$, where $\bar{x}_o$ is the electrical power required by the T_3 circuit to maintain a constant temperature when the chamber contains a dummy rod, and $\bar{x}$ is the electrical power required when the chamber contains a fueled rod. When a drift occurs in $\bar{x}_o$, the same drift occurs in $\bar{x}$, and the difference, $\bar{x}_{alpha}$, remains the same, as would be expected. The data control program is such that $\bar{x}_o$ is keyed into a memory register by the operator. So, periodic updating of $\bar{x}_o$ is all that was necessary to correct for a day-to-day change of $\bar{x}$ for a single rod.

Among the operating requirements detailed in the ANL Model IV Fuel-Rod Calorimeter Operating Manual (internal document, available on request from the authors), are minimum line voltage of 105V, a maximum of 35 V of continuous noise, and a maximum vibration of 450 in/sec^2. The

power measurement is also dependent on the position of the fuel rod along the measuring chamber. The calorimeter was found to perform optimally in a room that has temperature control of +1°C or less, with temperatures within the range of 18°C to 26°C. When the ambient temperature was kept to within a degree of 23°C, the zero-power intercept was uncertain by ±0.20 mW or no more than 0.07%. This implies an uncertainty of ±0.05 gm or 0.13% in the calculation of grams of radioactive material of a typical fuel rod containing 40 gm of plutonium.

G. Data Control and Precision after Stabilization

The data control system of the Model IV Fuel-Rod Calorimeter is designed so that the relative precision of the power measurement remains constant over time, provided that a minimum stabilization time has elapsed after the insertion of a rod. This is desirable for fast measurements because the first measurement of a rod after stabilization of the T_3 circuit is as precise as, say, the tenth measurement of that rod. The two features of the data control program which enable this situation are (1) the use of fifty measurements, rather than five or five hundred, to determine each mean value of power; and (2) the use of a maximum limit of standard deviation to ensure that the T_3 circuit has stabilized to a state that reflects a statistically normal distribution. A third feature will be investigated which would indicate on a 95% confidence level whether or not the power measurement was drifting toward a stabilization value within the 0.1% precision range. Even without this added precision, the mean value of the power measurements is uncertain by only 0.1% relative.

H. Conclusion

In conclusion, the greatest assets of the ANL Model IV Fuel-Rod Calorimeter have been shown to be an improved calorimeter design, operator control of input data (esp., the zero-power intercept), and an improved data control system. More specifically, the accuracy and speed demonstrated by the Model IV Fuel-Rod Calorimeter in the lab need not be sacrificed in less ideal environments because of the following features:

1) The environmental conditions which yield measurements with an accuracy of 0.1% generally are not difficult to maintain.

2) Updating the zero-power intercept is a fast and simple method of correction, in the event that these conditions are violated.

3) Choosing a small enough allowed standard deviation ensures that the first printed value will occur only after the T_3 circuit stabilizes, and that it will represent a value which cannot be improved by increasing the measurement time.

III. DESIGN AND CONSTRUCTION EFFORT FOR THE FOUR-METER FUEL-ROD CALORIMETER

A. Description of the Four-Meter Fuel-Rod Calorimeter

A sketch of the four-meter fuel-rod calorimeter system is shown in Fig. 5. The system consists of:

1) A preheater, 14 feet long, for preheating the fuel rods.

2) Electronics for controlling the preheater.

3) The calorimeter, 15 feet long, consisting of four concentric cylinders whose temperatures are precision controlled.

4) Electronics units for controlling the calorimeter including a microprocessor.

5) A "silent 700" typewriter as the I/O device, including dual cassette magnetic tape units.

B. Mathematical Model of the Calorimeter

A schematic of the temperature control circuits of the cylinders in the measurement chamber box is shown in Fig. 6. The outer (plastic)

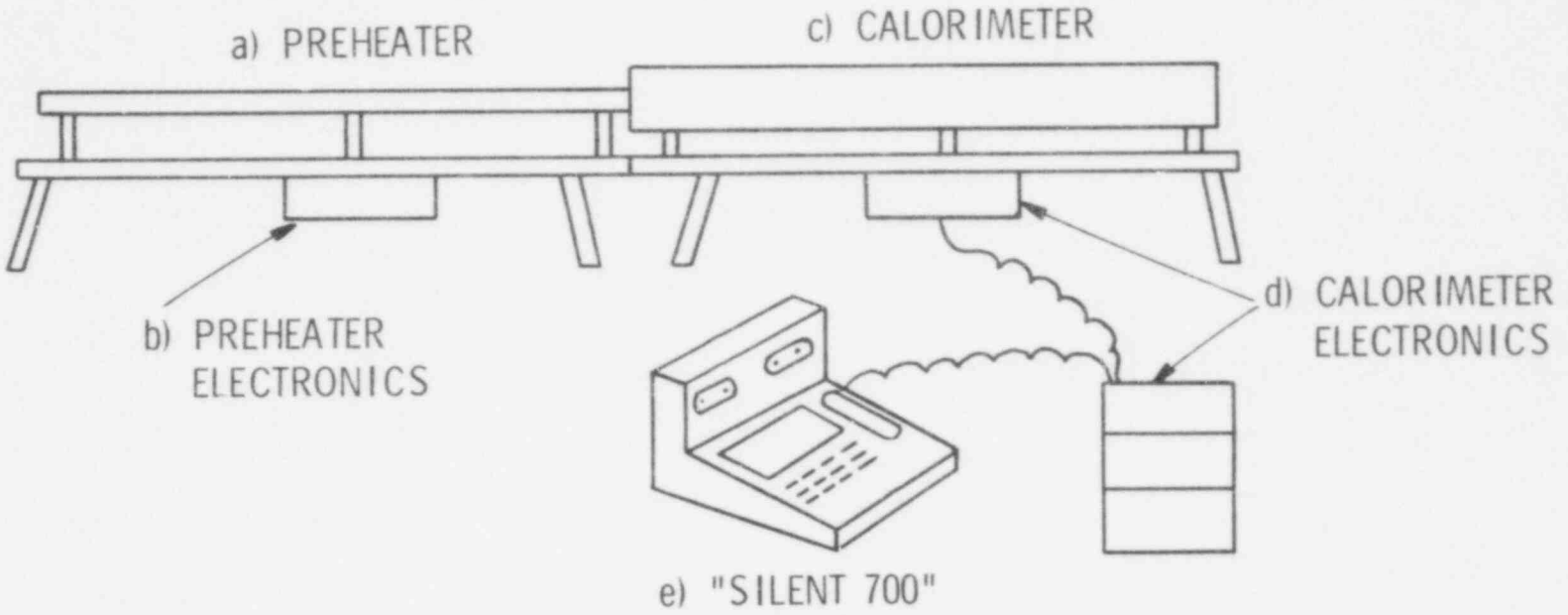
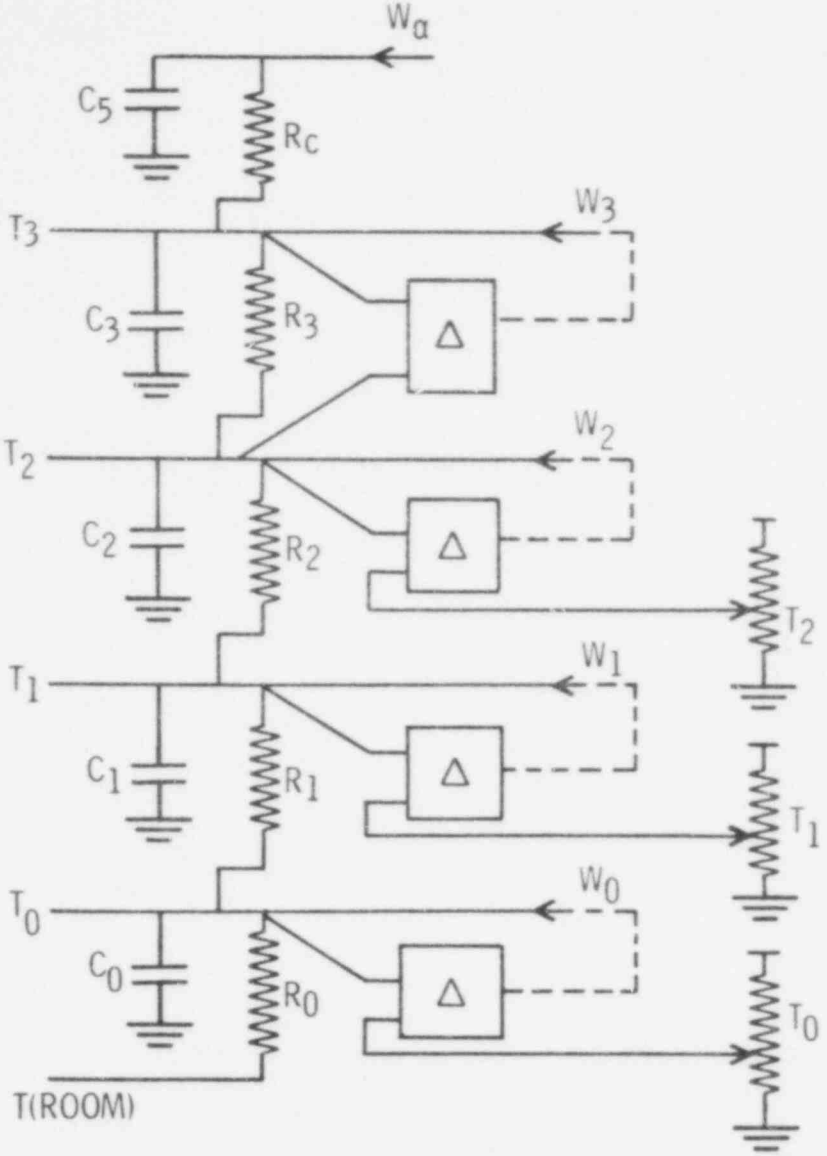


Fig. 5. Four-meter Calorimeter



W_α = HEAT DUE TO α ACTIVITY OF FUEL

C_S = THERMAL CAPACITY OF FUEL SAMPLE

R_C = THERMAL CONTACT RESISTANCE BETWEEN FUEL AND CHAMBER

C_3 = THERMAL CAPACITY OF SAMPLE CHAMBER

R_3 = THERMAL RESISTANCE BETWEEN T_3 AND T_2 CHAMBERS

Fig. 6. Five Temperature-control Circuits of the Four-meter Calorimeter

case is at room temperature. As in the one-meter calorimeter, the outermost cylinder is held at a higher temperature T_o by automatic control of the input current, or power, W_o. The potential is stored by the thermal capacity C_o while the current through the thermal resistance R_o causes heat to be lost to the room.

