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CRITICAL MASS STUDIES OF PLUTONIUM SOLUTIONS

BY

F. E. KRUESI, J. O. ERKMAN AND D. D. LANNING
TECHNICAL SECTION
ENGINEERING DEPARTMENT

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CRITICAL MASS STUDIES OF PLUTONIUM SOLUTIONS

By

E. Kruesi, J. O. Erkman and D. D. Lanning

Physics Sub-Unit
Applied Research Unit

May 19, 1952

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ABSTRACT

The chain reacting conditions for plutonium nitrate in water solution have been examined experimentally for a variety of sizes of spheres and cylinders. The effects on the critical mass of the displacement of hydrogen and the addition of poisons to the fuel were measured in water tamped and bare reactors.

In this report the data obtained in the investigation is presented graphically and in tables. Some preliminary analysis has been made yielding the results:

- i) the absorption cross-section of Pu^{240} is 925 ± 200 barns and
- ii) the minimum critical mass of Pu^{239} in water is 510 grams at a concentration of about 33 grams per liter.

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CRITICAL MASS STUDIES OF PLUTONIUM SOLUTIONS

INTRODUCTION

The criticality experiments reported here were designed primarily to determine safe and efficient operating limits for the Hanford separations processes from the standpoint of nuclear safety. Experimentation was directed toward the determination of the minimum critical mass of a light water tamped and moderated, homogeneous plutonium solution. The effects on critical mass of geometry, concentration, foreign atoms, isotopic content, tamping, and temperature were investigated. This information together with a study of the effects of various perturbations such as control rods, shafts, voids, etc. was required to evaluate the degree of safety afforded in the processing of these solutions. Some work has been directed toward the interpretation of data in terms of existing reactor theory. The intent of this report is to make available the critical mass data obtained during the course of the Hanford program and to describe the experimental facilities and procedures employed for this work.

EXPERIMENTAL FACILITIES AND PROCEDURES

Site Layout

These criticality studies were conducted in an isolated area, three miles from other project installations. The reactor assembly was remotely operated as it was felt that the greatest hazard to personnel in the event of the worst possible disaster would be that of high radiation levels and air borne plutonium contamination. The control and laboratory buildings were separated by a distance of 175 feet and so oriented that neither the generally prevailing nor force winds would carry air borne contamination towards the control building. This distance was acceptable from both radiation and contamination considerations and was not so great as to cause instrumentation difficulties.

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The control building housed the control equipment for the operation of the reactor assembly and provided facilities for instrument repair, air sample monitoring and office purposes.

The laboratory building was designed specifically for these studies. It contained two thermally insulated reactor assembly rooms, a chemical laboratory for the handling and storage of plutonium solutions, a room for the storage of radioactive material and wastes, a personnel change room, and work areas. Air exhaust and filtering equipment was provided to service the chemical laboratory and exhaust hood, reactor rooms, and work areas.

