Second United Nations International Conference on the Peaceful Uses of Atomic Energy

A/CONF.15/P/1543
Abstract (USA)
19 March 1958

ORIGINAL: ENGLISH

Confidential until official release during Conference

AQUEOUS HOMOGENEOUS TYPE RESEARCH REACTORS

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Abstract

Since the first International Conference on the Peaceful Uses of Atomic Energy, a number of aqueous homogeneous type research reactors have been constructed. The development, design, manufacture and testing associated with all of these reactors have been the responsibility of Atomics International. Units of basically different designs for operation at approximately five, five hundred, and fifty thousand watts, as well as for the use of uranium fuel enrichments of 20% and approximately 90% U^{235} at all three power levels, were built and their performance determined.

The "L-8" reactor design was developed for operation at 50,000 watts and use of 90% enrichment fuel. This reactor employs a new, closed cycle, wet type, forced circulation system for the recombination of radiolytically decomposed water. In this system an excess of oxygen at nearly atmospheric pressure is forced over the reactor core and then to a catalyst where the decomposition products are recombined. The water formed is carried on to the core vessel. Experiments on this recombining system were carried out under a variety of conditions up to a simulated power level of 120,000 watts. An L-8 reactor, incorporating a number of other new design features, was constructed and tested at the Armour Research Foundation.

The "L-54" reactor, 20% enrichment at 50,000 watts, was designed to use the same gas recombining system as the L-8. Experiments, however, showed the necessity of a large increase in core volume to avoid precipitation of uranium peroxide when using the 20% enriched fuel. An L-54 reactor was constructed for the Japanese Atomic Energy Research Institute. This unit was satisfactorily tested at 60,000 watts. Another L-54 installation has been put into operation at the University of Frankfurt, and two more are under construction for the Institute for Nuclear Studies of Berlin and the Polytechnic of Milan.

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DISCLAIMER

An entirely different design, the "L-6", was developed for use with 90% enrichment and 500 watts of power. This reactor employs a closed gas handling system depending upon the transport of the radiolytic gases to the catalyst by pressure differences automatically established by the recombination process. The system operates at very low pressure and uses no pump or other moving parts. The recombined water is condensed and returned to the core solution by gravity flow. An L-6 reactor was constructed for use in AEC programs at Santa Susana, California. It has been operated very successfully at powers up to 2,000 watts.

Another reactor, the "L-55", was designed for use with 20% enrichment and 500 watts of power. It uses the vapor pressure gas recombiner but differs from the L-6 in the arrangement for shielding, control rods, and experimental facilities. An L-55 reactor was constructed and satisfactorily tested for the Danish Atomic Energy Commission.

For very low powers, the "L-47" reactor, 90% enrichment and 5 watts, was designed and constructed for Atomics International's use in Canoga Park, California. This unit is especially interesting in that it uses only lead and borated water shielding, and is primarily lead reflected.

Another reactor, the "L-77", for 20% enrichment and nominally 10 watts, is being completed in Canoga Park. This unit uses a novel arrangement of several shielding materials. The core size was increased over that of the L-47 to permit the use of 20% enrichment fuel with a predominantly lead reflector. Both reactors use the vapor pressure recombiner, but with this system located inside of the core vessel.

In these various reactors a number of different experimental facilities have been provided, including exposure tubes, horizontal and vertical thermal columns, and gamma-ray exposure facilities near the gas recombiner. Neutron and gamma-ray flux distributions have been obtained in these reactors and their experimental facilities, and compared with theoretical predictions. Different shielding materials have been used and their effectiveness determined. Measurements have been made and compared with theory on critical mass and control rod worth, as well as mass, volume, temperature and power coefficients of reactivity. Extensive laboratory and operational data on the corrosion of austenitic stainless steels have been developed. Experiments have been carried out on the precipitation of uranium peroxide and the use of additives to retard this process. Many other tests were conducted on individual components for the different reactor installations, including especially their gas recombiner systems.

This development work, designs of the various reactors, and the measured performance characteristics of each installation will be described.

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U.S.A.
June 1958

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AQUEOUS HOMOGENEOUS TYPE RESEARCH REACTORS

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INTRODUCTION

At the time of the first International Conference on the Peaceful Uses of Atomic Energy, there were four aqueous homogeneous research reactors in operation. These were the SUPO reactor at Los Alamos, the NCSC reactor at North Carolina State College, and the WBNS and L-3 reactors, both located in California. These reactors were described in the proceedings of the first International Conference. Since that time there have been a number of aqueous homogeneous research reactors of improved design constructed and placed in operation. These designs and the associated experimental development work were carried out by Atomics International.

The aqueous homogeneous, or solution type, research reactor is of special interest because of its inherent safety characteristics. The large negative temperature and power coefficients of reactivity provide an effective shutdown mechanism for an otherwise uncontrolled power excursion. The fuel costs are minimized and fuel additions in any predetermined amounts can be easily and safely made. Versatile experimental facilities can be inexpensively provided, including high level gamma ray exposure facilities which are essentially free of neutrons. This type of reactor has a relatively low cost and only a small operating staff is required to safely and adequately maintain the reactor.

The solution type research reactors placed in operation during the past several years fall into three power range categories. The largest type is the L-54. It is designed to operate at 50 kw and has the advantages of a large graphite reflector, both horizontal and vertical thermal columns, and gamma ray exposure facilities in its sub-pile area. A second reactor, designated the L-55, has been designed to operate at power levels up to 1.5 kw. Its most unique feature is a large 5-1/2 feet square vertical thermal column which is especially useful for exponential type experimental work.

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The lowest power unit, designated the L-77, is designed to operate at 10 watts and is intended primarily as a training and minimum research facility. These reactors, and their operating characteristics will be further described in the sections which follow.

Despite the large variation in operating power, these reactors have several features in common. All of the three reactors described above are designed to operate with uranyl sulfate fuel, the sulfate being preferred to the nitrate because of the greater radiation stability, smaller parasitic neutron absorption, and high uranium concentrations possible. The conversion to the uranyl sulfate fuel solution from the uranium hexafluoride is very carefully controlled to minimize impurities. Specified allowable impurities based on the total uranium metal content are as indicated in Table I.

TABLE I

iron	less	than	200	ppm
boron	"	"	0.2	ppm
cadmium	"	"	0.2	ppm
cobalt	"	"	3	ppm
nickel	"	"	50	ppm
phosphorus	"	"	20	ppm
silicon	"	"	50	ppm
total gadolinium and rare earths	"	"	50	ppm
total halides (except fluorine)	"	"	5	ppm
fluorine	"	"	50	ppm

The primary system of all the solution type reactors is fabricated from type 18-8 columbium or titanium stabilized or extra low carbon stainless steels. All welding is accomplished by the shielded arc tungsten electrode method and controlled and inspected to precise procedures developed over the past ten years. Chemical cleaning, mass spectrometer helium leak checking and pressure checking to 300 psi are also performed in accordance with rigid procedure. The completed system is capable of withstanding a hydrogen and oxygen explosion without damage. In all cases the core vessels are spherical, being assembled from two hemispheres, each hemisphere having been spun from a 0.093-inch stainless steel sheet. The spinning process reduced the core vessel wall thickness to a minimum of 0.068 inch. Before welding the two hemispheres together by the shielded arc tungsten electrode process, they are annealed and cold sized. Other similarities in design will be apparent from the following descriptions of each reactor.

L-54 (AND L-8) 50 KW REACTOR

The L-8 reactor was designed and built for the Armour Research Foundation, Chicago, Illinois. It was installed and started up in 1956 and is presently operating at 10 kw. It is expected that 50 kw operation will begin this year.

At about the time the Armour reactor was completed, a second reactor of its type was being designed for the Japanese Atomic Energy Research Institute. It was apparent that modifications would be necessary due to the limitation of using 20 per cent enriched uranium fuel. This revised design became known as the L-54 reactor. The Japanese L-54, or JRR1, was installed and first operated in the summer of 1957. It has been operated at a maximum power level of 62 kw. A similar reactor installation was recently completed in Frankfurt, Germany, and one is nearing completion in Berlin, Germany. A picture of the Frankfurt reactor installation is shown as Figure 1. Another unit of this type is being fabricated and will be located in Milan, Italy. The sub-pile area for the Milan reactor has been redesigned to isolate the various components from each other so that replacement or repair, if necessary, will be more easily accomplished. This and other design changes which will be described in further detail have been made to improve the usefulness and operability of the reactor.

DESIGN

A detailed study of the Armour reactor 90 per cent enriched uranium fuel and core size indicated that if the uranium were enriched to only 19 or 20 per cent in the U^{235} isotope, the core volume would have to be increased. The Armour core consists of a 12-inch diameter sphere, containing approximately 12 liters of fuel solution with slightly more than 100 grams of total uranium per liter. The 20 per cent enriched fuel design has a fuel solution volume of approximately 26-1/2 liters containing approximately 250 grams total uranium per liter. This solution concentration was selected since it was believed to be well below the concentration (400-700 gms/liter) which might permit the formation of insoluble uranyl peroxide during reactor operation and results in a near optimum hydrogen to uranium ratio. In other essential respects, the cores for use with 90 per cent and 20 per cent enriched fuel are the same. Heat is removed during operation by circulating distilled water through 1/4-inch diameter cooling coils spaced equally throughout the core volume. The four vertical re-entrant control rod thimbles are located equidistant from each other on a 6-inch diameter circle. Immediately above each core vessel is an overflow chamber designed to temporarily retain any fuel solution which might be expelled from the core during a severe reactor transient. A cutaway view of the reactor core showing the internal cooling coils, control rod sleeves, and overflow chamber, is shown in Figure 2.

One of the primary considerations in designing the fuel and gas containing system for a reactor of this type and power level is to devise a reliable method for recombining the gases which are released due to the radiolytic decomposition of water in the fuel solution. Over 10 liters per minute of hydrogen and oxygen gases are released when the reactor is operating at 50 kw.

A system which has become known as the wet type gas handling system was developed and is shown schematically in Figure 3. This system consists primarily of a canned rotor pump which forces water through an exhauster causing an oxygen sweep gas to flow through a catalyst chamber of platinized alumina pellets, up to the region immediately above the fuel solution where

it thoroughly mixes with the evolving hydrogen and oxygen, and then back to the catalyst bed where the hydrogen and oxygen are recombined. The excess water thereby formed in the recombiner passes over a level regulating device and is carried back to the core by the sweep gas.

The gas recombiner, cooling system, fuel drain tank, piping, and valves are all located in a steel lined room below the reactor called the sub-pile room. During the early design phase of the L-8 and L-54 type reactors, it was believed that the primary system could be decontaminated if necessary for the repair or replacement of any of these components. However, it has since been found very difficult to obtain adequate decontamination, particularly of the platinized aluminum oxide catalyst; and repair or replacement of components is quite time consuming. Therefore, the sub-pile area was completely redesigned for the L-54 Milan reactor. The gas recombiner and gamma ray exposure facilities were located in a separate compartment. The canned rotor pump, fuel drain tank, valves, pressure transducers, gas flowmeter, catalyst heater, and thermocouples are all located in the adjacent area or extend into and are readily accessible for replacement from the adjacent area. The canned rotor pump, which also becomes quite radioactive due to fission products collecting on its inside surfaces, was isolated from the other equipment by lead shielding blocks. If necessary, it will be possible to remove most of these components without any decontamination of the primary system and the others with relatively little decontamination being required. The new arrangement of the sub-pile area is shown in plan view in Figure 4. Section and plan views of the reactor are shown in Figures 5 and 6. In all other important aspects, the Milan reactor is identical to the previous L-54's installed in Japan and Germany.

The steel lined sub-pile room and an aluminum enclosure surrounding some of the reflector graphite and the stainless steel core provide the secondary enclosure for the system. This enclosure is entered through a gas tight door which is normally closed and interlocked with the reactor system such that it cannot be opened when the reactor is operating.

The reactor room shell becomes the tertiary enclosure and is usually designed to be gas tight to the extent of permitting 1 to 5 per cent volume leakage per day when the reactor is located in a densely populated area. Arrangements are made such that all doors and ventilation ducts could be sealed off if an accident occurred which released gaseous radioactive materials into the reactor room.

The experimental facilities located above the reactor room floor level include the following:

One horizontal beam tube, 10.1-cm diameter (15.2-cm hole in shield)
Two horizontal beam tubes, 10.1-cm diameter
Four vertical beam tubes, 10.1-cm diameter
Two horizontal beam tubes, 7.16-cm diameter
One straight pneumatic tube, 5.1-cm diameter
One curved pneumatic tube, 3.8-cm diameter
One central exposure tube, 3.8-cm diameter

One horizontal thermal column, 1.52 m square by 1.60 m long
Two thermal-column access ports, 15.2-cm diameter
Two thermal-column access ports, 15.2-cm square
One vertical thermal column, 1.37 m in diameter at its upper surface

Space for the second horizontal thermal column is initially filled with removable dense-concrete shielding blocks.

In the sub-pile area, exposure facilities are designed to utilize the gamma rays emitted from the gas recombiner. This gamma source originates from the decay of gaseous fission products and is almost neutron free. Four 10.1-cm diameter ports and one 15.2-cm x 45.7-cm rectangular gamma port extend into the region near the gas recombiner. For the Milan reactor, a special 3.7-cm diameter sleeve extends through the gas recombiner and is expected to be in a gamma flux region of about 5×10^5 r/hr when the reactor is operating at a power level of 50 kw.

There are no particularly unique features in the electronic control and safety systems of this type reactor. Temperature, pressure and flow indicating and control systems, radiation monitoring units, and reactor safety, control and automatic power regulating equipment are mounted in the control console which is located in the control room adjacent to the reactor. The reactor safety and control channels receive signals from two fission chambers, two compensated ion chambers and one uncompensated ion chamber. A typical control console is illustrated in Figure 7.

Four control-safety rods, each controlling approximately 1.9 per cent reactivity, fall into the reactor at essentially the speed of gravity when the electro magnets holding them are de-energized by any of a number of scram signals. The rod drive units are of the rack and pinion type and are neatly compacted and housed above the reactor shield.

OPERATION

The Armour reactor has been in operation since the first part of 1957 and, as of April 1958, the machine had accumulated approximately 10,000 kw hr. Their maximum licensed power level is 10 kw but will be increased shortly to 50 kw. Details of the operational experience with the Armour reactor are reported more fully in a separate paper included in the proceedings of the second International Conference.

Performance of the Japanese L-54 has been very satisfactory since it was first placed into operation. Maximum power levels up to 62 kw have been obtained with all components functioning as designed. During initial power runs, there was noticed an initially unexplained decrease in oxygen pressure. Subsequent review of the data indicates the oxygen loss may be due to incomplete oxidation of the uranium from the plus 4 valance state to the plus 6 state during preparation. The uranium in the core could then have been

completely oxidized to the plus 6 state by the radiolytic oxygen generated during power operation, thus causing the measurable loss in pressure.

