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The Irradiation of Liquid Plutonium Fuels in a Thermal Reactor

by

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ABSTRACT

In-pile test loops for irradiating liquid plutonium fuel alloys have been designed and operated in the Omega West Reactor. These loops provide a sodium environment for test fuel capsules at temperatures corresponding to those expected for LMBFR. Seven fuel capsules have been irradiated with up to 2% burnup of plutonium. Irradiation of two fuel specimens is currently in progress. The report describes the design, construction and operation of the irradiation loops and handling of the irradiated test fuel capsules.

ACKNOWLEDGEMENTS

Many people at the Los Alamos Scientific Laboratory have contributed to the work reported here. Particular thanks are due D. L. Welch, T. C. Christman, D. I. Morris, and H. M. Ruess for their contributions to the design, assembly, and operation of the experimental apparatus.

INTRODUCTION AND SUMMARY

This paper describes an in-pile irradiation program which has been in progress at the Los Alamos Scientific Laboratory (LASL) since January 1964. The purpose of this program is to obtain preliminary irradiation data on the liquid plutonium alloy fuels and tantalum containers that will be used in the Fast Reactor Core Test Facility (FRCTF).[1,2] Thus, the irradiations are performed under operating conditions and environment simulating those expected in the FRCTF; i.e., fuel-container interface temperatures are maintained above the fuel freezing point of approximately 440°C and below 725°C during all phases of reactor operation and shutdown. The irradiations are accomplished in LASL's 5.0 MW MTR-type reactor, the Omega West Reactor (OWR).[3] The technique developed employs a natural convection sodium loop located in the OWR core where the fast and epithermal neutron flux components are highest. A tantalum fuel capsule containing 24 g of Pu-Co-Ce fuel (6.2 g Pu/cc) placed in the loop operates at an average specific power of 150 W/g plutonium. With the present reactor operating schedule, 1% burnup is achieved in about 10 weeks.

I. IN-PILE TEST ASSEMBLY

A. General

Figure 1 shows the in-pile test assembly which consists of two separate parts; the outer part, the environmental cell, is semipermanently installed in the reactor and contains the convecting sodium loop, thermocouples, heater units, and a gas annulus for heat removal and temperature control. The inner part, an experimental insert, contains the test fuel and additional thermocouples; it is installed and removed as a unit from the environmental cell.

During nuclear operation, sodium is heated by the fuel and heaters and convects upward within an insulating flow divider. It then descends outside the flow divider, transferring heat through a gas annulus to the OWR water coolant stream. The heaters are thermostatically controlled to stabilize temperatures during periods of nuclear operation; during shutdown, they keep the fuel above its melting point of approximately 440°C.

B. Environmental Cell

The environmental cell is a 2-in. o.d., 22-ft long stainless steel assembly containing a 13-in. long convecting sodium loop in its lower section. The convecting sodium is doubly contained and consists of 400 g of hot-trapped sodium. A stainless steel flow divider, containing an argon-filled gas gap, minimizes radial heat conduction between the convective legs of the sodium loop. The top and bottom of the convective sodium channels are rounded to smooth the reversal of the sodium flow.

A heat leak gas annulus is formed by the primary sodium container and the outer tube of the cell. The gap is 0.020-in. thick over the lower nine inches and 0.012-in. thick over the top four inches at operating temperatures. The reduction in gap thickness at the top increases the convective sodium flow. A continuously flowing gas mixture (helium and nitrogen) is admitted to the annulus at the top and exhausted at the bottom via a tube passing through the outer leg of the sodium stream.

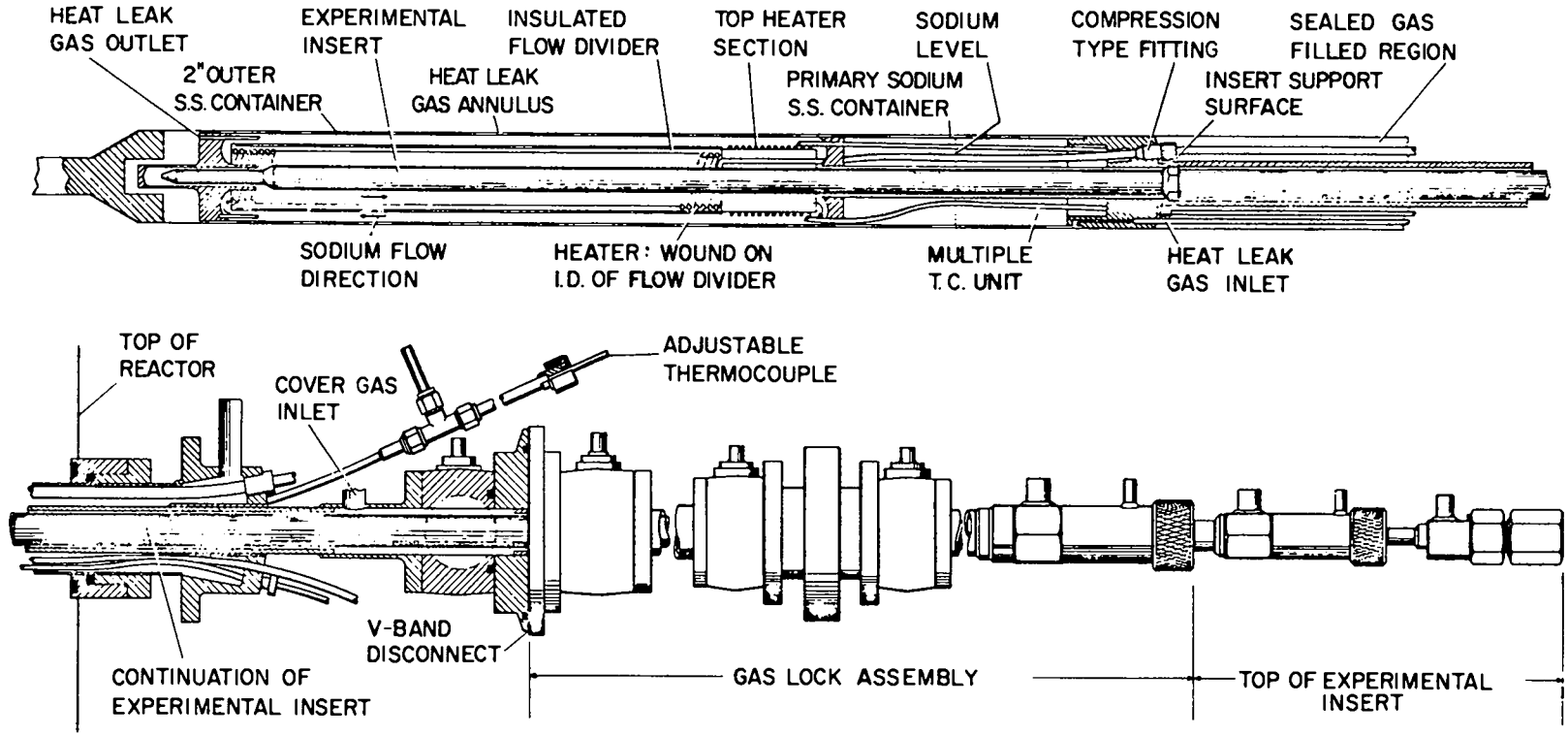

Fig. 1. In-Pile Test Assembly

The cell contains seven electrically powered heaters. Six of these are supported on the interior wall of the flow divider and are used for temperature control; the other is wound on the noninsulating upper section of the flow divider and is used only during sodium melting operations. The heater units (Fig. 2) were obtained commercially and are 27 ft long, Inconel-sheathed, MgO-insulated, Nichrome V cored, and have nickel lead wires. The heater section is swaged down to 0.093 in. o.d. over its 50-in. long active length. The six heaters are normally operated at a power of 5.0 kW, which gives a heat flux of 60 W/in.^{2} of sheath area. (The manufacturer suggests an operating range of 45-100 W/in.^{2} for the heaters if immersed in water or higher conductive media.) Prior to installation, the lower six feet of each heater are given a thorough radiographic and dye penetrant examination to determine sheath and heater integrity. Also, lead and insulation resistances are carefully measured. The 0.250-in. o.d. and 0.188-in. o.d. sections of each heater sheath are sealed to the cell with metal compression-type seals.

A multiple thermocouple unit, consisting of three stainless-steel-sheathed, chromel-alumel thermocouples swaged in an 0.125-in. o.d. stainless steel sheath, is located in the outer convecting sodium stream. The thermocouples are 0.031-in. o.d., MgO-insulated, and are ungrounded. A fourth sheathed thermocouple is located in the heat leak gas exhaust tube; an O-ring fixture at the cell top provides a sliding seal for this thermocouple so that the position of the junction is adjustable; this thermocouple can be used to obtain the axial temperature distribution in the outer convecting sodium stream.

