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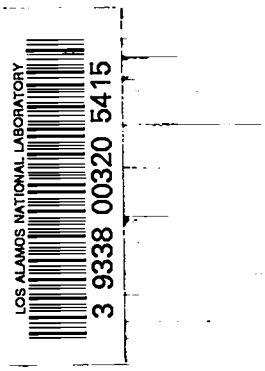
Thermal Irradiation of Liquid Plutonium-Alloy Fuels



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THERMAL IRRADIATION OF LIQUID PLUTONIUM-ALLOY FUELS

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ABSTRACT

In-pile test loops for irradiating liquid plutonium-alloy fuels were operated in the Omega West Reactor. These loops provided a sodium environment for test fuel capsules at temperatures similar to those expected for liquid-metal-cooled fast breeder reactors. Eleven fuel capsules were irradiated with up to 2.6 at.% burnup of plutonium. This report describes the design, construction, and operation of the irradiation loops and the handling of the irradiated test fuel capsules.

INTRODUCTION AND SUMMARY

An in-pile irradiation program was carried out at the Los Alamos Scientific Laboratory (LASL) from January 1964 to November 1966 to obtain additional irradiation data on the liquid plutonium alloy fuels and tantalum containers to be used in the Fast Reactor Core Test Facility (FRCTF).[1,2] The test capsules were irradiated in LASL's 5.0-MW MTR-type reactor, the Omega West Reactor (OWR).[3] Operating conditions simulated those expected in the FRCTF: fuel-container interface temperatures were maintained above the fuel freezing point of approximately 440°C and below 725°C during all phases of reactor operation and shutdown. A natural-convection sodium loop was located where the fast and epithermal neutron flux components are highest in the OWR core. A tantalum fuel capsule, containing Pu-Co-Ce fuel (24 g Pu - 6.29 g Pu/cc) and placed in the loop, operated at an average specific power of 150 W/g plutonium. With the reactor operating schedule, 1 at.% burnup was achieved in about ten weeks.

IN-PILE TEST ASSEMBLY

The in-pile test assembly consisted of two separate parts (Fig. 1): the environmental cell, semipermanently installed in the reactor; and the experimental insert, installed into and removed from the environmental cell as a unit. The cell contained the convecting sodium loop, thermocouples, heater units, and a gas annulus for heat removal and temperature control; the insert contained the test fuel capsule and additional thermocouples.

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

Environmental Cell

The environmental cell was 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 (hot-trapped, 400 g) was doubly contained. A stainless steel flow divider, containing an argon-filled gas gap, minimized radial heat conduction between the convective legs of the sodium loop. The top and bottom of the convective sodium channels were rounded to smooth the reversal of the sodium flow. Sodium was introduced into the loop in the following way: a 5/8-in.-i.d., 6-ft-long tube prefilled with hot-trapped sodium was placed in the cell; the lower 6 ft of the cell was placed in an electrical furnace containing a helium atmosphere. The cell was then

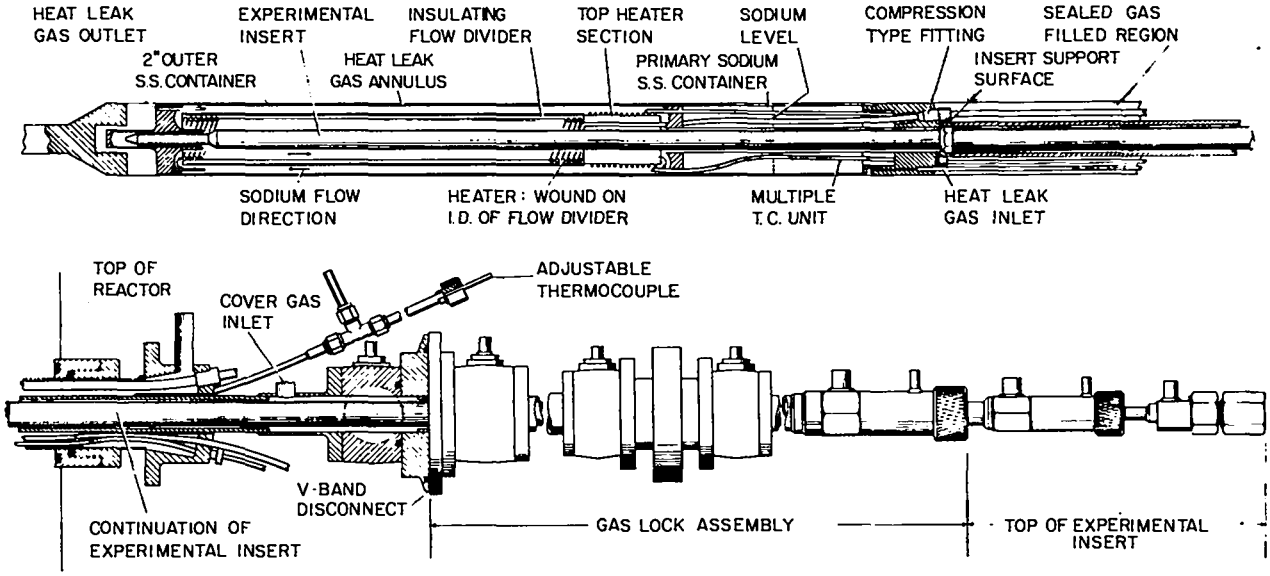

Fig. 1. In-pile test assembly.

evacuated and heated to approximately 140°C. Sodium filling was then observed by thermocouples located in the cell. After filling, the cell was pressurized with helium and its lower portion was radiographed to determine whether it had filled properly. The cell was initially over-filled with sodium; the desired level was reached by inserting a Zircaloy dipper tube into the cell, operating the cell at its normal operating temperature for a few weeks, and removing excess sodium through the dipper.

A heat-leak gas annulus was formed by the primary sodium container and the outer tube of the cell. The gap was 0.020 in. thick over the lower 9 in. and 0.012 in. thick over the top 4 in. of the gas annulus at operating temperatures. The reduction in gap thickness increased the convective sodium flow. A continuously flowing mixture of helium and nitrogen was admitted to the annulus at the top and exhausted at the bottom by a tube passing through the outer leg of the sodium stream.

The cell contained seven resistance heaters. Six of these were supported on the interior threaded wall of the flow divider and were used for temperature control; the other was wound on the non-insulating upper section of the flow divider and was used only for sodium melting. The heater units shown in Fig. 2 were obtained from the Aero Research Instrument Department of American Standard. They had Inconel sheathing, MgO insulation, Nichrome V cores, and Ni lead wires. The heater section of the unit was swaged down to 0.093-in. o.d. over its length of 52 in. The six heaters were normally operated at 5.0 kW, which corresponds to a heat flux of 60 W/in.^{2} of sheath area. (The manufacturer suggested 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 6 ft of each heater was examined by radiography and dye penetration to determine sheath and heater integrity. Lead and insulation resistances were measured. The

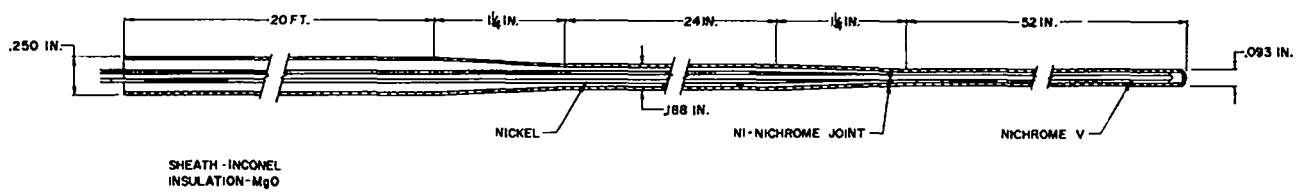

Fig. 2. Electrical heater.