The second concentric cylinder is held at temperature T_1 by automatic adjustment of power W_1 which flows through R_1 and also through R_o. The third concentric cylinder is held at temperature T_2 in a similar manner. The temperature difference $T_3 - T_2$ is controlled by automatic adjustment of the power W_3. The equation relating this power to the temperature difference in the steady state is $W_o = W_\alpha + W_3 = (T_3 - T_2)/R_3$, where W_o is the zero-power intercept, and W_α is the power attributed to alpha activity. Studies of the model indicate that this scheme will yield better performance of the instrument than the scheme used in previous models wherein the temperatures T_3 and T_2 were controlled independently of each other. Study of the model also reveals that T_2 can be made more stable if the sum $W_3 + W_2$ is constant after initial stabilization. This improvement is mechanized in the present instrument.

C. Temperature Control

Computer control of the critical temperatures will be accomplished by using a prediction technique. According to this technique, a set of error signals, ε_1, ε_2, ε_3, is made at time intervals Δt. The final error ε_F is predicted using measured system parameters. Then the input signal is modified by a step change calculated to achieve zero error at the end of time interval Δt. One such approach is described in Appendix A. To aid in setting up the computer programs, a simulator has been constructed and will be used to optimize methods of temperature control until construction of the calorimeter is completed.

D. Construction

The preheater has been wound and tested. Figs. 7 through 11 are drawings of sections of the four-meter calorimeter, and they provide