The physics of the L-8 and L-54 system are not directly comparable because of the change in core size and the superior gross reflector properties obtained in the L-54 design. The correlation of the experimental data by group diffusion and perturbation theory has not been satisfactory. It is felt this is due primarily to the complex reflector geometry of this type reactor. As is mentioned later, theoretical correlation of experimental data on the more simple geometries is much more satisfactory. Table II is a comparative tabulation of the physics of the L-8 and L-54 systems.

TABLE II

	L-8	L-54
Core diameter, inches	12.0	15.75
Fuel enrichment	88.14	19.91
Solution volume at critical, liters	11.92	24.36
Critical mass, gms U^{235}	1,218	1,172
Solution density, gms/cm^3	1.21	1.33
Temperature coefficient, %/°C	-0.024	-0.04*
Mass coefficient, %/gm U^{235}	~0.015	0.0195*
Water coefficient, %/gm	0.0013	~0
H/U^{235} ratio	~235	~490
Control rod worth, each, %	~1.7	~1.9

* based on listed rod worth

Initial measurements of the thermal neutron flux in the L-54 type reactor indicate that the level remains high throughout the graphite reflector. Measurements of the flux through the central exposure tube, 2-inch straight pneumatic tube, and the horizontal thermal column are presented in Figures 8 and 9. All flux data have been normalized to one (1) at the core center. Preliminary absolute flux measurements indicate the center core thermal flux is 1.3×10^{12} at 50 kw reactor power.

The control rods of the L-54 reactor were determined to have an individual reactivity worth of approximately 1.9 per cent. This is about 30 per cent higher than that predicted by simple perturbation theory. Theory also predicts large positive shadowing effects among the various rods, although no such effect could be detected experimentally.

L-55 (AND L-6) - 1500 WATT REACTOR

The second type aqueous homogeneous reactor, differing markedly from the previously described 50 kw unit, is the L-55. This reactor is rated for operation to power levels of 1500 watts. Two reactors of this type, differing only in the geometric arrangement of the concrete shielding, have

been constructed and are in operation. One unit is located at the Santa Susana, California field laboratory operated by Atomics International; the other unit, DR1, has been installed in the Danish Atomic Energy Commission laboratory at Risø, Denmark.

DESIGN

The reactor is basically a 12-1/2-inch diameter, type 347 stainless steel, spherical core vessel embedded in a right circular cylinder of reactor grade graphite, 5 feet in diameter by 54 inches high. At the rated power level, the core is adequate for fuel of 20 per cent as well as 90 per cent enrichment in U^{235}. The gas recombiner, further described in the following paragraph, is located outside and adjacent to the 5-foot diameter reflector tank which encloses the graphite. The recombiner connects to the core through a 2-inch line. The core, recombiner, and associated piping constitute the primary system which is all welded except for one stainless steel ring joint flange which allows convenient disassembly for shipping. During operation at power, the core temperature is maintained constant by a cooling system circulating distilled water. The water discharges the heat picked up in the core to a secondary refrigerant type cooler for eventual dissipation to either the atmosphere or other cooling water system. The entire core, reflector, and recombiner assembly is completely enclosed by a 46-inch thick octagonal heavy concrete shield. Figure 10 indicates the general orientation of the various reactor components as installed. Figure 11 illustrates details of the reactor core assembly.

The L-55 reactor utilizes a vapor pressure type recombiner. This type recombiner, which completely contains all the fission gases, is extremely simple and requires no moving parts for its operation. The unit relies upon a low total system pressure of the order of 2-1/2 cm mercury absolute for most efficient operation but will operate reliably up to 15 cm of mercury absolute. Radiolytic gases from the core, at a rate of approximately 0.2 liters/min/kw, are conducted through the 2-inch line connecting the top of the core vessel to the recombiner tank where the gases impinge upon a platinized aluminum oxide catalyst and are recombined. Cooling coils located within the recombiner condense the water vapor, the condensate returning to the core by gravity drain. The recombination process automatically establishes a pressure differential between the core and the recombiner, ensuring the transportation of gases to the catalyst bed. Laboratory and operational experience conclusively demonstrate no danger exists from hydrogen-oxygen explosions as long as the low system pressures are maintained.

Reactivity is controlled by four horizontal control rods which move tangentially to and outside of the core vessel, one on either side and the other two immediately underneath. The rods are driven by reversible electric motors. Two of the rods are designated as safety rods and are designed to decouple from the drive assembly by means of an electromagnet. The two safety rods are driven to the "in" position by gravity weights located in wells under the drive assembly. The four rods are similar in construction, the neutron absorbing section consisting of a flat stainless steel tube 50 inches long with a cross section of 3-3/4 by 1/2 inch. Each tube is filled with one kilogram of compacted boron carbide powder. The two safety rods are provided only with "in" and "out" light indicators. The other two units,

designated as the shim and regulating rods, are provided with selsyn transmitters which furnish remote rod position indication at the reactor control console.

The reactor core is accessible for experimental purposes through a 1-inch diameter exposure tube which penetrates the central region of the core. Additionally, 4-inch square graphite stringers adjacent to the core can be removed from the reflector tank. The stringers are in the fourth, sixth, eighth and tenth graphite layers. In the sixth and eighth layers, the stringers can be removed from both sides of the reflector tank, making four through holes available. Above the core, the top shield concrete plugs can be removed and the resultant void space stacked with graphite, making available a 5-1/2-foot square vertical thermal column. With the vertical thermal column, the reactor has adequate capacity to drive a normal exponential experiment.

The instrumentation and control system is conventional in every respect and composed of proven components. The neutron flux is monitored by four channels: one channel consisting of a fission chamber monitoring to three decades above source level and reading out on a log count rate meter; two channels, monitoring from just above source level to full power using compensated ionization chambers driving linear and log micro-microammeters; and a fourth channel using an uncompensated ionization chamber reading out on a microamp movement meter relay unit. Additional instrumentation monitors the temperature and pressure at various points and provides information on fuel solution level. All the instruments are mounted at the control console.

OPERATION

The machine operated by Atomics International, the L-6, was placed into operation as a 1500-watt reactor early in 1957. The critical mass with 93 per cent enriched fuel was determined by multiplication measurements to be 756 grams U^{235} with a total of 12.45 liters of core solution. Operation of the L-55 reactor located in Denmark, commenced in August 1957. The critical mass of this unit, fueled with 19.90 per cent enriched uranium, was found to be 984 grams U^{235} with 13.05 liters of core solution. During critical assembly, loading of the reactor core with highly enriched material involves only small changes in core volume, whereas loading with 20 per cent enriched material involves large changes. The influence of this on the inverse multiplication vs. mass curve is shown in Figure 12. The distinct departure from linearity for the lower enrichment can be explained by the larger geometric buckling change as the critical experiment progressed. A summary of the core solution composition at critical for each reactor is given in Table III.

TABLE III

CORE SOLUTION COMPOSITION AT CRITICAL

		Reactor using 93% enriched fuel	Reactor using 19.90% enriched fuel
U^{235}	gms	756	984
U^{238}	gms	58	3,962
H_2SO_4	gms	231	404
UO_2SO_4	gms	1,256	7,614
H_2O	gms	12,033	12,712

For all enrichments, the experimental critical mass agreed within approximately 5 per cent with that calculated using two-group diffusion theory and fitted group constants. A typical flux distribution through the core and reflector is shown in Figure 13. Measurement of the maximum thermal flux in the reactor fueled with the 93 per cent enriched material yields a value of approximately 7×10^{10} neuts/cm^2-sec at 1500 watts. The flux in similar reactors of different enrichments is inversely proportional to the mass of U^{235} present. The gamma intensity in the core during operation is approximately 10^5 r/hr.

A summary of the various physics parameters associated with the reactor is listed in Table IV, both for the case of 93 and 19.90 per cent enriched fuel.

TABLE IV

PHYSICS CONSTANTS

	L-6 Reactor using 93% enriched fuel	L-55 Reactor using 19.90% enriched fuel
Critical mass, gms U^{235}	756	984.4
Solution volume, cubic centimeters	12,447	13,050
Solution density, gms/cm^3	1.085	1.505
Temperature coefficient, %/°C	-0.027	-0.033
Mass coefficient, %/gm U^{235}	0.044	0.016
Water coefficient, %/gm H_2O	0.0015	0.00168
H/U^{235} atom ratio	395	318
Reflector	graphite	graphite
Fast fission factor*	~1.0	~1.0
Resonance escape probability*	~1.0	0.946
Thermal utilization*	0.76	0.83

* calculated

Differences in the values can be adequately correlated with the difference in fuel enrichment.

The reactivity worths of the control rods have been measured to be approximately 1.5 per cent on the reactor fueled with 19.9 per cent enriched material. Values of rod worth calculated by perturbation theory satisfactorily check the experimental numbers for the case of the higher enrichment but consistently underestimate the worth of the rods for the 19.90 per cent enriched case. Underestimation of rod worth through perturbation theory is unusual; normally this theoretical procedure overestimates the reactivity controlled by a poison rod.

In use, the L-55 reactor system has proven to be dependable and simple to operate, requiring a minimum permanent staff of only one operator and a part-time electronics technician. The reactor can be readily started up or shut down without regard to the past operating history. Power levels from a fraction of a watt to 1500 watts can be adequately controlled to a precision approaching one per cent by means of the automatic control system. The fluxes are sufficiently high to permit a wide variety of experimental work, yet are not so high as to require elaborate and extensive handling systems.

L-77 (AND L-47) 10 WATT REACTOR

The 10-watt reactor was developed to fulfill the need for a low power, extremely safe reactor intended primarily for training in the field of nuclear engineering. It is equally desirable that the reactor possess sufficient flexibility to be useful as a low flux research tool. These restrictions dictate a reactor of minimum size, one that can be transported and easily installed in existing buildings; would require a minimum of maintenance; could be operated by a small staff of limited experience, and, above all, a reactor of demonstrated safety.

Initial work was started in late 1956. The inherent safety characteristics of the solution type reactor, as demonstrated in the KEWB experiments, led to the selection of this type unit for a training reactor. Also, it was early decided to utilize the vapor pressure recombiner, previously described, because of its extreme simplicity. A second major reason for selecting the vapor pressure type recombiner is that the required operating point of the recombiner, namely, a low total system pressure, is exactly the condition which yields greatest safety from the standpoint of a nuclear excursion.

On the basis of early critical experiments at Oak Ridge National Laboratory, it appeared that lead would be equally satisfactory as water as a neutron reflector for the core. Lead also has the obvious advantage of being a compact gamma ray shield.

The initial conceptual design reduced, therefore, to an integral water boiler type core-vapor pressure recombiner completely surrounded by a lead reflector-shield. Because of the uniqueness of this concept, it was decided to construct a prototype unit, designated the L-47, with the power level set at 5 watts.

The design of the prototype is shown in Figure 14. The core consists of a 12-1/2-inch diameter sphere on which is located a canopy assembly containing the recombiner. The entire unit is surrounded by a 6-inch thick, 3500-pound lead shield. To obtain adequate neutron shielding during operation, the entire core reflector assembly is supported in the middle of an 8-foot diameter by 8-foot high shield tank which is filled with water. Reactivity control is obtained through two 0.980-inch diameter vertical control rods operating in reintrant thimbles spaced diametrically opposite each other 3 inches on either side of the core center. The poison material is a 10-inch long sleeve of .032 inch thick cadmium. The rods were found to control approximately 1.2 per cent each in reactivity. The only experimental facilities provided are a series of five through tubes in the core plus two additional beam tubes terminating at the lead surface. The reactor instrumentation, kept as simple as possible consistent with the safety of the reactor and the needs of the experimental program, is conventional in every respect.

The reactor was first placed in operation in August 1957. The initial loading of the fuel was of 89 per cent enrichment in U^{235}. Criticality was reached when 1196 grams of U^{235} had been loaded into the core. This number is somewhat lower than that predicted by two-group three-region diffusion theory. The initial operation of the reactor was satisfactory except for the expected high radiation levels outside the shield tank at 5 watts power. Analysis indicated the predominant components of the radiation consisted of capture gammas originating from the steel and the water outside the primary lead reflector. To suppress water capture gammas, the shield tank water was poisoned with a boron compound, sodium octaborate, with boron concentrations up to 10 grams per liter. Each increase in the boron concentration required the addition of further U^{235} to the reactor core to maintain criticality, indicating that the six inches of lead did not function as an infinite reflector. The effect of the boron loadings on reactivity is shown in Figure 15. Throughout the boronation experiments, a series of neutron flux traverses were made. Thermal neutron flux was determined by means of indium and gold foils and miniature BF_3 counters. Fast neutron flux data were obtained with NTA-type film. These data are presented in Figure 16. A summary of the pertinent reactor parameters is given in Table V.

TABLE V

L-47 REACTOR CHARACTERISTICS

Reflector	6" lead followed by water
Critical mass, gm U^{235}	1196
Solution volume, cm^3	12,110
Solution density, gm/cm^3	1.146
Mass coefficient, %/gm U^{235}	0.014
Water coefficient, %/gm H_2O	0.0023
Temperature coefficient, %/°C	-0.02*
H/U^{235} atom ratio	243

* approximate due to uncertainty in the temperature measurements

The operation of the prototype reactor proved the feasibility and safety of operating the water boiler type core and vapor pressure recombiner in close proximity to each other in the same vessel, the desirability of using lead within the reflector region, and the adaptability of the water boiler core to this application.

Utilizing the data obtained on the prototype reactor, design was started in late 1957 on a production type reactor possessing the requirements of a versatile training and educational tool. In addition to the requirements imposed on the prototype unit, the production model reactor was to be made suitable for use with fuel of 20 per cent or greater enrichment. The power level was increased to 10 watts. This reactor has been designated the L-77 Laboratory Reactor.

To better accommodate the use of 20 per cent enriched fuel, the core of the L-77 was increased to 15-3/4-inch diameter, the same size successfully used on the L-54 type reactor. This core size increase was considered necessary on this reactor but not on the 1500-watt L-55 because of the poorer neutron properties of the L-77 reflector. The location of the recombiner assembly in the canopy above the fuel solution was retained, as were the five experimental through tubes in the core, these having proved extremely useful during the L-47 experimental program. A cutaway of the core is shown in Figure 17.

The shield design was optimized to permit the operation of the reactor at power levels of 10 watts with low accompanying radiation levels external to the reactor tank. The core is surrounded by a three-region composite shield assembly. Referring to Figure 18, it is seen the inner shield is composed of a mixture of pelletized lead and a relatively radiation stable organic compound. This inner region is a minimum of six inches thick and serves a dual function as the primary core reflector. Completely surrounding the inner shield is an intermediate region, a minimum of two inches thick, composed of normal paraffin mixed with a soluble organic boron compound. A third solid shield, composed of borated paraffin and pelletized lead, encloses the inner and intermediate solid shields on the sides and top. The core composite shield assembly is canned in a stainless steel vessel 43 inches in diameter and 56 inches high.