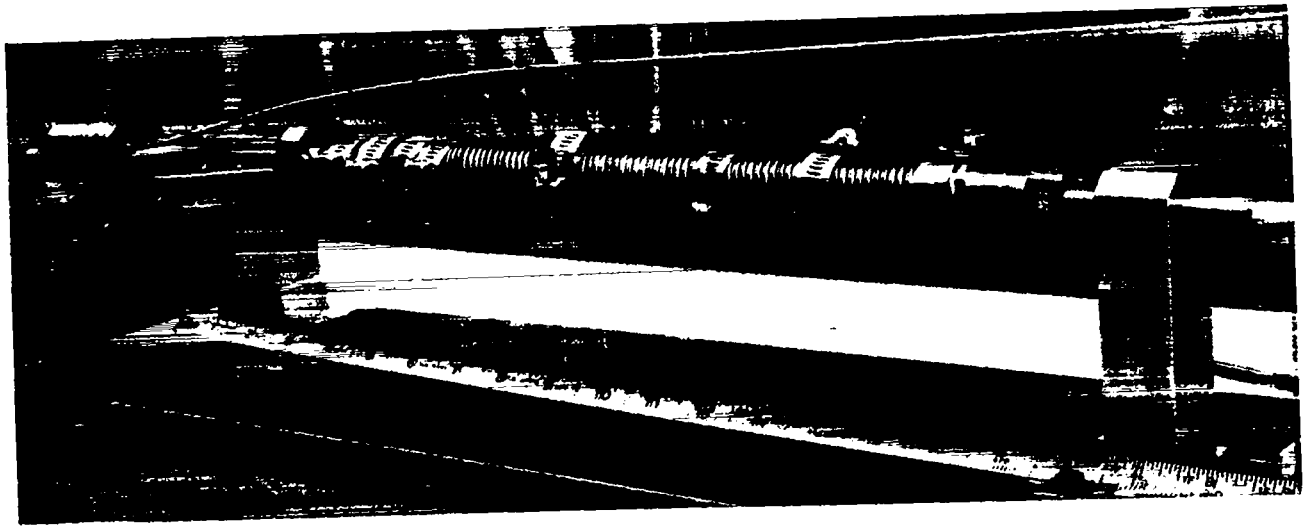

Fig. 3. Heater unit wound on mandrel.

0.250-in.-o.d. and 0.188-in.-i.d. sections of each heater sheath were sealed to the cell with metal compression-type seals.

For installation, the heater units were first wound on a mandrel, as shown in Fig. 3. The flow divider was then threaded over the heaters; the small clamps were removed as the operation progressed. After the flow divider was in position, the mandrel was unscrewed from the assembly, and the top heater was installed as shown in Fig. 4.

Three stainless-steel-sheathed, Chromel-Alumel thermocouples swaged in an 0.125-in.-o.d. stainless steel sheath were located in the outer convecting sodium stream. The thermocouple junctions were located at the top and bottom of the sodium loop and at the point where the gas gap changed thickness. The thermocouples were 0.031-in. o.d., MgO-insulated, and ungrounded. A fourth sheathed thermocouple was located in the heat-leak gas exhaust tube; an O-ring fixture at the cell top provided a sliding seal for this thermocouple, so that the position of the junction was adjustable. This thermocouple was used to obtain the axial temperature distribution in the outer convecting sodium stream.

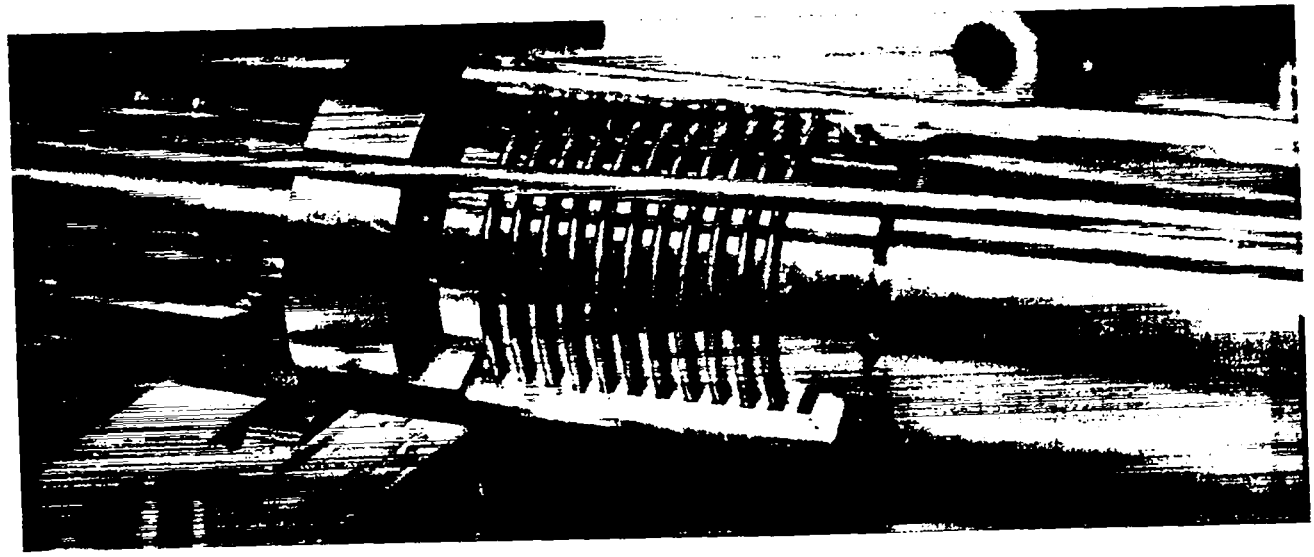

Fig. 4. Installation of top heater.

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

A 3-ft-long, heavy-walled steel cylinder, slotted to accommodate heater and gas lines, was located in the lower part of the sealed gas-filled region. This cylinder provided a neutron scattering medium to prevent neutron streaming to the top of the reactor.

The usual inspection procedures of radiography, dye penetration, and helium leak testing were employed during the construction of the cell. All stainless steel tubing used for the cell was degreased and cleaned by the manufacturer. Common solvents were used to clean other cell components.

After the cell was filled with sodium, it was operated at temperature for a few weeks in a nonnuclear environment closely simulating that of the OWR. The cell was placed in a dummy fuel element held in a 6-ft-long standpipe. The cell was inclined 1.25° from the vertical, as it would be in the OWR. A Zircaloy dipper tube was inserted in the convecting sodium to simulate the lower portion of an insert; this tube adjusted the sodium level and hot-trapped the convecting sodium. Water to cool the cell was circulated downward through the dummy fuel element, passed out the bottom of the element into radiators where it was cooled, and returned to the top of the standpipe. Electrical heaters in the standpipe adjusted the water temperature. A manometer continuously indicated the pressure drop through the dummy fuel element. During operation, the water flow was approximately 12 gal/min, producing a 1.0-psi pressure drop through the dummy element with water temperatures of 45-50°C.

Insert

Figure 5 shows an insert typical of those installed and operated in the cell. The insert was 28 ft long and contained a test fuel capsule in its lower section. The tantalum capsule (0.420-in. o.d., 0.020-in. wall, 8 in. long) was filled to a height of about 2 in. with Pu-Co-Ce fuel (24 g Pu),