Helium cover gas, at a pressure slightly above atmospheric, is maintained over the convecting sodium and in the upper sealed annular region of the environmental cell.

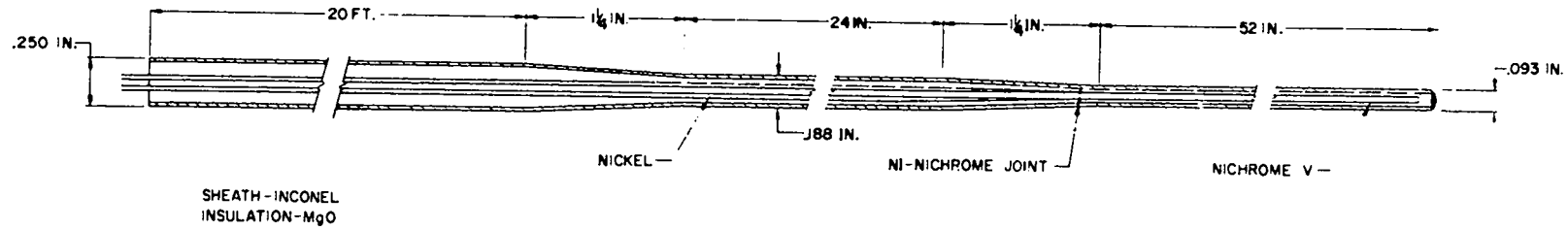

Fig. 2. Electrical Heater Unit

C. Insert

Figure 3 shows an insert typical of those which have been installed and operated in the cell. The insert is 28 feet long and contains a test fuel element in its lower section. The tantalum fuel element (0.420-in. o.d., 0.020-in. wall, 8-in. long) is filled to a height of about two inches with Pu-Co-Ce fuel (24 g Pu). The closed space above the fuel is filled with helium at atmospheric pressure. The fuel element is placed in an 0.575-in. o.d., 18-in. long tantalum container to provide secondary containment for the fuel. Sodium is used in the resulting annulus to provide a thermal bond. The junctions of eight 30-ft long chromel-alumel thermocouples are located at various axial positions in the annulus and are sealed at the top end of the insert by a multiple-penetration, compression-type fitting.

A Zircaloy tube located above the capsule is used for introducing the bonding sodium into the annulus. This tube is prefilled with hot-trapped sodium and installed in the insert during assembly. After final leak testing and radiography, the insert is evacuated and brought to a temperature of about 140°C to melt the sodium in the Zircaloy tube. During operation, this tube also continuously hot traps the bonding sodium.

To monitor the integrity of the fuel capsule, a helium sweep stream flows down the annulus between the 3/4-in. o.d. tube and the 3/8-in. o.d. tube, passes through the volume above the capsule, and is exhausted via the 3/8-in. o.d. tube through a molecular sieve. If gaseous fission products were released, they would be delayed for several hours in the sieve and detected by a nearby scintillation detector. The helium sweep stream is obtained from a diffusion cell to minimize contamination of the bonding sodium.

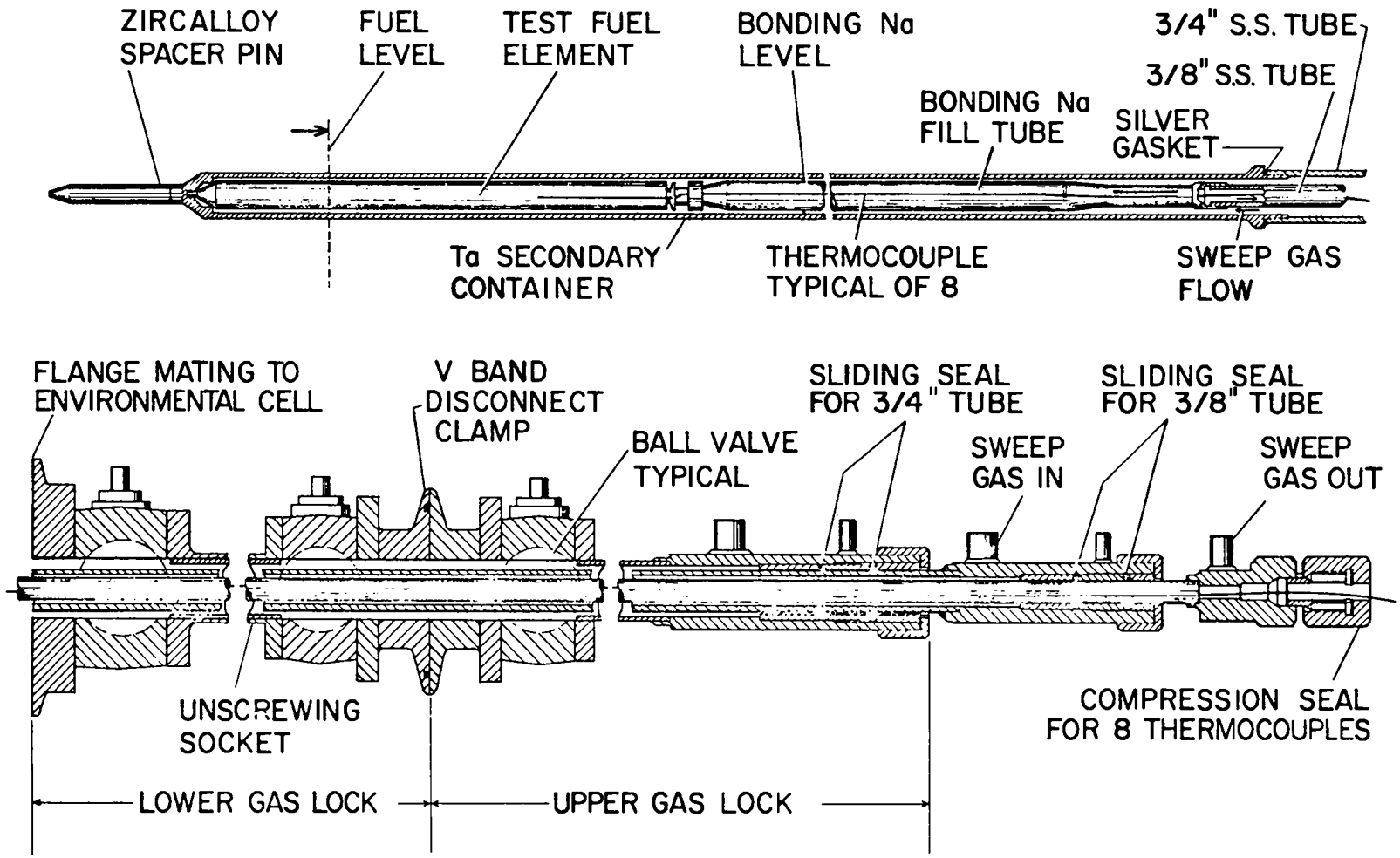

Fig. 3. Typical Experimental Insert

The 3/4-in. stainless steel tube is attached to the secondary tantalum container with a threaded joint which is sealed by a silver gasket. This joint provides a separation point during removal of the experimental insert from the environmental cell when the irradiation has been completed.

Two containers which act as gas locks are used in connection with the insert and remain in place at the top of the environmental cell during an irradiation experiment. These maintain a helium atmosphere over the convecting sodium during installation and removal of the insert from the cell and confine any activity which might be released by a fuel capsule. A double O-ring fixture on the top container provides a sliding seal for the 3/4-in. tube. A similar fixture seals the 3/8-in. o.d. tube to the 3/4-in. tube.

D. Control Circuitry and Instrumentation

The six heater units are electrically separated into two groups of three, each having identical control circuits. Figure 4 is a simplified schematic of the control circuit for one group. A variable autotransformer supplies power to the heaters from a 208 V source. The input to the transformer is controlled by mercury relays which are actuated by temperature control instruments. A wattmeter indicates the power drawn by the heaters. Each heater is separately fused and is fed by its own isolation transformer to prevent arcing in case of a heater lead-to-sheath short. Indicator lamps are incorporated between the heater leads and ground to give visual warning of insulation resistance deterioration.