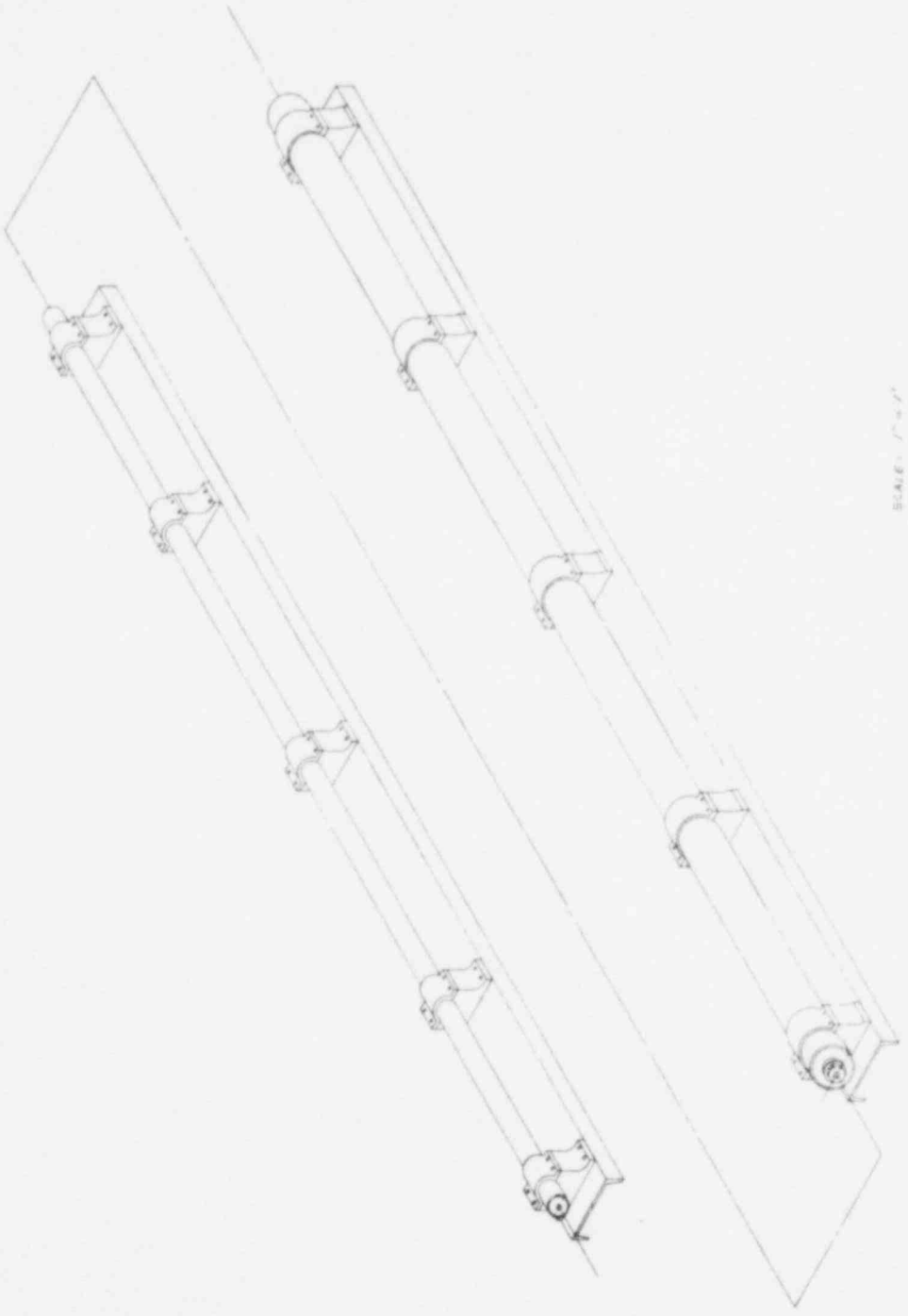


Fig. 7. Four-meter Calorimeter with Preheater

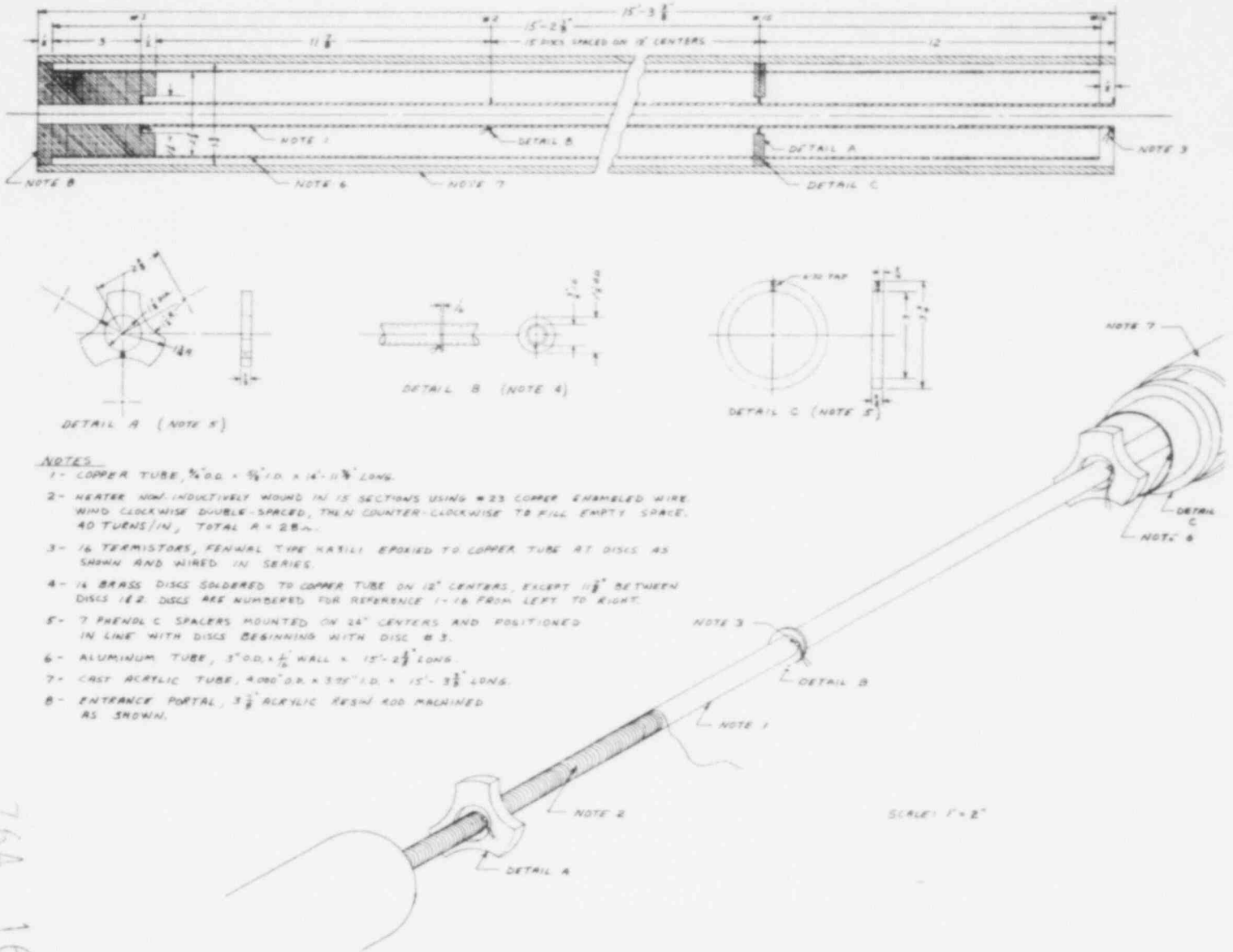


Fig. 8. Preheater for the Four-meter Calorimeter

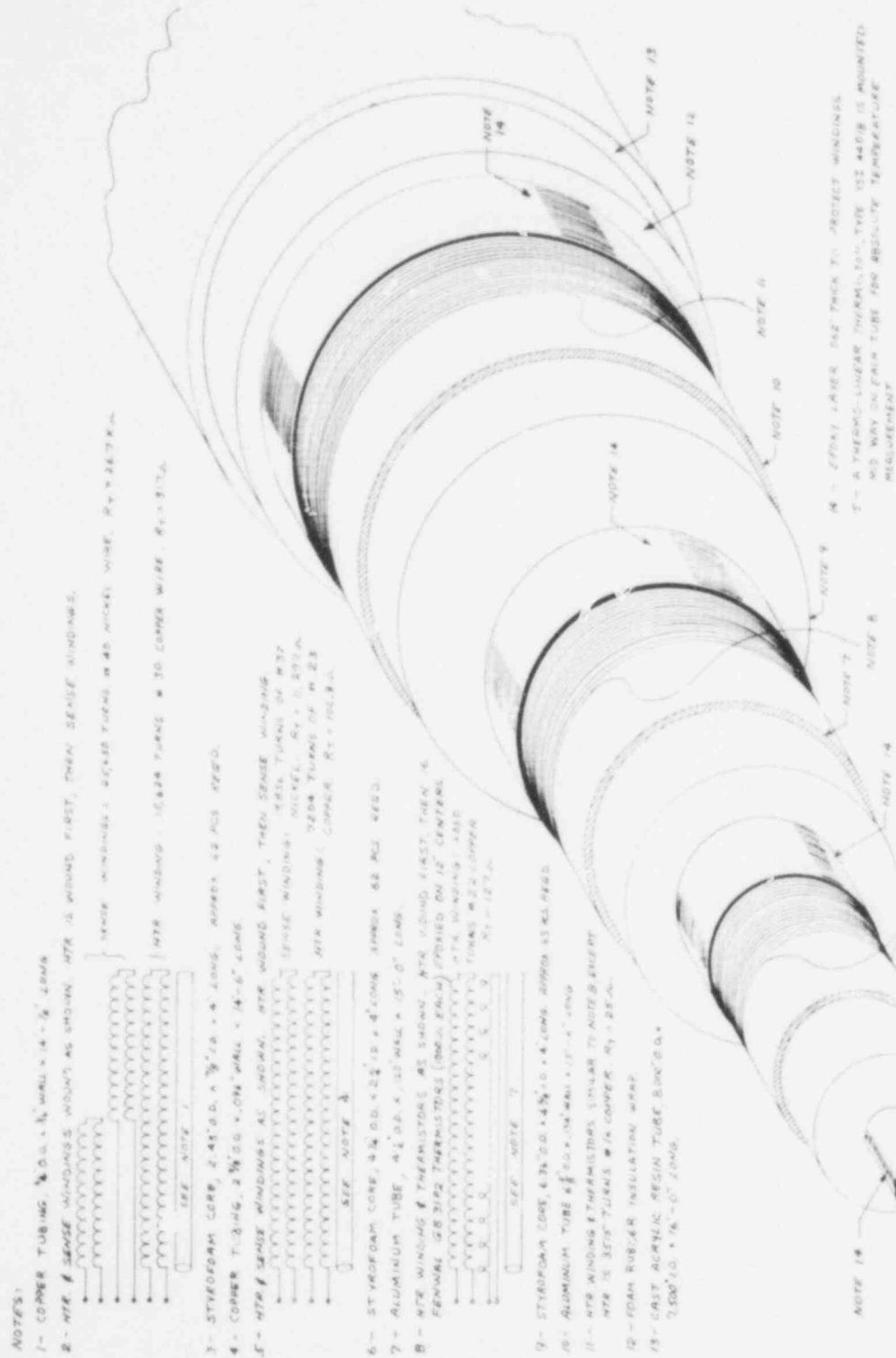
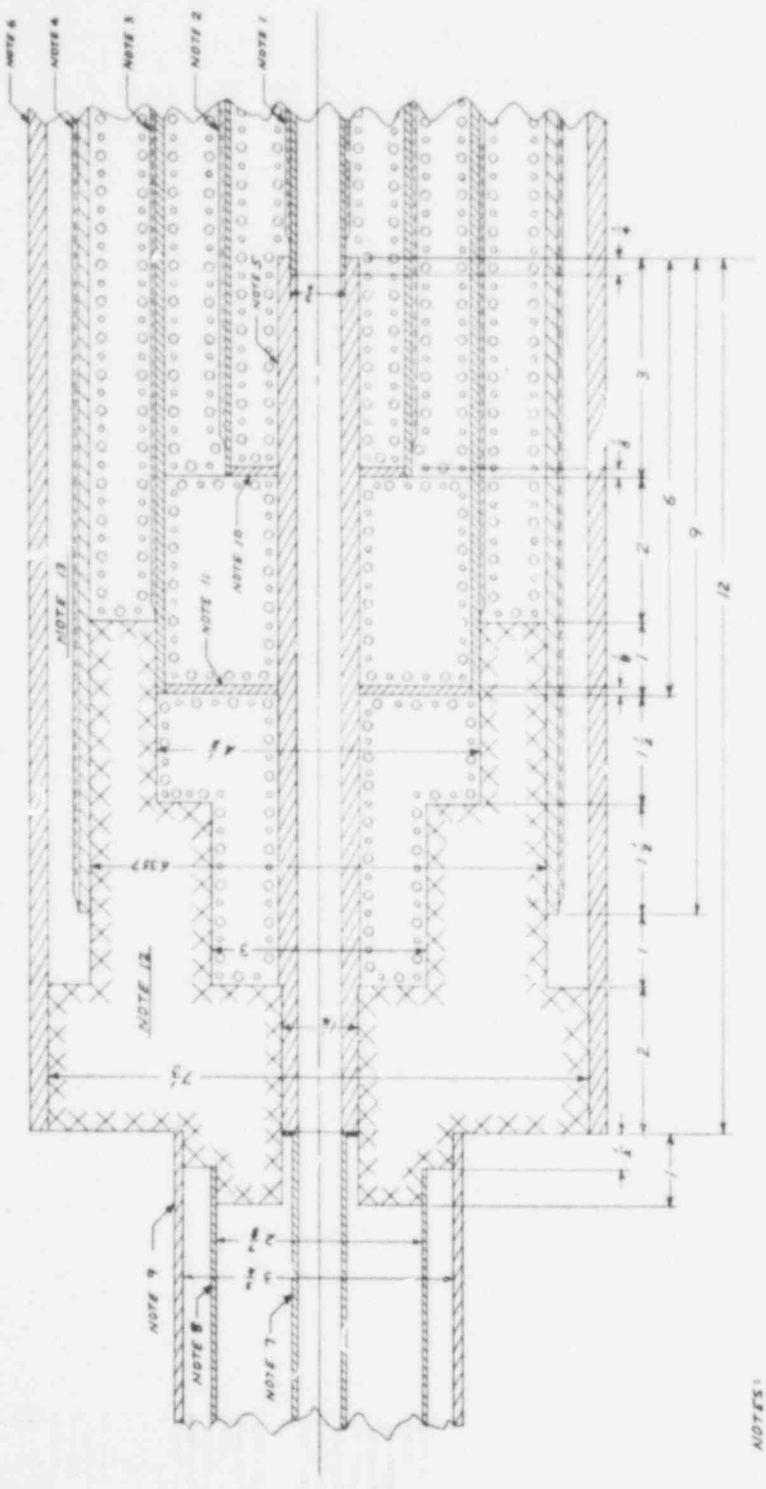


Fig. 9. Cylinders of the Four-meter Calorimeter



NOTES:

1. COPPER TUBING, ¾" O.D. × ½" WALL × 14' ½" LONG. HEATER & SENSE WINDINGS WOUND AS DETAILED ON SHEET 3, NOTE 2.
2. COPPER TUBING, 2 ½" O.D. × .134" WALL × 14'-6" LONG. HEATER & SENSE WINDINGS WOUND AS DETAILED ON SHEET 3, NOTE 5.
3. ALUMINUM TUBING, 4 ½" O.D. × .120" WALL × 15'-0" LONG. HEATER WINDING & THERMISTORS MOUNTED AS DETAILED ON SHEET 5, NOTE 8.
4. ALUMINUM TUBING, 6 ⅝" O.D. × .134" WALL × 15'-6" LONG. HEATER WINDING & THERMISTORS MOUNTED AS DETAILED ON SHEET 3, NOTE 11.
5. NYLON BUSHING, 1⅜" O.D. 1⅝" I.D. × 12" LONG. ENDS MACHINED AS SHOWN.
6. CAST ACRYLIC PLASTIC TUBING, 8.000" O.D. × 2.500" I.D. × 48" LONG, 4 REQ'D.
7. SAME TUBING AS NOTE 1 EXCEPT WITH 1¾" O.D. × ⅛" THICK SPACERS SPACED 12" APART. HEATER WINDING AS DETAILED ON SHEET 2, NOTE 2.
8. ALUMINUM TUBING, 3" O.D. × ⅛" WALL × 16'-0" LONG
9. CAST ACRYLIC PLASTIC TUBE, 4" O.D. × ⅛" WALL × 48" LONG, 4 REQ'D
10. COPPER TERMINATING DISC, 2.453" O.D. × 1.25" I.D. × ⅛" THICK.
11. ALUMINUM TERMINATING DISC, 4.24" O.D. × 1.25" I.D. × ⅛" THICK.
12. CAST ACRYLIC PLASTIC ROD, MACHINED TO DIMENSIONS SHOWN.
13. THIS SPACE FILLED WITH FOAM RUBBER INSULATION TAPE WRAPPING
14. SPACERS INDICATED BY (○○○○) DENOTES STYROFOAM FILL
15. SCALE: 1" = 1'-0"