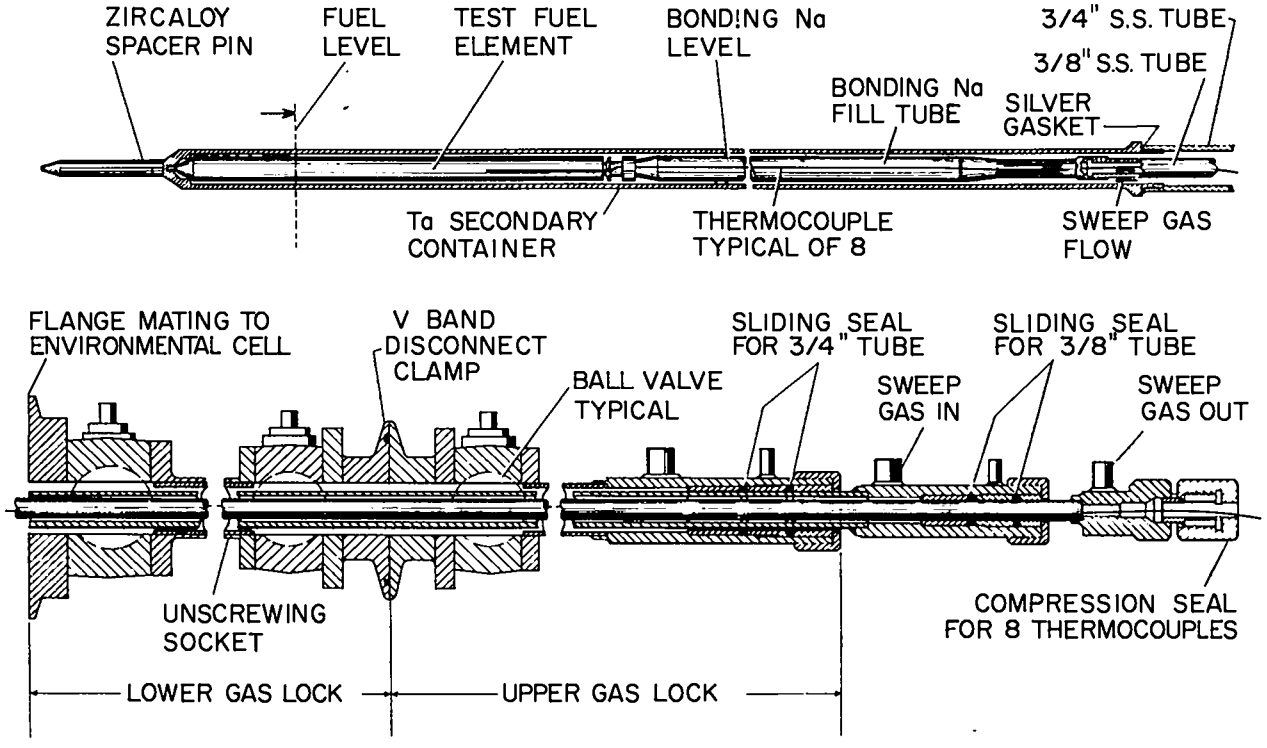

Fig. 5. Experimental insert.

and the closed space above the fuel was filled with helium at atmospheric pressure. The capsule was placed in a 0.575-in.-o.d., 18-in.-long secondary tantalum container to provide containment in case the fuel penetrated through the capsule. Sodium provided a thermal bond between the two containers. The junctions of eight 30-ft-long Chromel-Alumel thermocouples were located at various axial positions in the annulus and were sealed at the top end of the insert by a multiple-penetration, compression-type fitting. The thermocouple junctions were accurately positioned by a slotted nickel tube in the annulus.

The bonding sodium was introduced into the annulus by a Zircaloy tube located above the capsule. This tube was prefilled with hot-trapped sodium and was installed in the insert during assembly. Figure 6 shows the installation of the sodium-filled Zircaloy tube and eight thermocouples in the insert. Because the lower end of the Zircaloy tube had been opened, exposing the sodium, assembly was done in a plastic bag containing an inert atmosphere. After final assembly, the lower portion of the insert was placed in a furnace, evacuated, and heated to 140°C. The insert thermocouples indicated the progress of filling. After filling, the insert was pressurized with helium and radiographed. The Zircaloy tube hot-trapped the bonding sodium during operation at temperature.

A 3/4-in. stainless steel tube and a 3/8-in. stainless steel tube formed the passageway for the sweep-gas flow (Fig. 5). The 3/4-in. tube was attached to the secondary tantalum container with a threaded joint which was sealed by a silver gasket. During the removal of the insert from the cell, this joint made it possible to separate the highly activated part of the insert from the less activated part, making shielding simpler.

Two containers which acted as gas locks were used in connection with the insert and remained in place at the top of the cell during an irradiation experiment (Fig. 7). They maintained a helium atmosphere over the convecting sodium during installation and removal of the insert from the cell and confined any radioactivity which might have been released by a fuel capsule. Double O-ring fixtures provided sliding seals between the 3/4-in. tube and the top container and between the 3/8-in. and the 3/4-in. tubes.

Fig. 6. Installation of sodium-filled Zircaloy tube.

Fig. 7. Insert in reactor, showing gas locks.

ENVIRONMENTAL CELL AND INSERT HANDLING

Environmental cells or inserts were handled with the aid of a winch on a platform above the reactor. The winch hook could rise high enough to suspend either unit over the reactor top.

Cell handling was relatively straightforward; the most complicated part was shielding the bottom end during its removal from the reactor. Insert handling was more complicated. The cell had to be penetrated without admitting air into the helium cover gas. When the insert was removed from the cell, the highly radioactive fuel capsule and its tantalum container had to be separated from mildly activated steel tubing and thermocouples. Finally, all activated materials had to be shielded for gamma radiation and sealed in containers.

Environmental Cell

Insertion into the Reactor. The cell was vertically suspended over the reactor by the winch. A gland seal was installed on a reactor-top experiment port, and the cell was lowered through it until the bottom end came to rest on a pin in a dummy fuel element in the reactor core. Gas lines, heater leads, and thermocouple leads were then connected. After the sodium was melted, the cell was ready to receive an insert.

Removal from the Reactor. For the experiments described in this report, two environmental cells were installed in the reactor tank. One has now been removed from the reactor, and the other is being adapted for the test irradiation of solid fuel elements. The irradiated cell was removed without difficulty.

The insert was taken out before the cell was removed. Although enough shielding was provided to remove the cell with the insert inside if the insert could not be removed, this situation never occurred.

The cell sodium was frozen, and all gas and electrical lines were disconnected. The cell was raised 7 ft and clamped, and a split shield (9-in. o.d., 2-in. i.d., 40 in. high) of depleted uranium was installed around the top of the cell (Figs. 8 and 9). When both cells were in place, the shield could not be installed until the adjacent cell was raised 7 ft, because of the closeness of the cells. Therefore, the other cell was also raised, with its gas lines disconnected and its electrical leads extended.

After the shield was in place, the cell to be removed was raised an additional 2 ft, so that the cutoff saw could be installed. This device is a portable grinder, mounted in a hinged frame, driving a carborundum wheel. One side of the hinge clamps to the cell, and the other side holds the grinder. The cutoff saw must first be installed with its cell clamps loose, so that the cell can be raised with the saw resting on the shield top.

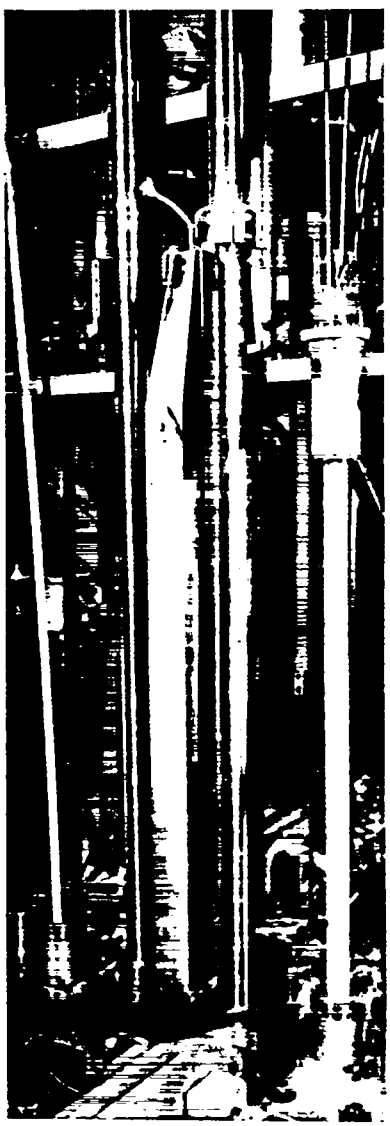

Fig. 8. Half of shield installed.

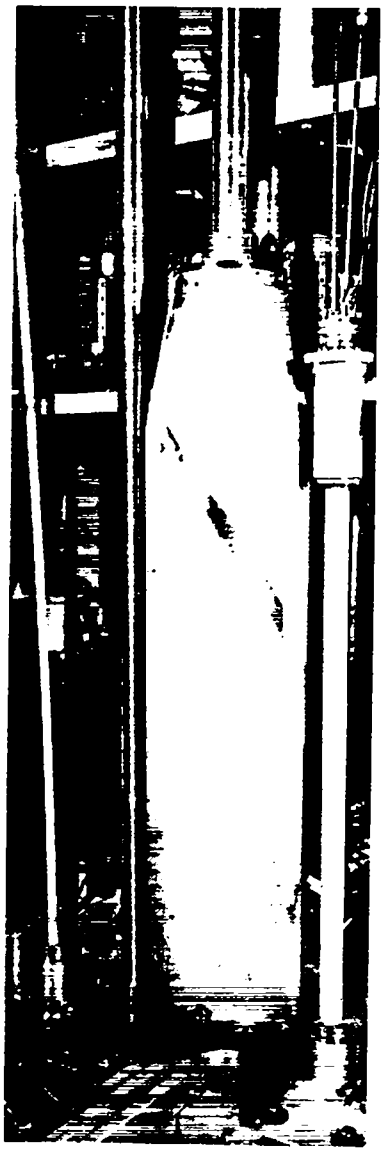

Fig. 9. Entire shield installed.