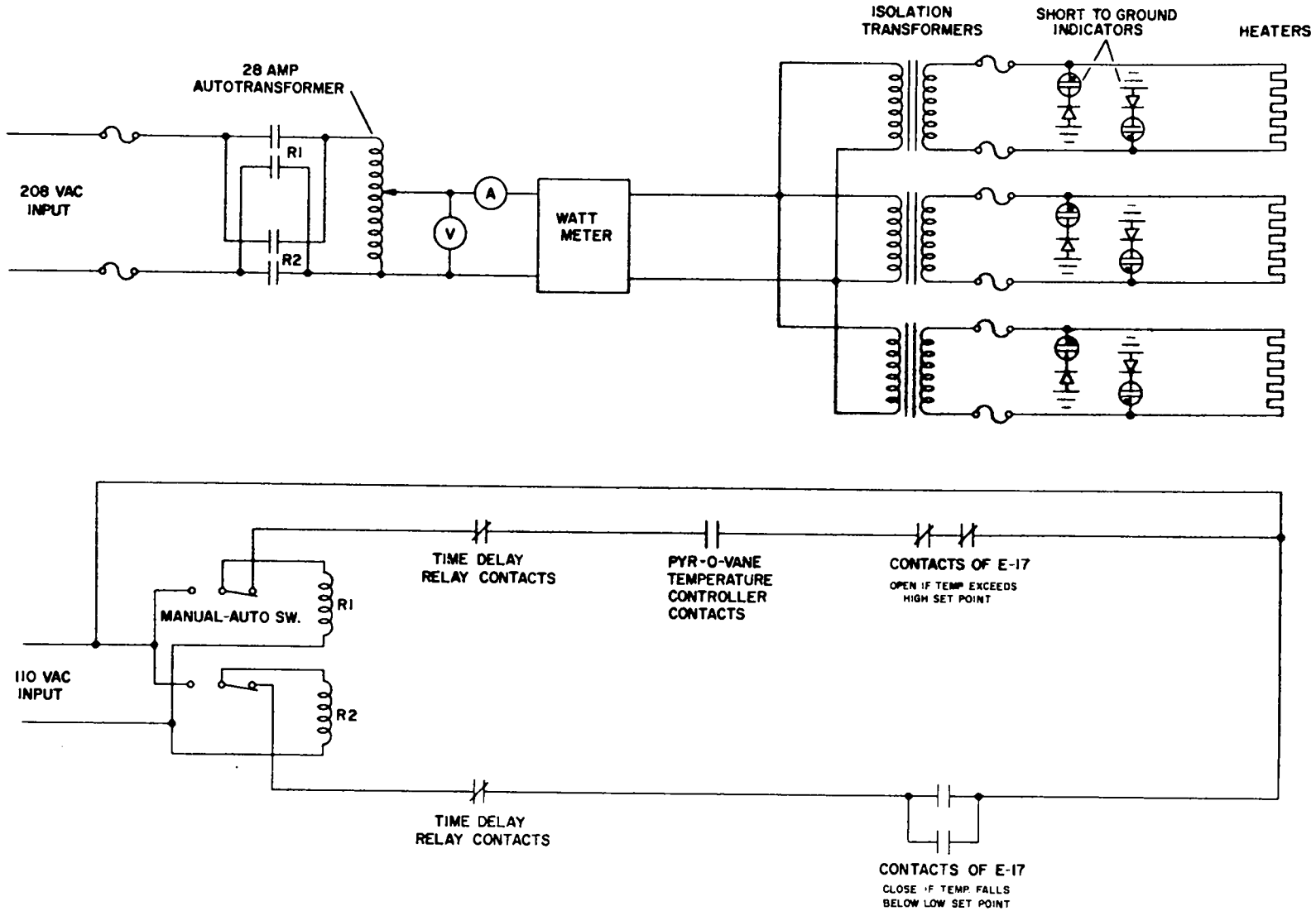

Fig. 4. Simplified Heater Control Circuit

Six of the thermocouples are connected to recording-only equipment; the remaining six thermocouples are connected to indicating- and recording-type control equipment. High and low temperature alarms obtained from these instruments are fed into the main reactor alarm system. If the temperatures exceed the control set-point, a 2-out-of-3-type network will scram the reactor.

Other annunciator circuits indicate the existence of high or low flow in the sweep gas and heat leak gas flowmeters, high or low radiation level from the sweep gas activity monitor, and loss of instrument power.

II. DESIGN AND PERFORMANCE

A. Thermal

This section of the paper discusses the thermal design and performance characteristics of the convecting sodium loop which has just been described and the neutronic characteristics of OWR fuel irradiation experiments.

Some desirable features of this design (Fig. 1) are as follows:

1. High heat fluxes, and resulting high temperature gradients, are restricted to the immediate neighborhood of the fuel. Consequently, heat fluxes across the gas gap are low, permitting larger gaps and giving a low film drop to the reactor coolant stream.
2. The heaters need not be confined to a small space, nor is thermal conduction through them a factor. Hence, commercially available heaters may be used, and they may be operated conservatively.
3. A wide range of control over temperature and power operating characteristics is available at any time through variation in the composition of the gas mixture flowing in the heat leak annulus.

During reactor operating periods, the heat source (fuel) is near the bottom of the rising leg of the convection loop, while the major heat sink (thinner gas annulus) is at the top of the descending leg. These locations favor a high convective flow, which is desired to minimize the vertical temperature gradient in the fuel sample. During periods of reactor shutdown, the heat source (electrical heaters) is not as well localized, so the convective flow is reduced.

The calculated temperatures around the convecting loop are shown in Fig. 5 for an assumed 5 kW fission or electrical source. The corresponding turbulent convective flows are 0.90 gal/min for fission heat and 0.66 gal/min for electrical heat. Measured temperatures and convective flow, based on power and temperature measurements, are consistent with these calculations.

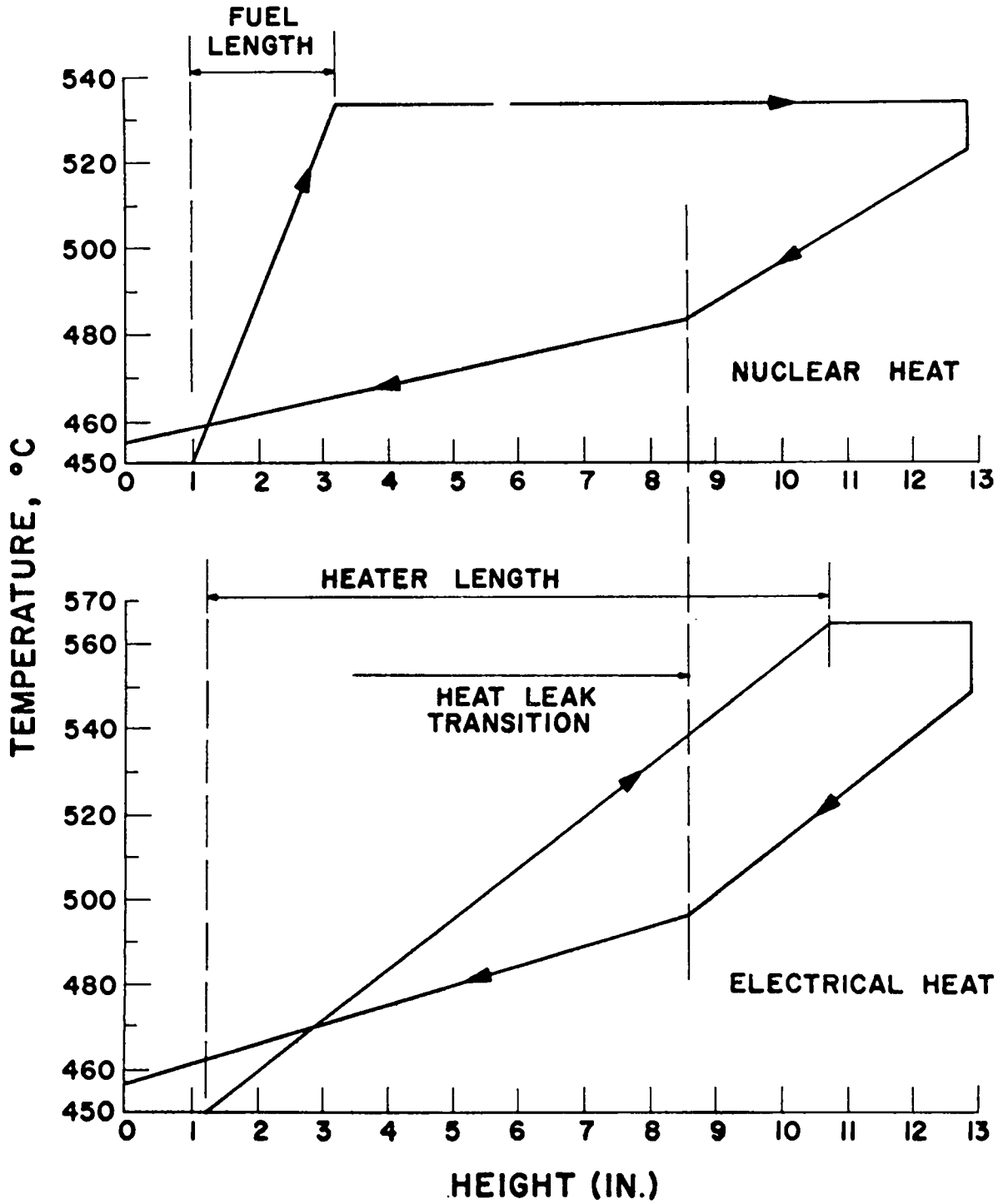

Fig. 5. Temperatures in Convecting Sodium Loop

The vertical temperature rise along the fuel during the above nuclear operation is about 80°C, and during electrical operation it is much less (about 30°C). The 80°C figure is comparable to that expected for the liquid plutonium fuel elements of proposed reactor designs, so these designs are well simulated in the present test samples. This temperature rise would increase, less than linearly, with test sample power; at a power several times present levels, the temperature at the top of the fuel would become excessive for long-term operation.