Fig. 10. Entrance Portal of the Four-meter Calorimeter

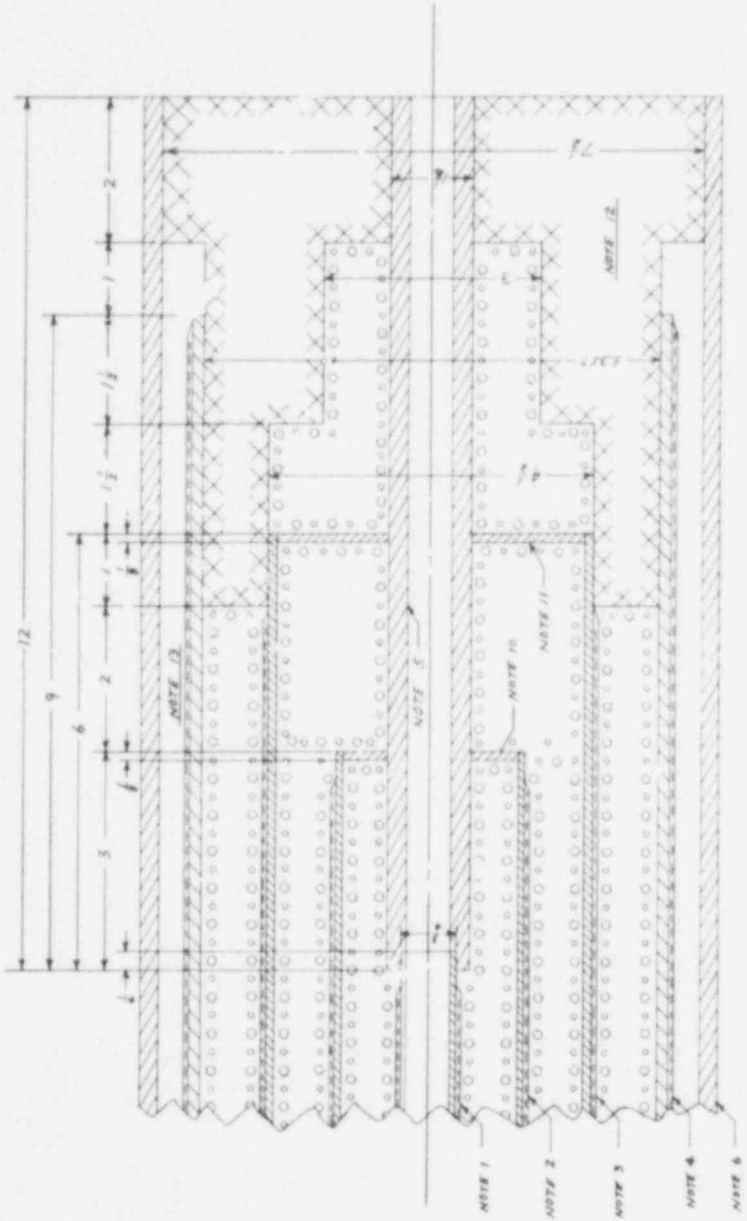


Fig. 11. Exit Portal of the Four-meter Calorimeter

material specifications and dimensions of the cylinders and the sense windings. The inner cylinders which maintain temperatures T_3 and T_2 are being wound. The preamplifiers for the critical temperatures T_3 and T_2 are under test. The nickel-wire bridges produce a 5-microvolt signal for a temperature error of 10^{-4}°C. This provides an S/N of about 10. Drifting of the preamp is about 3 microvolts/month. Since $W = K(T_3 - T_2)$ and $dW = KdT$, the long-term stability dW is less than or equal to 0.1 mW for K = 1. Components for the electronics units have been procured and initial construction is under way.

The microprocessor unit has been received and is currently being used to assemble programs for data control. The necessary modifications have been made to the microprocessor to make it operational from the "silent 700" terminal. The design has been started and parts have been ordered for the analog-to-digital and digital-to-analog interfaces to the processor. These interfaces are needed for data acquisition, automatic troubleshooting, and automatic calibration systems. The programming for these systems has been started.

E. Preliminary Studies for Bulk and Kinetic Calorimetry

Preliminary studies for the bulk calorimeter is under way. A second bulk calorimeter is being constructed on a different project, and detailed measurements of its performance will aid in the study report.

Preliminary studies for kinetic calorimetry have begun. The programming studies are sufficiently similar to bulk calorimetry that advantage is being taken to consider both problems simultaneously.

IV. DESIGN AND CONSTRUCTION EFFORT FOR THE ANALYTICAL SMALL-SAMPLE CALORIMETER

The analytic calorimeter under construction is being designed for measurement of solid plutonium compounds or mixtures encountered in plutonium recycle fuel fabrication. The sample size will be 1-2 grams of contained plutonium. The sample volume will be 7.58 cm^3. The inside diameter of the sample capsule will be large enough to accept power reactor fuel pellets up to 1.47 cm in diameter. The calorimeter will be used in conjunction with chemical analysis to determine the specific power of plutonium samples. A goal of this project is to develop an instrument capable of a single-measurement precision of 0.05% relative standard deviation with a systematic error in thermal-power measurement of 0.01% relative, or less.

A thermal circuit model for this calorimeter has been developed under a subcontract to South Dakota School of Mines and Technology. A status report on this work was given at the INMM meeting in June, 1976.[7] A computer program has been developed to utilize the thermal circuit model, and has been used to generate axial temperature profiles for the calorimeter to determine the optimum power settings for the thermal guards. The program was also used to determine the optimum length of the insulating spacers between the center coils and guard coils --information which is difficult and time-consuming to obtain experimentally.

The coils for the calorimeter, shown in Fig. 12, have been constructed and their control circuits have been breadboarded. Tests of temperature and power stability are now being conducted. A microprocessor and a "silent 700" page printer will provide control and readout for the calorimeter.

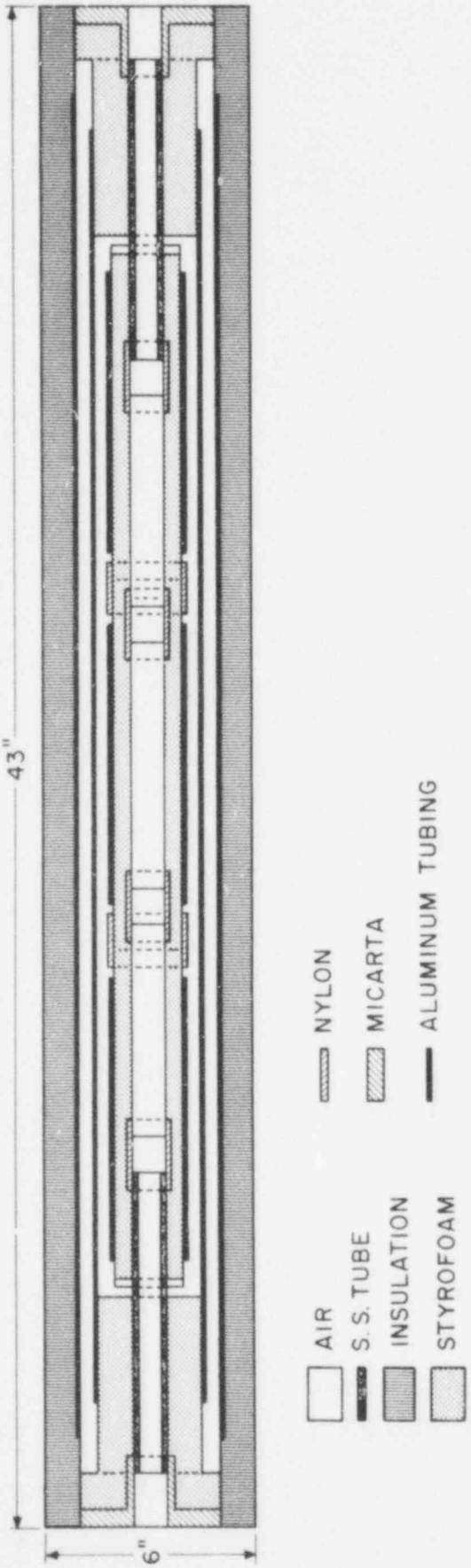


Fig. 12. Coil Assembly for the Analytical Calorimeter

APPENDIX TO PART I.

Computer Control of Calorimeter Temperature

The error prediction technique to be used to control critical temperatures is described by the following derivation. A single time-constant calorimeter section can be represented by the circuit in Fig. 13. By a current-node analysis, the response to a constant input power W is

$$T - T_{REF} = RW(1 - e^{-t/RC}) + (T_0 - T_{REF})e^{-t/RC}, \qquad (1)$$

where T, T_{REF}, R, and C are as defined in Figs. 13 and 14, and $T_0 - T_{REF}$ is the initial condition. As shown in Fig. 14, $T_3 - T_{REF} = \Delta T$ and $T_3 = T + \varepsilon$, so $T - T_{REF} = \Delta T - \varepsilon$ and $T_0 - T_{REF} = \Delta T - \varepsilon_0$. Then Eq. 1 becomes

$$\Delta T - \varepsilon = RW(1 - e^{-t/RC}) + (\Delta T - \varepsilon_0)e^{-t/RC}.$$

After a time interval Δt this can be written as

$$\Delta T - \varepsilon_1 = RW(1 - e^{-\Delta t/RC}) + (\Delta T - \varepsilon_0)e^{-\Delta t/RC}, \qquad (2)$$

where $\Delta T - \varepsilon_0$ is the initial error at $t = 0$. Similarly, after a second time interval Δt, the output is

$$\Delta T - \varepsilon_2 = RW(1 - e^{-\Delta t/RC}) + (\Delta T - \varepsilon_1)e^{-\Delta t/RC}, \qquad (3)$$

where $\Delta T - \varepsilon_1$ is taken as the initial error at $t = \Delta t$. The general expression for this output is

$$\Delta T - \varepsilon_n = RW(1 - A) + (\Delta T - \varepsilon_{n-1})A, \qquad (4)$$

where $A = e^{-\Delta t/RC}$. From Eq. 4,

$$\Delta T - \varepsilon_2 = RW(1 - A) + (\Delta T - \varepsilon_1)A, \text{ and}$$