Plastic bagging was then placed over the cell and was fastened to the lower part of the cell (Fig. 10) to contain possible alpha contamination inside the cell. All personnel left the reactor top while the cell was raised until the radioactive part was positioned in the shield. The cell was then clamped to the top of the shield, and the non-active part was cut off by turning on the grinder and closing the hinge. After cutoff, the upper part of the cell was pulled up, and the bag was tied twice between the two parts and cut between the two ties. The shielded part of the cell was removed for examination.

Fig. 10. Shielding and bagging installed, with cell ready to be raised.

Experimental Insert

Insertion into the Environmental Cell. The insert was vertically suspended over the reactor. The lower gas lock was attached to the cell by a V-band clamp. An O-ring between the lower gas lock and the cell made a gas-tight seal. After the gas lines had been connected, the gas locks were purged of air and filled with helium. They were then opened to the cell.

The insert was lowered through the sliding O-ring seal at the top of the upper gas lock (Fig. 5). This seal consisted of two O-rings in the annulus between the seal body and the movable 3/4-in. tube. The O-rings were separated by metal rings, and the volume between the O-rings was evacuated. The sliding tube was lubricated by a thin film of vacuum grease.

Finally, the insert was lowered until the fuel capsule was just above the cell sodium, at which time the sweep-gas lines were connected and the insert was evacuated. Only then was the insert lowered to its final position so that the sodium in the insert would be melted under vacuum. The helium sweep-gas flow was then started.

Removal from the Environmental Cell after Irradiation. The thermocouple train was raised 2 ft to pull the thermocouples out of the secondary container. All the insert thermocouple extension leads and gas lines were removed, except the line for evacuating the sliding O-ring seal. A plastic sleeve was installed at the connection between the two gas locks to contain radioactive contaminants when the two gas locks were separated.

Gamma shielding was then assembled around the two gas locks. This consisted of:

1. The main capsule shield of depleted uranium (9-in. o.d., 1-in. i.d., 25-1/2 in. high), which split into two halves and was assembled around the lower gas lock (Fig. 11).
2. A smaller split shield of depleted uranium (5-in. o.d., 1-in. i.d., 28 in. high), which was assembled around the upper gas lock to shield the irradiated steel parts (Fig. 12).
3. A 2-in.-thick shield of lead, which was placed around the front of the ball valves between the two uranium shields (Fig. 13).
4. Bags of lead shot, which were stacked around the valves at the bottom of the capsule shield (Fig. 13).

The insert was raised 2 ft higher, so that the cutoff saw could be attached. If alpha contamination of the insert was expected, plastic bagging was put over the assembly and attached to the top of the upper gas lock.

When everything was in place, all personnel left the reactor top, and the insert was raised until the hexagonal top of the secondary container engaged a socket in the top of the gas lock. After the ball valves between the cell and the lower gas lock were closed, the outer sweep-gas tube was detached from the secondary container.

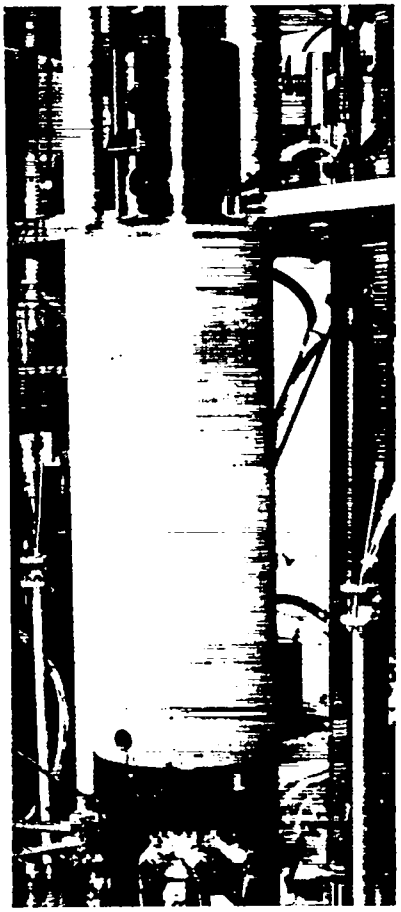

Fig. 11. Main capsule shield in place.

The tubing was raised out of the lower gas lock, and the valves between the two locks were closed. The tubing was then cut off above the upper gas lock, and the unirradiated part was removed, leaving the radioactive parts in two shielded gas-tight containers which were separated and removed one at a time. If bagging was used, it was gathered and tied twice between the cut ends, then severed between the ties. The shield containing the capsule was put into a large lead pig for transportation to the hot cells.

Handling Experience. Eleven inserts of various types were installed and removed successfully. The highest gamma radiation dose received by

Fig. 12. Main capsule shield and smaller split shield in place.

personnel during an insert removal was 0.2 rem. Usually, the maximum dose received by any one person was less than 0.1 rem.

Minor complications arose during the removal of some inserts. Several of the inserts had thermocouples that could not be removed from the fuel region. When this happened, the insert was raised with the thermocouples in place. When the 3/4-in. tube was unscrewed from the tantalum secondary container, the thermocouples either sheared off or wound around each other. As the tubing was pulled away from the tantalum, the unbroken thermocouples were allowed to slip down through the thermocouple seal at the top of the insert. The middle gas-lock valves were closed on these thermocouples.

Fig. 13. All shielding in place.

After the two gas locks had been separated and the bagging between them had been tied, the bagging and the thermocouples were severed by the same cut.

The outer sweep-gas tubing was scored by other stainless steel parts in the first inserts. The scored tubing galled the compressor rings in the sliding O-ring seal, which caused the tube to jam. Scoring was prevented by installing brass bushings in the gas locks where the tubing would otherwise rub on stainless steel.

GAS CONTROL SYSTEM

The gas system maintained a helium atmosphere over the cell and insert sodium volumes during operation, as well as during the insertion and removal of an insert from a cell. Helium was continuously swept over the capsule and exhausted through a molecular sieve to provide a means for detecting gaseous fission products if the capsule cladding failed. Heat removal from the cell could be adjusted by a continually flowing He/N_2 gas mixture. Figure 14 is a diagram of the control system.