The effect of varying the composition of the He + N_2 gas mixture flowing through the heat leak annulus is shown in Fig. 6. No dependence on rate of flow was observed for pure gases or for a mixture over the range tested, 0 to 40 cc/min. The thermocouple indicating approximately the mean temperature in the descending (outer) leg was held at 540°C during these measurements. Early design calculations for pure N_2 and for pure He at a mean temperature of 487°C are indicated. A power range of about 3 to 12 kW can be spanned.

During operation, the gas mixture is adjusted to give approximately the desired conditions, after which fine control is maintained by the thermostatically controlled electrical heaters. The only automatic change in gas mixture is the rapid introduction of N_2 under certain emergency conditions to bring power requirements within the range of the electrical heaters (5 kW).

At full reactor power, the radiation heating produced in the loop containing a fuel capsule with 24 g plutonium is 5.6 kW -- 2.0 kW gamma heat and 3.6 kW fission heat. Gamma heat (1 W/g) was measured by placing a stainless steel dummy capsule in the sodium loop and observing the difference in electrical power required to maintain a constant average temperature drop across the heat leak annulus with the reactor at zero power and at 5.0 MW. The fission heating was measured in a similar manner. The value obtained for the fission heating by this method agrees within 15% with the values obtained by radiochemical analysis.

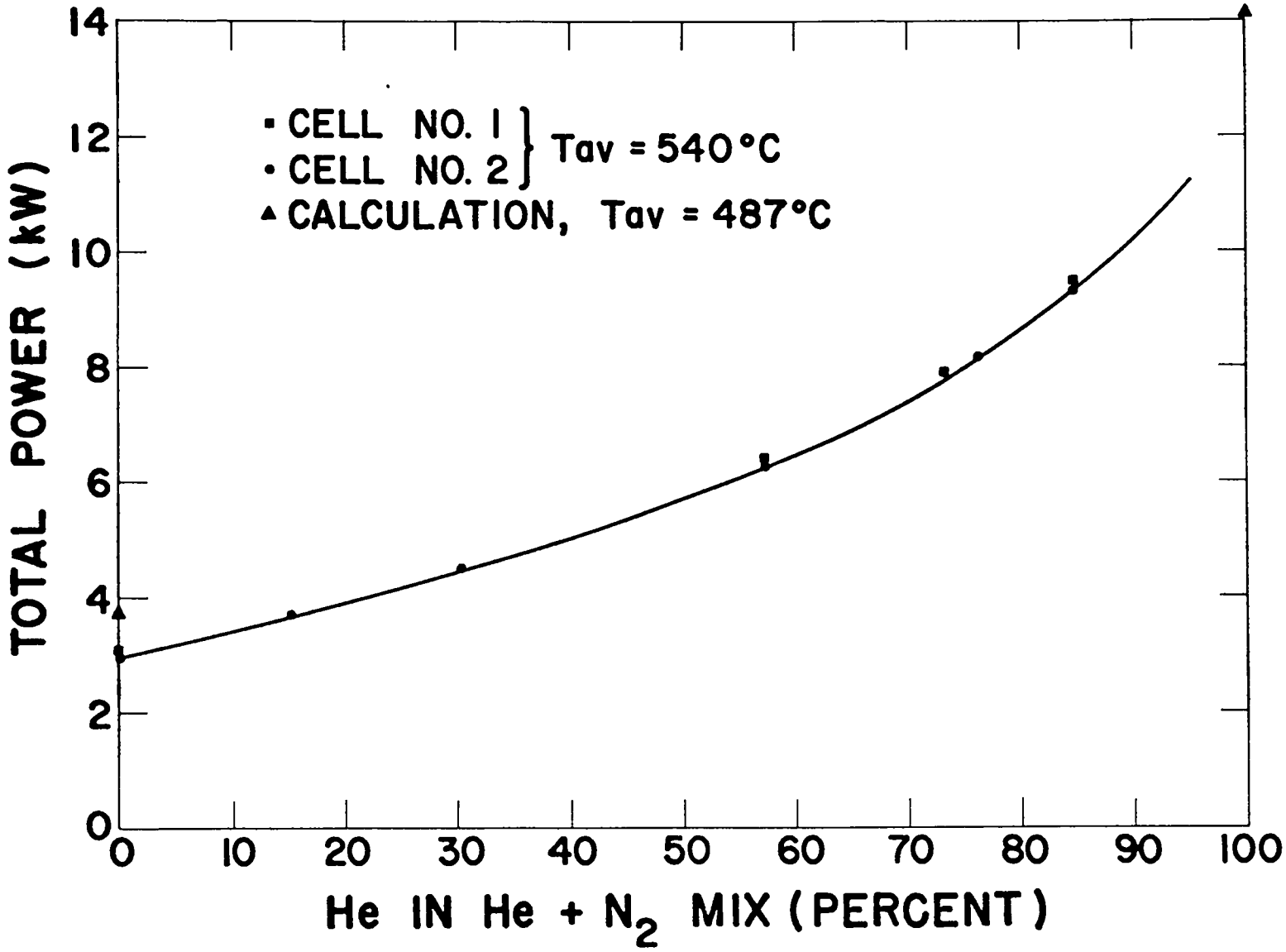

Fig. 6. Effect of Gas Mixture on Power

B. Neutronic

Before starting detailed design and construction of the irradiation experiment, preliminary neutronic calculations indicated that useful data could be expected from such irradiations. The calculations were made using a simplified one-dimensional cylinder model of the OWR reactor with a cylinder of fuel along the axis. Material compositions were realistically specified in 65 mesh intervals which were closely spaced within the fuel and in its neighborhood. The DTK code used was of the Carlson[4] type, based on transport theory in S_4 approximation. The Hansen-Roach[5] 16-group cross section library was used.

Flux distribution for the present design, containing fuel of 5.7 g Pu/cc, is indicated in Fig. 7. Here the thermal flux and the fast flux (combined three top groups, energy over 0.9 MeV) are shown. The thermal flux is depressed by about a factor of five at the fuel surface.

The corresponding power distribution is indicated in the upper curve of Fig. 8. The lower curve is for a dense fuel sample surrounded by a thick cadmium neutron filter. This calculation was compared with experimental results obtained in an early (1960) experiment in OWR, using LAMPRE I fuel (Pu-Fe eutectic, 16 g Pu/cc). This comparison was used to obtain a normalizing factor for use in this and other calculations. The two curves drawn represent the normalized power distributions, while the points shown were obtained by radiochemical analysis of solid specimens of fuel which were irradiated for short times at low power. Samples for the latter were taken at various radii by machining on a lathe. The horizontal lines on the points indicate the radial extent of the samples where it is significant. The experimentally determined edge-to-average ratio for the 6.2 g Pu/cc fuel was 5.1, while the calculated value for the 5.7 g Pu/cc fuel, the nearest calculation available, was 5.7. Respective average specific powers were 143 and 158 W/g Pu. The lower curve was normalized to the experimental value of 35 W/g Pu. The edge-to-average ratios were 1.4 (calculated) and 1.5 (experimental).