$$\Delta T - \varepsilon_3 = RW(1 - A) + (\Delta T - \varepsilon_2)A.$$

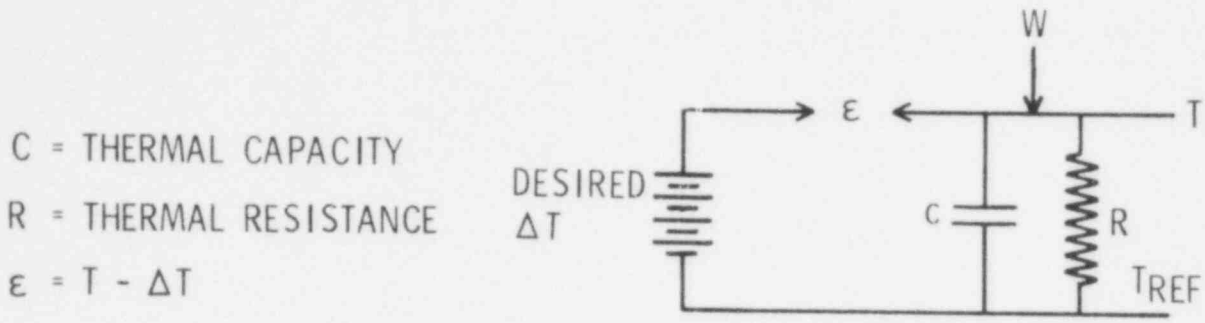


Fig. 13. Temperature-control Circuit--Simplified

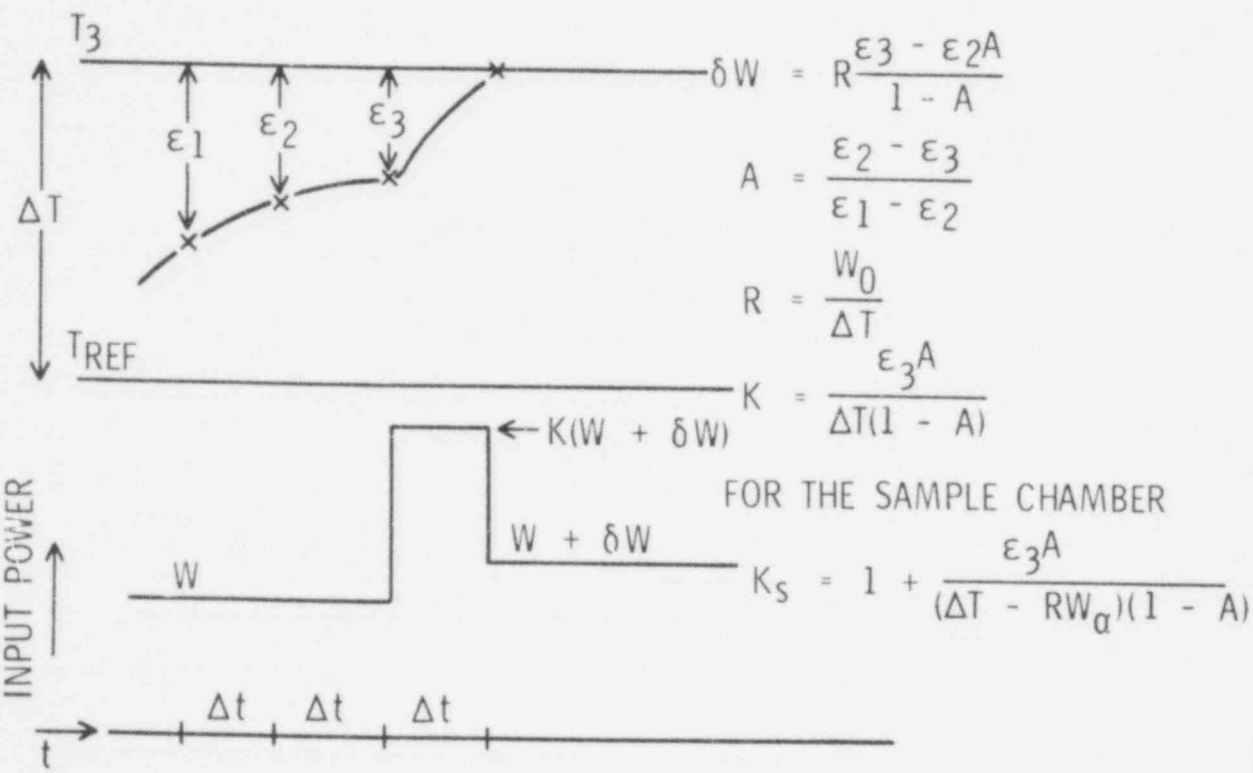


Fig. 14. Error Prediction Technique of Temperature Control

Simultaneous solution yields

$$A = \frac{\varepsilon_2 - \varepsilon_3}{\varepsilon_1 - \varepsilon_2}, \text{ and}$$

$$RW = \frac{\Delta T(1 - A) + \varepsilon_2 A - \varepsilon_3}{1 - A}. \qquad (5)$$

Assume for now that equilibrium is achieved at $t = t_4$; that is, $\varepsilon_4 = \varepsilon_F$. At equilibrium, the exponential A has decayed to zero, and the final error ε_F can be determined from Eq. 4 as

$$\Delta T - \varepsilon_F = RW.$$

Using Eq. 5, this becomes

$$\Delta T - \varepsilon_F = \frac{\Delta T(1 - A) + \varepsilon_2 A - \varepsilon_3}{1 - A}.$$

Solving for ε_F yields the expression

$$\varepsilon_F = \frac{\varepsilon_3 - \varepsilon_2 A}{1 - A}.$$

From this expression of final error can be determined the additional amount of power δW required to make the final error equal to zero. That is,

$$\delta W = R\varepsilon_F = R\frac{\varepsilon_3 - \varepsilon_2 A}{1 - A},$$

where R is evaluated as

$$R = \Delta T/W0, \text{ and}$$

W_0 = zero-power intercept.

Applying an input power of $W + \delta W$ will thus ensure a correct final temperature difference, although this value may not be reached for several time constants. The signal $W + \delta W$ may be magnified to a value $K(W + \delta W)$ such that the correct temperature is achieved in Δt. From Eq. 4 we can write:

$$\Delta T - \varepsilon_4 = RW(1 - A) + (\Delta T - \varepsilon_3)A, \text{ or}$$

$$\varepsilon_4 = \Delta T - RW(1 - A) - (\Delta T - \varepsilon_3)A. \tag{6}$$

To make $\varepsilon = 0$ in the time interval Δt, we let $W = W + \delta W$ and increase the power by a factor of K:

$$0 = \Delta T - KR(W + \delta W)(1 - A) - (\Delta T - \varepsilon_3)A.$$

So,

$$K = \frac{\Delta T(1 - A) + \varepsilon_3 A}{R(W + \delta W)(1 - A)} \cdot \tag{7}$$

Since $R(W + \delta W) = \Delta T$, this factor becomes

$$K = 1 + \frac{\varepsilon_3 A}{\Delta T(1 - A)} \tag{8}$$

The program is illustrated in Fig. 14. During the interval from t_1 to t_3 the errors ε_1, ε_2, and ε_3 are acquired, and calculation of δW and K is made. During the interval from t_3 to t_4 the correction $K(W + \delta W)$ is calculated. For $t > t_4$, $K(W - \delta W)$ is applied to the input power, and new error values ε_1, ε_2, ε_3 are measured to determine more precise values for correction of the input power.

For the fuel sample chamber, W is comprised of W_α, the heating due to alpha activity of the fuel; and W_e, the auxiliary electrical heating; that is,

$$W = W_{\alpha} + W_{e} \ . \qquad (9)$$

When Eq. 9 is used in Eqs. 6 and 7, the expression for K in Eq. 8 becomes

$$K_{s} = 1 + \frac{\varepsilon_{3}A}{(\Delta T - RW_{\alpha})(1 - A)} \ ,$$

where W_{α} is the anticipated heating by alpha activity. If W_{e} is the historical average value of required electrical power for a large number of samples, then the value δW will be an immediate indication of deviation from the mean for that particular sample.

PART II. SNM HOLDUP MEASUREMENTS

SUMMARY OF ACTIVITIES AND ACCOMPLISHMENTS

I. State-of-the-Art Review

A review has been conducted of the state-of-the-art of in situ assay of plutonium residues. There is a dearth of written information on this subject, but results from a few measurements indicate that large quantities of plutonium holdup can occur in processing equipment.[8-13] Most of these measurements have been made using gamma-ray detectors.[8,10-13] Passive neutron measurements have also been used.[8,9] Measurement uncertainties, where estimated, ranged from ±16%[8] to ±50%.[9] Other nondestructive assay techniques have also been reviewed to determine if methods used for other applications might be applied to residue measurements. Subjects reviewed included gamma-ray, active- and passive-neutron, and thermal measurement techniques. A computer search of *Nuclear Science Abstracts* has been set up to provide a continuing list of relevant new reports and publications.

II. Techniques Using Gamma Rays

Accurate and precise gamma-ray techniques for quantitative determination of plutonium content in a sample have been successfully used for many years. For measurements of homogeneous material of uniform thickness, such as fuel elements, one can correlate the number of gamma rays in a particular energy interval with amount of material by calibrating with standard samples of known Pu content. However, a major problem with using such a technique for measurements of Pu holdup is the self-absorption of the gamma rays in thick deposits. Since the thickness of Pu deposits encountered in holdup situations can vary greatly, a measurement which simply determines the mass of Pu from the number of gamma-ray counts based on a calibration standard of a particular thickness without a separate sample thickness determination can be greatly in error. In order to improve on such inherently inaccurate methods, a technique has been investigated which includes a sample thickness determination in addition to a conventional

gamma-ray count-to-gram conversion. Both thickness and count-to-gram are calibrated with the same standard. The technique is based on measuring the intensity ratio of high- and low-energy gamma rays. Matrix attenuation corrections based on such intensity ratios have been used previously in measuring plutonium contained in scrap and waste.[14] A combination of matrix attenuation and self-attenuation effects[15] and self-attenuation effects alone[16] were studied for waste and scrap containers by this technique as was self-attenuation in plutonium-containing solutions.[17]

For a constant external absorber, this intensity ratio is only a function of source thickness if single peaks of the same isotope are compared (high-resolution Ge(Li) detector). If a low-resolution NaI(Tl) detector is used, several high-energy peaks of different isotopes are compared to several low-energy peaks of different isotopes. Thus the isotopic composition must be held constant in order to obtain thickness data from peak group intensity ratios. For either detector, the intensity ratios are independent of source-detector geometry and collimator configuration.

The errors introduced by self-absorption in a count-gram measurement without thickness determination increase with increasing sample thickness and decreasing gamma-ray energy. The fraction of self-absorption of 413 keV and 129 keV gamma rays for various thickness of plutonium are shown in Table I.

