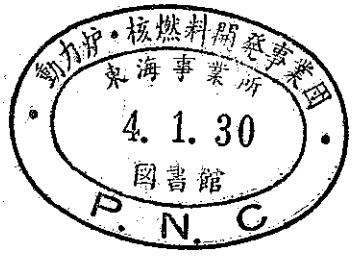
PNCT841-75-34

Non-destructive Assay Equipment for Quantitative Determination of the Nuclear Material in Plutonium Fuel Fabrication Facility

September, 1975



TOKAI WORKS

POWER REACTOR & NUCLEAR FUEL DEVELOPMENT CORPORATION

NON-DESTRUCTIVE ASSAY EQUIPMENT FOR QUANTITATIVE DETERMINATION OF THE NUCLEAR MATERIAL IN PLUTONIUM FUEL FABRICATION FACILITY

K. Onishi, H. Akutsu, T. Itaki, K. Miyahara, Y. Tokoro
and M. Tsutsumi

September 1975

To be presented to the IAEA International Symposium
on the Safeguarding of Nuclear Material
October 20-24, 1975, Vienna, Austria

TOKAI WORKS
POWER REACTOR & NUCLEAR FUEL DEVELOPMENT CORPORATION

PNCT841-75-34
September, 1975

NON-DESTRUCTIVE ASSAY EQUIPMENT FOR QUANTITATIVE DETERMINATION OF THE NUCLEAR MATERIAL IN PLUTONIUM FUEL FABRICATION FACILITY

K. Onishi, H. Akutsu, T. Itaki, K. Miyahara, Y. Tokoro
and M. Tsutsumi

Tokai Works
Power Reactor and Nuclear Fuel Development Corporation,
Ibaraki-ken Japan

Abstract

NON-DESTRUCTIVE ASSAY EQUIPMENT FOR QUANTITATIVE DETERMINATION OF THE NEUCLEAR MATERIAL IN PLUTONIUM FUEL FABRICATION FACILITY. In plutonium fuel Fabrication Facility (PFFF) of PNC, nuclear materials are received in the form of oxide powder both for plutonium and uranium as raw material, and are shipped to reactor site as the sub-assembly after passing through fabrication processes such as pellet preparation, fuel rod fabrication and assembling. This production line has some strategic points where the nondestructive assay is available for the safeguards techniques. In our facility, four strategic points for nondestructive assay were established as follows. (1) Receiving of nuclear material, (2) Shipping of subassembly, (3) Fuel rod, (4) Contaminated waste and scrap of the fuel power or pellets. This paper reports the result obtained from a callorimeter, γ scanner, neutron coincidence meter that was prepared for these strategic points.

1. INTRODUCTION

In Plutonium Fuel Fabrication Facility (PFFF) of PNC, PuO_2-UO_2 mixed oxide fuel for Heavy Water Reactor (HWR) and Farst Breeder Reactor (FBR) are being fabricated. Flow diagram of pellet process and fuel rod, sub-assembly process are shown in Figure 1 and 2. Nuclear materials are received in the form of oxide powder for both uranium and plutonium as raw material and are shipped in the state of the sub-assembly after passing through the fabrication process composed of pellet preparation, fuel rod fabrication and assembling. These production line has some strategic points where the non destructive assay is available for the safeguards.

They would be receiving of PuO_2 powder and UO_2 powder as raw material, scrap pellets or powder of PuO_2-UO_2 mixed oxide, solid waste from all the process, fuel rod loading the mixed oxide, and sub-assembly that is to be shipped to reacter site. We have been making some research and development on the utilization of nondestructive assay for the above strategic points for a few years. Among them, γ scanner for a fuel rod of an experimental reactor "Joyo" and a drum scanner for solid waste are in practical use, and now an effort is succeeded to calorimeter for the sealed PuO_2 powder and scrap of the PuO_2-UO_2 mixed oxide, γ scanner for the HWR fuel rod of low plutonium content, neutron coincidence meter for the sub-assembly and drum scanner for the solid waste sealed in the drum can. For next step, we are planning to prepare the carton scanner for the solid waste, filter scanner for absolute filter and active interogration method for PuO_2-UO_2 mixed oxide scrap. The paper describes the results of experiments on the calorimeter, γ scanner for the HWR fuel rod, and neutron coincidence meter for "Joyo" sub-assembly.

2. CALORIMETER

Non destructive assay method of plutonium by calorimeter has been developed in some countrys. Our calorimeter was prepared by remodeling of the small twin conduction type calorimeter utilized generally for other purposes in Japan, in order to determine the plutonium content in the mixed oxide pellets or plutonium dioxide powder as the raw material. Drawing of the calorimeter and details of sample cell portion are shown in Figure 3 and 4. As known from Figure 3, this calorimeter consists of a polystylene foam to cut off the room temperature fluctuation, a pipe circulating the constant temperature water, a polystylene foam (6 cm thickness) which reduces the temperature fluctuation of the circulating water and keeps

an aluminum block at a constant temperature, and two units of the cell for the sample and reference side. The circulating water is controlled at ±0.05°C, and aluminum block is beeing kept at ±0.02°C.

Both cells are attached closely to aluminium block by 8 piaces of the thermomodule as shown in Fig. 3. The thermal conductivity of one thermomodule is about 0.04 cal/°C sec and electromotive force is about 6.5 mv/°C. The 8 thermomodules of each cells are conected in series, and both of these 8 thermomodules are conected in the oposite polality in order to subtract the fluctuation effect of aluminum block temperature. The generated volatage is determined by the microvoltmeter and Recorder, V-F converter, scaller and printer. The electrical circuit of this calorimeter is shown in Figure 5.

Experiment

(1) Stability of the zero point (Back ground)

The twin design of this calorimeter seems to reduce the effect of the room temperature fluctuation, but when the room temperature fluctuates in a wide range, the small difference of the thermal conductivity between the two side cells might given an effect to the electro motive force as back ground. The electro motive force was measured in the state of the empty cell, and its fluctuation was found less than ±2 μV as shown in Fig.6. This experiment was done in Fabruary, room temperature in daytime was controlled in 20°C by heating, and at night about 10°C. This back ground value seems to be sufficiently small for the determination of plutonium sample.

(2) Calibration curve by the standard heater

The calorimeter has standard heater of 7 range (0.05W ∿ 5W) for calibration. The obtained calibration curve is shown in Figure 7. This calibration curve is considered to be satisfactory for plutonium assay. The relation of the time and voltage is shown in Fig. 8. The time required to reach the equilibrium was approximately 10 hours, and this steady state condition was continued longer than 3 days. The fluctuation of the voltage was not detect, and reproducibility was better than 0.1 %.

(3) Determination of plutonium content in the plutonium dioxide powder

The plutonium dioxide powder with known Isotopic composition,

plutonium content and Am-241 content, was sampled in the stainless steel vessel by using an accurate balance and sealed in a plastic bag. This sample was loaded in the sample side cell and the measurement was started. The data obtained is shown in Table I. Calculated value is obtained from isotopic composition, plutonium content, Am-241 content and Specific power of isotopes, and measured value is the data obtained by our calorimeter. The major source of error for the measured value is uncertainty in the ^{238}Pu isotopic composition and its error will be estimated about 2 % of calculated value for used plutonium samples. The specific power of isotope used data of MLM-1798 in Mound laboratory.

(4) Determination of the plutonium content in PuO_2-UO_2 mixed oxide

The scrap of PuO_2-UO_2 mixed oxide pellets was sampled in the plastic bottle, and this mixed oxide contains 17.7 % PuO_2 and 82.3 % UO_2 which is enriched in ^{235}U by 23 %. Prior to this measurement, Specific powder of 23 % enriched Uranium was determined, and its result was 6.85×10^{-6} watt/g. When the above mixed oxide sample was measured by the calorimeter, measured value showed 0.6135 watts, and the calculated value was 0.6102 watts. Confidence of calorimeter value with the calculated value was very good.

3. γ SCANNER FOR THE FUEL ROD

With γ-scanning method, we have checked all the fuel rods fabricated for Fast Breeder Reactor. All fuel rods have the same plutonium content mixed oxide pellet stack in the middle and blanket pellet stack (depleted uranium dioxide pellet) on both side. Therefore it is necessary to check whether both kinds of pellet stack are loaded in the right position. This quality control is the reason why we make use of the γ scanning method. Here, gross γ ray scanning method is closen, so it is impossible to determine the plutonium content accurately, but it can distinguish core mixed oxide pellet from blanket pellet in a fuel rod clearly. Explanation of this γ scanner is not described in this paper.