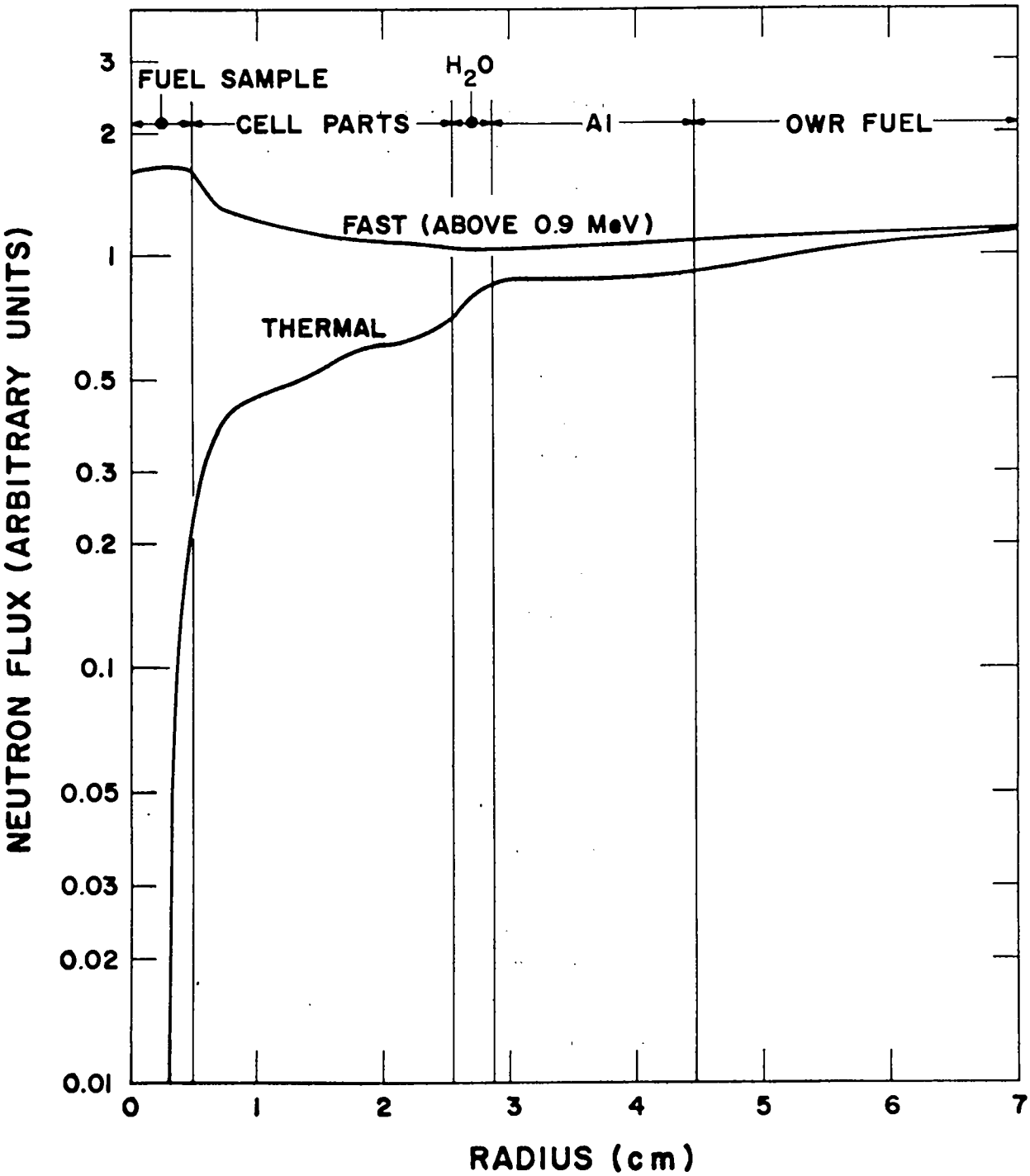

Fig. 7. Flux for 5.7 g Pu/cc Fuel in Present Design

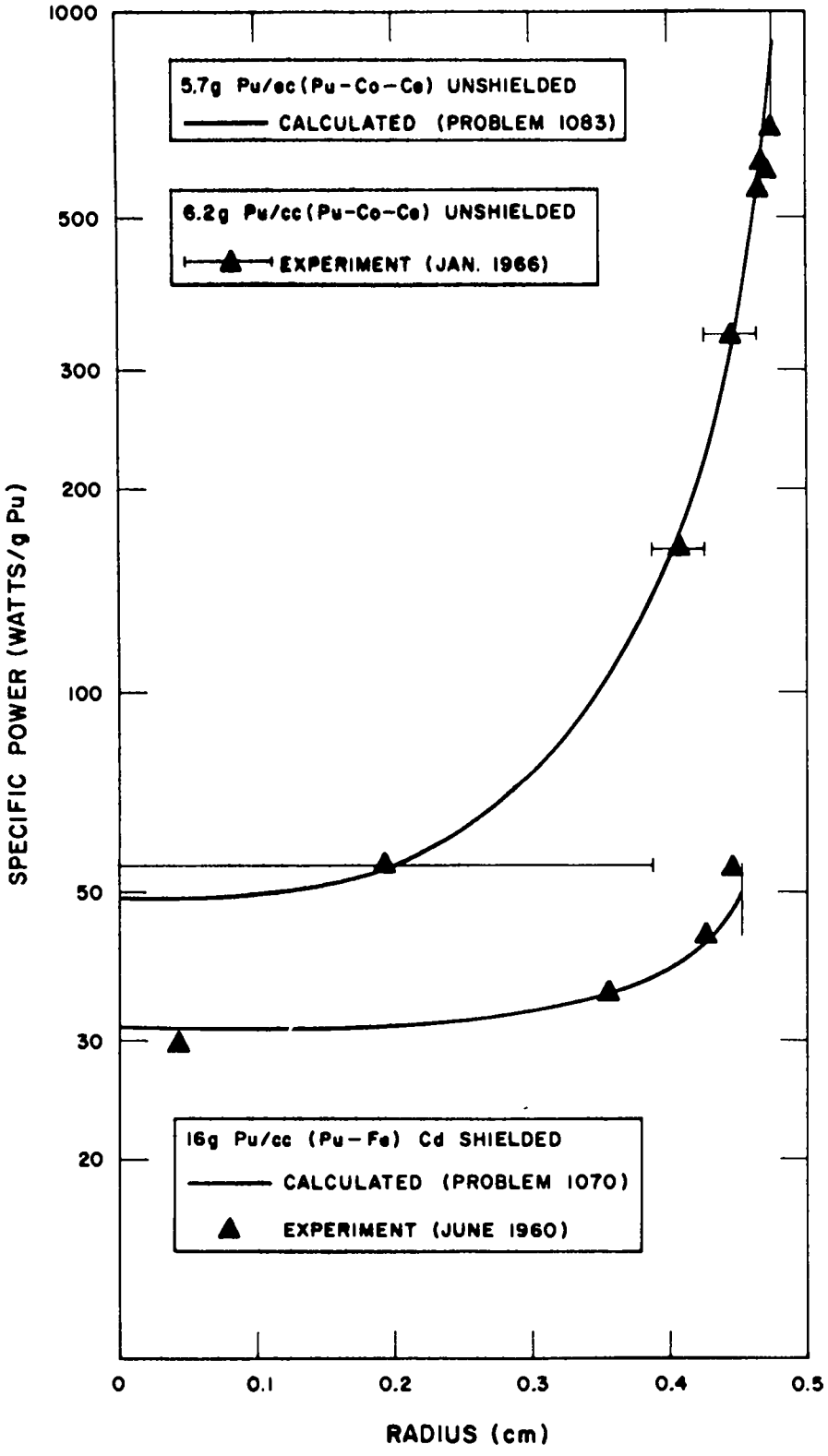

Fig. 8. Radial Power Distributions

A calculational study was made of the effect of varying the thickness of a natural boron filter for the case of 3.0 g Pu/cc fuel. Results (Fig. 9) indicate that a serious loss in average power is involved for even a moderate improvement in edge-to-average ratio.

The edge-to-average ratio without filtering is a function of the plutonium content of the fuel, as shown in Fig. 10. Analysis of the related fuel temperature distributions indicated that, for liquid fuels in the range from 3 to 8 g Pu/cc fuel, the power distribution of unfiltered test samples was satisfactory. Comparison was made to samples with the same average power density but of uniform radial power generation. Both central temperature rise and the force inducing fuel convection were found to be about 40% (for 8 g Pu/cc fuel) to 50% (3 g Pu/cc fuel) of those for the uniform distribution. Hence, it was decided to omit neutron filters in the design. Actually, the high power density at the tantalum wall may be considered advantageous in overtesting certain interface effects such as fission product recoil damage.