Table I. Fraction of Gamma Rays Self-absorbed

Plutonium Thickness, Inch	413 keV	129 keV
0.001	0.007	0.09
0.002	0.014	0.17
0.015	0.10	0.33
0.060	0.33	0.91
0.120	0.55	0.96

These values were calculated according to the treatment of Evans.[18]

In order to make a thickness determination, a calibration curve was generated for high- to low-energy intensity ratios (I_{413}/I_{129} for Ge(Li) detector, I_{High}/I_{Low} for NaI(Tl) detector) as a function of sample thickness. The samples used were plutonium foils 2 inches wide, 3 inches long, and clad in stainless steel. The curve from a Ge(Li) detector calibration experiment is shown in Fig. 15. A similar curve for an NaI(Tl) detector using the low-energy region between 179 and 239 keV (the ^{237}U 208 keV peak) and the high-energy region between 300 and 460 keV is shown in Fig. 16. The ratios are normalized to an arbitrary thickness, 0.020 inch, so that for the thickness x, the ordinate is $(I_{412}/I_{129})_x/(I_{413}/I_{129})_{0.020}$ or $(I_{High}/I_{Low})_x/(I_{High}/I_{Low})_{0.020}$. Using this curve, it is only necessary to make a single calibration measurement of a sample of known thickness incorporating the external absorber to be encountered in a holdup situation. The *relative* change in intensity ratio from one thickness to another indicated in Figs. 15 and 16 is independent of external absorber, but, in order to determine thickness in a holdup configuration, a calibration measurement must be made using the expected external absorber. After this, all ratios can be compared to that for the sample of known thickness and an unknown thickness determined from the calibration curve. The calibration curve should cover the range of thickness expected. The thinnest sample included can be that below which the errors from self-absorption are not considered important for a particular application.

As an example of this procedure, a thick layer of Pu in a heavy-wall pipe was simulated. The pipe was stainless steel, 2.88-inch od with an 0.214-inch thick wall. Using a Ge(Li) detector, a calibration measurement was made for an 0.020-inch thick Pu sample inside the pipe. The intensity ratio now becomes the reference ratio $(I_{413}/I_{129})_{Ref}$. A second Pu sample, an "unknown," was then placed in the pipe and counted. The ratio $(I_{413}/I_{129})_x/(I_{413}/I_{129})_{Ref}$ was 1.831. From the calibration experiment, the ratio $(I_{413}/I_{129})_{0.040}/(I_{413}/I_{129})_{0.020}$ was 1.787. Thus, the "unknown" would be identified as slightly thicker than 0.040 inches. In fact, the "unknown" was 0.040 inches thick and the accuracy of the determination was within 2.5%. The statistical precision was 2%. For an 0.060-inch thick sample, the accuracy was within 4.2% and the precision 1%. Using an NaI(Tl) detector, both the 0.040-inch and the 0.060-inch samples

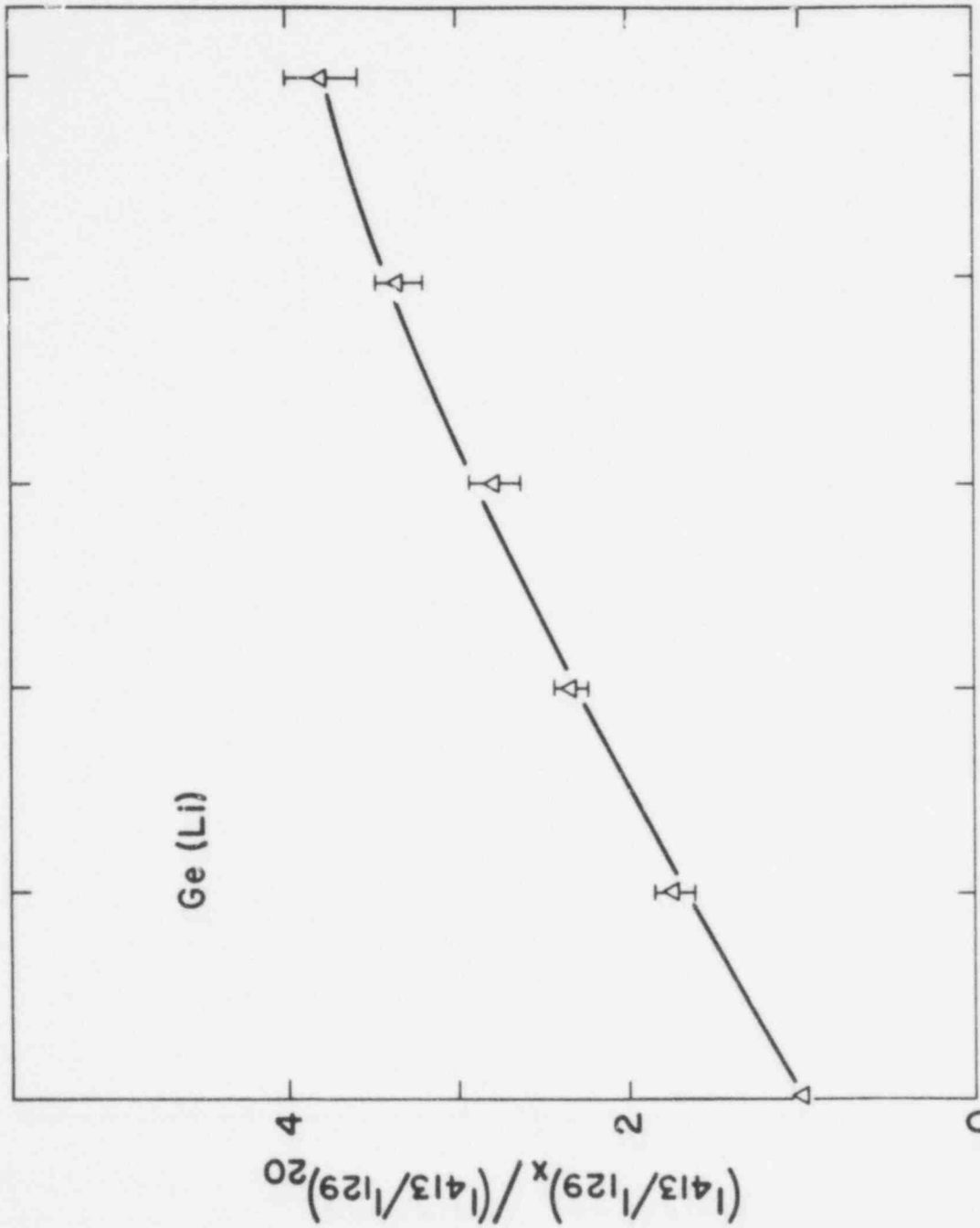


Fig. 15. Gamma-ray Intensity Ratio as a Function of Plutonium Thickness Using a Ge(Li) Detector

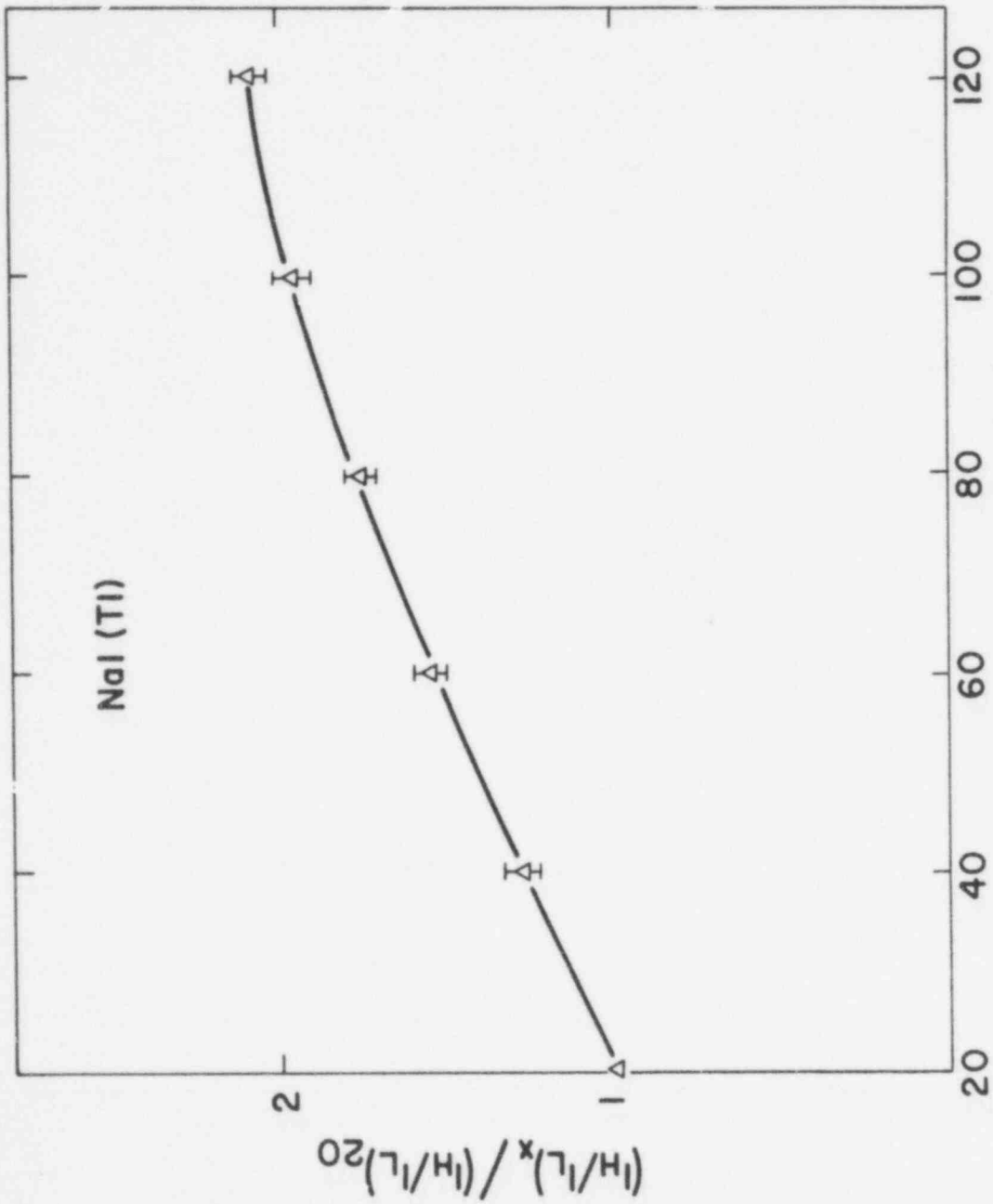


Fig. 16. Gamma-ray Intensity Ratio as a Function of Plutonium Thickness Using a NaI(Tl) Detector

were identified within 1% with a 1% statistical uncertainty. In an actual holdup measurement, the 0.020-inch thick sample would be used to determine both $(I_{413}/I_{129})_{Ref}$ and the count-to-gram conversion. If, upon counting an unknown sample, the intensity ratio results indicated an 0.040-inch thick material, the number of grams reported would simply be twice that indicated by the 0.020-inch thick calibration sample. These experiments are idealized since the foils are of uniform thickness. In an actual holdup situation, uniform thickness cannot be assumed, and the effect of such nonuniformity should be investigated.