In another project for the development of Heavy Water Reactor (HWR project), we prepared the new γ-scanner for the fuel rod. Because the HWR fuel assembly consists of two kinds of rod which contains different plutonium enriched pellet (0.55 w/o fissile Pu for outer region fuel rod and 0.8 w/o fissile Pu for inner region fuel rod.). These two kinds of rod must be distinguished from each other for the quality control by this γ - scanner. The content of ^{239}Pu is determined from measured γ intencity

of Pu (384 keV complex), while low energy γ ray (mainly 60 keV of ^{241}Am) is reduced in intencity by Al-Cu alloy plate of 3 mm thickness set at the surface of detectors. The block diagram of electrical circuit is shown in Figure 9. γ ray is detected by 3 NaI(Tl) γ ray detector set in the equipment, because the inspection room for this canner is too limited in space to scan the total length of fuel rod by one detector. The detected signals are passed through independently each other, Pre-amplifier, Single channel pulse hight analyzer, Rate meter, Recorder and Scaller 1.2.3, and total counts of the Scaller 1.2.3 is recorded by Scaller 4. Pu content is calculated from this total counts. γ ray spectrum of plutonium is shown in Figure 10. But we could not obtain the complete data because of shortness in period from preparation of this γ scanner. However it was found that the acculacy is about 1 % for the Pu content in the fuel rod of the plutonium isotopic composition belonging to a single lot and may be better than 4 % for the plutonium with the fluctuation of its isotopic ratio which is generally used for a fabrication compaign of a series of fuels. A fuel rod containing completly different plutonium gave a very different result, and this may limit the availability of this γ scanner for the safegurds.

4. NEUTRON COINCIDENCE METER

Neutron coincidence technique was applied to determine the plutonium content in FBR Type fuel sub-assembly. The drawing in this equipment and electrical circuit are shown in Fig. 11 and 12. As shown in Fig. 11, a part of detector was constructed by arranging 6 ^{3}He proportional counters (active length; 10", pressure; 4 atm, diameter; 1") and cadmium metal as a neutron absorber in the polyethylene modelator (450 mm in outer diameter, 600 mm in hight) which has a hole of 100 mm diameter in the center in order to insert the sub-assembly as the specimen.

The sub-assembly of FBR Type includes 91 fuel rods in hexagonal wrapper tube which has outer face to face distance of 74.7 mm between the parallel side. The over all length of the sub-assembly is 2970 mm and total weight is approximately 60 kg. Some special sub-assembly that include 84 fuel rods was prepared for monitering of neutron flux in reactor. The sub-assembly that include 91 fuel rods, or 13 kg PuO_2-UO_2 pellet, in which is contained 2 kg Pu metal or about 380 g ^{240}Pu.

380 g ^{240}Pu.

The signals detected by 6 ^{3}He counters, is classified into 3 pattern by a gate generator, and count each pattern signals by 3 scaller. Scaller

1 records all signals of neutron. Scaller 2 records the signals that occur within a set time interval which is programmed by the gate generator, after a first signal (Trigger Puls) act on scaller 2. Scaller 3 records the signals that occur in a time interval identical to that in scaller 2, after the set intermediate time is spent. These set time is programmed by gate generator, and decided by the die away time of the neutron in the polyethylene moderator of detector.

EXPERIMENT

(1) Calibration curve

The response of the detector was calibrated by the actual FBR Type fuel rods and sub-assembly that is well known for the plutonium content, isotopic composition and PuO_2-UO_2 mixed oxide weight. Figure 13 shows the calibration curve of neutron coincidence counting and Figure 14 shows calibration curve for total neutron counting, Calibration curve for the data of neutron coincidence counting was not linear, due to the dead time of counter which needs to be corrected the counting loss by the next formula.

$$N_S = K \times (Sc-Sb) \{1 + \frac{St \cdot tg}{T}\}$$

N : Spontaneous fission neutron yield
Sc : Coincidence counts by scaller 2
Sb : Delayed coincidence counts by scaller 3
St : Total counts by scaller 1
tg : Gate generator cycle time
T : Counting time

Calibration curve for total neutron counts has good linearity as shown in Figure 14, since isotopic composition and chemical composition of nuclear material is relatively constant in this FBR fuel fabrication compain. When 1 sub-assembly is counted for period of 2000 seconds, acculacy is ±2 % for the neutron coincidence count and ±0.1 % for gross neutron count.

5. CONCLUSION

Results of the experiments on the three non-destructive assay equipments were discribed in this paper. These equipments were prepared not only for requirement of the safeguards but for quality assurance for the fuel production. All of the three equipments were found to be effective

for the safeguards by its accuracy, simple structure, easiness of maintenance and low cost.

Calorimeter will be used for plutonium dioxide contained in can, scrap from the process with very good accuracy. It should be mentioned that the calorimeter takes only minimum time for verification of an inspector by the use of sealing, in spite of its long measurement time.

Neutron coincidence meter is characterized by good accuracy, less than 0.1 % by gross counting and 2 % by neutron coincidence counting for FBR type fuel sub-assembly. This will be applied not only to verification purpose, but to quality assurance of a final fuel product for the reactor site.

The γ-scanner for fuel rod is widely used in many facilities for the purposes of the quality control rather than the safeguards. The accuracy of this scanner using NaI detectors may limit availability for inventry taking of the safeguards, although it works well for the quality control and verification for the safeguards.

RERERENCES

1) F. A. O'Hara, J. D. Nutter, W. W. Rodenburg, M. L. Dinsmore, "Calorimetry for safeguards purposes" MLM-1798.

2) F. L. Oetting, "Determination of plutonium in plutonium oxide by calorimetry" J. Inorg Chem. 1965, Vol.27.

3) C. Weitkamp, H. Beißwenger, V. W. Schneider, "Accuracy of calorimetric plutonium determination", KFK-1299.

4) R. H. Augustson, H. O. Menlove, R. B. Walton, L. V. East, A. E. Evans, M. S. Krick, "Development of techniques for active and passive assay of fissionable materials." IAEA-SM-133/51.

List of Tables and Figures

Table I Analytical data of PuO_2 powder

No.	Sampling Weight PuO_2 (g)	Pu Metal Weight (g)	Calcurated Value $(watt/g_{Pu})_{W_1}$	Calorimeter Value		Error
				Measured Value(watt)	$(watt/g_{Pu})_{W_2}$	$\frac{W_1-W_2}{W_1} \times 100$ %
1	5.098	4.45	0.00371	0.0165	0.00371	0.00
2	11.010	9.61	0.00371	0.0356	0.00371	0.00
3	22.067	19.26	0.00371	0.0714	0.00371	0.00
4	148.01	128.86	0.00376	0.4890	0.00379	-0.80
5	295.00	256.85	0.00378	0.9659	0.00376	+0.53
6	1517.0	1320.85	0.00375	4.981	0.00377	+0.53
7	1630.0	1416.80	0.00379	5.359	0.00377	+0.53
8	1839.0	1598.46	0.00380	6.045	0.00378	+0.53