The average power density of Fig. 10 increases rather slowly with the plutonium content of the fuel. For the denser fuels, it appears to consist of a constant part, about 1000 W/cc, plus a linear part having a slope of about 42 W/g Pu. These are probably attributable to nonpenetrating and to penetrating components of the flux, respectively.

Figures 9 and 10 are for a preliminary design of the irradiation experiment. The final design, used for Figs. 7 and 8, contains more tantalum and gives lower power densities -- presumably at the expense, primarily, of the nonpenetrating component of flux.

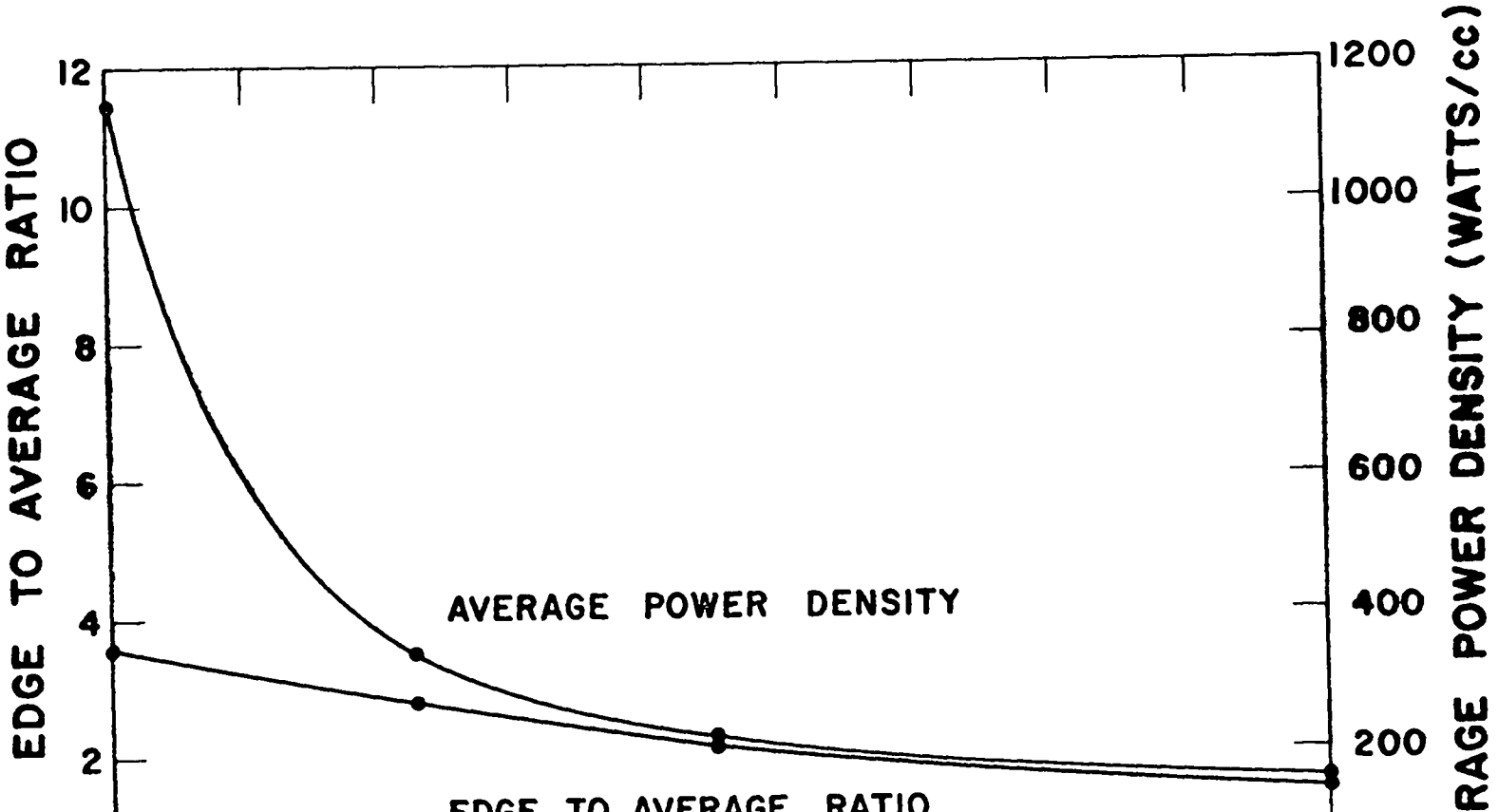

Fig. 9. Effect of Natural Boron Neutron Filters

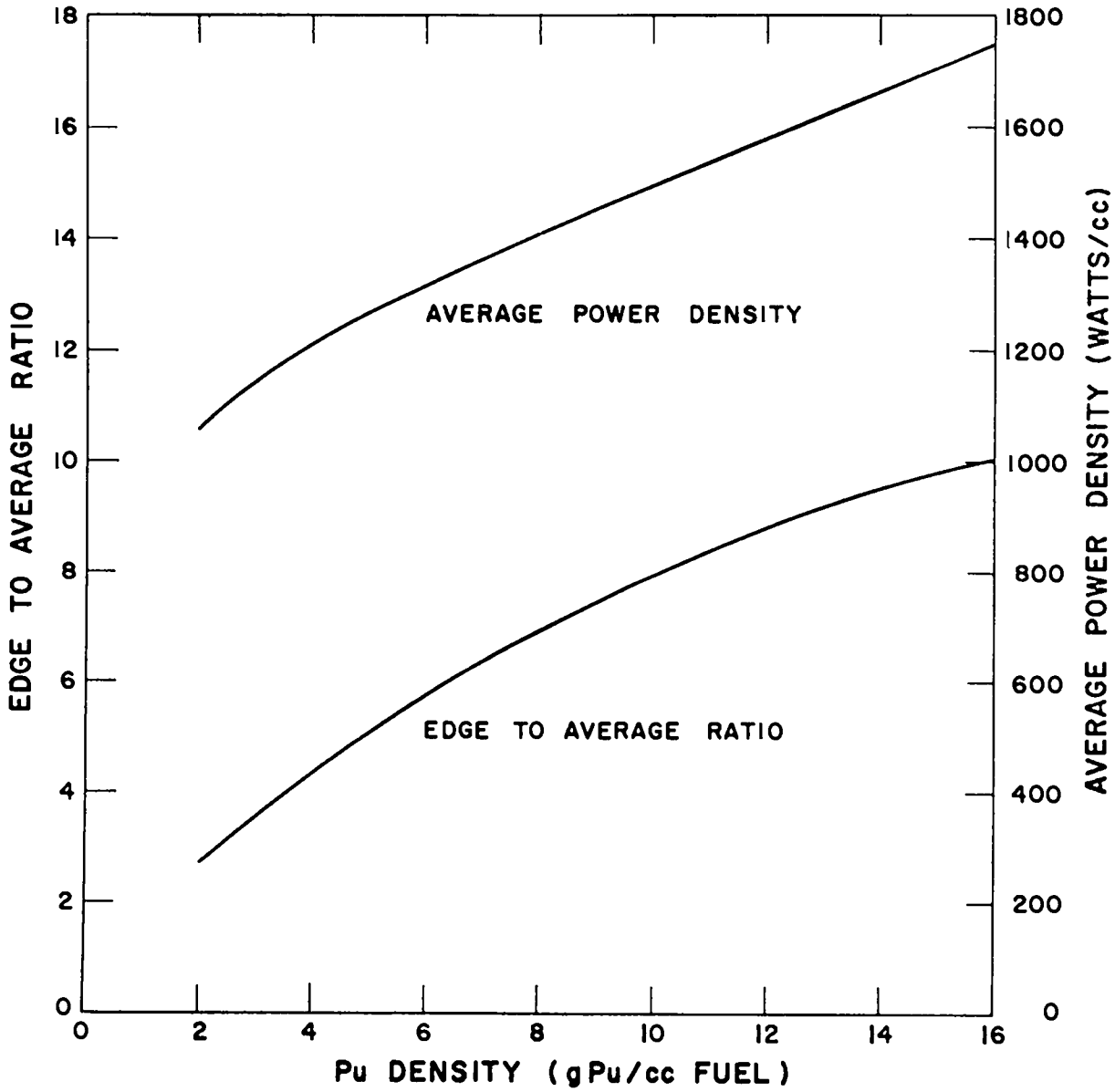

Fig. 10. Effect of Plutonium Content of Fuel

III. LOOP OPERATION AND HANDLING

A. General

There are two environmental cells installed in the OWR at the present time. The first cell was installed in March 1965, and the second in December 1965. Each cell has been operated with a variety of inserts as follows:

1. A nonfueled insert containing a stainless steel capsule to measure gamma heating,
2. A solid-fueled insert to measure the fission power distribution,
3. A liquid-fueled insert for system checkout.