III. Collimator Design

Almost any gamma-ray holdup measurement will require detector collimation to define the sampling region and to eliminate effects of background radiation. A collimator has been designed to give good spatial resolution while maintaining high sensitivity for gamma rays emitted from plutonium. The collimator is designed for use with a 2-inch diameter NaI(Tl) detector but can be easily modified for use with other detectors. It consists of a 1-1/8 inch thick lead-alloy shield around the sides of the detector and a cylindrical insert over the front of the detector. This insert contains 37 parallel holes 4 inches long and 3/16 inch in diameter. The high length-to-diameter ratio allows high resolution. At ten feet from the collimator, gamma rays will be collected from a one-foot diameter circle. The large number of holes allows a large fraction of the detector crystal area to be used--32%--to maintain high sensitivity. This collimator is currently being fabricated by ANL shops. Also, different collimator designs are under consideration for use with sources of different geometries, such as pipes which approximate line sources.

IV. Techniques Using Neutrons

Neutrons emitted in (α, n) reactions and in spontaneous or induced fission of plutonium isotopes have long been used as a basis of well-established nondestructive assay methods. In general, techniques utilizing neutrons have the advantage of being relatively insensitive

to container attenuation and self-absorption problems. However, self-multiplication due to induced fission can be a problem for large masses of Pu. The techniques also suffer from being sensitive to background neutrons from material in the detector vicinity, especially if total neutrons are counted. Also, total neutron-counting methods are sensitive to the chemical composition of the Pu environment. Coincidence methods detect only neutrons from fission, and thus are independent of the chemical composition of the Pu matrix. This method is also less sensitive to background neutrons. however, coincidence methods lack sensitivity, since the detector efficiency for coincidence neutrons is the square of the efficiency for total neutrons. This lack of sensitivity results in higher statistical uncertainties or unreasonably long counting times.

For holdup applications, a neutron assay system must be able to adapt to various dimensions and geometries depending on the location and geometry of the holdup container. With this in mind, a two-slab "sandwich" configuration was chosen. The slabs are made of 3-inch thick polyethylene and contain two 2-inch diameter 12-inch active length BF_3 neutron proportional counters. The slabs are simply placed on opposite sides of the suspected holdup container. With this system, one can count either total neutrons (α, n plus fission) or coincidence neutrons (fission only). As in the gamma-ray experiments, a 2.88-inch od, 0.214-inch-wall stainless-steel pipe with various Pu samples inside was used to simulate holdup. Again, the plutonium samples were 2 by 3 inches and clad in stainless steel.

To study the sensitivity of the system, the response of the detector for coincidence neutrons was measured as a function of slab separation from a Pu sample centered between two slabs. The results are shown in Fig. 17. As shown, the response falls rapidly with separation distance d; roughly, as $1/d^4$. This is not surprising since the count rate for total neutrons decreases roughly as $1/d^2$ (for a point source), and the coincidence efficiency varies as the square of the total neutron efficiency. This rapid decrease in sensitivity with slab separation is potentially a serious problem for coincidence counting of holdup. Typical sensitivities for Pu in the 2.88-inch od pipe (2.88-inch slab separation)

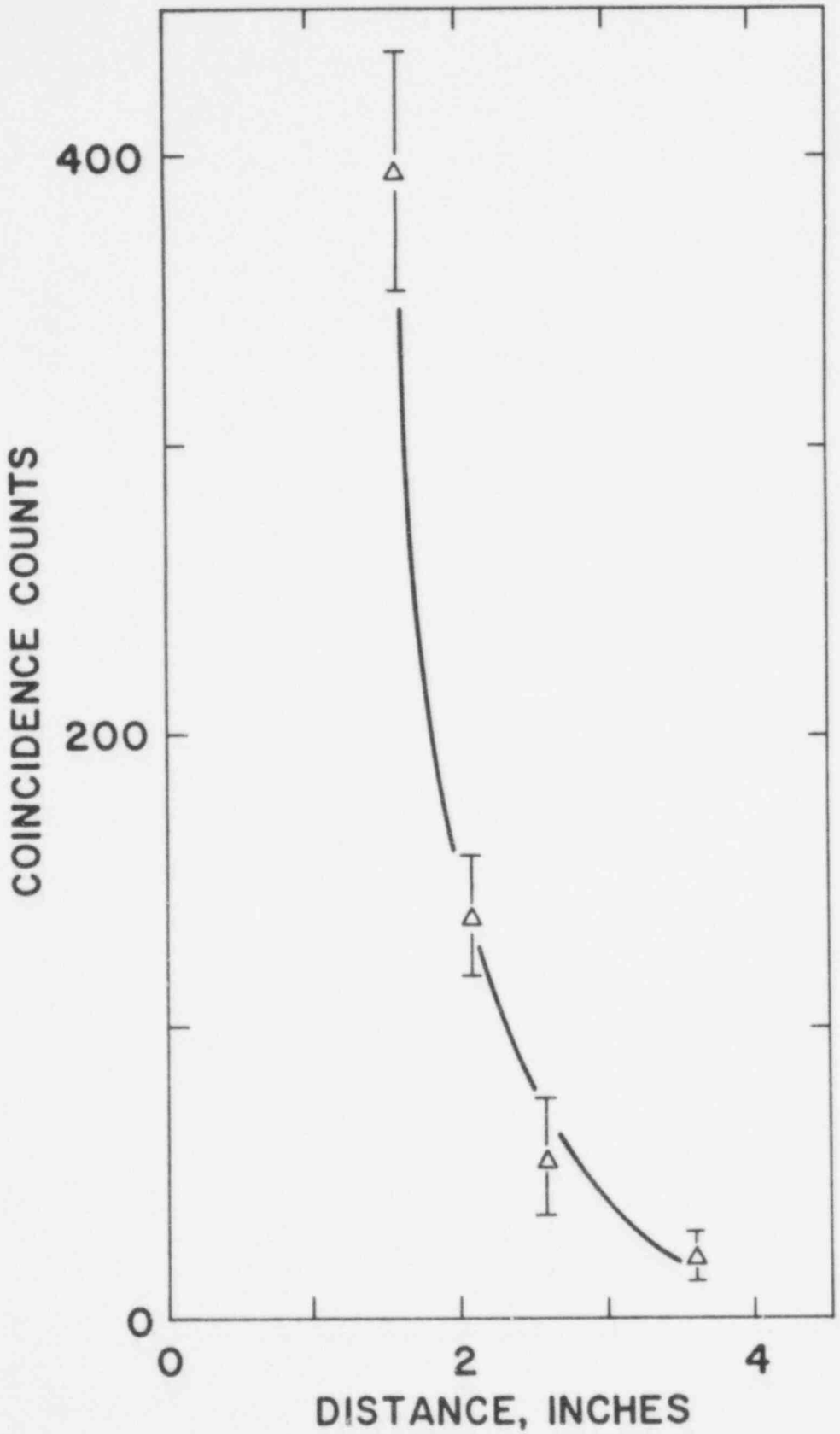


Fig. 17. Neutron Coincidence Counts as a Function of Detector Slab Separation

were about 0.2 coincidence counts sec^{-1} $(gm\ ^{240}Pu)^{-1}$, and the standard deviation of several measurements was typically 10%.

Measurements were also made with Pu samples in the pipe and various amounts of Pu outside the detector slabs but in the vicinity of the detector. An example of these results is shown in Fig. 18. The pipe contains 47 gm Pu, and 360 gm of Pu is located outside the detector at various distances from the face of one slab. As indicated, the Pu outside the detector has very little effect on the coincidence count rate once the material is 8 inches or more away away from the detector. The total neutron-count rate shows a relatively greater effect continuing out to greater distances. Both methods do approach the count rates for no external Pu at large distances. Error bars are not shown for the total neutron counts, since their relative statistical uncertainty is quite small (approximately 1%). This compares quite favorably with the statistical uncertainties of up to 21% for the coincidence points. Figure 18 does illustrate, however, that the more rapid decrease in coincidence counts with increasing source-detector distance makes coincidence techniques less sensitive to background contributions.

One additional problem with the neutron-counting technique is the dependence of the coincidence- and total-count rates on the spatial arrangements of the sample with respect to the detector slabs. For example, a Pu sample centered between the slabs was found to give a higher count rate than the same sample near an edge. This could lead to greater uncertainty when measuring samples of unknown location or spatial distribution. Also, it should be noted that, since the detector response is a function of slab separation, a new count-to-gram calibration will be necessary for each slab arrangement.

Although there are difficulties, preliminary experiments suggest that neutron-counting methods hold promise for use in holdup measurements. They could be especially useful in applications where attenuation by heavy containers or self-absorption make gamma-ray methods impractical. Neutron-counting methods could also provide a useful check on other assay results. Further work, including efforts to reduce the effect of background neutrons, is planned.