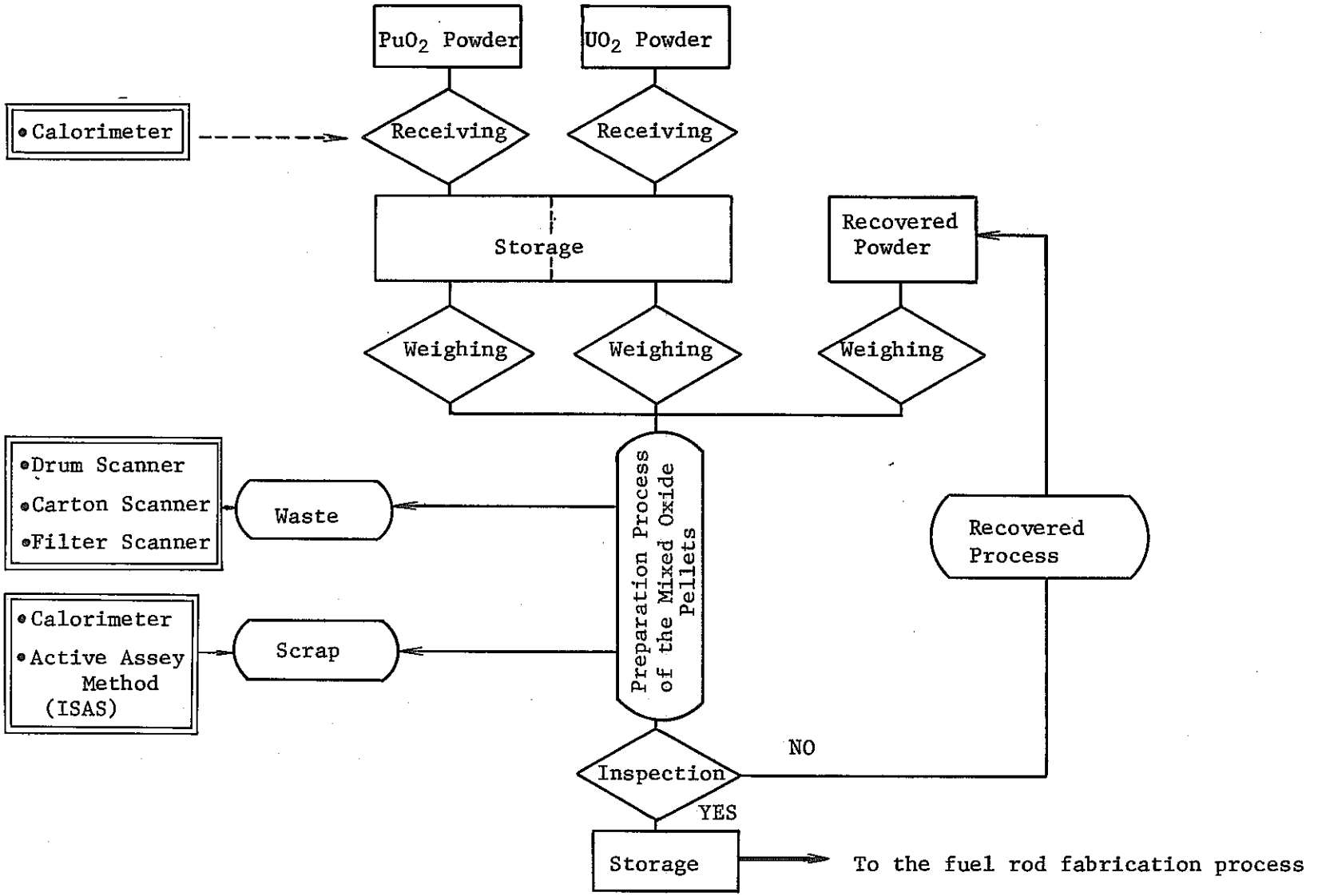


Fig. 1 Flow diagram of mixed oxide pellets fabrication process

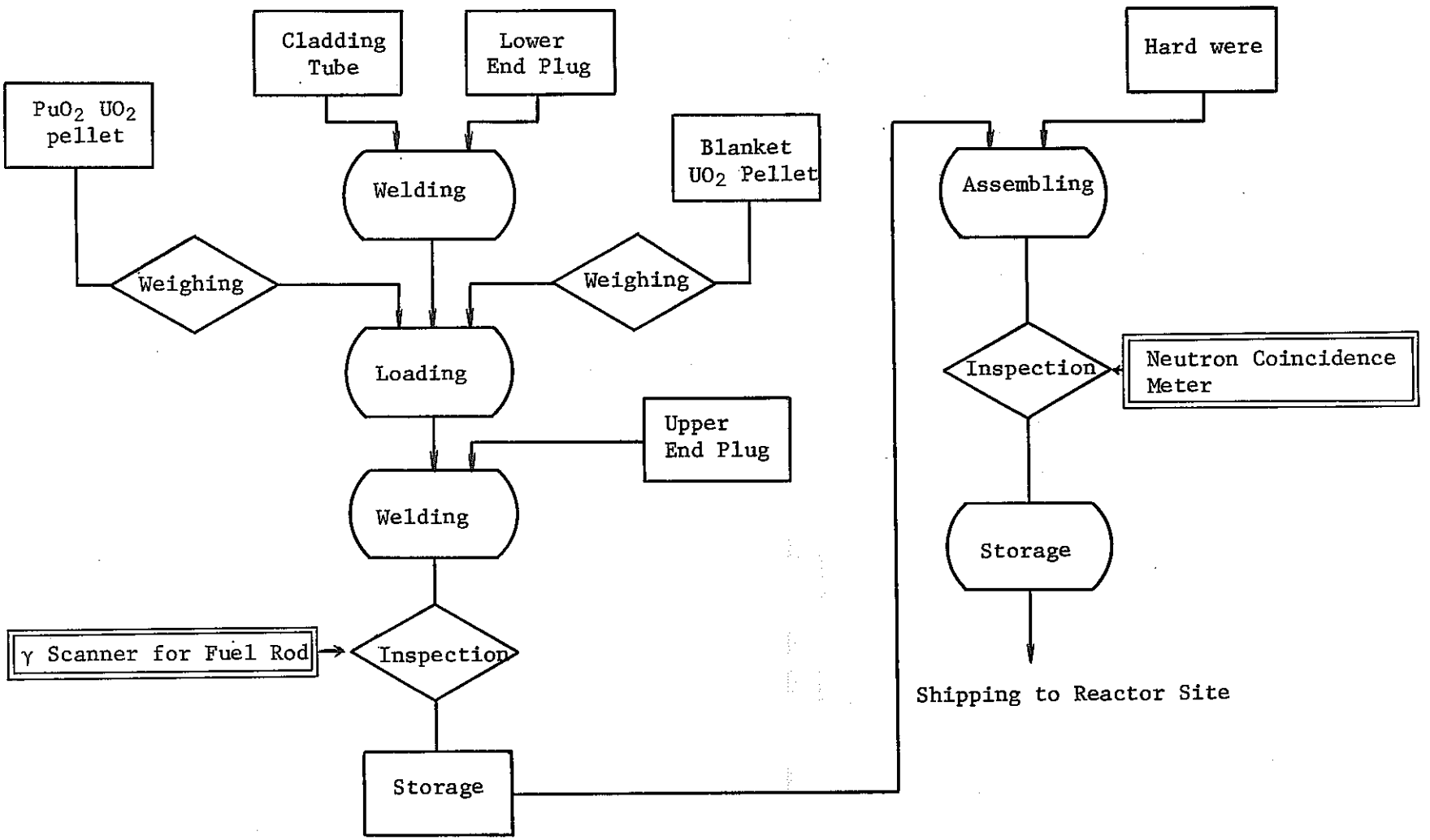


Fig. 2 Flow diagram of the fuel assembling process

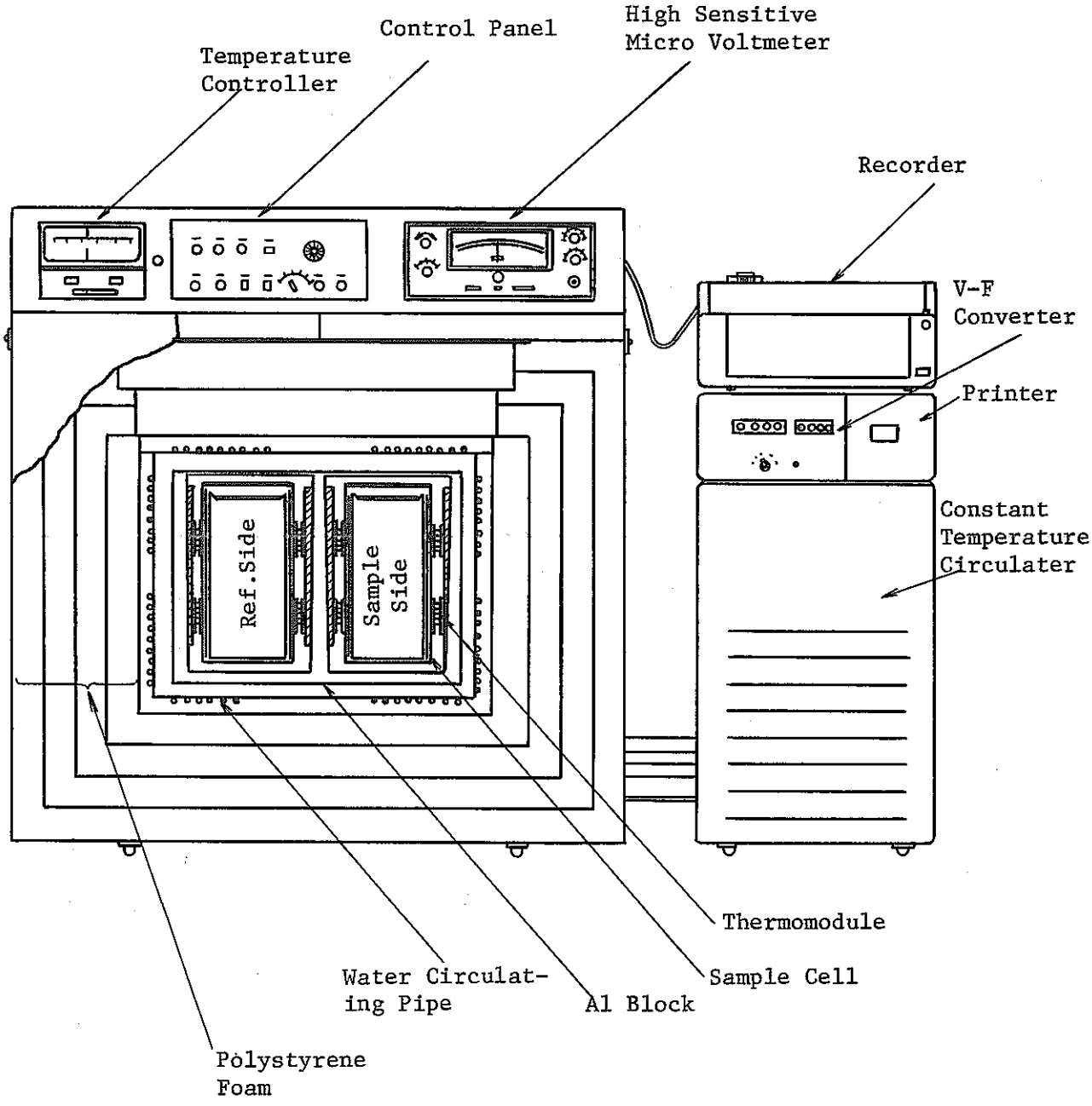