The entire core-shield assembly is contained in an 8-foot diameter by 7-foot high outer shield tank which is filled with normal water during reactor operation. Access to the experimental facilities is from outside the reactor tank after unlocking and removing the shield plugs provided. The experimental facilities are more fully described as follows:

One main exposure tube	1.37 in. (3.48 cm) inside diameter
Four auxiliary exposure tubes	0.75 in. (1.90 cm) inside diameter
Two beam tubes	1.875in. (4.75 cm) inside diameter
Two spare instrumentation thimbles	2.75 in. (7.00 cm) inside diameter
One radioactive gas tube	0.15 in. (0.38 cm) inside diameter

Reactor power is monitored from source level to full power, a range of approximately five decades, by two neutron channels. Both channels utilize

uncompensated ionization chambers, located in the pelletized lead-organic shield under the reactor core, to drive micro-microammeters located on the control console. One micro-microammeter furnishes a linear readout of reactor power; the other, the log of power and its derivative, or period. Additional instrumentation indicates rod position, temperature, and pressure. An annunciator furnishes information on the cause of all reactor shutdowns. The operating controls are grouped conveniently on the turret immediately in front of the operator.

It is planned to add the experimental data from the L-77 operation as a supplement to this paper.

DEVELOPMENT PROGRAM

The development of components and information pertaining to aqueous solution type research reactors has been a continuing program at Atomics International since early 1952. Major work has been concentrated in those areas which prior work with aqueous homogeneous reactors had indicated major deficiencies existed; namely, the gas handling system, fuel stability, and corrosion.

GAS HANDLING SYSTEMS: Work at Atomics International was early directed toward the development of a gas handling system capable of operation at reactor powers of 50 kw or above. Earlier type gas handling systems had all operated on the "dry cycle", some of which required the undesirable periodic release of gaseous activity from the system. In addition, a gas blower was necessary in the dry system and difficulties had been encountered in maintaining good seals or in developing a hermetically sealed blower with a long expected life. Such a system was not considered suitable for a reactor located in a populated area.

The Wet Type Gas Handling System (WTGHS), which was developed, eliminates the necessity of discharging radioactive gases from the system, automatically maintains the system water balance between the recombiner and core, and cools the circulating pump and catalyst bed. The WTGHS consists basically of a tank containing approximately 12 liters of distilled water in which is located a submerged catalyst bed. Water is circulated from the recombiner tank sump in a closed cycle through a heat exchanger, the pump, a gas ejector by which the pressure differential for circulating the gases is established, and back to the recombiner sump. Cooling water circulated through the secondary side of the heat exchanger removes the heat generated by the pump and the recombination of the radiolytic gases. The radiolytic gases, in a carrier stream of oxygen, are pulled from the core by the water ejector, the gases being discharged to the space above the water level in the recombiner tank. From here, the gases are drawn through an entrainment eliminator, the catalyst bed, and thence back to the core. Excess water in the recombiner sump, due to recombination of the hydrogen and oxygen, overflows into a wier from which it is picked up by the gases returning to the core. A sketch of a typical mechanical arrangement of the recombiner system is shown in Figure 19.

The upper limit of operation of the WTGHS is set by the hydrogen concentration as determined by the diluent gas volume flow rate and the amount of water vapor contained therein. The lower limits of hydrogen concentration, resulting in inflammation in a hydrogen-oxygen system, are approximately 4.5 per cent; for detonation, approximately 15 per cent. Measurement of the hydrogen concentration in the operating WTGHS shows a maximum level of approximately 6 per cent entering the catalyst bed and 1.5 per cent leaving. The above figures are based on a gas generation rate at 50 kw for a reactor operating on fully enriched fuel. For reactors utilizing 20 per cent enriched uranium, the gas generation rates are approximately 30 per cent less, thereby permitting the recombination system to operate at higher equivalent reactor powers. A series of test runs have been performed on a developmental WTGHS recombiner unit in which stoichiometric mixtures of hydrogen and oxygen were charged to the system. Only mild explosions or flashbacks were audible with a corresponding maximum recorded pressure of 22 psi. Most of the explosions originated from the catalyst bed, in which case the entrainment eliminator and large recombiner tank volume effectively prevent flashbacks into the region above the core. During operation, the recombiner system automatically transfers back to the core any solids or fuel which might have been carried over due to entrainment. At 50 kw steady state operation, the equilibrium concentration of uranium in the recombiner sump water is well under 0.1 grams.

Development tests of the WTGHS were carried out to equivalent reactor powers of 120 kw (155 kw for reactors utilizing 20 per cent enriched fuel) and the system functioned normally in every respect. At present, four reactors employing the WTGHS were operating satisfactorily, with two additional units scheduled to go into operation soon. Typical operating conditions with the system operating at 50 kw are shown in Table VI.

TABLE VI

WTGHS OPERATING CONDITIONS AT 50 KW

Sweep Gas Flow, CFM	7.6
Recombiner Pressure, psia	12
Recombiner inlet gas temperature, °C	50
Recombiner outlet gas temperature, °C	36
Catalyst bed upper temperature, °C	240
Catalyst bed center temperature, °C	270
Catalyst bed gas outlet temperature, °C	170
Recombiner tank gas temperature, °C	33
Recombiner tank water temperature, °C	27

A second type recombiner developed at Atomics International, using the same platinized aluminum oxide catalyst but operating on a different principle, is satisfactory for use on solution type reactors operating at somewhat lower power levels than the L-54. This unit, called the vapor pressure recombiner, has been previously described. Laboratory tests conducted on the behavior of this recombiner system demonstrate that no explosion danger exists from

the stoichiometric hydrogen-oxygen mixture, particularly so when the total system pressure is maintained at one or two psia.

In a series of tests, a number of capsules were prepared by evacuation and filling with stoichiometric mixtures of hydrogen and oxygen. In summarizing the results of these tests, the following points should be noted:

a) That in the absence of a radiation field, it appears that only an active dry catalyst such as platinum can cause ignition of a stoichiometric hydrogen and oxygen mixture.

b) That even a dry platinum catalyst (at up to 100°C initial temperature) can not cause ignition if the system pressure is below 10 inches Hg absolute.

c) That a high gamma field (about 10^5 r/hr) not only did not ignite the gas mixture, but did not produce an increase in the rate of recombination.

Apparently a threshold pressure exists for the combination of hydrogen-oxygen mixtures. A capsule containing a glowing filament was filled with different initial quantities of hydrogen and oxygen. It was found that below approximately two to three inches Hg absolute, there was no detectable ignition or explosion.

Laboratory experiments demonstrate the capacity of the vapor pressure recombiner system is limited only by the size of the catalyst bed and the heat removal capacity. The unit installed on the L-55 type reactor has a 1400 gm catalyst bed and has been operated in the laboratory to equivalent reactor powers exceeding 10 kw. Limitation on power level was due to the heat removal capacity resulting in high temperatures and, therefore, high total system pressures.

The vapor pressure recombiner installed in the L-77 reactor, containing 80 gms of catalyst pellets, has similarly been laboratory tested to 400 watts equivalent reactor power. Because of these low anticipated reactor powers, there is no cooling system on this unit other than natural radiation, conduction, and convection to the surrounding structure.

FUEL STABILITY: The operation of a low temperature aqueous solution type reactor core results in radiolysis of the water to give hydrogen and oxygen gases. One of the intermediate steps in the process involves the formation of hydrogen peroxide which can react with the soluble uranyl ion to give the insoluble uranyl peroxide. Atomics International has investigated several aspects of the problem toward reducing the equilibrium concentration of hydrogen peroxide in the fuel solution. Primary emphasis has been on the interrelation between hydrogen peroxide concentration and reactor specific power, solution temperature, uranium concentration, and catalytic effects.

Results of this investigation indicate the hydrogen peroxide concentration to be proportional to specific power, inversely proportional to

temperature and a complex function of uranium concentration and other catalytic effects. Temperature is found to decrease the hydrogen peroxide concentration by a factor of approximately 10 for every 20°C rise in temperature. The effect of catalyst in the fuel solution is somewhat more complex. For instance, iron is known to have a catalytic effect on the decomposition of hydrogen peroxide, the magnitude of which depends upon the uranium concentration. A measurement of the decomposition specific reaction rate constant at various uranium and iron catalyst concentrations is shown in Figure 20. It can be seen the rate constant is a maximum at a uranium concentration of approximately 300 grams/liter. Over the range of iron concentrations investigated (0-300 ppm), the catalytic effect had not saturated. Copper ion also has been used as a solution catalyst, but its effects are not so pronounced as that of iron. The presence of sulfuric acid also influences the rate constant considerably, increasing with increasing acid concentrations up to 0.5 M acid and then decreasing.

The effect of high radiation intensities on catalyzing the decomposition of hydrogen peroxide is unknown. The correlation of the experimentally determined hydrogen peroxide rate constants with reactor power level and known solubility data has not been too satisfactory, the results always being conservative by a factor of 10 or better. For this reason, it is felt that radiation may play some part in establishing the hydrogen peroxide equilibrium values. There has been no operating problem with uranyl peroxide precipitation in any of the reactors discussed here.

CORROSION: Extensive long term corrosion tests have been conducted at Atomics International on various stainless steels of interest in the solution type reactor program. These tests, coupled with information developed at Oak Ridge National Laboratory and other installations, conclusively demonstrate that corrosion of stainless steels by uranyl sulfate solutions at temperatures under 100°C is not a problem.

Coupons of commercial stainless steels of the following types: 304 (19% Cr, 10% Ni), 309s (23% Cr, 14% Ni), 310 (25% Cr, 21% Ni), 316 (18% Cr, 12% Ni), 321 (19% Cr, 10% Ni, Ti), and 347 (19% Cr, 11% Ni, Cb), have been tested in baths of uranyl sulfate solutions up to 2.5 molar with 1.0 molar added sulfuric acid. The averaged results are summarized in Table VII.

TABLE VII

CORROSION TEST OF STAINLESS STEEL COUPONS

Stainless Steel Type	Uranium Concentration gms/liter	Molarity H_2SO_4	Corrosion Rate mils/yr
304	270	0.5	.102
309s	270	0.5	.043
310	270	0.5	.043
316	270	0.3	.102
321	250	0.5	.165
347	270	0.3	.086
304	600	1.0	.178
310	600	1.0	.074
316	600	1.0	.130
316	450	0.5	.097
321	450	0.5	.113
347	450	0.5	.080

Before placing in the test baths, all coupons were pickled in a 1:1 HNO_3 solution containing 1.5 per cent hydrofluoric acid, followed by passivation in 1:1 HNO_3 and a distilled water rinse. As can be seen by inspection of the above table, all corrosion rates are extremely low, never exceeding 0.2 mils/yr. Metallographic examination of the test coupons indicates no intergranular or pitting type of corrosion attack.

Coupon surfaces were found to be free of discolorations or other unaccounted for blemishes. Because the fuel used in these reactors originates as uranium hexafluoride, there has been some concern about excessive amounts of fluorides carrying through the chemical processing into the finished sulfate. Corrosion checking of coupons exposed in solutions with up to 50 ppm fluorine has indicated no accelerated corrosive action. Coupons containing sample welds have similarly been checked, and weight losses are consistent with the results reported for unwelded samples.

After operation for three years, the primary system of the solution type reactor at the Radiation Laboratory of the University of California was opened up for inspection of the inside surfaces. Micro-techniques of chemical analysis were also utilized to analyze the fuel solution for corrosion products, iron, chromium, and nickel. The ratio of these metals was found to correspond roughly to the composition of the 18-8 stainless steel (type 316L). It was determined that not more than 2 grams of stainless steel total had been removed from the core vessel. This corresponds to an average corrosion rate of approximately 0.02 mils/yr, a factor of two less than the lowest laboratory results. Metallographic examination of the core vessel again showed no sign of intergranular or pitting type of attack.

It is known, from experience at Oak Ridge National Laboratory and with the North Carolina State homogeneous reactor, that the presence of chlorides in the fuel solution may under certain conditions accelerate sharply a localized or pitting type attack. For this reason, it is recommended that the chloride concentration in the fuel solutions be maintained under 5 ppm. Under these conditions it is believed that for all practical purposes the primary systems of these solution reactors have an unlimited lifetime.

CONCLUSION

Solution type research reactors have a longer history of operation than any other type research reactor utilizing enriched fuel. They have proven capable of operation at power levels of at least 50 kw. A wide variety of experimental facilities can be incorporated into the reactor design, including unique neutron free sources of gamma radiation. The reactors are relatively inexpensive to install and can be maintained by a minimal operating staff. There are no problems associated with fuel replenishment because of the very low burnup, the initial fuel charge lasting the life of the reactor.

The homogeneous solution type research reactor has been experimentally demonstrated to be among the safest of all the various types of research reactors now in use. This permits the installation of these units with a minimum of expense at locations in densely populated areas. In the lower power versions, they are ideally suited for training purposes because of the very effective safety mechanism inherent in this type reactor.