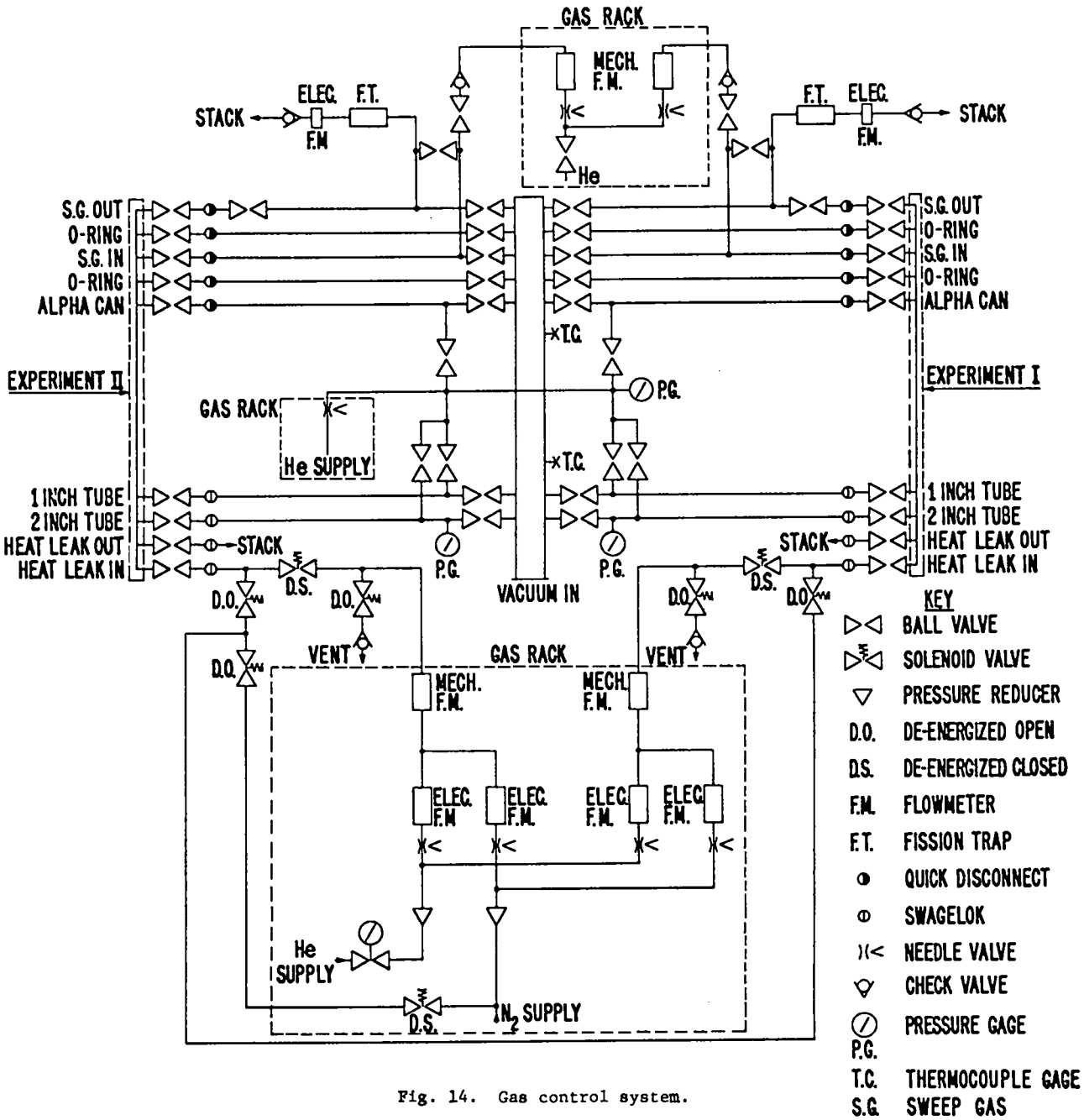

Fig. 14. Gas control system.

Vacuum and Helium Gas Service

A vacuum manifold consisting of a 2-1/2-in.-o.d., 5-ft-long stainless steel tube was located at the reactor top. It was equipped with thermocouple gauges for pressure indication and with flexible lines for connection to the cell and insert. The manifold was evacuated by a mechanical vacuum pump, and the pump exhaust was fed into the reactor vent system. By means of this manifold, the cell and insert were evacuated for sodium melting, the double O-ring seals were evacuated during insert installation and removal, and air trapped during installation was evacuated.

Helium containing less than 10 ppm impurities was used as an inert atmosphere for the cells and inserts.

Sweep Gas System

Capsule leaks were detected by the sweep-gas system. High-purity helium, obtained from a diffusion cell,[4] was directed through the insert to a molecular sieve and was exhausted to the reactor vent system. The molecular sieve (fission-gas trap) was monitored with a scintillation detector system. The helium flow to the insert was monitored with a flowmeter located in the exit line from the molecular sieve to the OWR exhaust system.

The helium diffusion cell purifier was obtained from Marshall Products Division of NRC. It produced ultrapure helium from commercial quality helium without periodic attention or regeneration. The performance of the diffusion cell was tested with the use of a gas chromatograph. Table I presents the impurities of a sample gas input to the diffusion cell and the impurities of the chromatograph carrier gas (Bureau of Mines helium).

TABLE I

COMPARISON OF DIFFUSION CELL SAMPLE GAS IMPURITIES WITH CHROMATOGRAPH CARRIER GAS IMPURITIES

Impurity	Sample Helium (ppm)	Chromatograph Carrier Helium (ppm)
O_2	8	0.1
N_2	30	0.7
Kr	36	Undetected
Xe	36	Undetected

Two tests were conducted with this sample gas as input to the diffuser. In both cases, the impurities in the output gas from the diffusion cell were no greater than the carrier gas impurities. The throughput of the cell depended on the input pressure and the temperature of the cell. For a flow of 20 cc/min, the input pressure was 500 psi, and the cell temperature was 250°C. The cell was heated by a coil furnace. Water for cooling the output seal of the cell was supplied from a tap at 0.5 gal/min. If the water flow dropped to 0.4 gal/min, an interlock switch turned off the furnace electrical power and activated an alarm.

The flow of sweep gas was indicated over a range of 0-100 cc/min by a Hastings-Raydist electrical flowmeter, which was independent of pressure and temperature over a wide range. The transducer had no moving parts and was weatherproof. Each flowmeter had high and low set points which could be adjusted to provide alarm signals.

The molecular sieve contained 100 g of Linde 5-A sieve material to delay fission-product gas, thus enhancing detection capability. The breakthrough time was 40 min for krypton and 20 h for xenon with a helium flow rate of 40 cc/min at 25°C. The molecular sieve was monitored for radioactivity by a scintillation detector (NaI) feeding an alarm. An output signal from the alarm was fed into a recorder for continuous recording of the radiation levels. Two set points on the alarm provided signals for high radiation levels or alarm failure. A scintillation system was used for obtaining spectral analysis of any observed increase in radioactivity.

Heat-Leak Gas System

The heat-leak gas consisted of a mixture of helium and nitrogen. Any mixture of the two gases, or either gas alone, could be obtained by a simple adjustment. Since helium conducts heat about five times as readily as nitrogen, varying the mixture provided control of the heat loss from the cell. The helium and nitrogen gas flows were individually controlled by throttle valves and were fed to a commercially purchased mixer. The flows of nitrogen and helium were measured by electrical flowmeters (Hastings-Raydist, Inc.). Set points on each flowmeter produced alarms for abnormal flow. The mixture then passed through the cell and was exhausted to the OWR vent system.

When the reactor was shut down, nitrogen was kept in the gas annulus, and about 3.0 kW of electrical heat maintained the system at operating temperature. When the reactor was at full power and the cell was loaded with 2 in. of fuel, about 5.6 kW of radiation heating (2.0 kW of gamma heat plus 3.6 kW of fission heat) was produced in the loop. An additional 2.5 kW of electrical heat stabilized the system temperature. A gas mixture consisting of ≈75% helium and ≈25% nitrogen was required to remove this 8.1 kW of heat.

The heat-leak mixture was automatically changed during reactor startup and shutdown (Fig. 15). A typical cycle started with the reactor down. As the reactor power was increased, less electrical heat was required to maintain temperature. When reactor power was 3.0 MW, 3.4 kW of nuclear heat was produced in the loop and no electrical power was required. At this point, the solenoid valves were manually energized, and the preset gas mixture began to flow through the heat-leak annulus, causing a 5.0-kW electrical heat demand. Also, the nitrogen dump line was filled to a pressure of 20 psig. As the reactor power was raised to 5.0 MW, less electrical heat was required, until at full reactor power about 2.5 kW of electrical heat was used for temperature control. When the reactor power dropped below 3.0 MW, the solenoid valves were 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 entered the annulus in about 3 sec. Valve V-5 was closed when de-energized to prevent continuous nitrogen flow during shutdown.

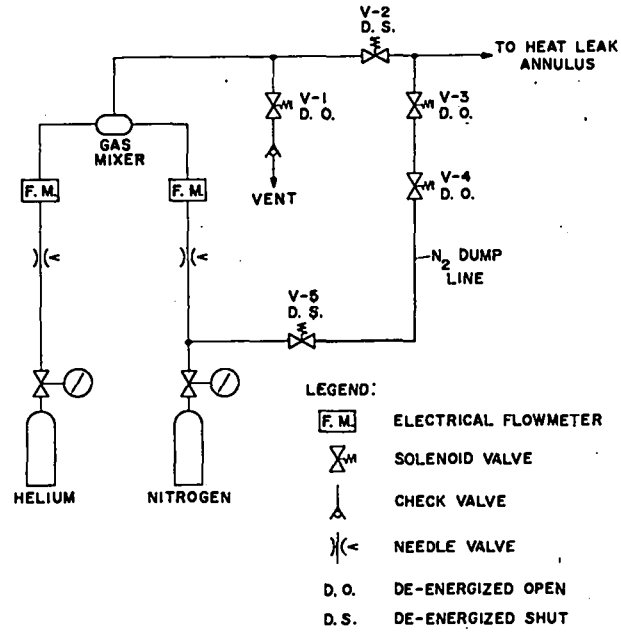

Fig. 15. Heat-leak gas control system.