Fig. 3 Drawing of calorimeter

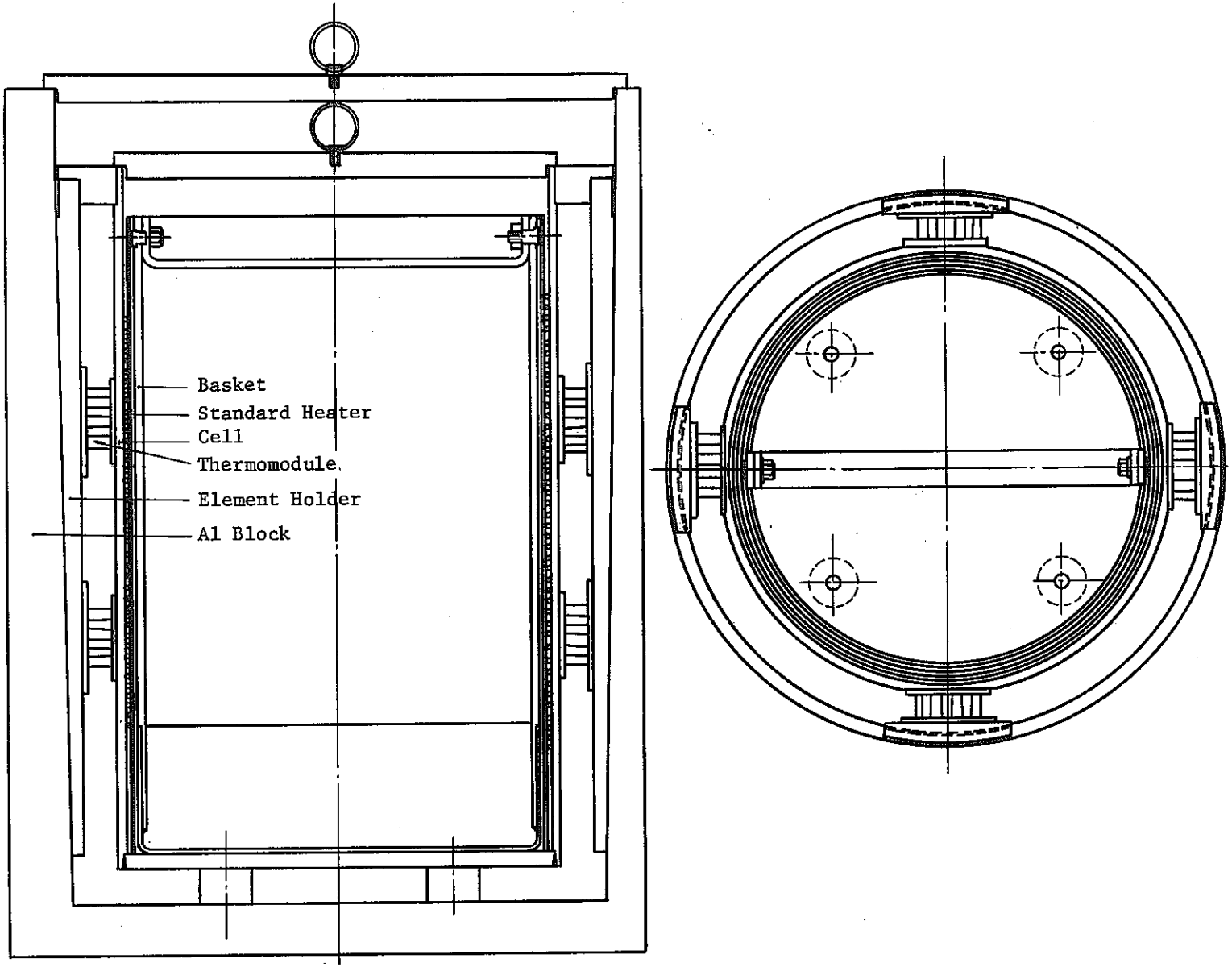


Fig. 4 Details of sample cell portion of the calorimeter

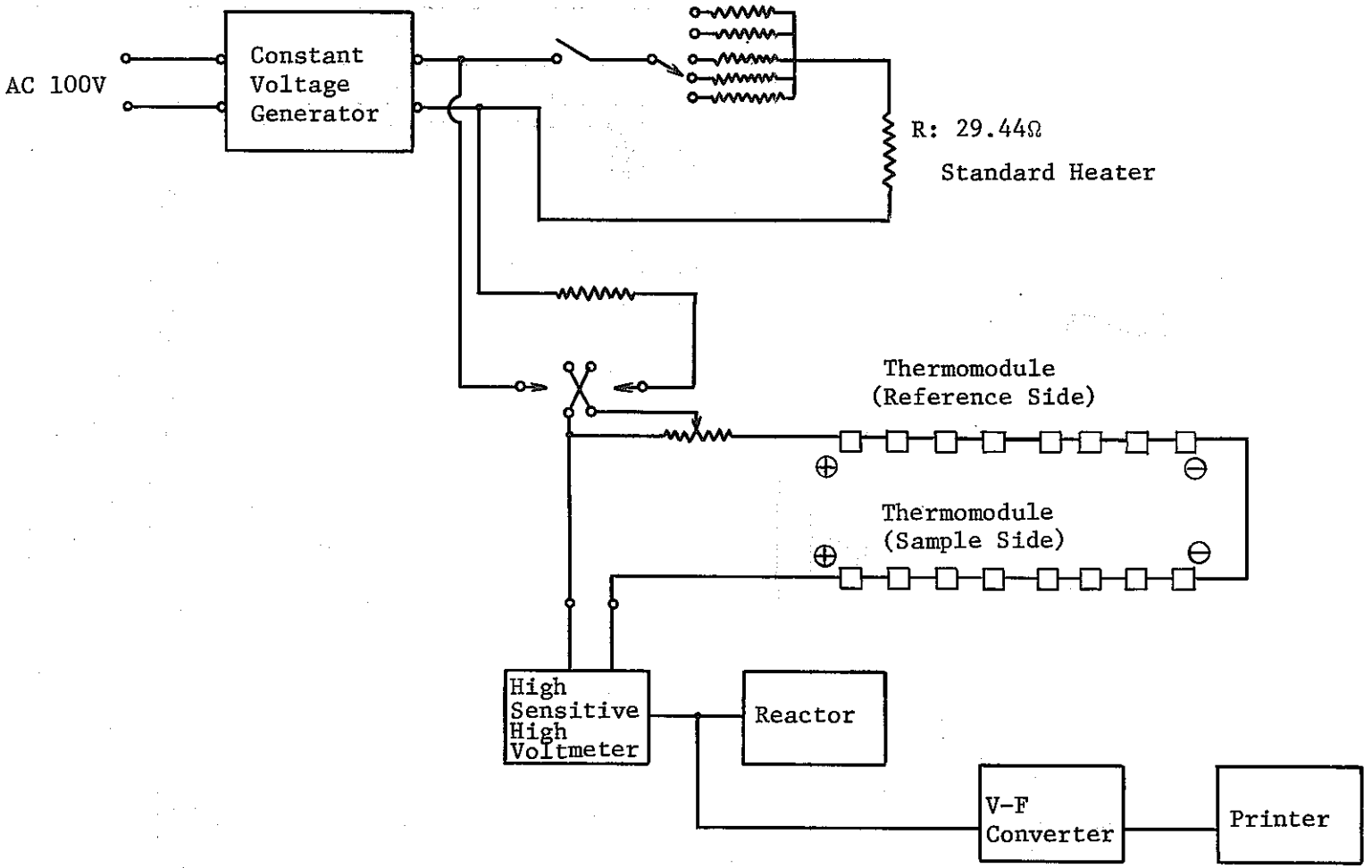


Fig. 5 Diagram of electrical circuit of calorimeter