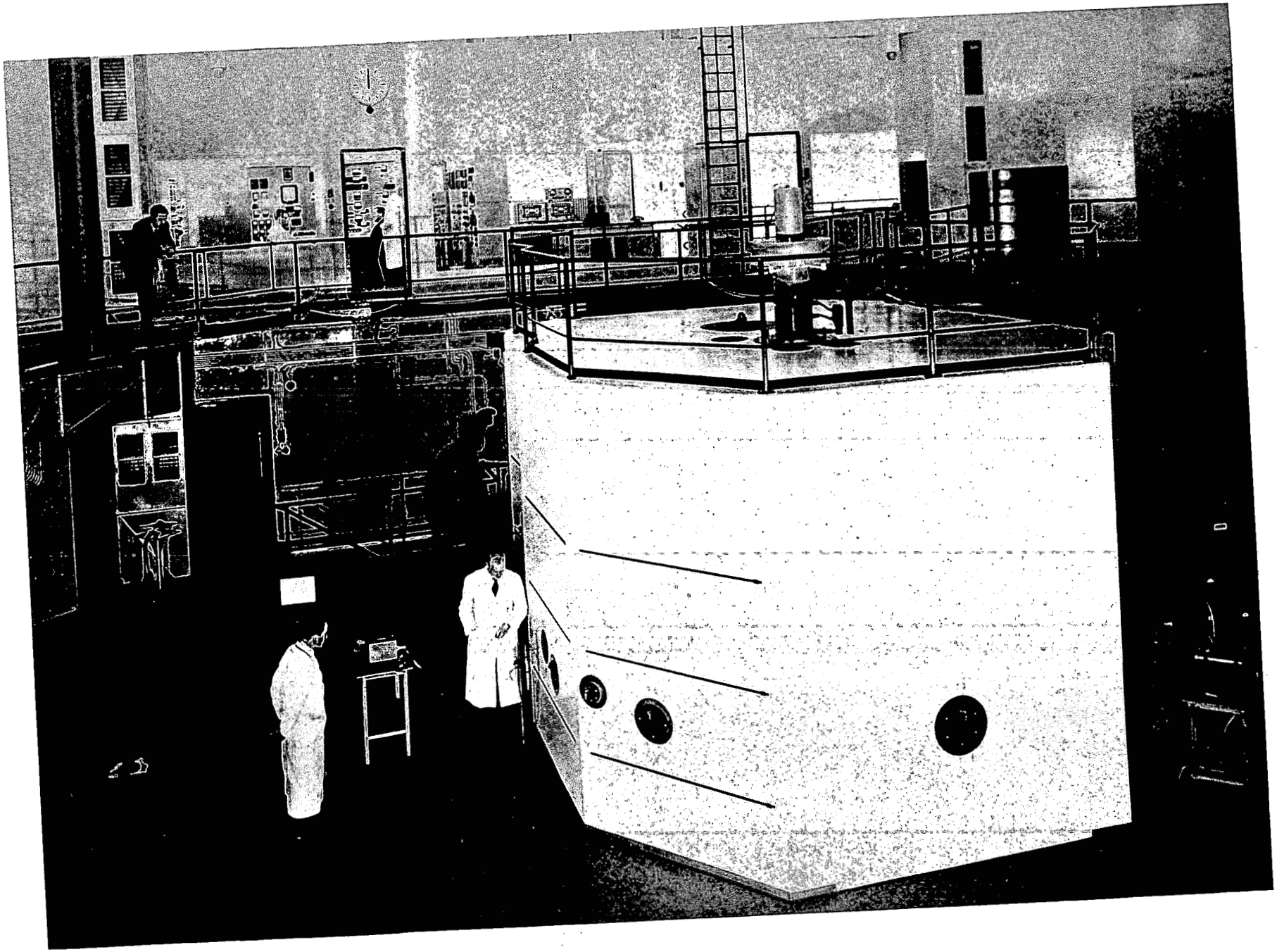

Figure 1 L-54 Reactor Installation, Frankfurt, Germany

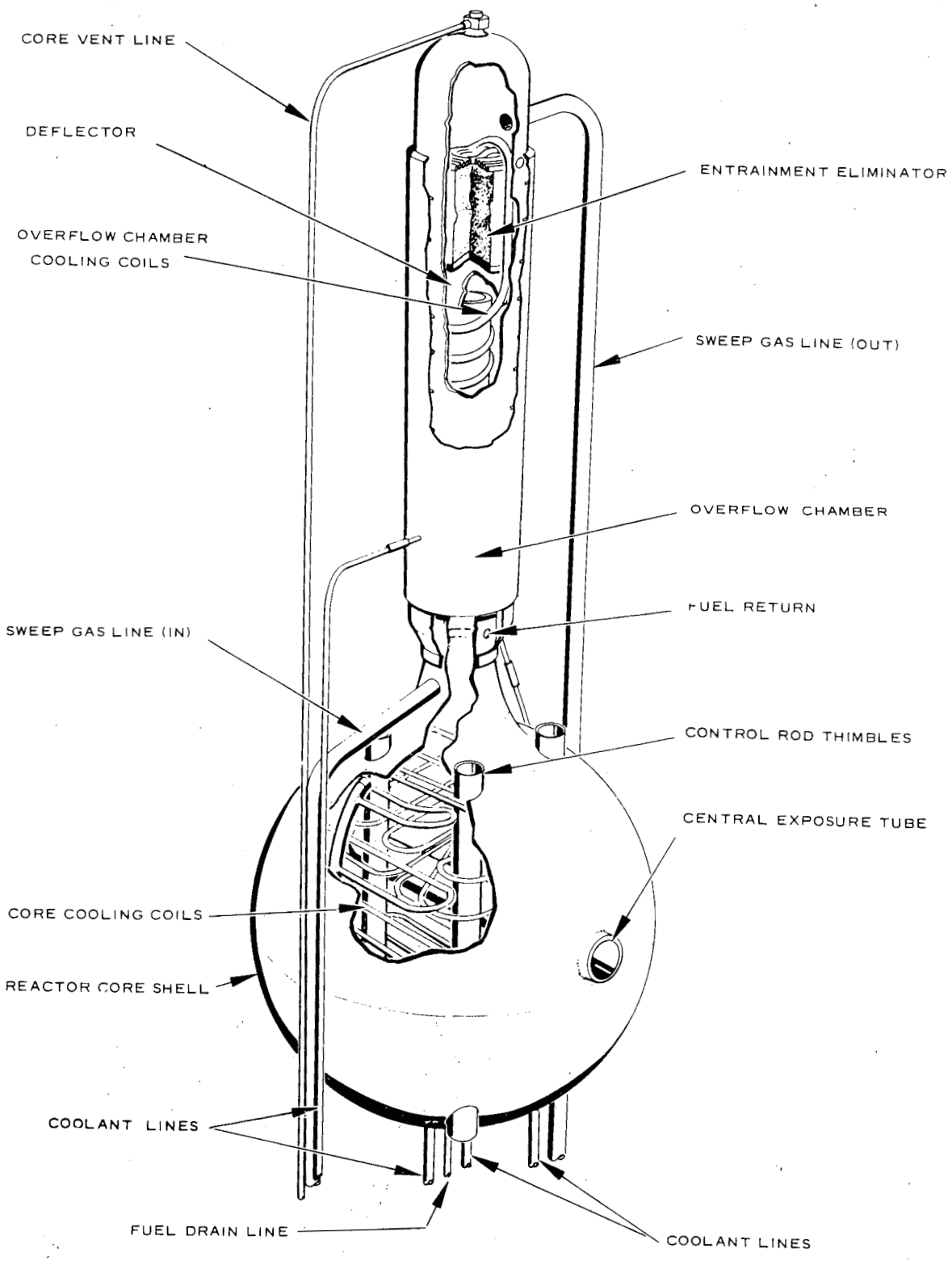

Figure 2 L-54 Reactor Core and Overflow Chamber

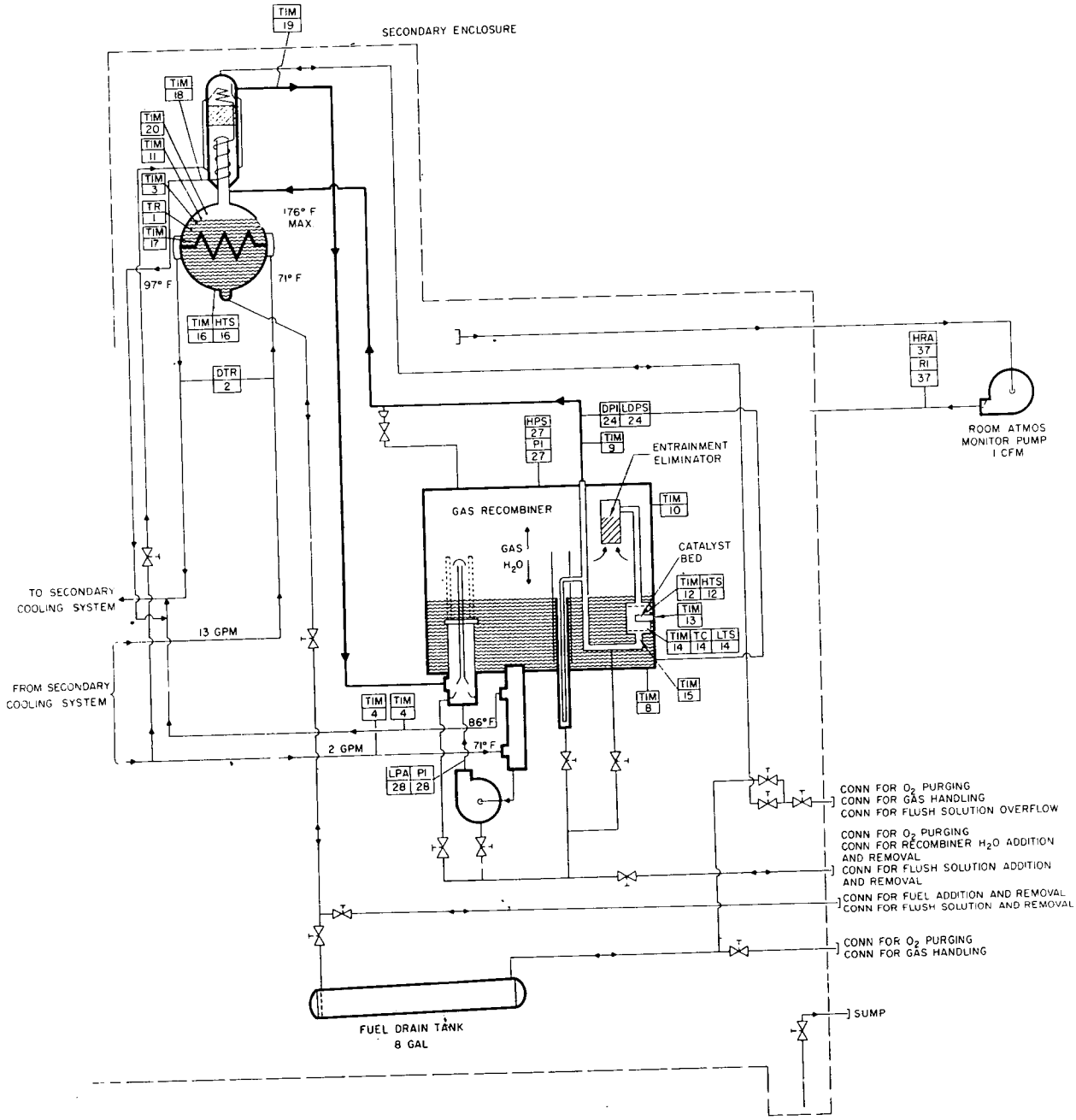

Figure 3. Wet Type Gas Handling System Flow Diagram

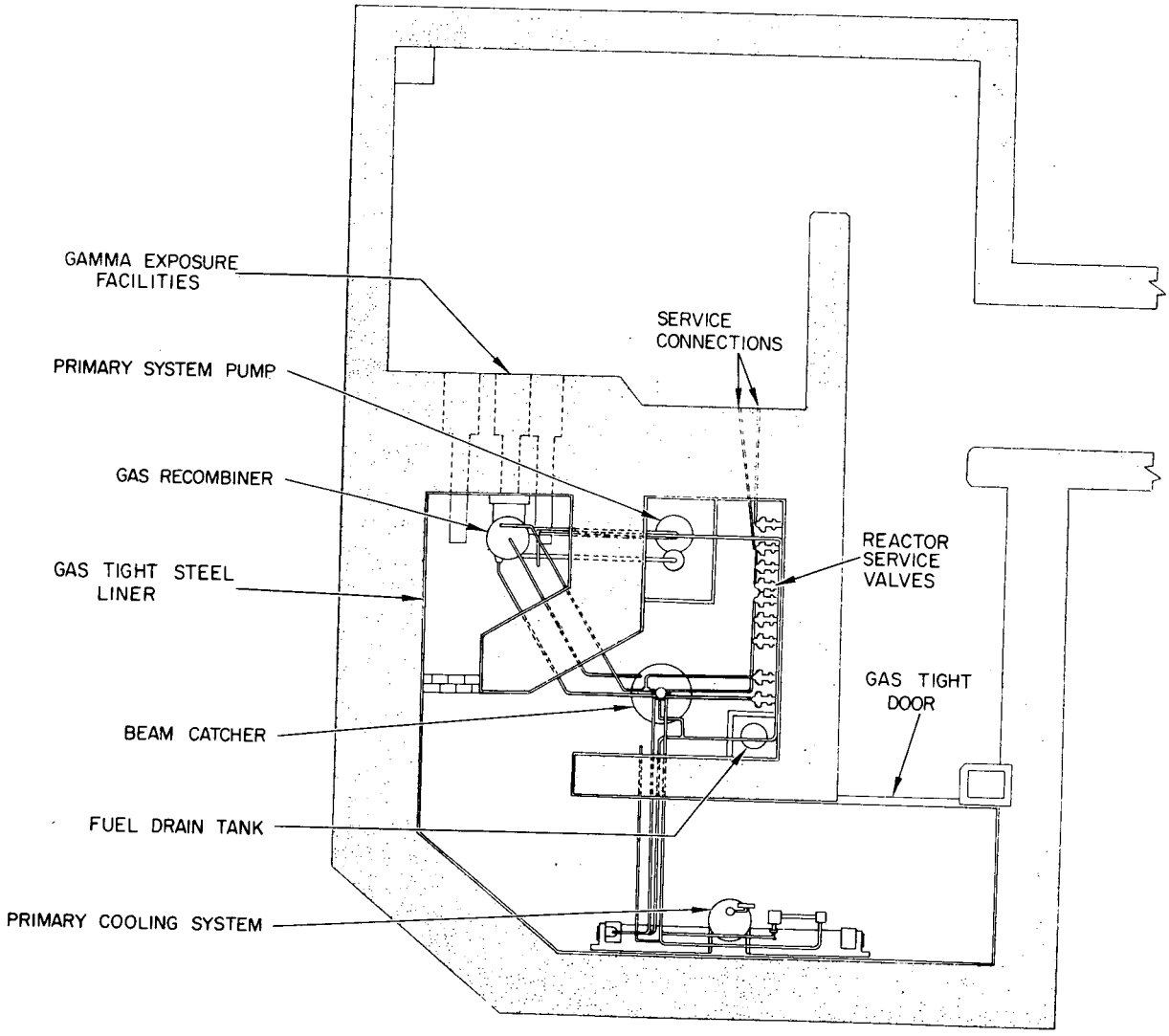

Figure 4. Revised Subpile Room for the L-54 Milan Reactor Installation.