Other irradiations included a fourteen-week irradiation to achieve 1% burnup. Presently one cell contains a fuel element which has achieved 2% burnup to date, and the other cell contains a fuel element which has achieved 1% burnup.

The electrical and gas control systems have been designed so that the loop operation requires very little manual attention during all phases of reactor operation and shutdown. Daily readings of the system parameters are taken and weekly checks of the control and alarm circuitry are performed.

B. Operation

When the reactor is shut down, nitrogen is kept in the gas annulus and about 3.0 kW of electrical heat is required to maintain the system at operating temperature. When the reactor is at full power and the cell is loaded with 24 g Pu of the 6.2 g Pu/cc alloy, about 5.6 kW of radiation heating (2.0 gamma heat + 3.6 fission heat) is produced in the loop. An additional 2.5 kW of electrical heat is used to stabilize the system temperature. A gas mixture consisting of helium and nitrogen (≃75% He) is required to remove this 8.0 kW of heat. A gas system automatically changes the heat leak mixture during reactor startup and shutdown (Fig. 11). During shutdown, all solenoid valves are de-energized and the preset gas mixture is vented through solenoid valve V-1. As the reactor power is increased, less electrical heat is required to maintain temperature. When reactor power is 3.0 MW, 3.4 kW

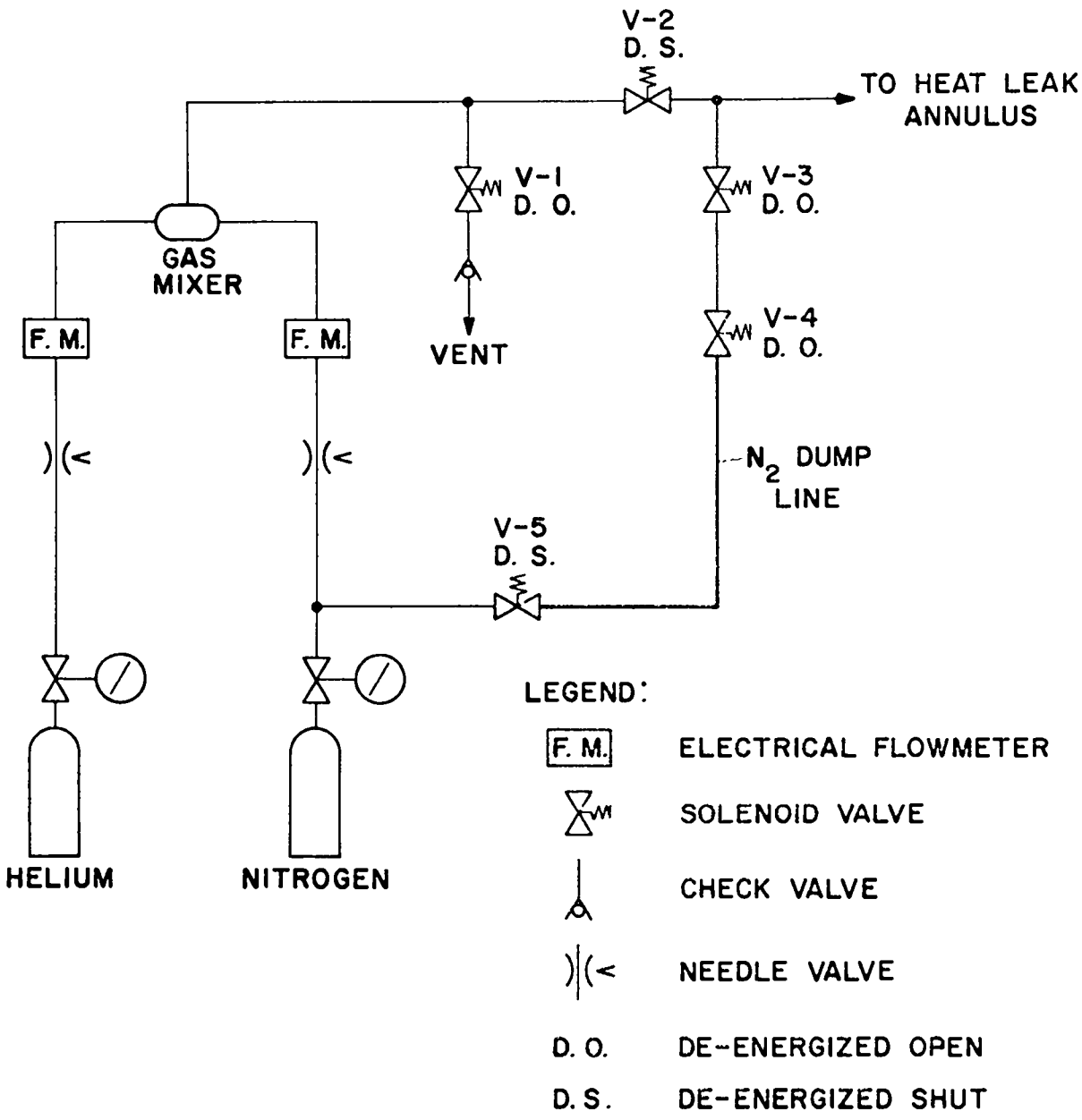

Fig. 11. Heat Leak Gas Control System

of radiation heat is produced in the loop and no electrical power is required. At this point, the solenoid valves are manually energized, and the preset gas mixture begins to flow through the heat leak annulus, causing a 5.0 kW electrical heat demand. Also, the nitrogen dump line is filled to a pressure of 20 psig. As the reactor power is raised to 5.0 MW, less electrical heat is required until at full reactor power, about 2.5 kW of electrical heat is used for thermostatic control. When the reactor power drops below 3.0 MW, the solenoid valves are automatically de-energized, resulting in the gas mixture being vented and nitrogen from the dump line sweeping out the helium-nitrogen mixture from the annulus. The nitrogen enters the annulus in about three seconds. Valve V-5 is closed when de-energized to prevent continual nitrogen flow during periods of shutdown.

The operation of the control system has proven to be entirely satisfactory and reliable. The heater control circuitry holds the system to within $\pm 5^{\circ}C$ during nuclear operation at power or while the reactor is shut down. There are twelve mercury plunger-type relays in the two systems; three of these have stuck in the off position and have had to be replaced. This is not unusual, considering that some of them switch 15 A currents $\approx$20 times per minute. The heater controls are divided into two independent groups as mentioned earlier. When each of the above failures occurred, the remaining group continued to operate, and there was little change in temperature.

There are fourteen heaters in the two systems. Some of these have operated at temperature for over a year, and all are still working. None has shown any significant change in heater resistance or heater-to-sheath insulation resistance. There are four thermocouples in each environmental cell, and each of the inserts has had up to eight thermocouples. Of the total of about 65 thermocouples used, all worked without trouble, except one which failed before installation of the insert into the environmental cell.

The nuclear reaction, $Na^{23}(n,p)Ne^{23}$ produces significant amounts of Ne^{23} in the system. During normal operation, the gas flow rate is adjusted so that the Ne^{23}, which has a half-life of 38 sec, decays to an undetectable level before reaching the monitor at the top of the reactor. However, this reaction permits a check of the sweep gas monitoring system. To make such a check, one increases the sweep gas flow. A response on the radiation monitor indicates proper operation. A standard source can also be used to check and calibrate the system.

C. Handling

The in-pile test assembly, as previously described, consists of two parts, a semipermanent environmental cell and a replaceable insert containing the test capsule.

To install the insert, the gas-tight container assembly (Fig. 3) is attached to the top of the cell with an O-ring seal. The containers are pumped out, filled with helium, and opened to the cell. The insert is then lowered into the cell through the sliding seal located at the top of the container assembly (Fig. 1). The convecting sodium temperature is held at approximately 250°C during these operations. After the insert has been satisfactorily installed, the system temperature is increased to melt the fuel.