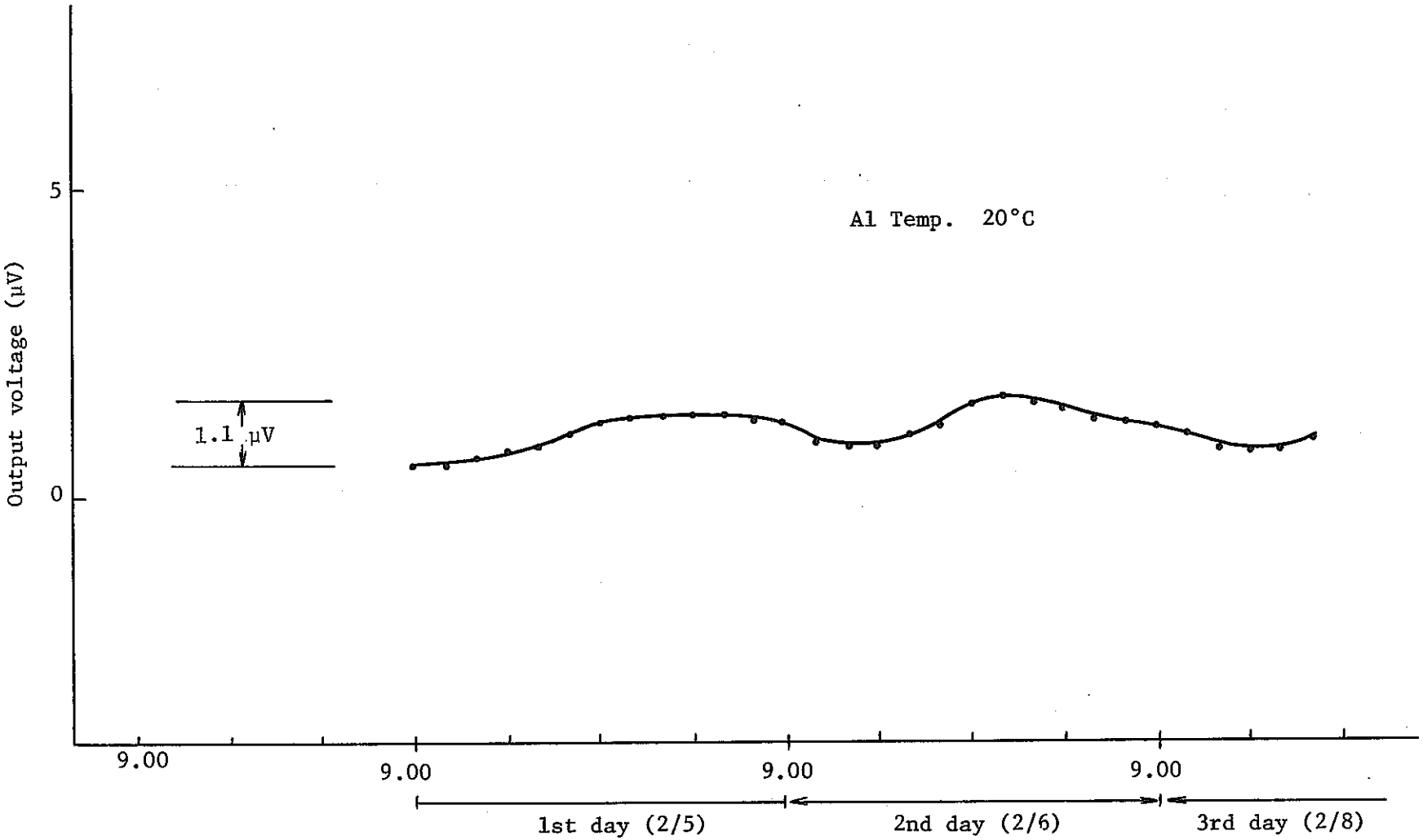


Fig. 6 Stability of zero point for calorimeter

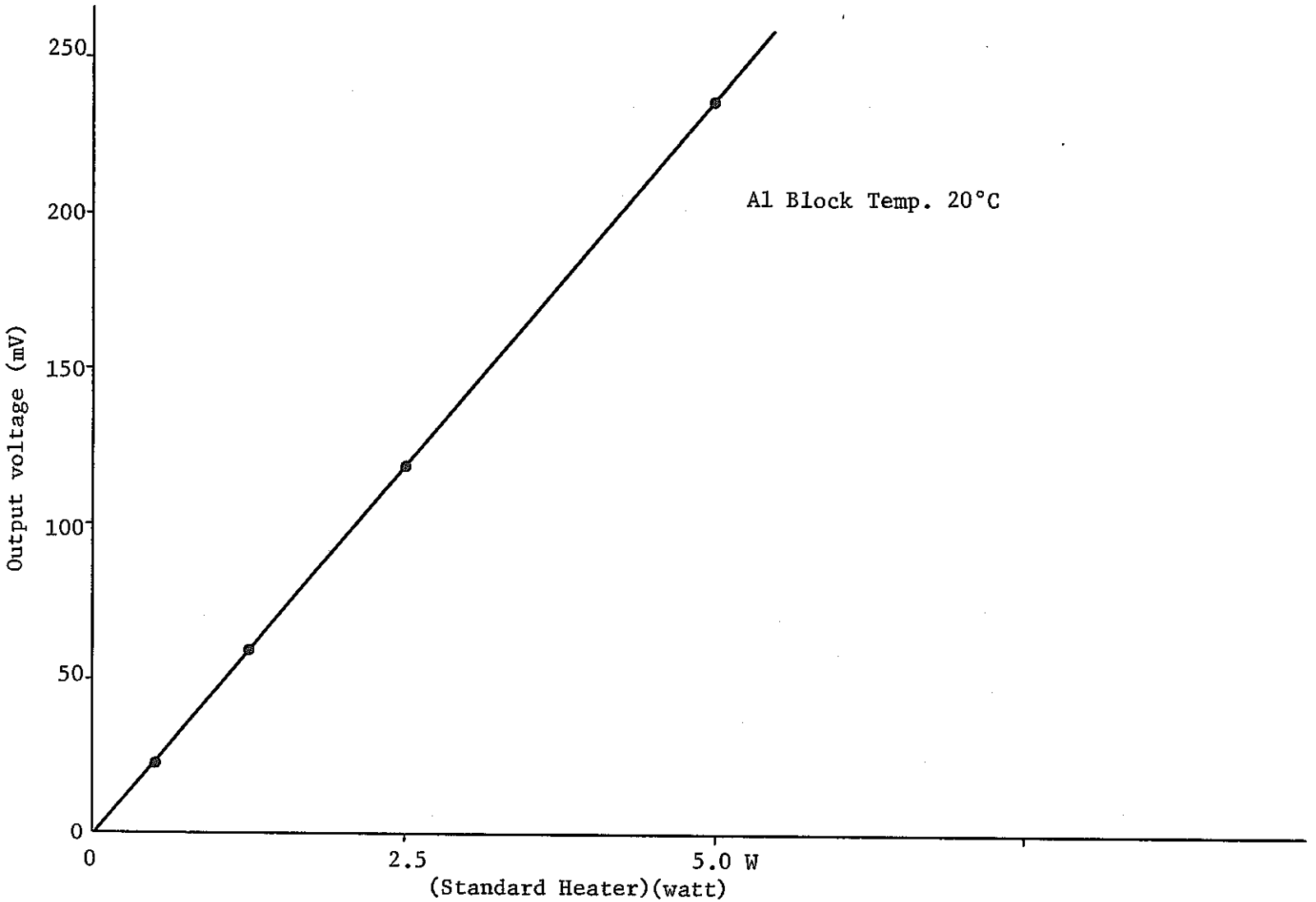


Fig. 7 Calibration curve by standard heater