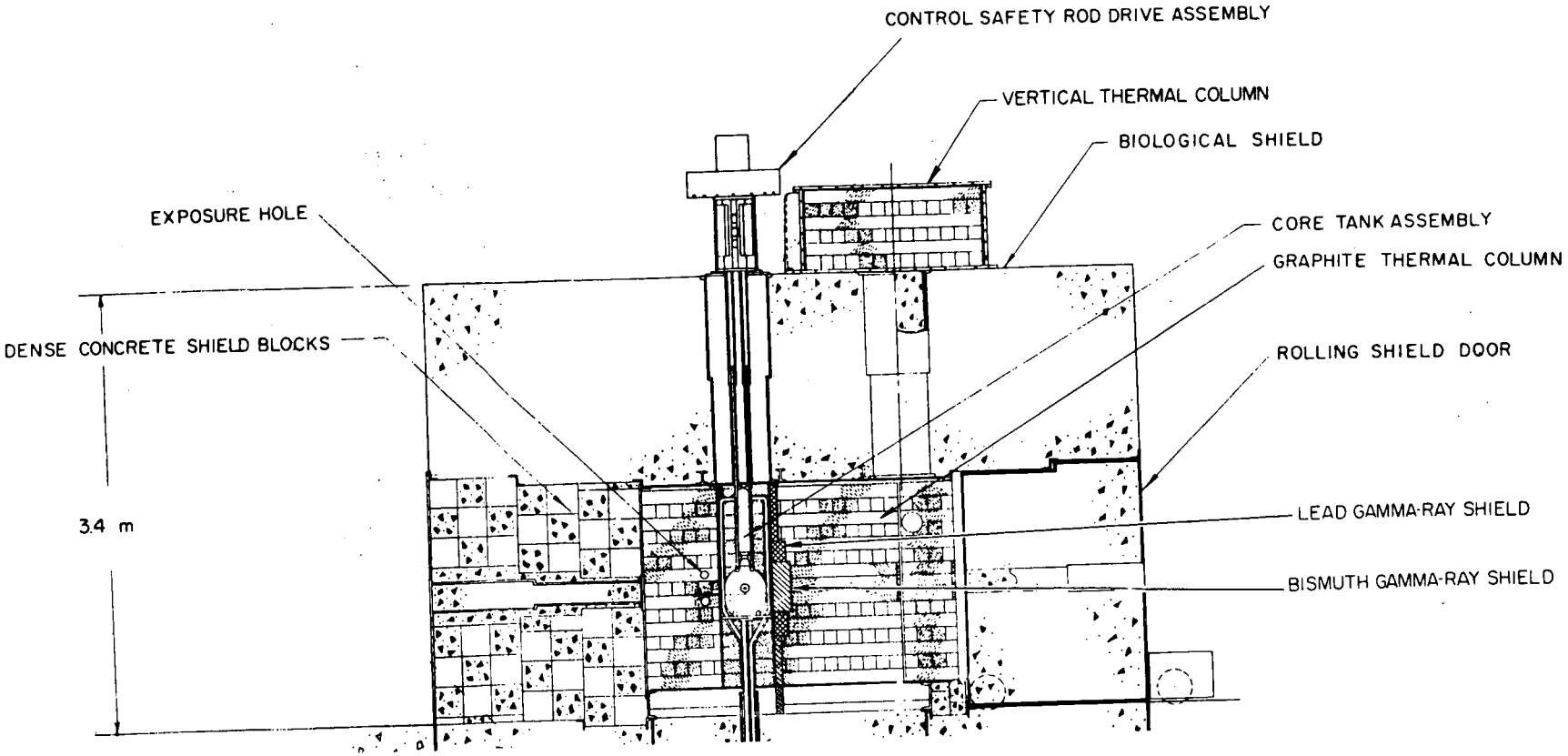

Figure 5. L-54 Reactor, Sectional View

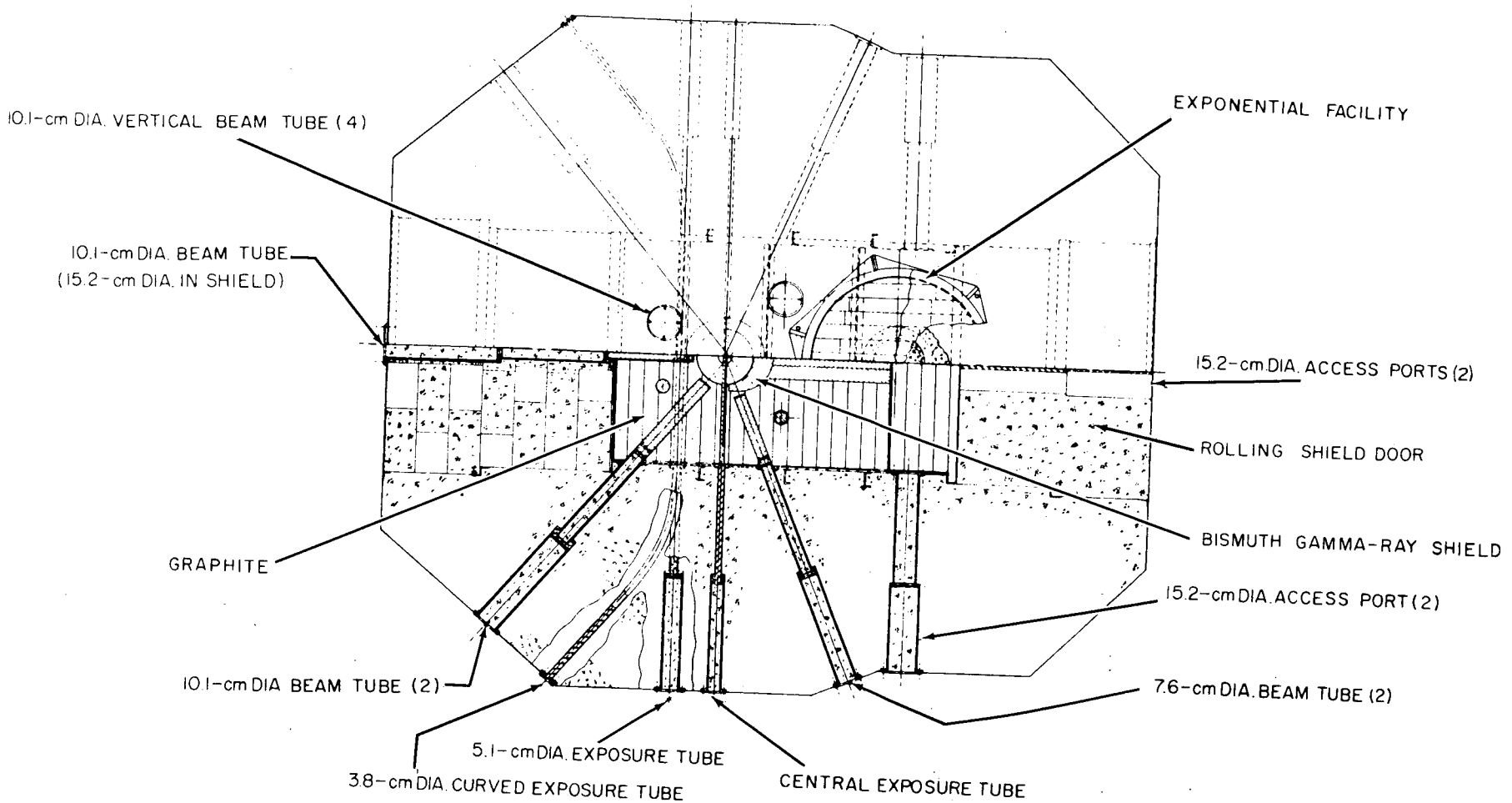

Figure 6. L-54 Reactor Plan View

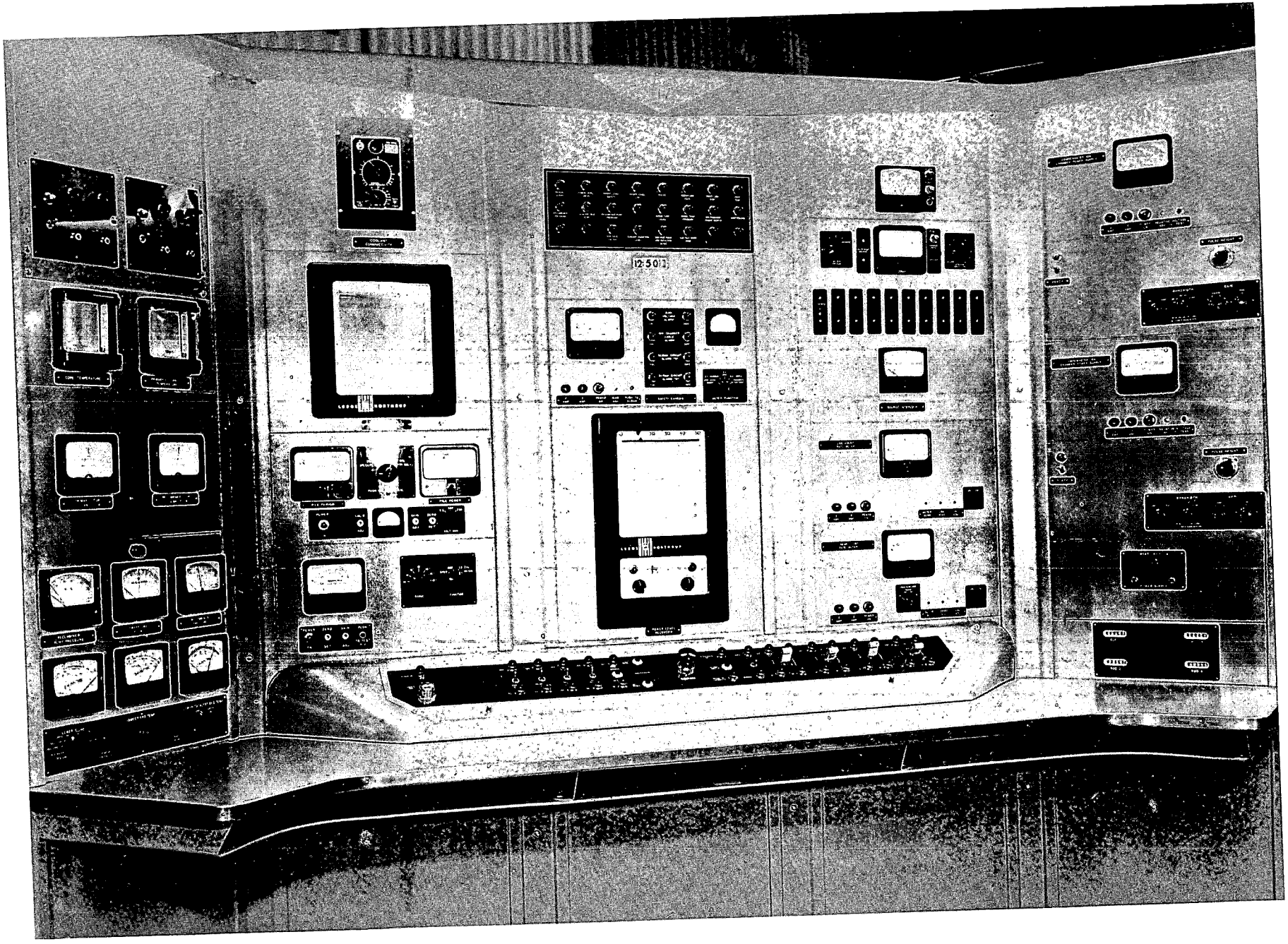

Figure 7 L-54 Reactor Control Console

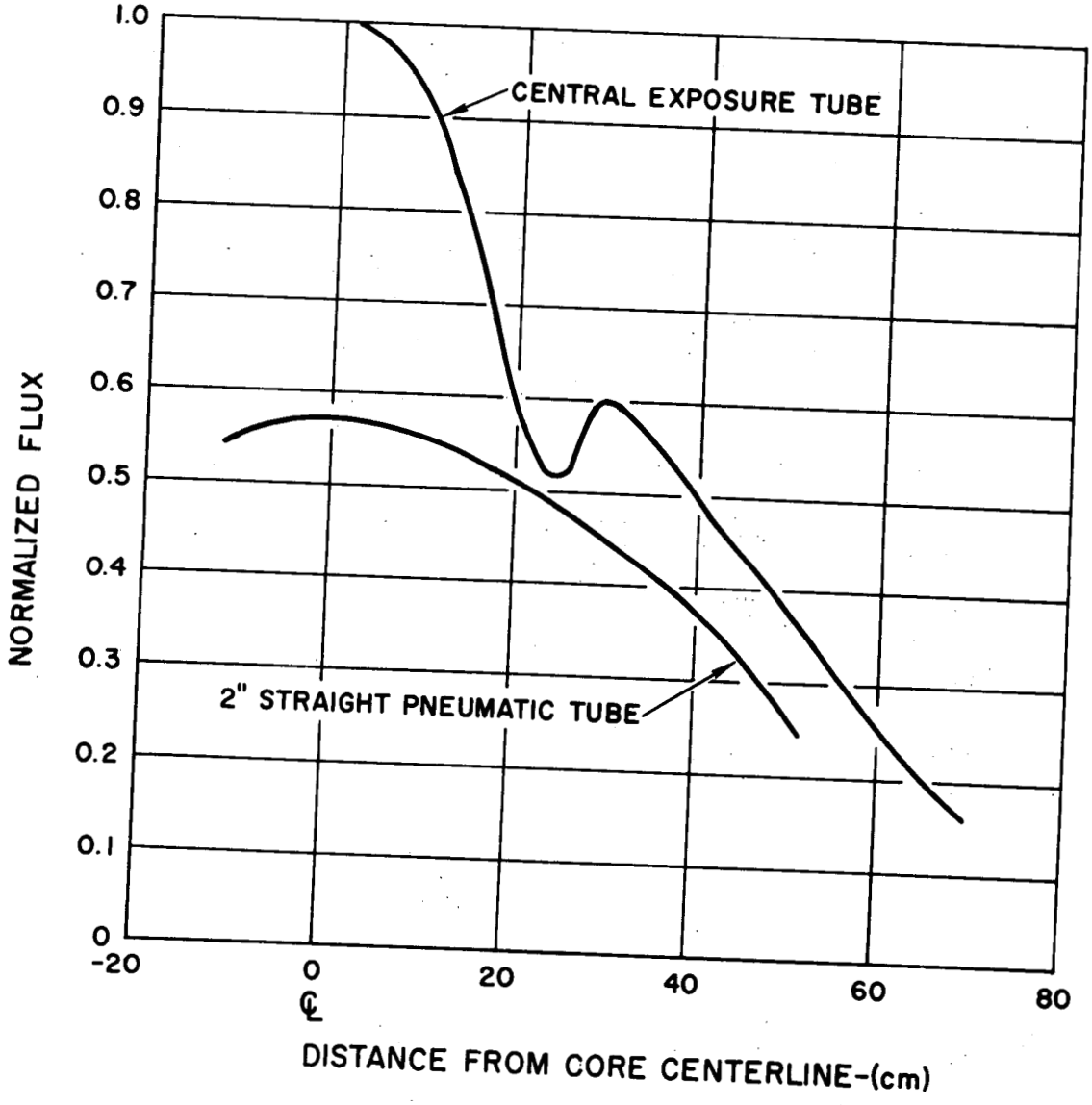

Figure 8
Thermal Neutron Flux Distribution - L-54 Reactor

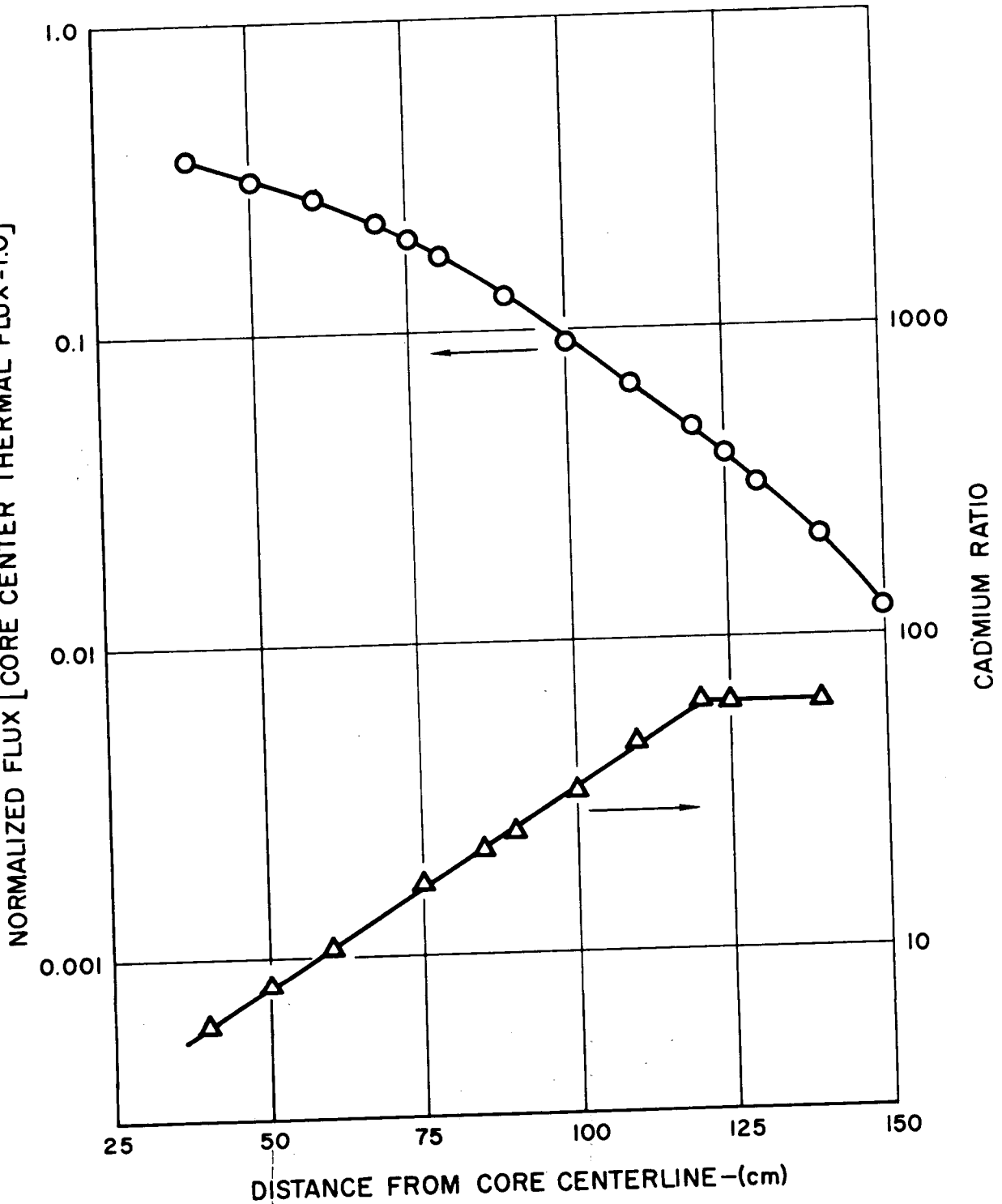