INSTRUMENTATION AND CONTROL CIRCUITRY

Temperature Control

The six main heater units in the cell were electrically separated into two groups of three, each group having identical control circuits. Figure 16 is a simplified schematic of the control circuit for one group. A variable autotransformer supplied power to the heaters from a 208-V source. The input to the transformer was controlled by the contacts of mercury relays R_1 and R_2 actuated by a Pyr-O-Vane unit with an Electronik-17 as an auxiliary controller. A wattmeter indicated the power drawn by the heaters. Under normal operating conditions, the autotransformer was adjusted so that the three heaters drew 2.8 kW at 170 V and 16.5 A when they were turned on. Each heater lead was separately fused by 8-A fuses and was fed by its own isolation transformer to prevent arcing in case of a short circuit between the heater lead and the sheath. Indicator lamps were incorporated between the heater leads and ground to indicate deterioration of the insulation resistance. A time delay relay whose contacts open 40 sec after a power outage prevented automatic restoration of power to the heaters, preventing uncontrolled remelting of the fuel.

Normally, a cell contained four Chromel-Alumel thermocouples, and the insert, eight. One of these was connected to a recorder in the OWR control room, and five were connected to a multipoint recorder on the mezzanine of the OWR reactor room. Of the remaining six thermocouples, two were connected to the Pyr-O-Vane controllers, and four were connected to two dual-pen Electronik-17 recorders.

Each Electronik-17 recorder served a threefold purpose. It was an auxiliary temperature controller in the event of a Pyr-O-Vane malfunction, a continuous temperature recorder, and a temperature alarm device. Each pen had four adjustable set points; one was used for low-temperature alarm, one for low-temperature control, one for high-temperature alarm and control, and the remaining set point was used in a two-out-of-three system for scramming the reactor if temperatures rose abnormally. These set points were:

low temperature	
control	490°C
alarm	510
high temperature	
alarm and control	690
reactor scram	710

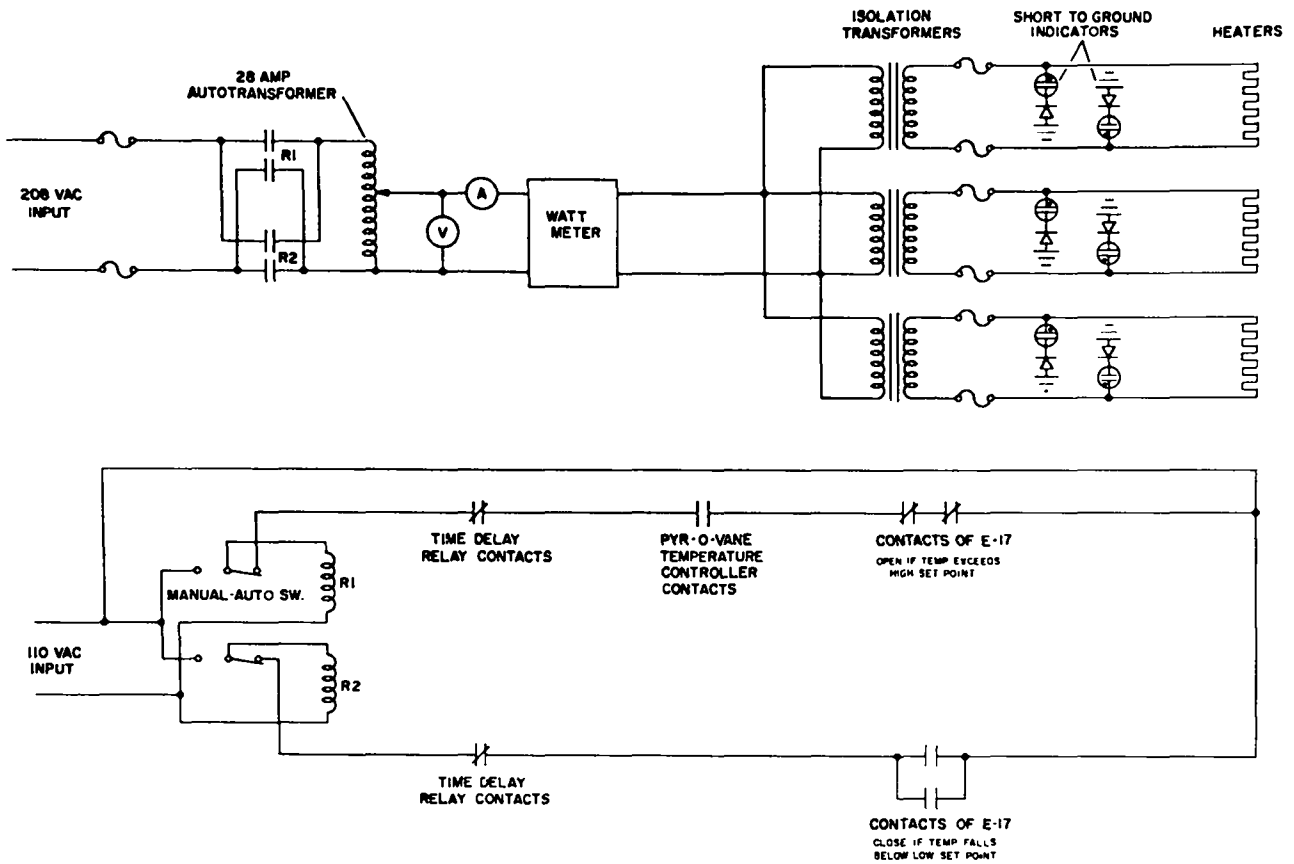

Fig. 16. Simplified heater control circuit.

The division of the heater control circuitry into two independent groups provided reliability and control flexibility. If a component in one group failed, the other group assumed control and prevented major temperature variations. The heater control circuitry held the system temperatures to within $\pm 5°C$ at all times. The operation of the control system was reliable and required little attention. However, the control and alarm circuitry were checked weekly to ensure proper operation of the system.

There were 12 mercury plunger-type relays in the two systems for switching heater currents; six such relays operated by Pyr-O-Vane controllers stuck in the "off" position and had to be replaced. This was not unusual, considering that some of them switched 15-A currents about 20 times a minute. When each of these failures occurred, the remaining group continued to operate, and there was little change in temperature.

There were 14 heaters altogether in the two systems. Some of these were operated at temperature for over a year, and none showed any significant change in heater resistance or heater-to-sheath insulation resistance. Of approximately 80 thermocouples used, all but one worked without trouble.

Alarms

Visual and audible alarms were located both at the control rack and in the OWR control room. These alarms indicated high or low temperatures, high or low flow in the sweep-gas and heat-leak gas systems, high or low radiation level for the sweep-gas activity monitor, scram conditions, and loss of instrument power.

LOOP OPERATION

Two cells were installed in the OWR: one was installed in March 1965 and the other in December 1965. Each cell was tested with three inserts:

1. an unfueled 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.

The general features and operating parameters of other fueled inserts that were operated primarily to achieve high fuel burnup were:

fuel material	Pu-Co-Ce (6.29 g Pu/cc)
Pu content	$\leq$32 g
container material	Ta, Ta-5W
burnup	$\leq$2.6 at.%
average power density	930 W/cc
average specific power	150 W/g
heat flux	170 W/cm^2
fuel element performance	17.8 kW/ft
maximum container temperature	700°C

At the completion of each irradiation, the fuel specimens were rapidly frozen by simultaneously scramming the reactor and turning off electrical heater power to preserve the operating distribution of the fission products. After removal, the fuel elements were radiographed to determine the fuel configuration at the time of freezing, examined by gamma-ray spectroscopy to determine fission-product distribution, and examined by metallographic techniques.

With one insert, an attempt was made to determine the behavior of the volatile fission products during reactor operation. Changes in the fuel column height were measured by five thermocouples placed just outside the fuel capsule in a slotted nickel tube. With this arrangement, the five thermocouples were accurately positioned at various heights relative to the top surface of the fuel. Changes in fuel height were determined from the temperature distribution changes observed with the thermocouples. Figure 17 gives results observed during the first five weeks; following weeks were similar to weeks 4 and 5. The initial sharp decrease in height was attributed to release of gross bubbles of cover gas trapped in the initial fuel melting (the vertical temperature distribution in the cell was such that melting occurred from the top downward). This entrapment made it impossible to determine directly the bubble-free fuel height. By extrapolation of the "no gas release" line in Fig. 17, a bubble-free height of 2.73 $\pm$ 0.03 in. can be inferred. Because calculations based on fuel loading, fuel density, and capsule diameter gave 2.85 in., the values plotted may be low by 0.12 in. absolute value, but relative values are consistent to about 0.01 in. The fuel level on Mondays, after a weekend of reactor shutdown, was significantly lower than that of the previous Saturday, just before shutdown. The decreases were as large as 0.080 in. and may have resulted from fuel thermal cycling during reactor shutdown and startup. Equilibrium fission-gas bubble content of the fuel appears to be about 3 or 4%.