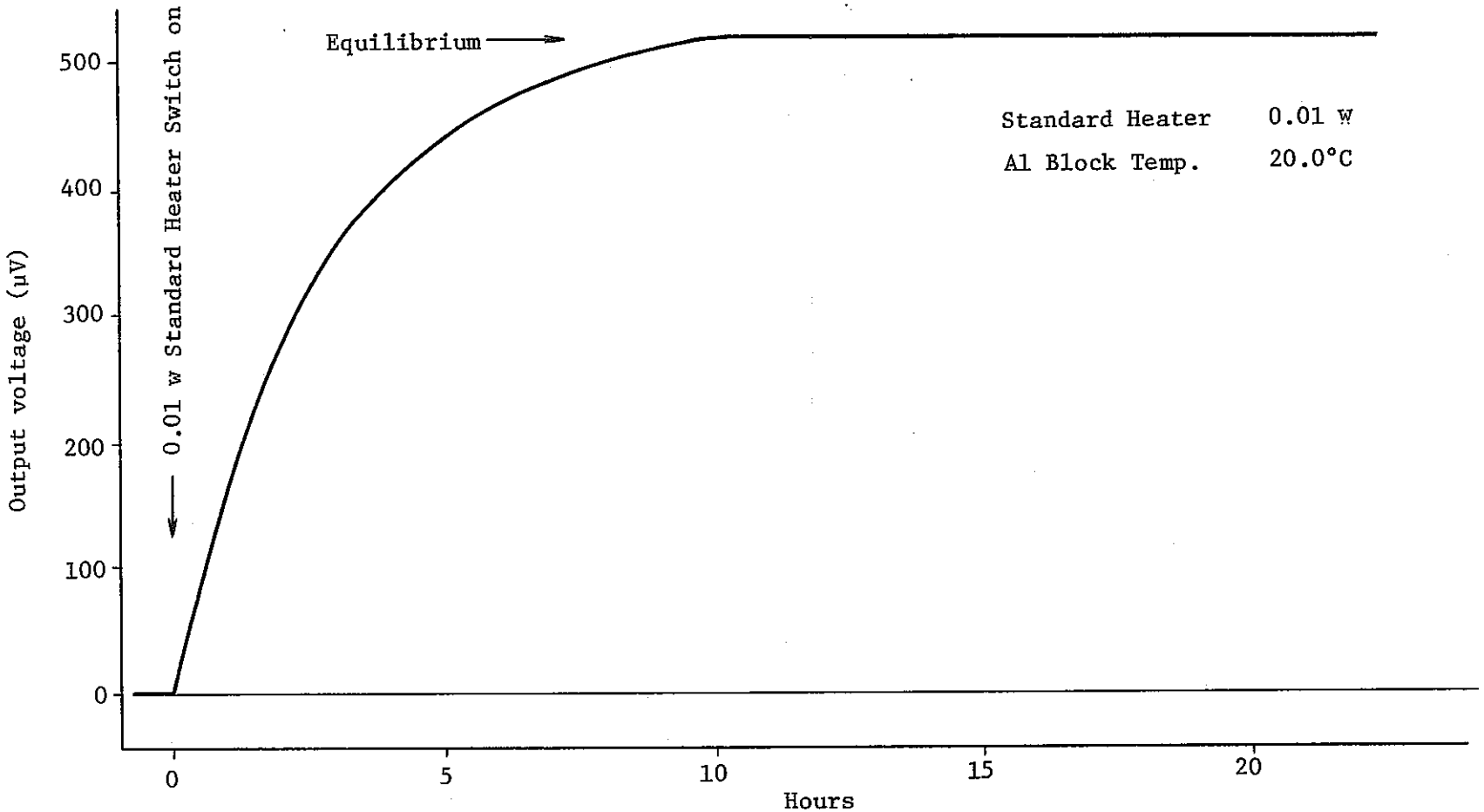


Fig. 8 Time to equilibrium by 0.01 w standard heater

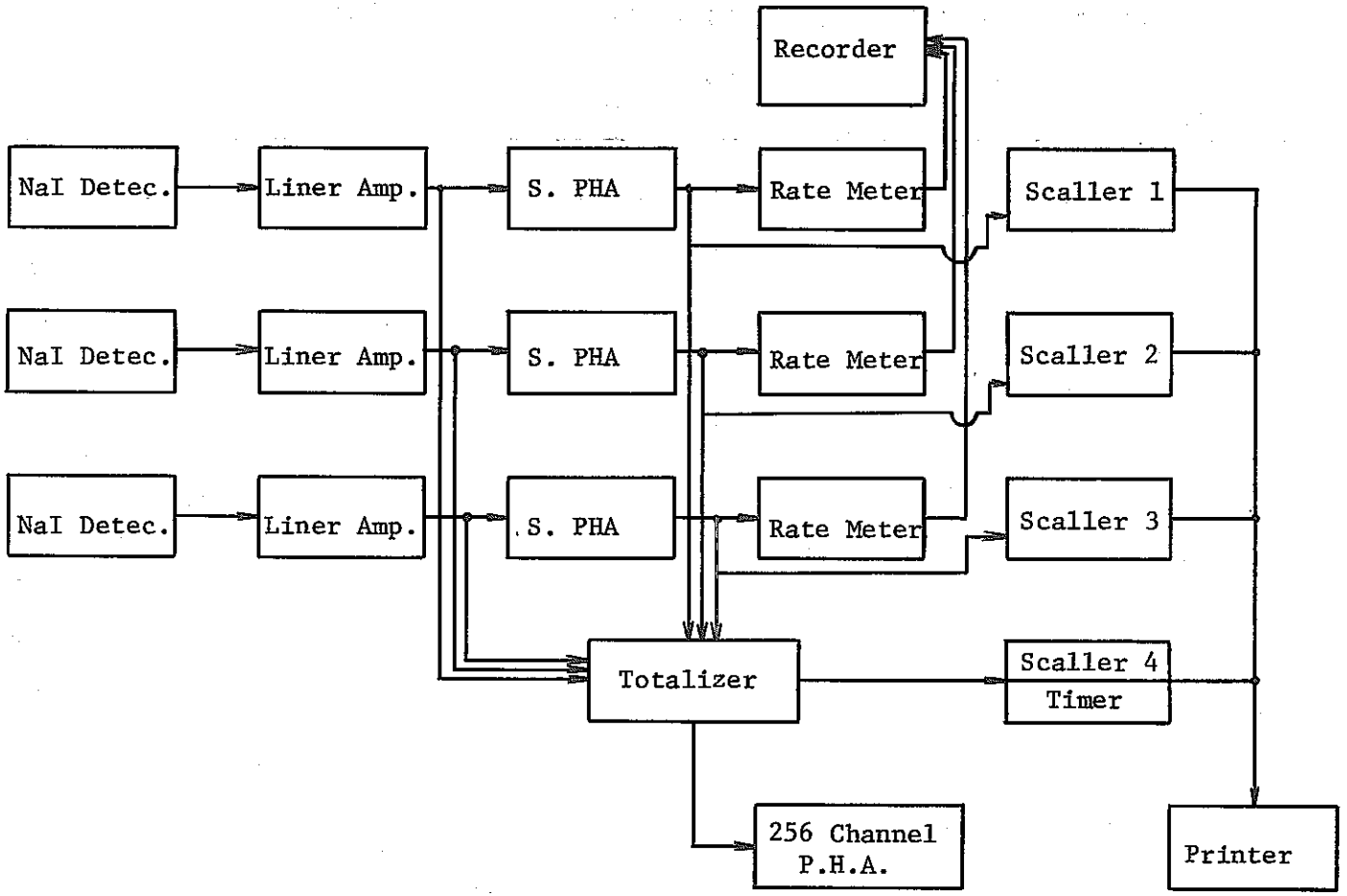


Fig. 9 Block diagram of electrical circuit for a γ-scanner

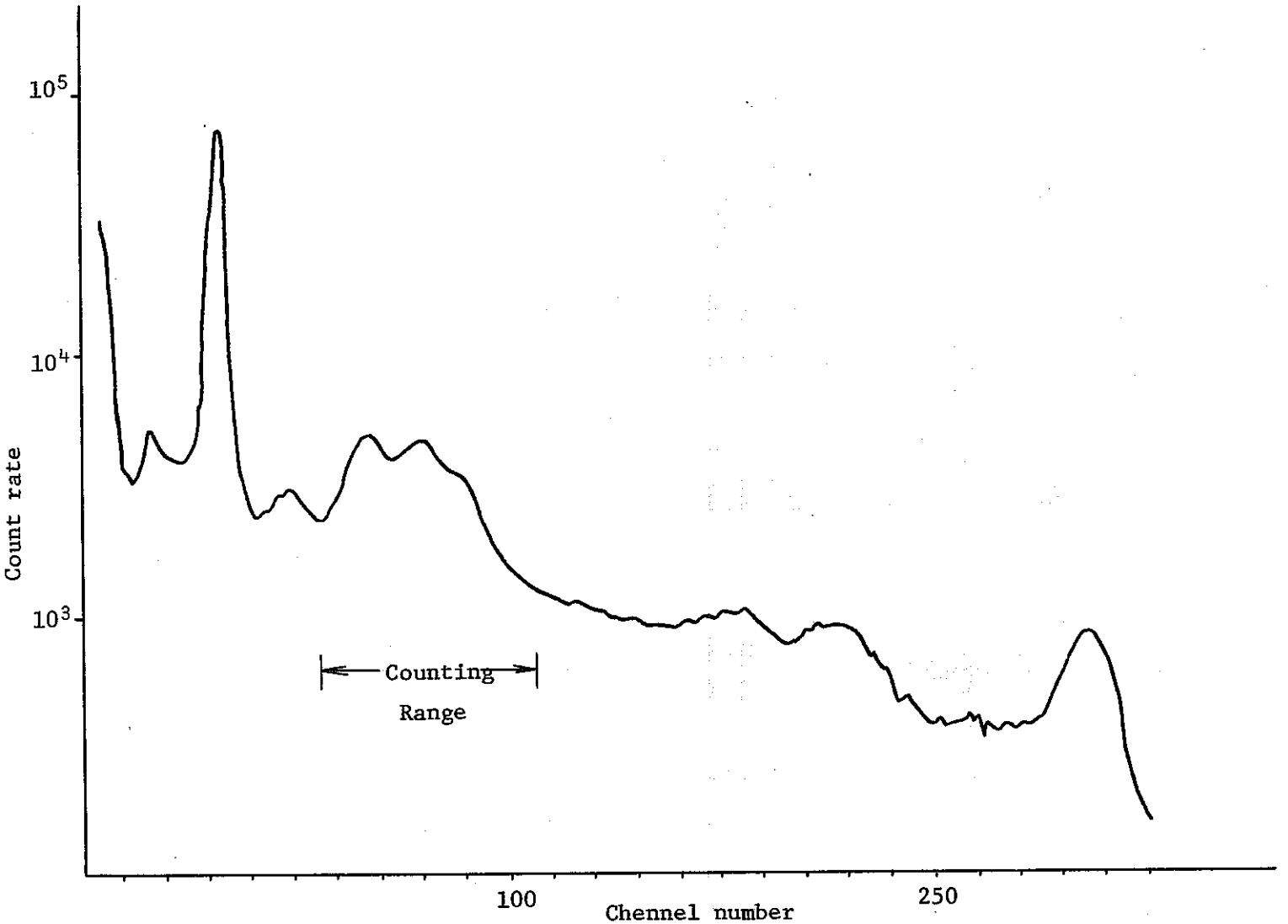