Figure 9
Thermal Neutron Flux Distribution Through L-54 Horizontal Thermal Column

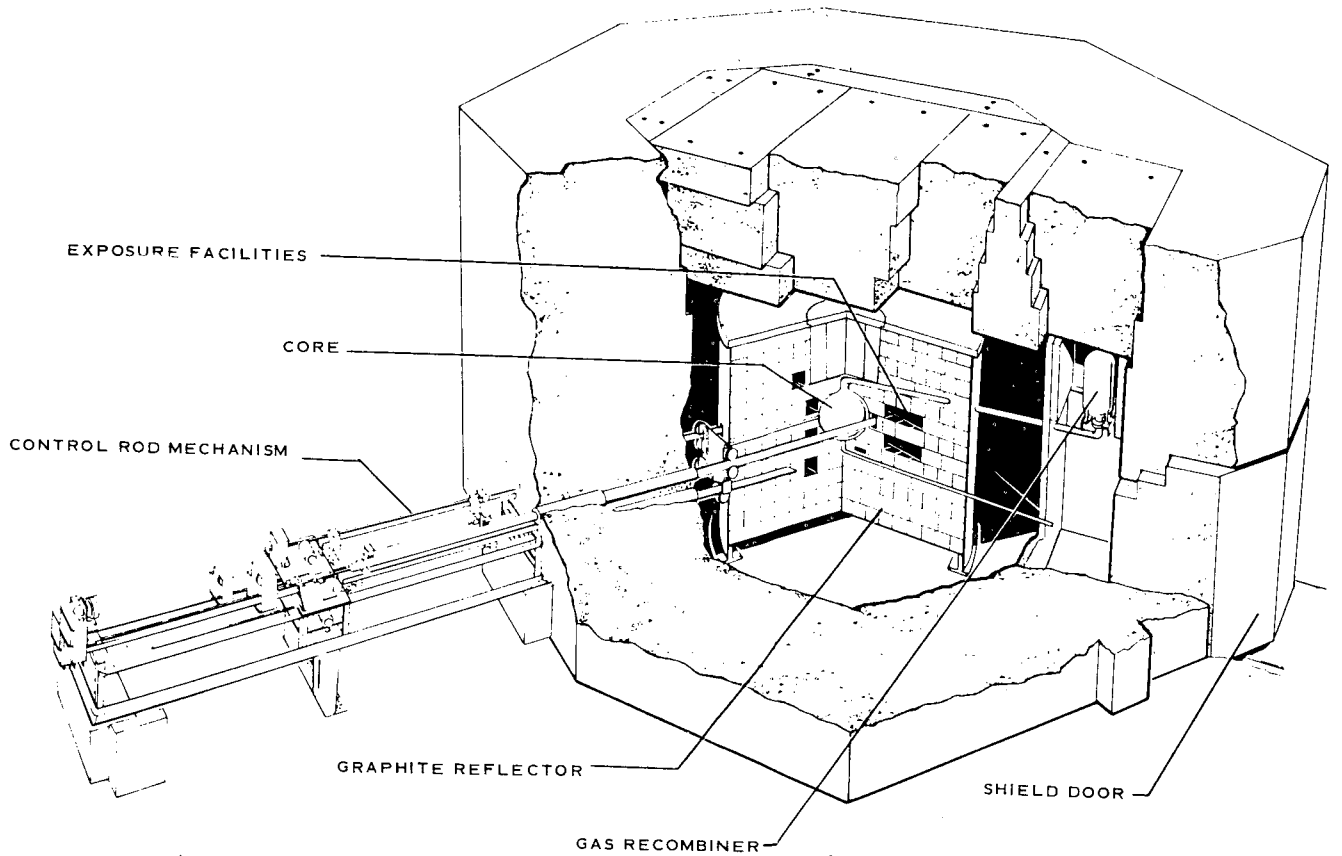

Figure 10. L-55 Reactor Installation

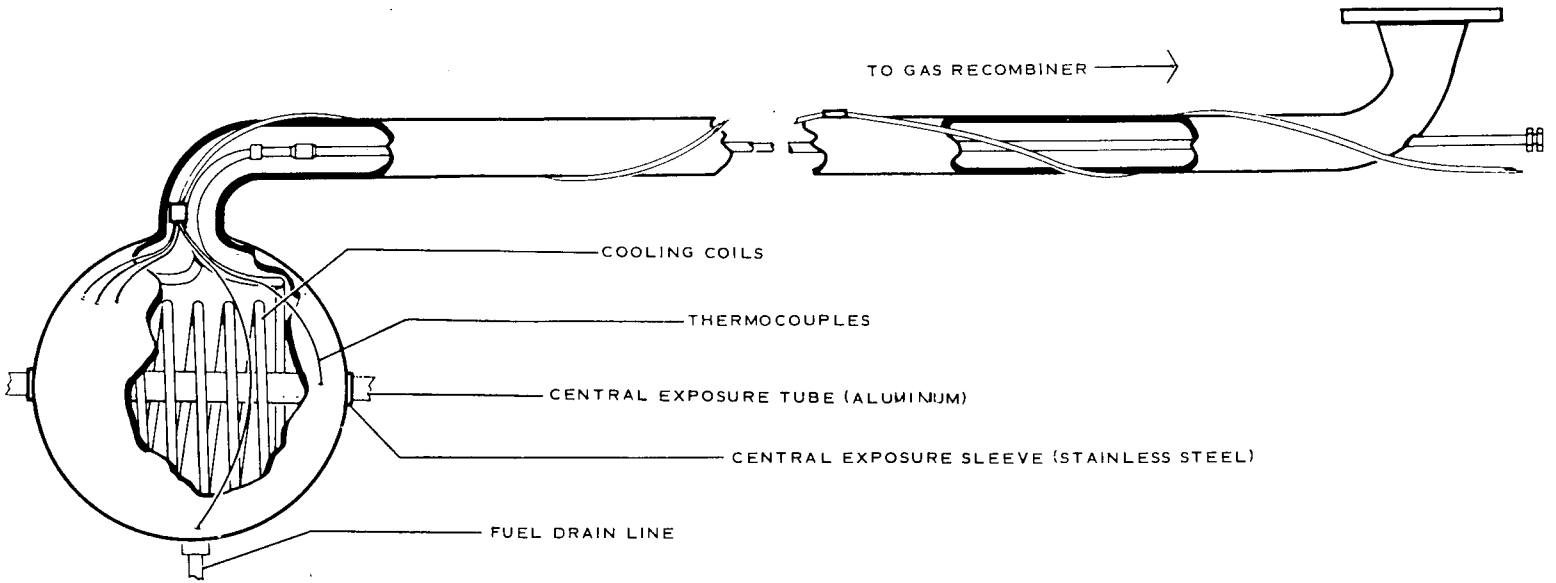

Figure 11. L-55 Reactor Core Assembly

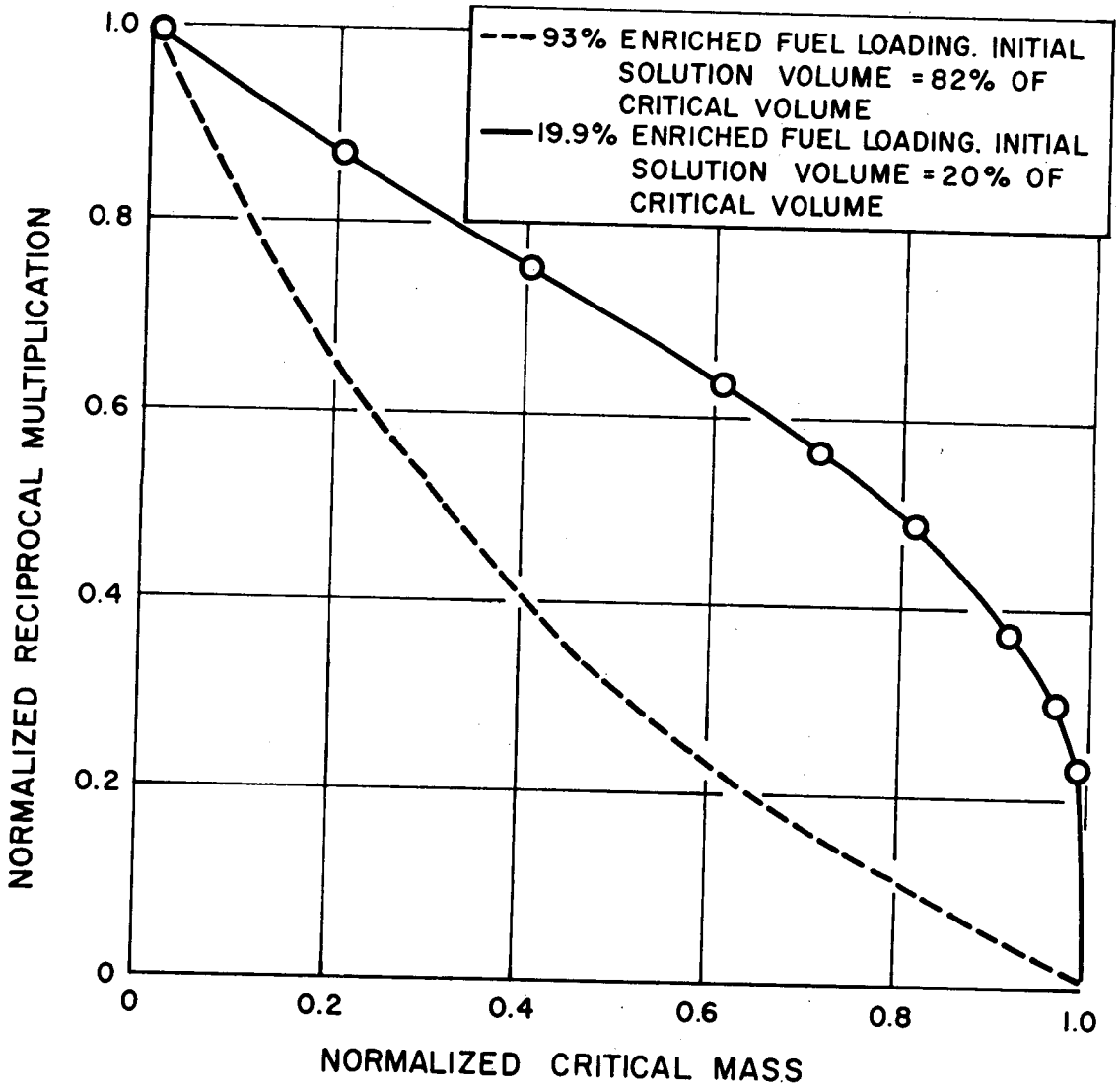

Figure 12. Typical Inverse Multiplication Versus Mass Curves.