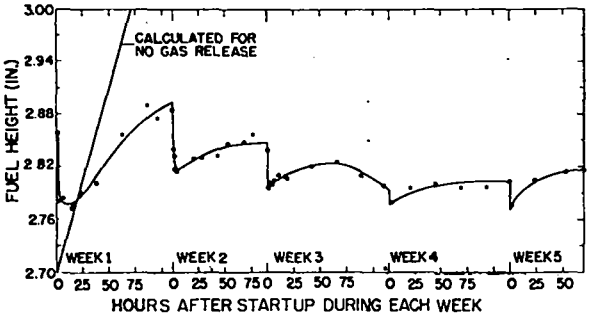

Fig. 17. Changes in fuel height.

Operation of three of the inserts was terminated because of the release of volatile fission products from the fuel capsules. The behavior of the system was different in each case.

In one, the sweep-gas monitor showed a sharp increase in radioactivity (ten times normal background). No other system perturbations were observed. The reactor was shut down and gamma spectra of the molecular sieve were obtained. These data showed existence of primarily short-lived gaseous fission products, indicating that plutonium rather than capsule plenum gas had leaked. Subsequent metallography showed capsule wall thinning at the fuel surface and a crack in the bottom of the capsule.

In the second, the sweep-gas monitor showed a small radioactivity increase (about background) with no other system perturbations. Gamma spectra of the molecular sieve indicated a plutonium leak. Because the radioactivity increase was small, the capsule was allowed to continue operating. The radioactivity increased gradually over the next two days at a rate of 0.25 mR/h^2. On the third day, the radioactivity increased rapidly to 50 times background, and the experiment was terminated. Later examination of the capsule showed about 30% wall thinning at the fuel surface.

In the third, operations personnel were alerted by a low-sweep-gas-flow alarm. Further investigation showed that all system temperatures had risen 20-30°C, except for two located at the fuel-level position. Their indicated temperatures had dropped 20-30°C. Also, all electrical heater power was turned off by the control circuitry, indicating an increase in nuclear heating. All of this occurred with no significant increase in radioactivity level of the molecular sieve. Although the events suggested a capsule failure, an attempt was made to restore sweep-gas flow to obtain definitive information before terminating the experiment. By alternately pressurizing and venting the annular space communicating with the outside of the capsule, radioactive gas was brought to the top of the reactor, resulting in fields of about 1 R/h at contact with the capsule. Subsequent investigation showed that a blockage had occurred in an inaccessible portion of the sweep-gas tubing. Examination of the capsule showed excessive wall thinning and plutonium on the outside of the capsule.

One of the inserts developed a leak at the top of the tantalum secondary container, which was sealed to the 3/4-in.-o.d. tube with a silver gasket. When a pressure differential existed between the insert and the cell, ^{41}Ar was observed from an (n,p) reaction with potassium contamination in the convecting sodium.

The reaction $^{23}Na(n,p)^{23}Ne$ produced significant amounts of ^{23}Ne, which could be brought to the reactor top by the sweep gas. During normal operation, the gas flow rate was adjusted so that the ^{23}Ne, which has a half-life of 38 sec, decayed to an undetectable level before reaching the monitor at the top of the reactor. However, this reaction permitted a check of the sweep-gas monitoring system. If an increased sweep-gas flow gave a response on the radiation monitor, the monitoring system was working properly. A standard source was also used to check and calibrate the system.

DESIGN AND PERFORMANCE

Thermal

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

While the reactor was operating, the heat source (fuel) was near the bottom of the rising leg of the convection loop, and the major heat sink (thinner gas annulus) was at the top of the descending leg. These locations favored a high convective flow, which minimized the vertical temperature gradient in the fuel sample. During reactor shutdown, the heat source (electrical heaters) was not as well localized, and thus the convective flow was reduced.

The calculated temperatures around the convecting loop are shown in Fig. 18 for a 5-kW fission and electrical source. The corresponding turbulent convective flows are 0.90 gal/min for fission heat

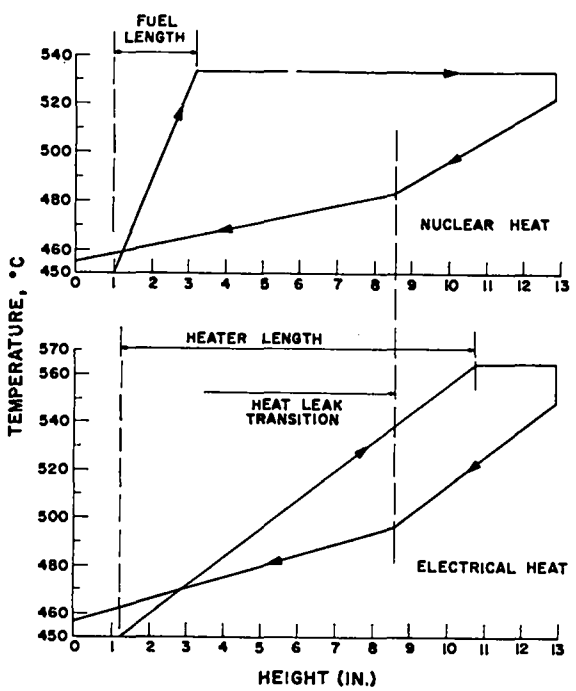

Fig. 18. Temperatures in convecting sodium loop.

and 0.66 gal/min for electrical heat. Measured temperatures and convective flow, based on power and temperature measurements, are consistent with these calculations.

The vertical temperature rise along the fuel during nuclear operation was about 80°C, and during electrical operation it was about 30°C. A vertical temperature rise of about 80°C was expected for liquid plutonium fuel elements of proposed reactor designs, so these designs were well simulated in the test samples. This temperature rise would increase, less than linearly, with test sample power; at a power several times the levels used, the temperature at the top of the fuel would become too high for long-term operation.

The effect of varying the composition of the helium-nitrogen mixture flowing through the heat-leak annulus is shown in Fig. 19. 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 mean temperature in the descending (outer) leg was held at approximately 540°C during these measurements. Early design calculations for pure nitrogen and for pure helium at a mean temperature of 487°C are indicated. A power range of about 3 to 12 kW can be spanned.

Gamma heating (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. Fission heating was measured in a similar manner; the value obtained by this method is within 15% of the values obtained by radiochemical analysis.

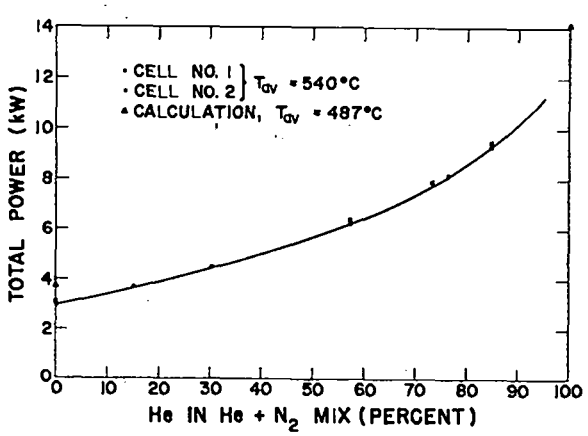

Fig. 19. Effect of gas mixture on power.

Neutronic

Before the detailed design and the construction of the irradiation experiment were started, preliminary neutronic calculations had indicated that useful data could be expected from such irradiations. The calculations were made with 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 closely spaced within and near the fuel. The DTK code used was of the Carlson[5] type, based on transport theory in S_4 approximation. The Hansen-Roach 16-group cross-section library[6] was used.

Flux distributions for the present design, containing fuel of 5.7 g Pu/cc are shown in Fig. 20. 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 5 at the fuel surface.

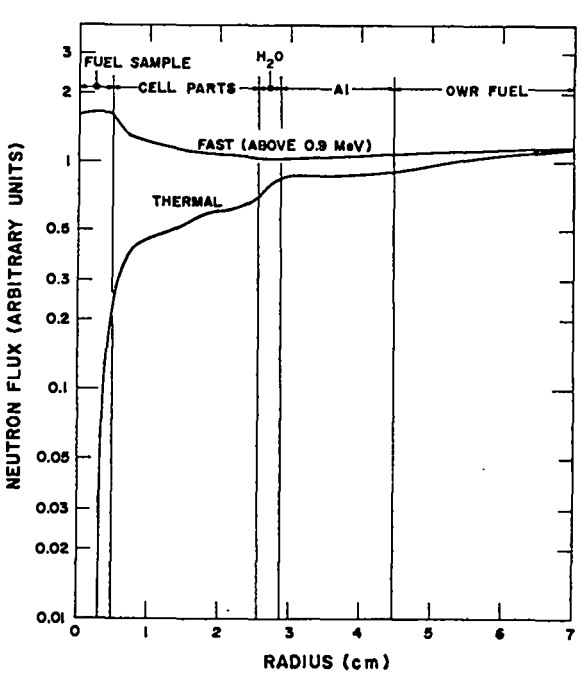

Fig. 20. Flux for 5.7 g Pu/cc.