Fig. 10 γ ray spectrum for a PuO_2-UO_2 pellet loading fuel rod

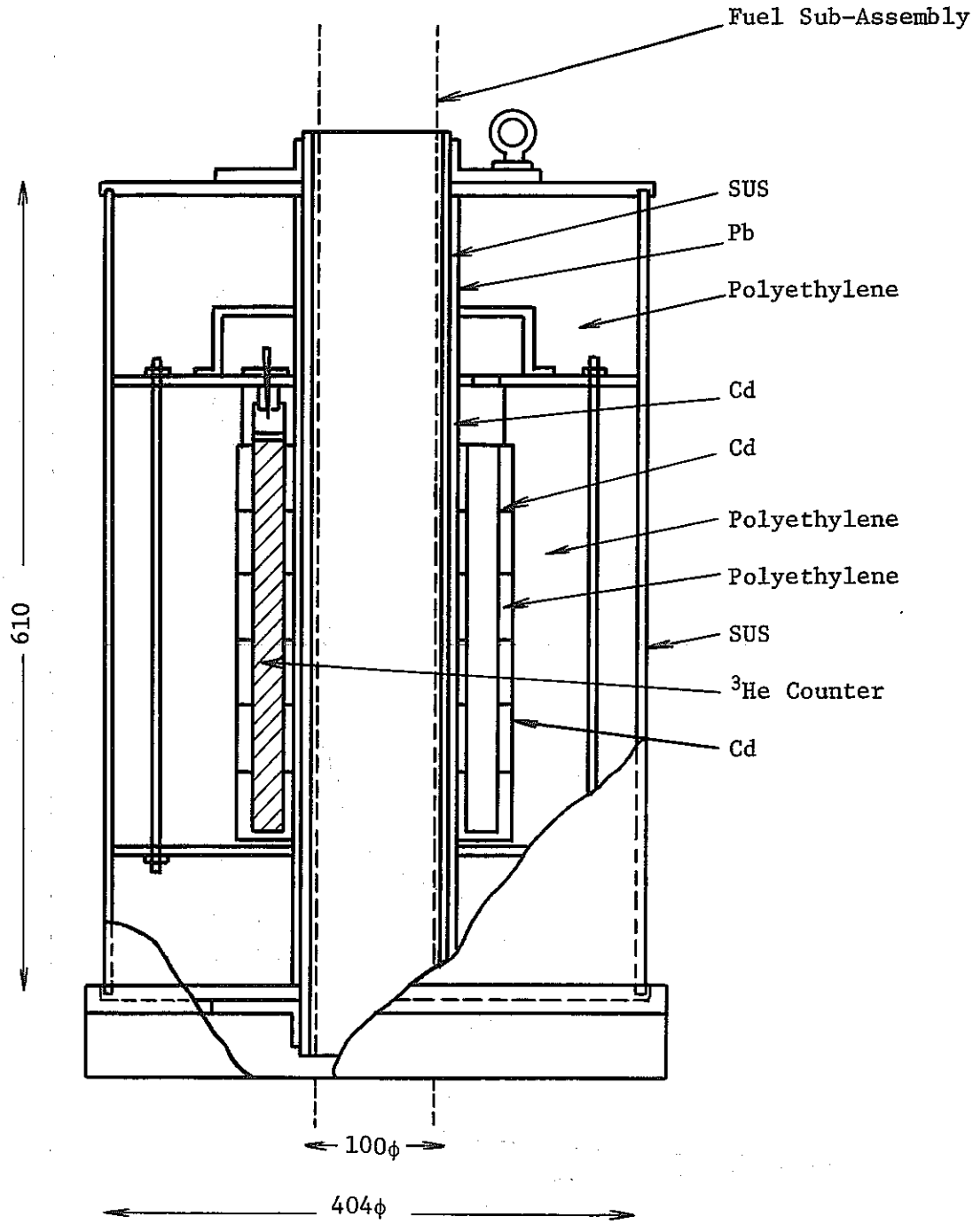


Fig.11 Detector assembly of neutron coincidence meter

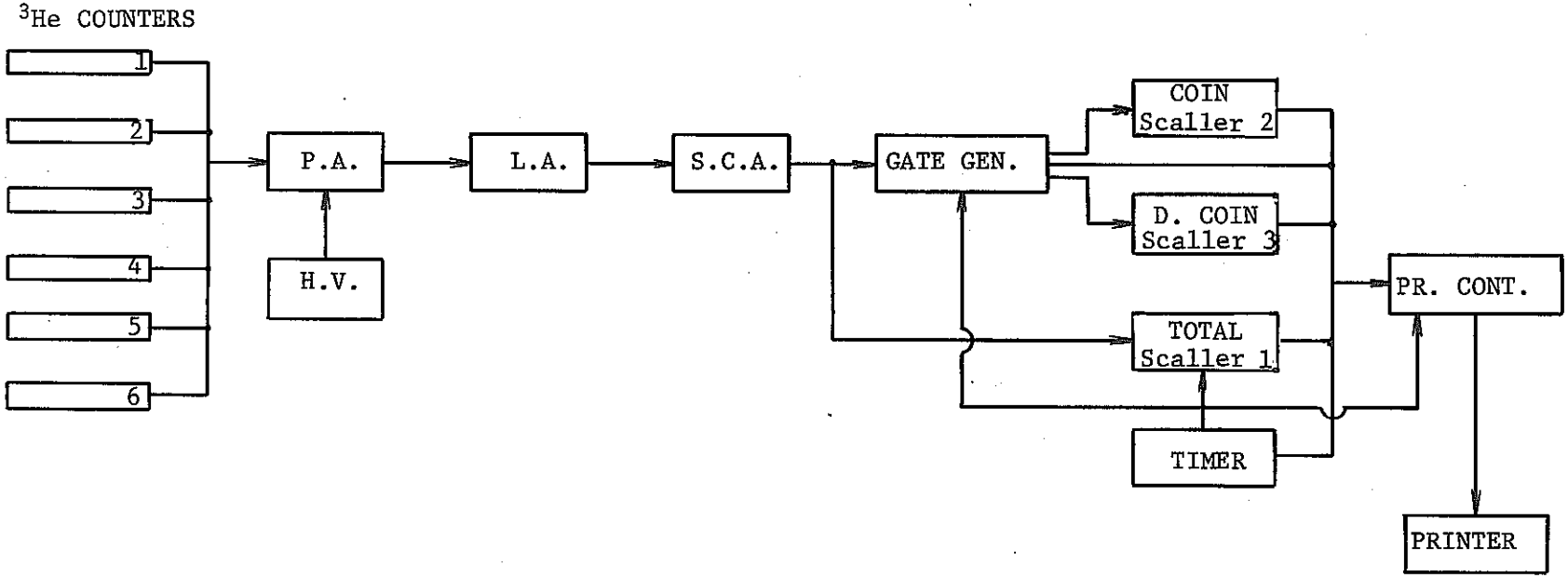