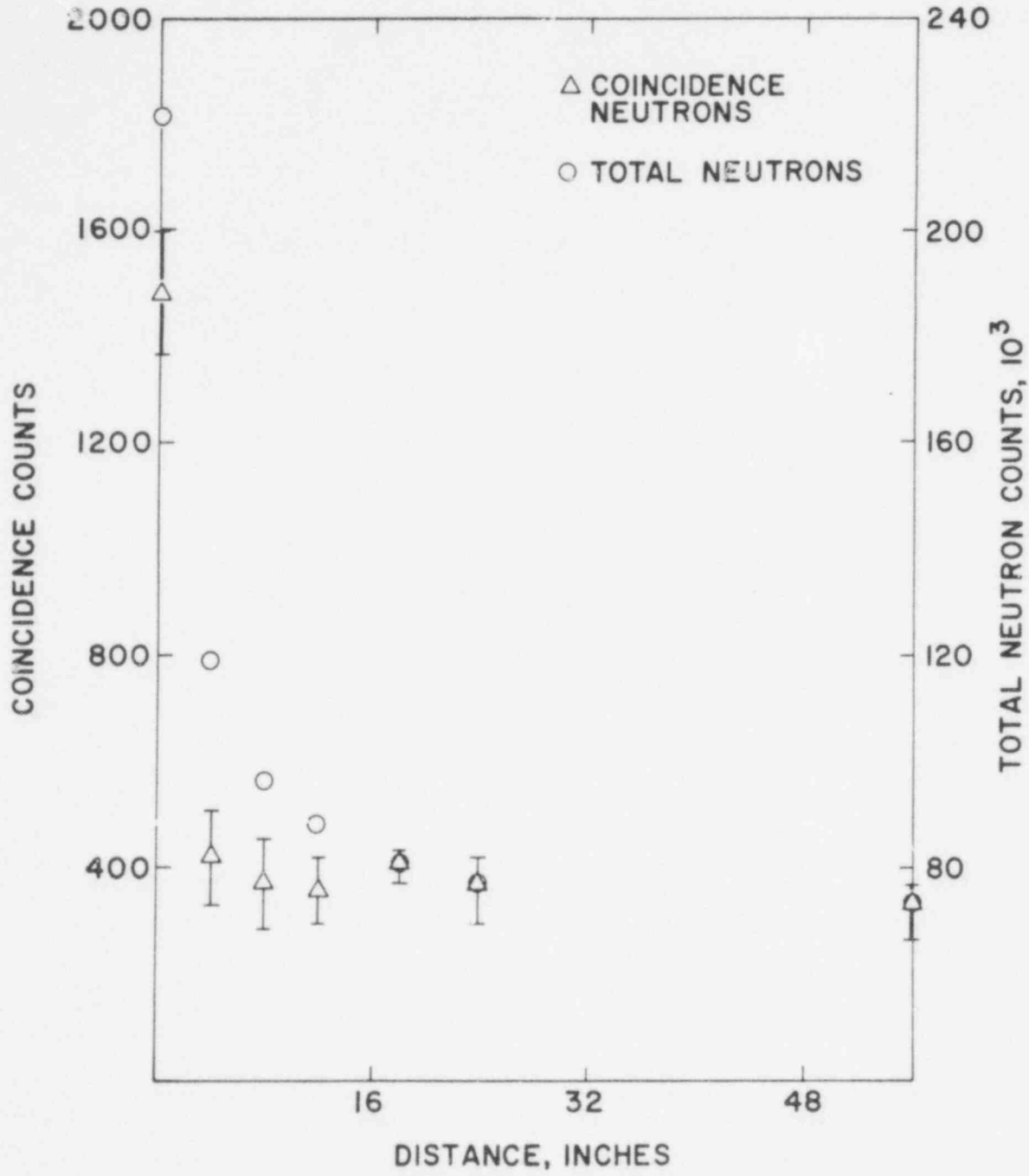


Fig. 18. Neutron Coincidence Counts and Total Neutron Counts as a Function of Distance Between Detectors and External Neutron Source

V. Methods Using Infrared Radiation

In addition to neutrons and gamma rays, the radioactive decay of plutonium also produces alpha-particles which result in self-heating of the plutonium. The measurement of this heat has been utilized in the calorimetric assay technique. As a result of the self-heating phenomenon, a plutonium sample will be at a slightly higher temperature than its environment. This temperature difference will depend on the amount and isotopic composition of the plutonium and the various heat-loss mechanisms operative. Small temperature differences can be detected by measuring the intensity of long wavelength infrared radiation emitted by the sample and its surroundings. Present infrared equipment is capable of resolving temperature differences of 0.1°C. Calculations have been carried out to estimate the temperature differences expected for various amounts of plutonium and different heat-loss mechanisms. For instance, a ZPPR "F" rod, which loses heat only by natural convection, is expected to be approximately 0.5°C warmer than its surroundings. These calculations were based on the work of Renken, Cox, and Sather.[19]

Figure 19 illustrates the use of an infrared temperature-difference measuring technique for detecting plutonium. The photograph shows seven ZPPR fuel rods. Three contain no plutonium, three are "F" rods containing approximately 10 gm plutonium, and one is a "G" rod containing approximately 20 gm plutonium. In the photograph darker areas are warmer than lighter areas. As shown in Fig. 19, the infrared scan can easily distinguish rods of different plutonium content based on their temperature difference. All the rods have been given a coat of flat black paint to provide uniform emissivity which is necessary for these measurements. The apparatus used was a Barnes T-4 infrared scanner.

The applicability of this method to in situ residue measurements was tested by placing an electrical resistor on the inner wall of a stainless steel pipe to simulate a lump of plutonium in a process line. The pipe had a 2-inch od with a 1/16-inch thick wall. The resistor was 0.4 inch in length. The results for three different power inputs are shown in Fig. 20. One inch on the photograph corresponds to approximately 3.25 inches of pipe. Again, the pipe

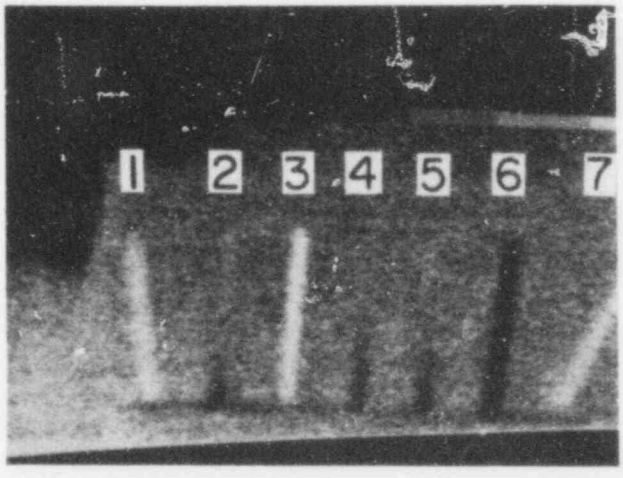


Fig. 19. Infrared Scan of ZPPR Fuel Rods. Rods 1, 3, and 7 Contain no Plutonium: Rods 2, 4, and 5 Contain 10 gm Plutonium; Rod 6 Contains 20 gm Plutonium. Darker Corresponds to Higher Temperature.

has been painted black for uniform emissivity. A typical plutonium isotopic mixture found in fuel rods will have a specific power of about 4 mW per gram. Thus, Fig. 20a represents the case of an approximately 25-gm lump of plutonium on the wall of a pipe. The smallest power input which could be detected was about 52 mW, which corresponds to about 13 gm of plutonium. Calculations are also in progress to determine temperature differences expected as a function of power input and heat-loss parameters expected in holdup situations. It is hoped that such infrared scanning methods will be useful in detecting local large concentrations of plutonium. The amount of plutonium in such a concentration can then be determined in a separate measurement.

In addition to applications for holdup location, the infrared technique has a potential application for making fast attribute checks of plutonium-containing items such as reactor fuel rods and plates. Experiments are planned to use a compact, lightweight infrared-imaging device for such attribute checks.

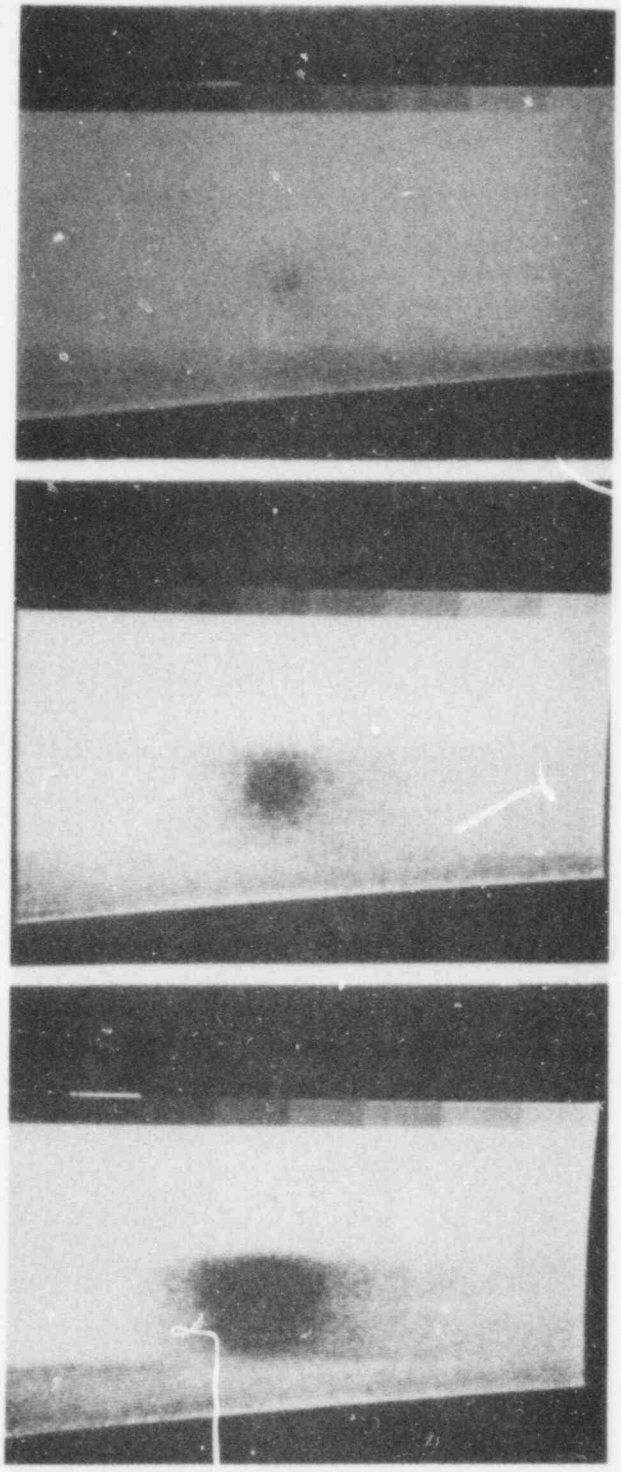


Fig. 20. Infrared Scans of Stainless Steel With Various Power Input to Electrical Resistor Attached to Inner Pipe Wall. Scan A, 104 mW; Scan B, 213 mW; Scan C, 360 mW.

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