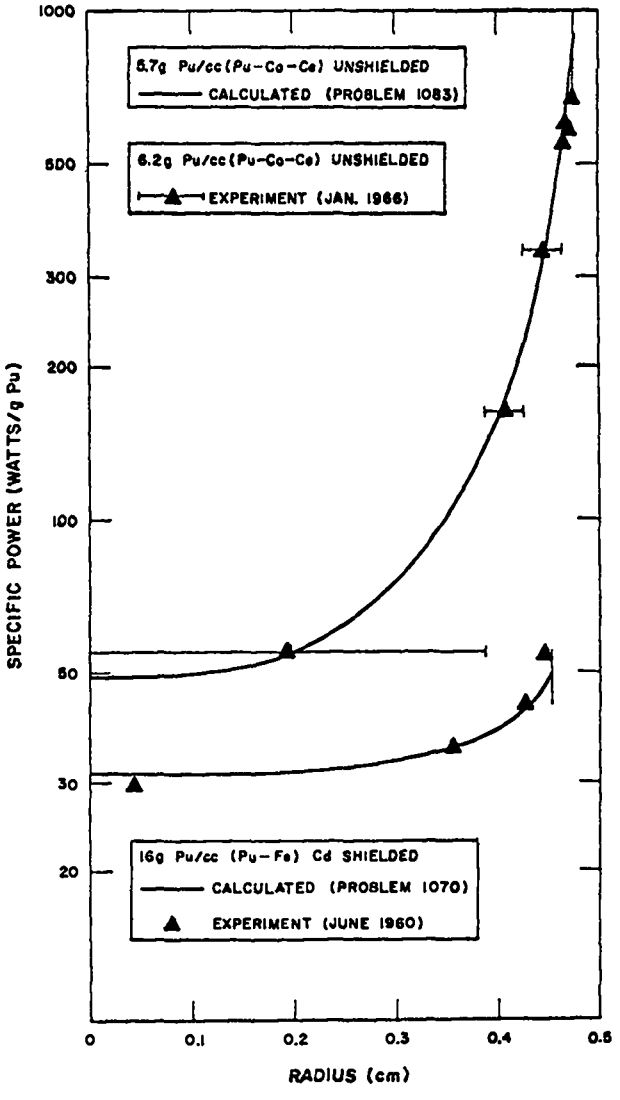

Fig. 21. Radial power distributions.

The corresponding power distribution is given by the upper curve of Fig. 21. The lower curve is for a dense fuel sample surrounded by a thick cadmium neutron filter. This calculation was compared with results obtained in an early (1960) experiment in OWR with LAMPRE I fuel (Pu-Fe eutectic, 16 g Pu/cc). From this comparison, a normalizing factor was obtained. The two curves represent the normalized power distributions; data points were obtained by radiochemical analysis of solid specimens of fuel which had been irradiated for short times at low power. Samples for radiochemical analysis were taken at various radii by machining on a lathe. The horizontal lines on the points indicate the radial width of the samples where it is significant.

The experimentally determined edge-to-average ratio for the 6.29 g Pu/cc fuel was 5.1, and 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).

The effect of varying the thickness of a natural boron filter was studied calculationally for 3.0 g Pu/cc fuel. Results (Fig. 22) 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. 23. Analysis of the related fuel temperature distributions indicated that, for liquid fuels having 3-8 g Pu/cc, the power distribution of unfiltered test samples was satisfactory. These samples were compared 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 2 (3 g Pu/cc fuel) to 2.2 (for 8 g Pu/cc fuel) times as large as those for the uniform distribution. Hence, neutron filters were omitted in the design. 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. 23 increases slowly with the plutonium content of the fuel. For the denser fuels, it appears to consist of a constant

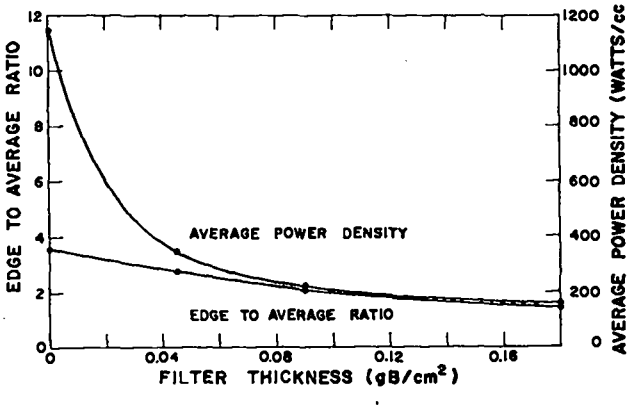

Fig. 22. Effect of natural boron neutron filters.

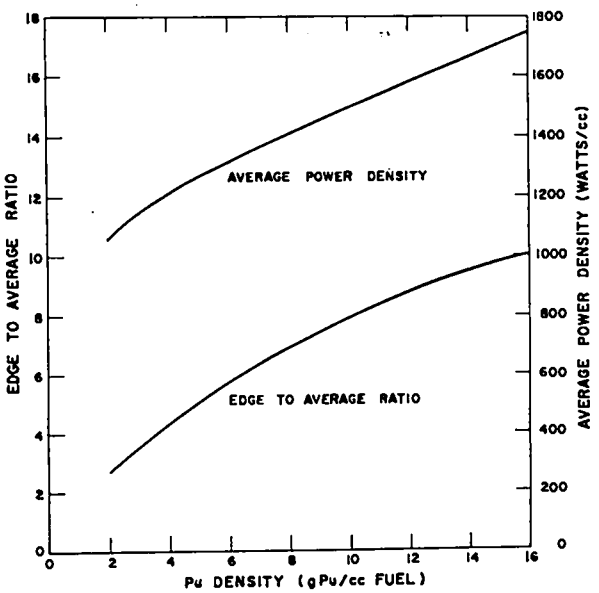

Fig. 23. Effect of plutonium content of fuel.

part, about 1000 W/cc, plus a linear part having a slope of about 42 W/g Pu. These components of the power density can probably be attributed to nonpenetrating and penetrating components of the flux, respectively.

Figures 22 and 23 are for a preliminary design of the irradiation experiment. The final design, used for Figs. 20 and 21, contains more tantalum and gives lower power densities--presumably at the expense of the nonpenetrating component of flux.

CONCLUSIONS

Liquid plutonium fuels have been irradiated to moderate fuel burnups. These irradiation tests have simulated the projected sodium environment and temperatures typical of a large central-station Liquid Metal Fast Breeder Reactor (LMFBR). The thermal stress and power densities are within a factor of 2 of the requirements for LMFBR's. It is not feasible in the OWR to investigate the properties of the fuel at and beyond anticipated LMFBR power densities because of the low available flux.

If this work were to be continued, the following areas should be investigated:

1. Dynamic behavior of the fission products in relation to fuel column height and the response of these fission products to mechanical shock.
2. Fluid dynamics of the fuel in relation to fuel column height and power density.
3. Axial temperature gradient effects, including operation with impressed temperature gradients.
4. Melt-freeze cycling of irradiated capsules.
5. The accumulation and disposition of gross quantities of fission products.
6. Effects of high fuel burnup.

These studies should be made with fuel columns up to 18 in. long. Such prototype fuel element tests would lead to a significant extension of understanding the utility of these fuels for LMFBR application.

Eventually, it would be necessary to irradiate prototype fuel elements in a fast flux spectrum. This could be done in a high-flux thermal reactor with appropriate flux filters for hardening the spectrum or, of course, in a fast reactor test facility. However, irradiation results other than those investigated in this work could be obtained in the OWR, even though the flux is limited. For example, fuel columns up to 6 in. long could be tested by using the environmental cell design with one-half the present heat-leak gas gap thickness. Even this modest increase in fuel height would enhance the fuel surface level detection--an invaluable measurement in the evaluation of items 1 and 2 above.

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