A minimum of two days cooling time is usually allowed before an insert is removed from the cell. There have been up to 2500 curies of gamma activity in inserts that have been removed. Prior to insert removal, two split shields of depleted uranium are assembled around the gas-tight containers at the top of the cell. These are shown in Fig. 12. The lower shield is nine inches outer diameter and four inches thick; the upper shield is five inches outer diameter and two inches thick. With the sodium liquid, the removal process

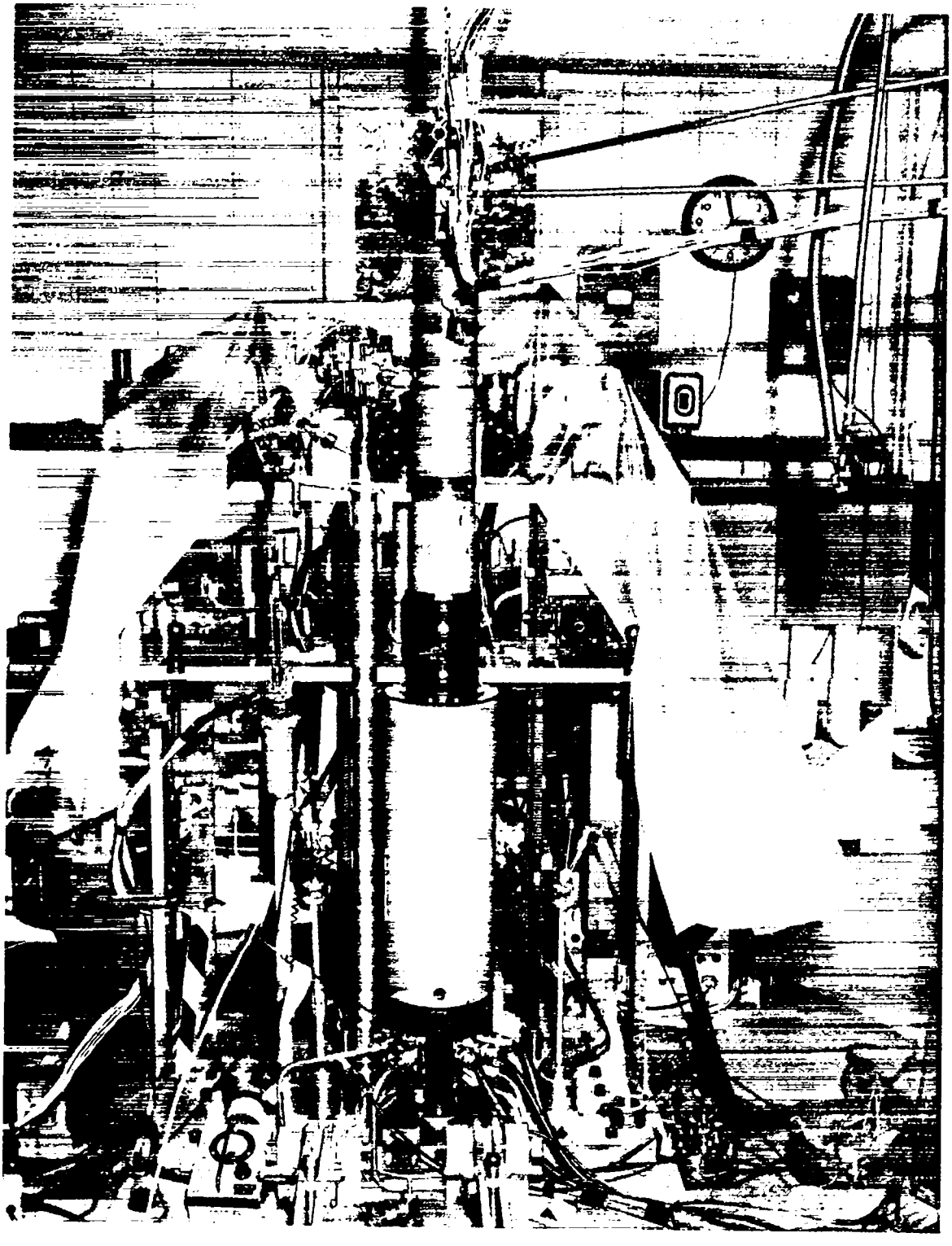

Fig. 12. Shields for Insert Removal

is started by raising the 3/8-in. o.d. inner tube two feet, pulling it through the upper seal (Fig. 3). This removes the thermocouples from the region of the fuel element. The insert is then raised until the tantalum secondary container is positioned in the lower container. In this position, the hexagonal top of the tantalum engages a mating socket in the container. Valves at the top of the cell and the bottom of the lower container are then closed. Next, the steel tubing is unscrewed from the tantalum secondary container and raised into the top shield. The valves between the two containers are then closed. The unactivated part of the steel tubing is now cut away, leaving the radioactive parts in two shielded, gas-tight containers which are separated and removed. Plastic bags would be used to seal the ends of the gas-tight containers, if the sweep gas indicated the presence of activity. No irradiated environmental cells have been removed from the reactor thus far. However, a uranium shield and other necessary components exist for this eventuality.

Figure 13 shows the radiation fields which can be expected through four inches of uranium from the fission product-tantalum source. Somewhat higher fields than this were actually experienced due to scattering from beams coming from the unshielded ends of the large gamma shield.

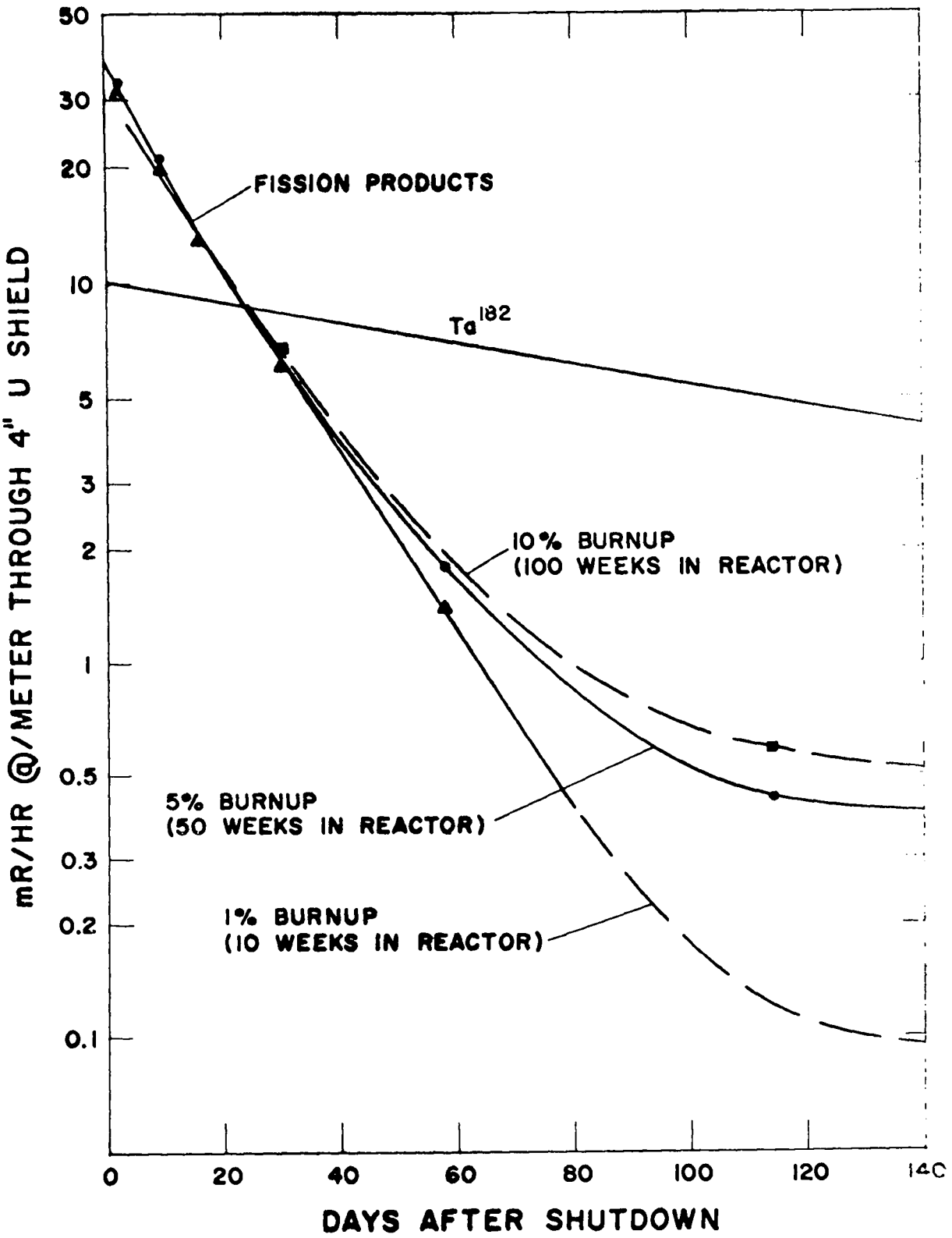

Fig. 13. Radiation Fields from Fission Products & Tantalum

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