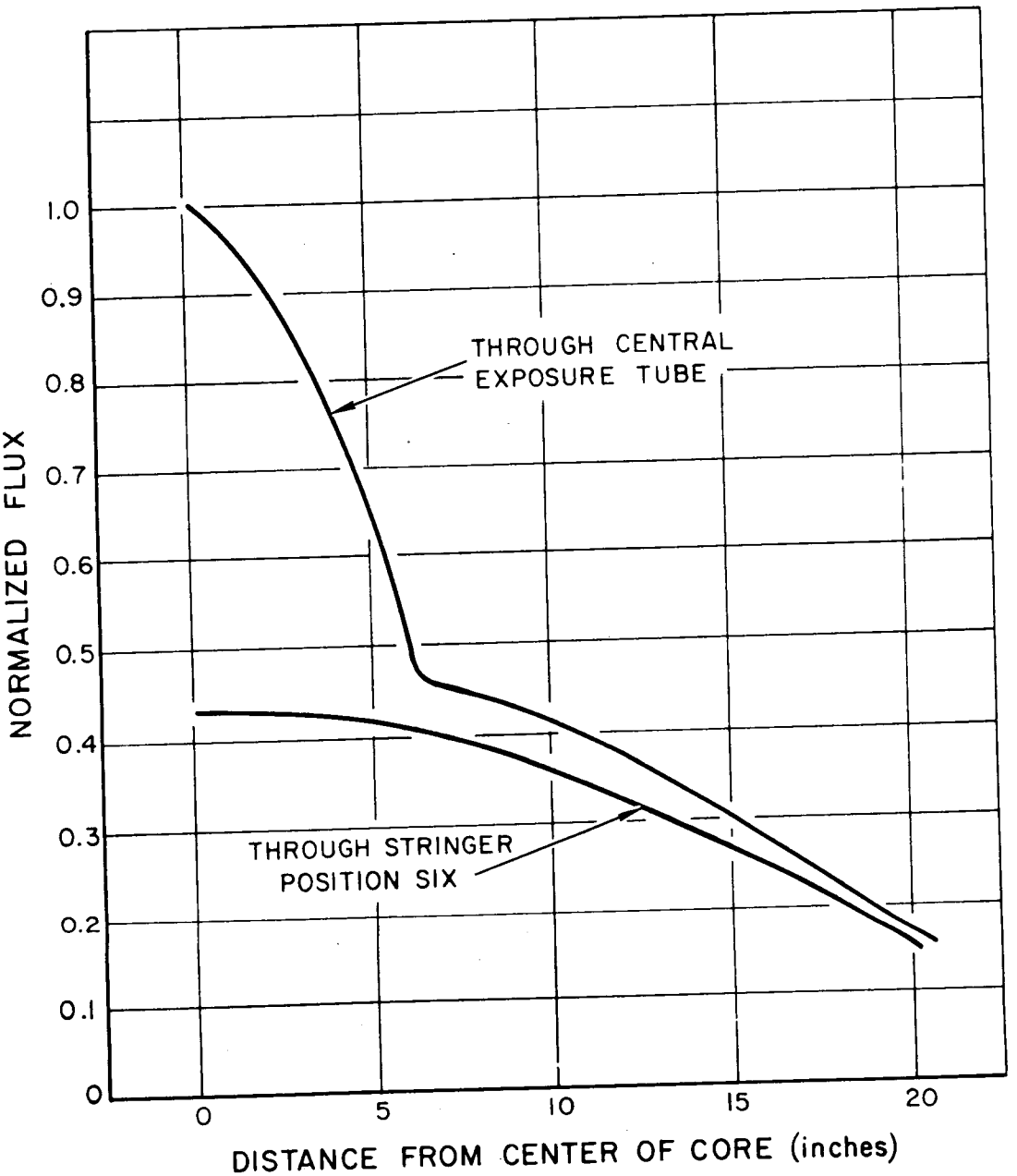

Figure 13. L-6 Reactor Thermal Flux Distribution, 93% enriched fuel

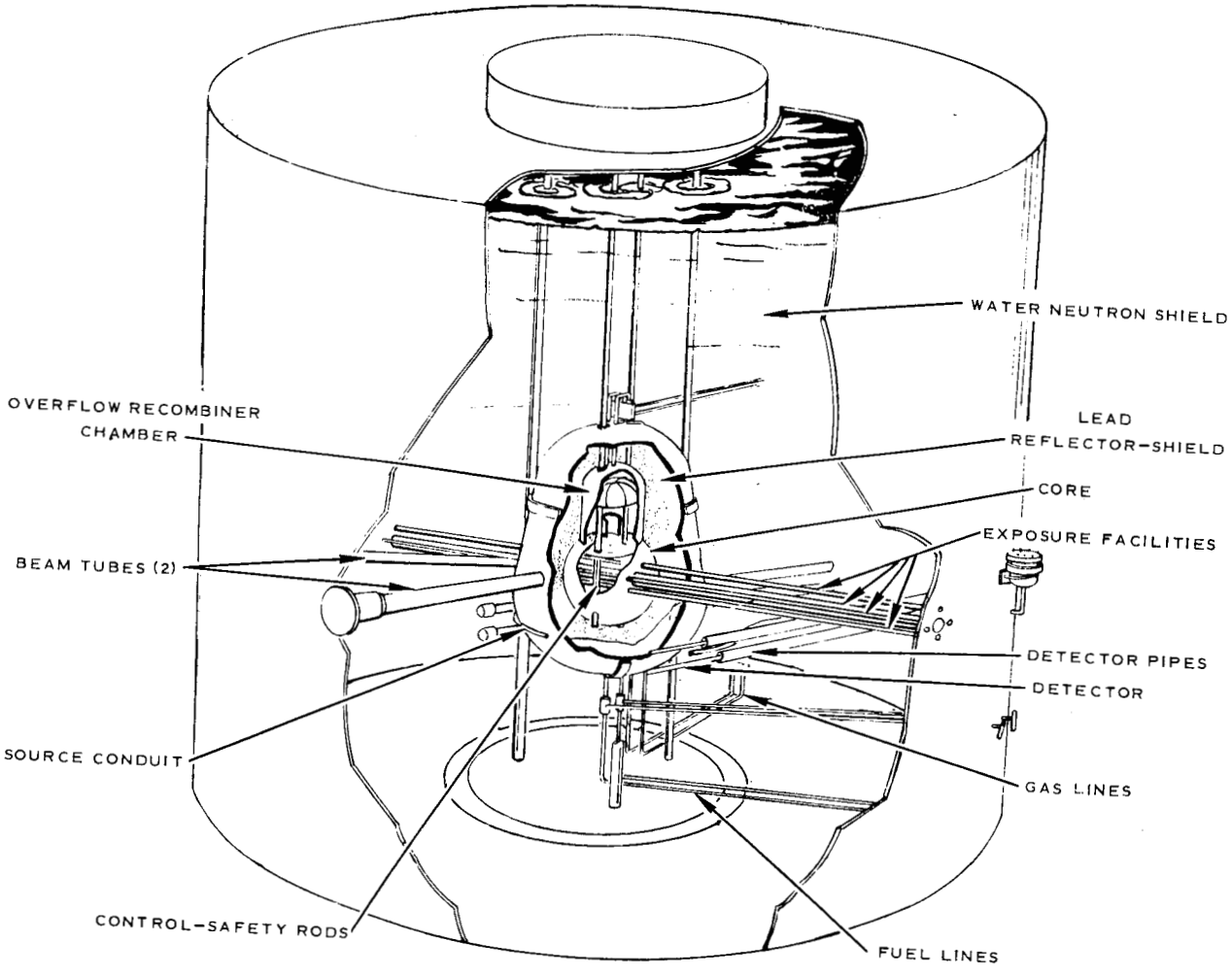

Figure 14. L-47 Reactor

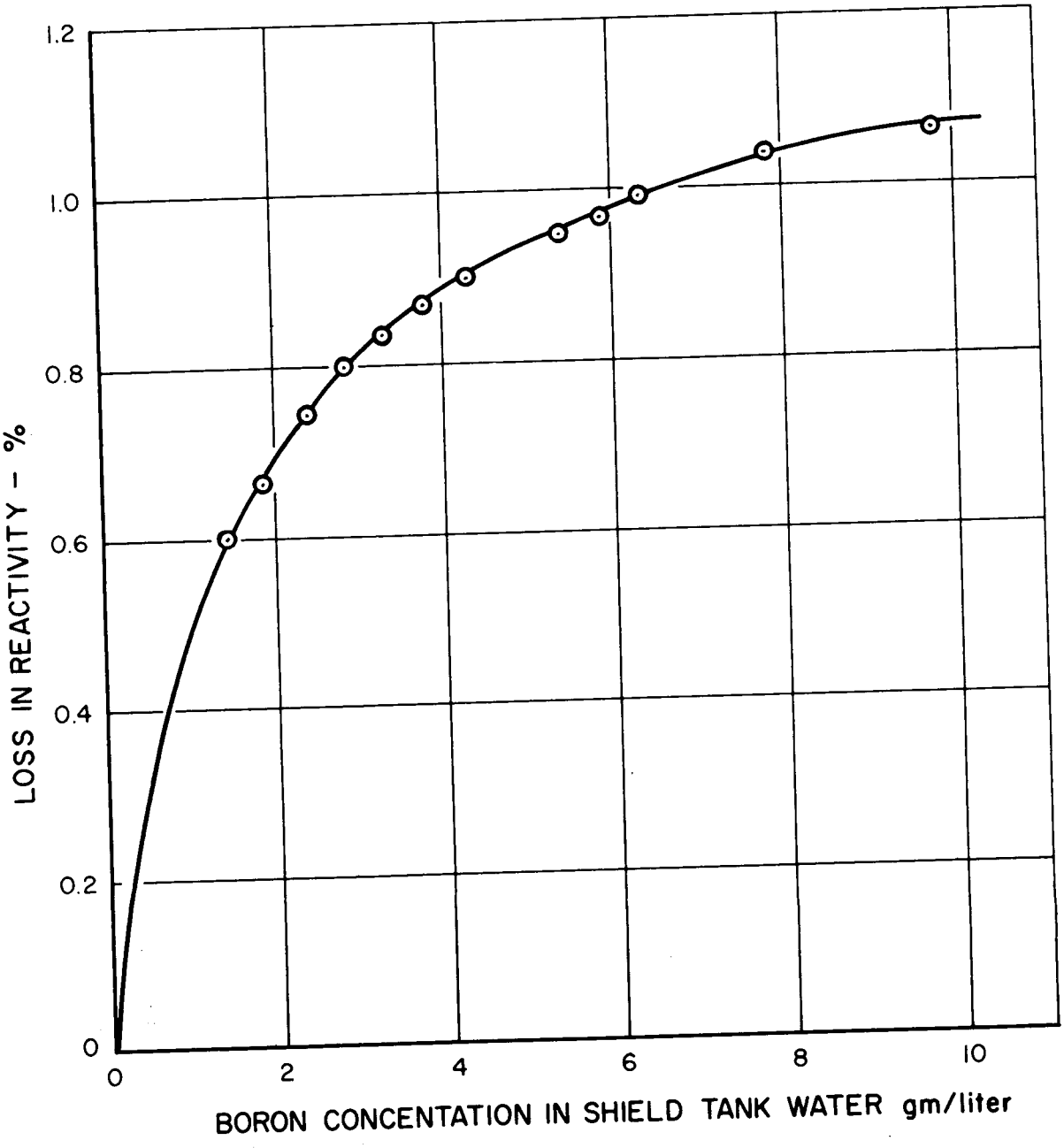

Figure 15 Effect on Reactivity of Adding Boron to the L-47 Shield Tank Water.

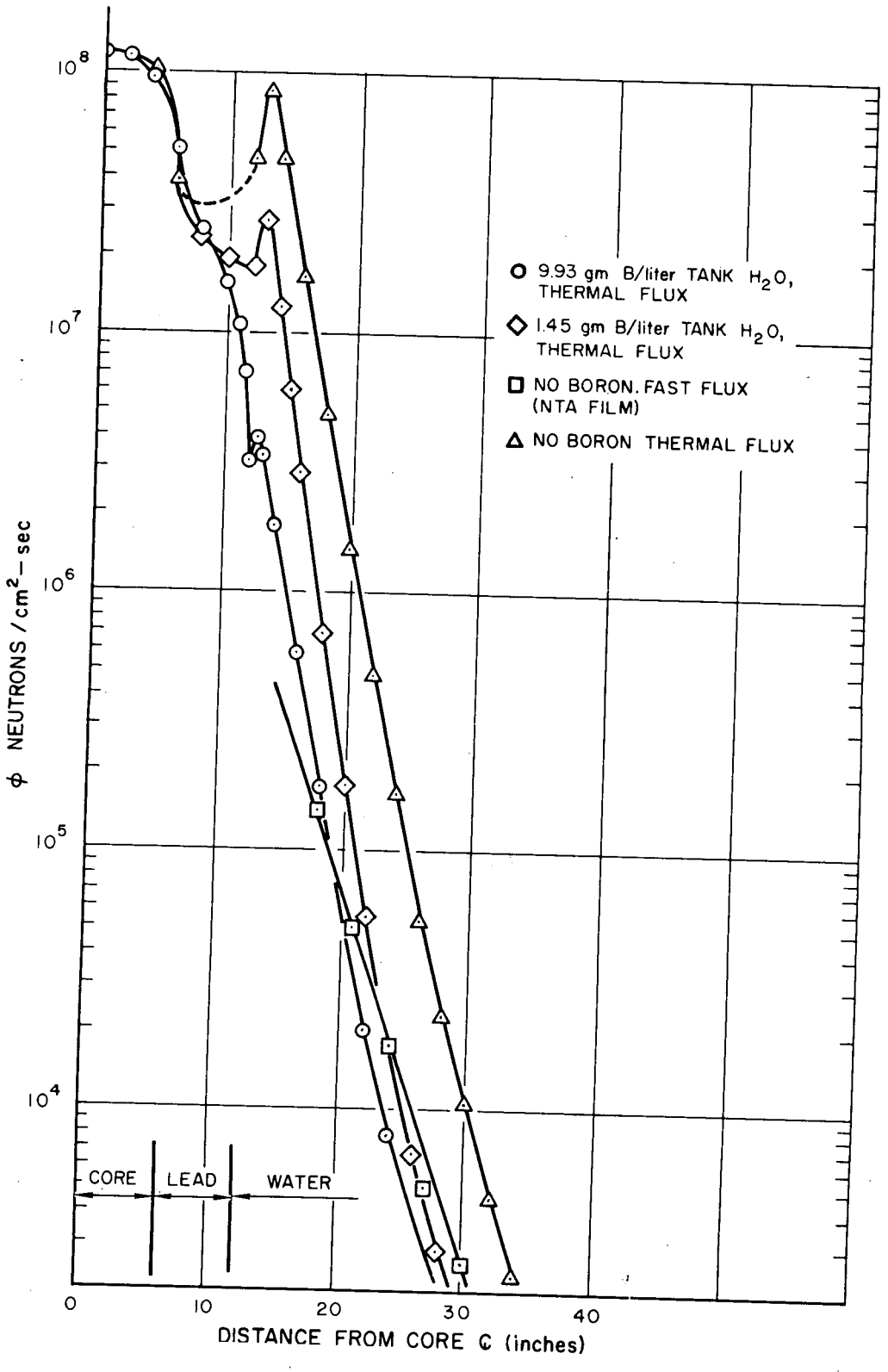

Figure 16. L-47 Reactor Neutron Flux Distribution at 5 Watts.