Fig. 12 Electrical system of neutron coincidence meter

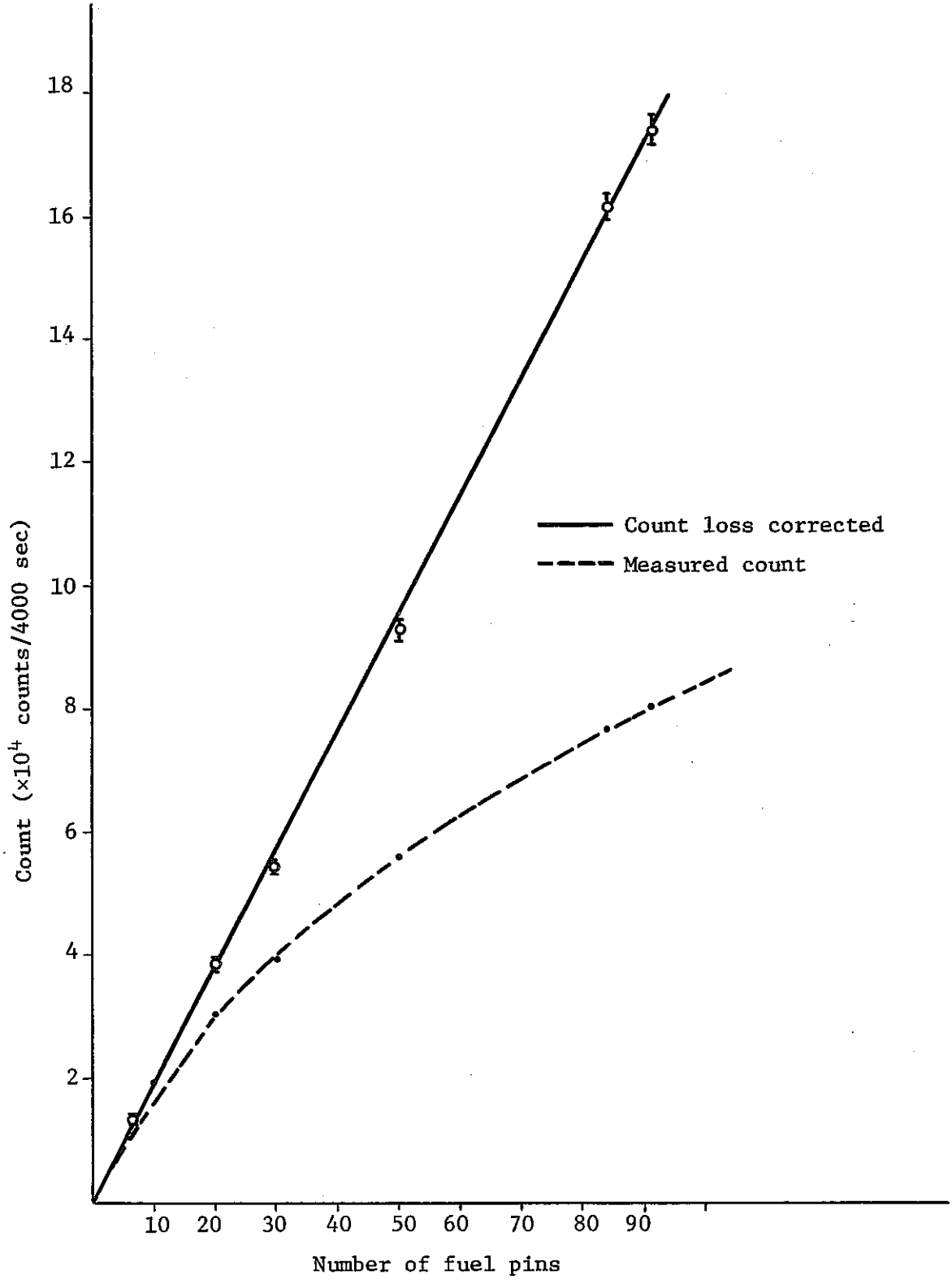


Fig. 13 Calibration curve by coincidence count

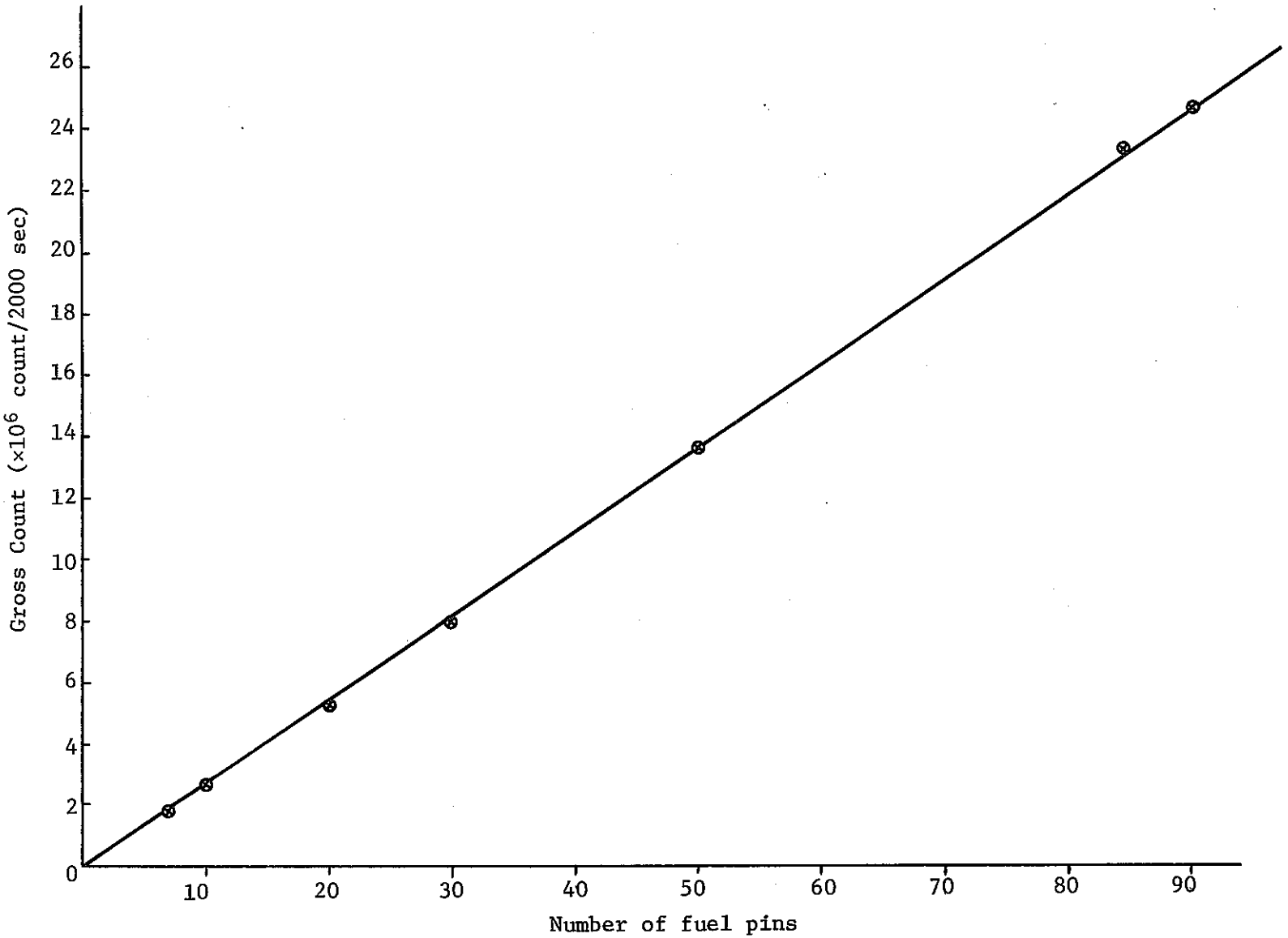


Fig. 14 Calibration curve by gross neutron counting