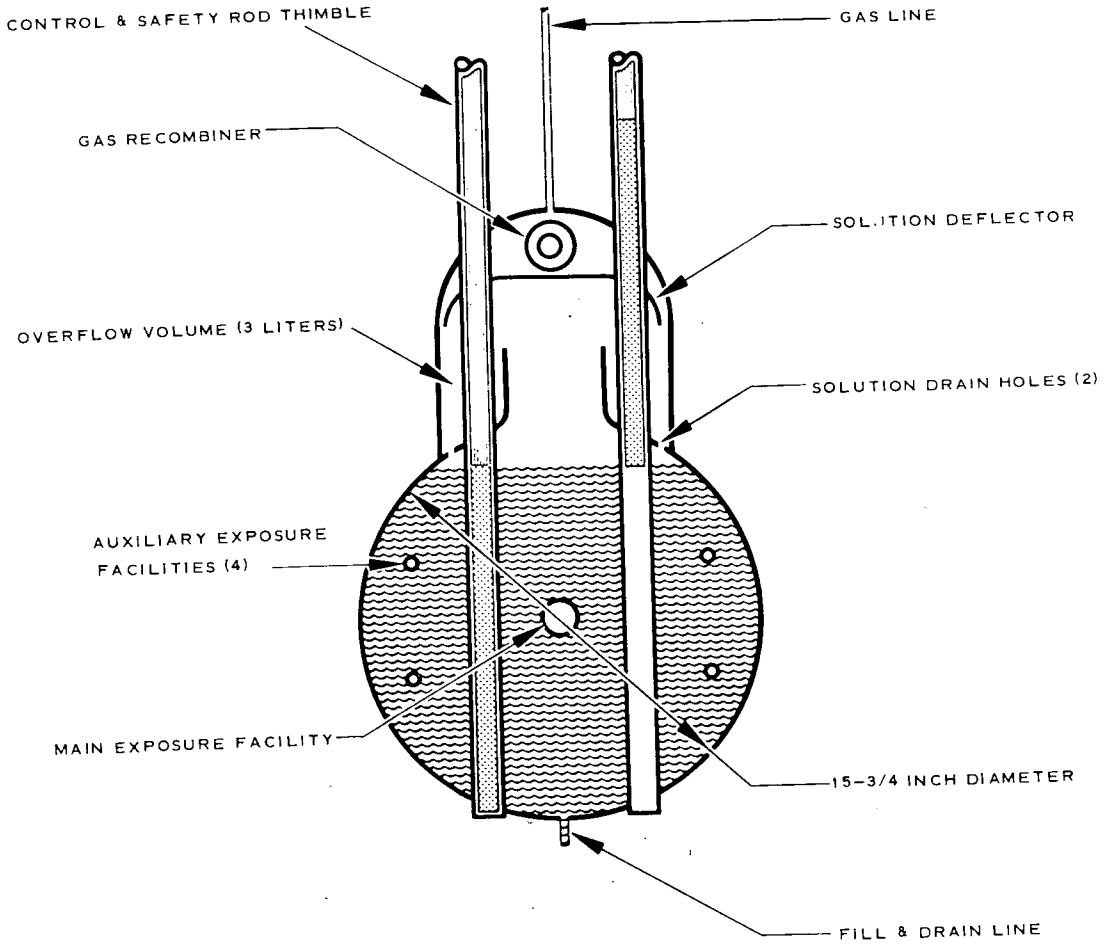

Figure 17. L-77 Reactor Core

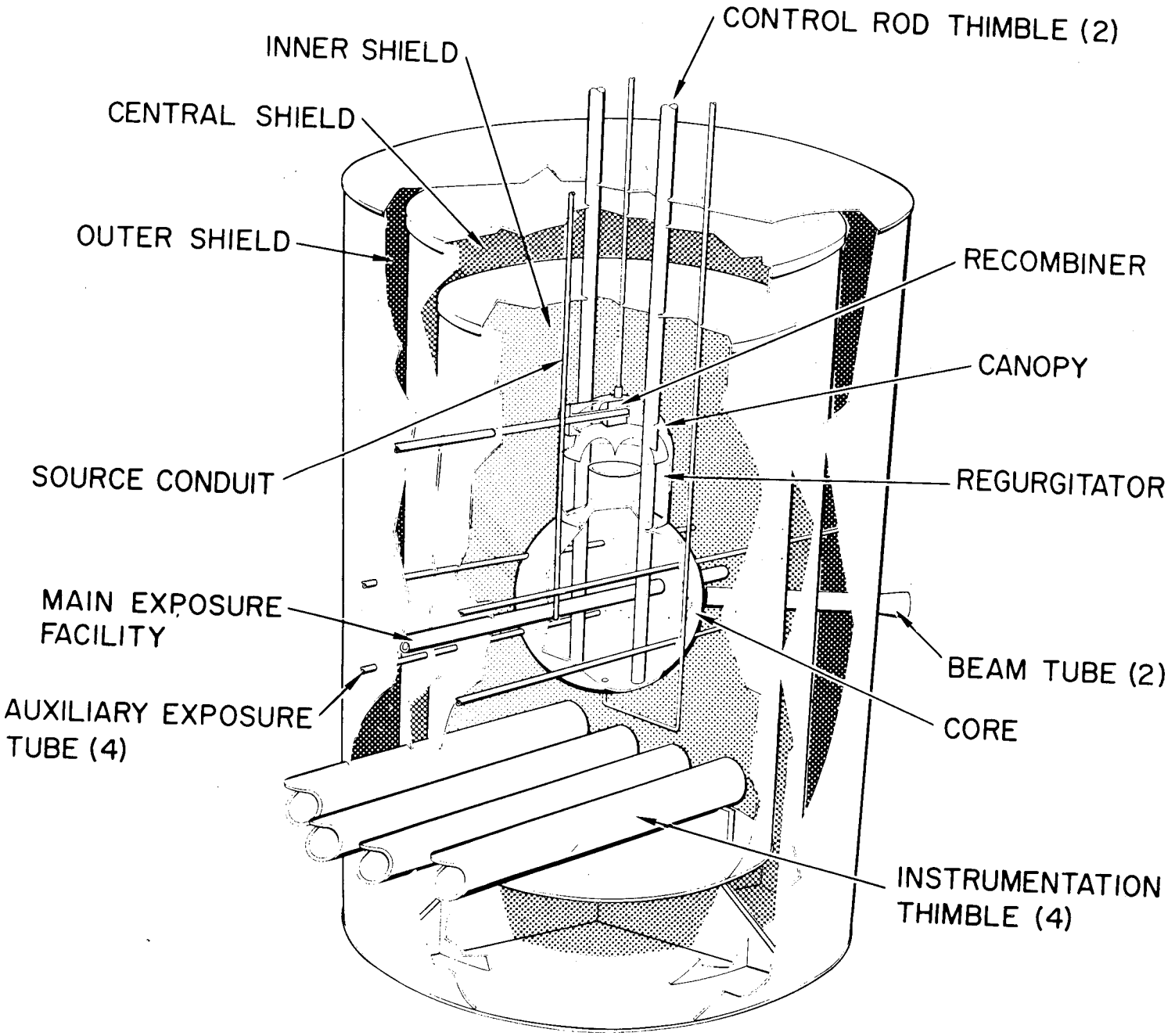

Figure 18 L-77 Reactor Solid Shielding

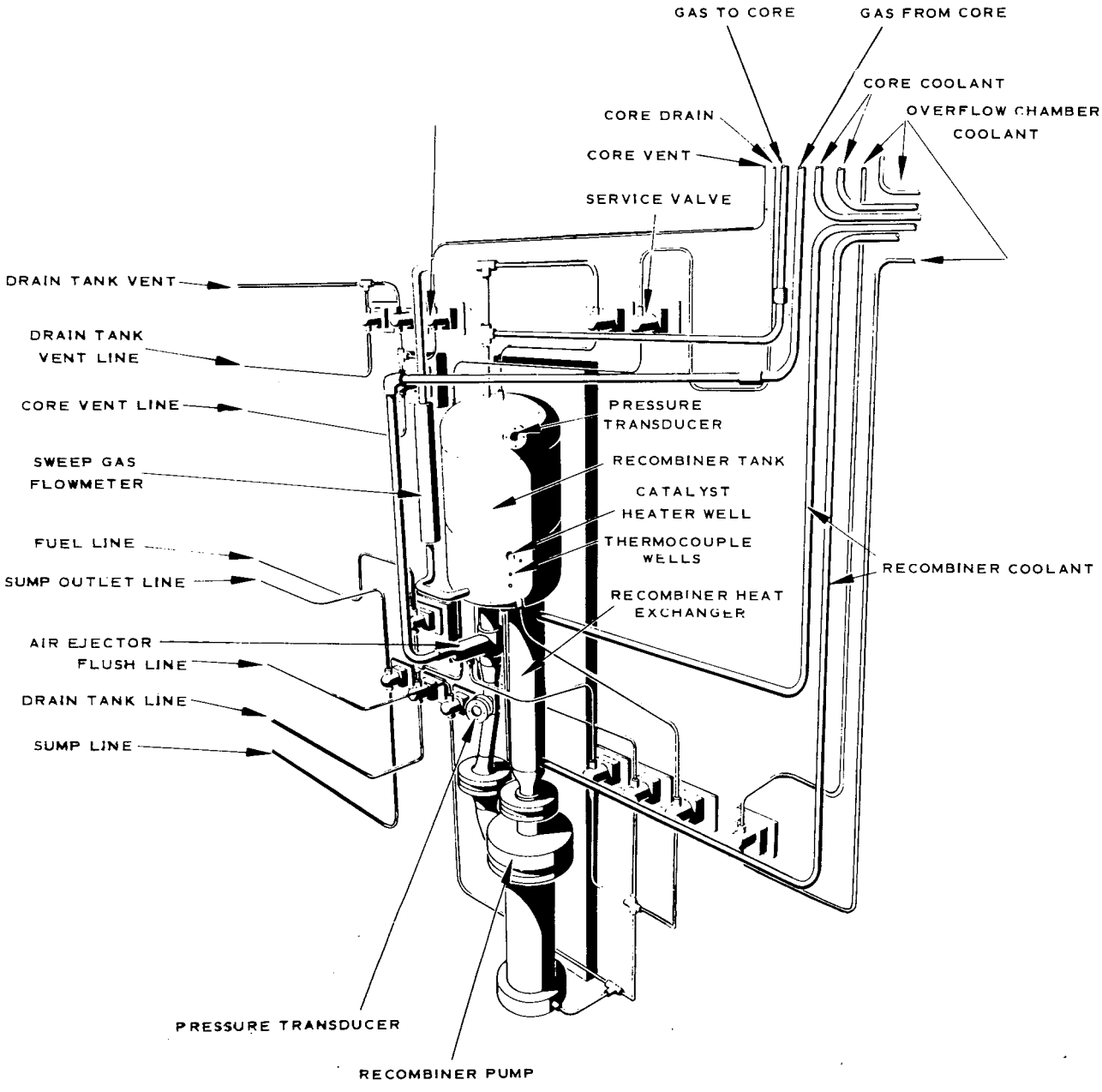

Figure 19 Recombiner System, German and Japanese L-54 Reactor Installation.

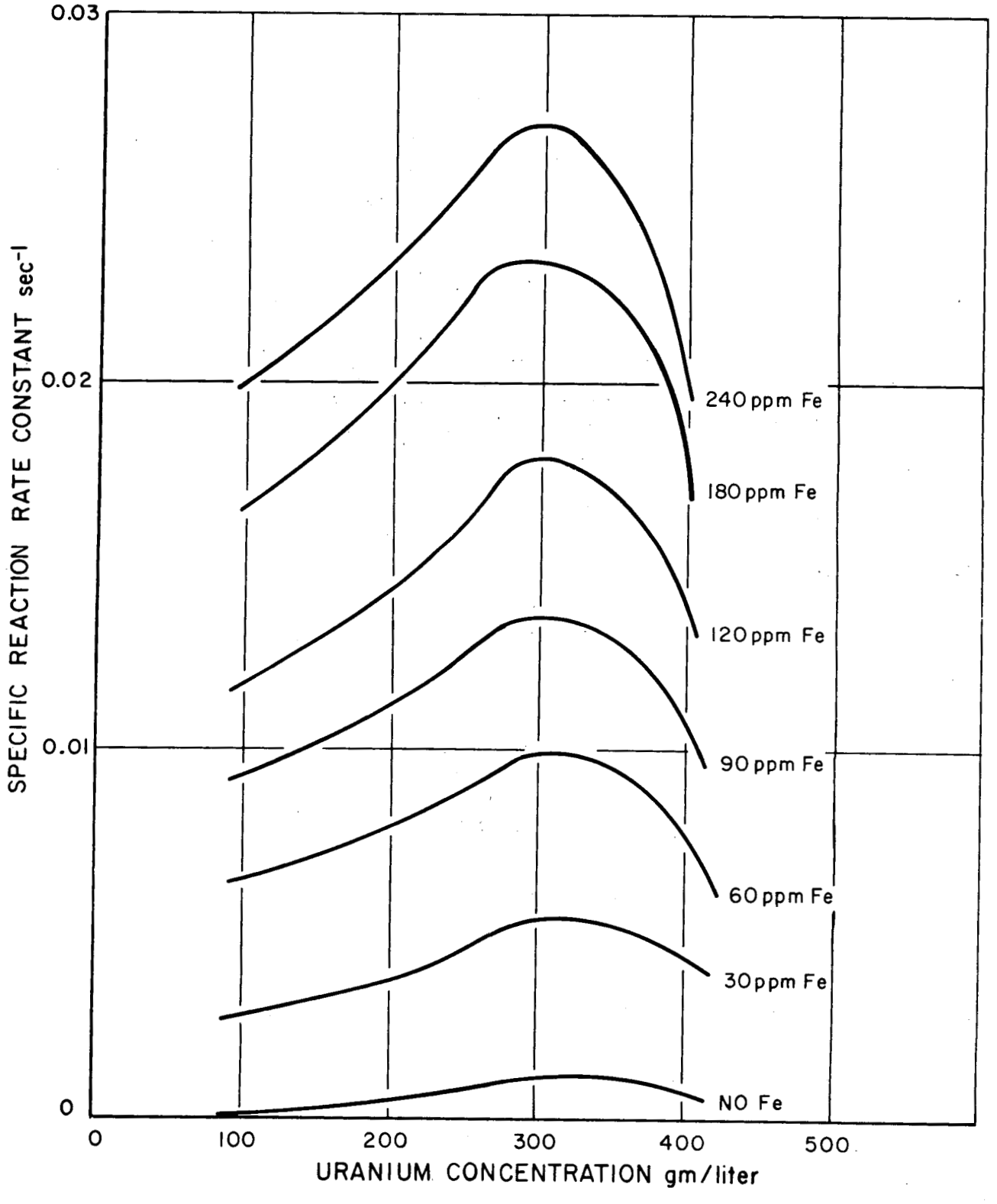

Figure 20. Hydrogen Peroxide Decomposition Specific Rate Constant.

* U. S. GOVERNMENT PRINTING OFFICE : 1958 O— 468967 (J-1313)