Fluid System

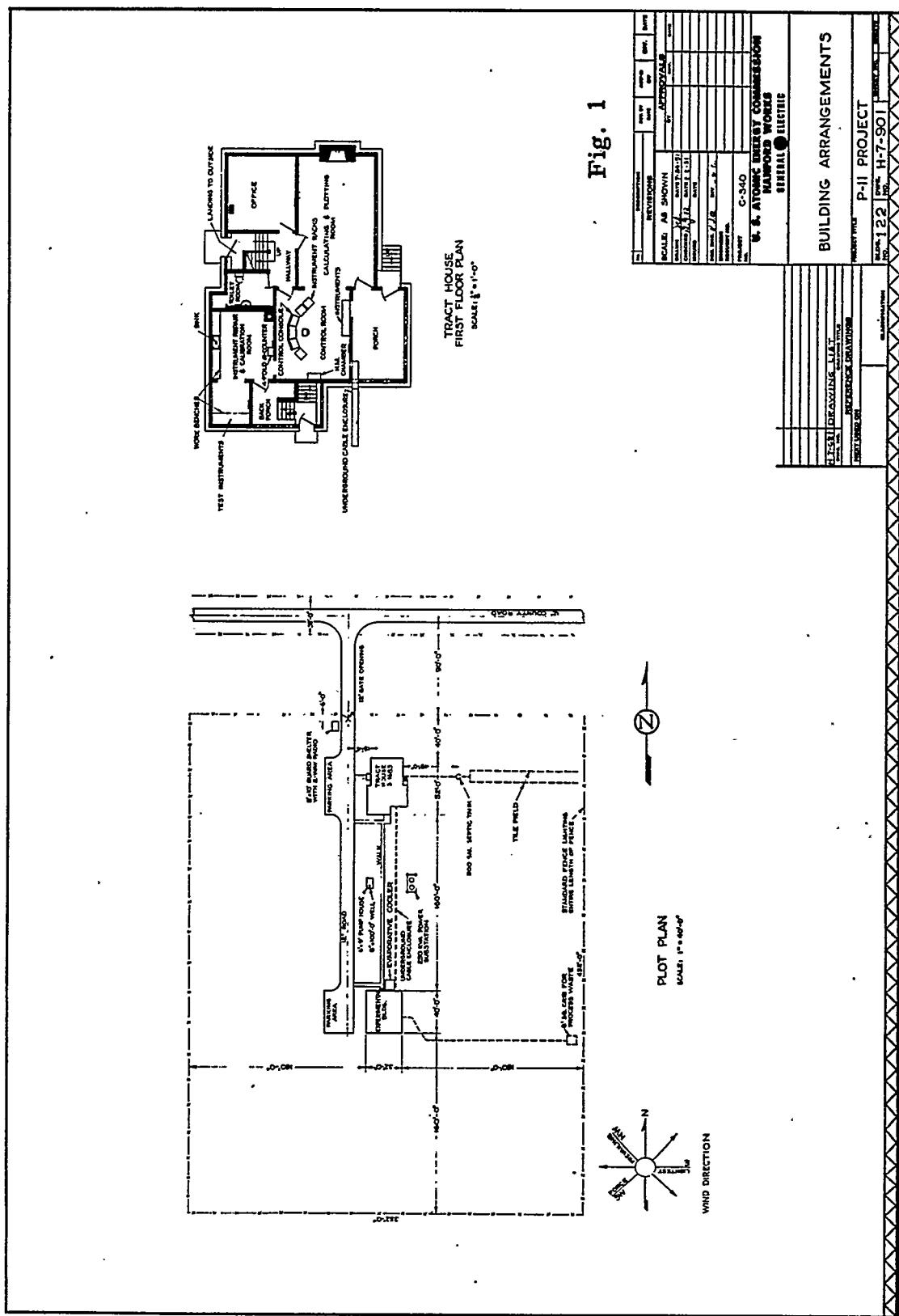
In the design of the plutonium fluid handling system special consideration was given to problems of nuclear safety, plutonium contamination, and corrosion resistance. Gravity flow was selected for the transfer of solution through the system as it was felt that plutonium contamination problems precluded the use of pneumatic pressure in this type of experimental apparatus. Solution flow rates were limited by pipe size and fluid head restrictions to about two liters per minute. Calculations had shown that inadvertent additions of fuel at this rate after criticality had been attained would not give rise to an explosive reaction. As the conditions of criticality for plutonium solutions were not known at the outset of these studies, vessel sizes and spacings were deliberately made overly conservative. All vessels except the reactors were made with diameters not exceeding five inches and were wrapped with 0.020 inch cadmium to reduce the effectiveness of possible tamping. All vessels, including the reactors, were vented to a common manifold which was in turn filtered to the atmosphere to compensate changes in barometric pressure.

Figures 1 and 2 show the P-11 Area plot plan and building arrangement.

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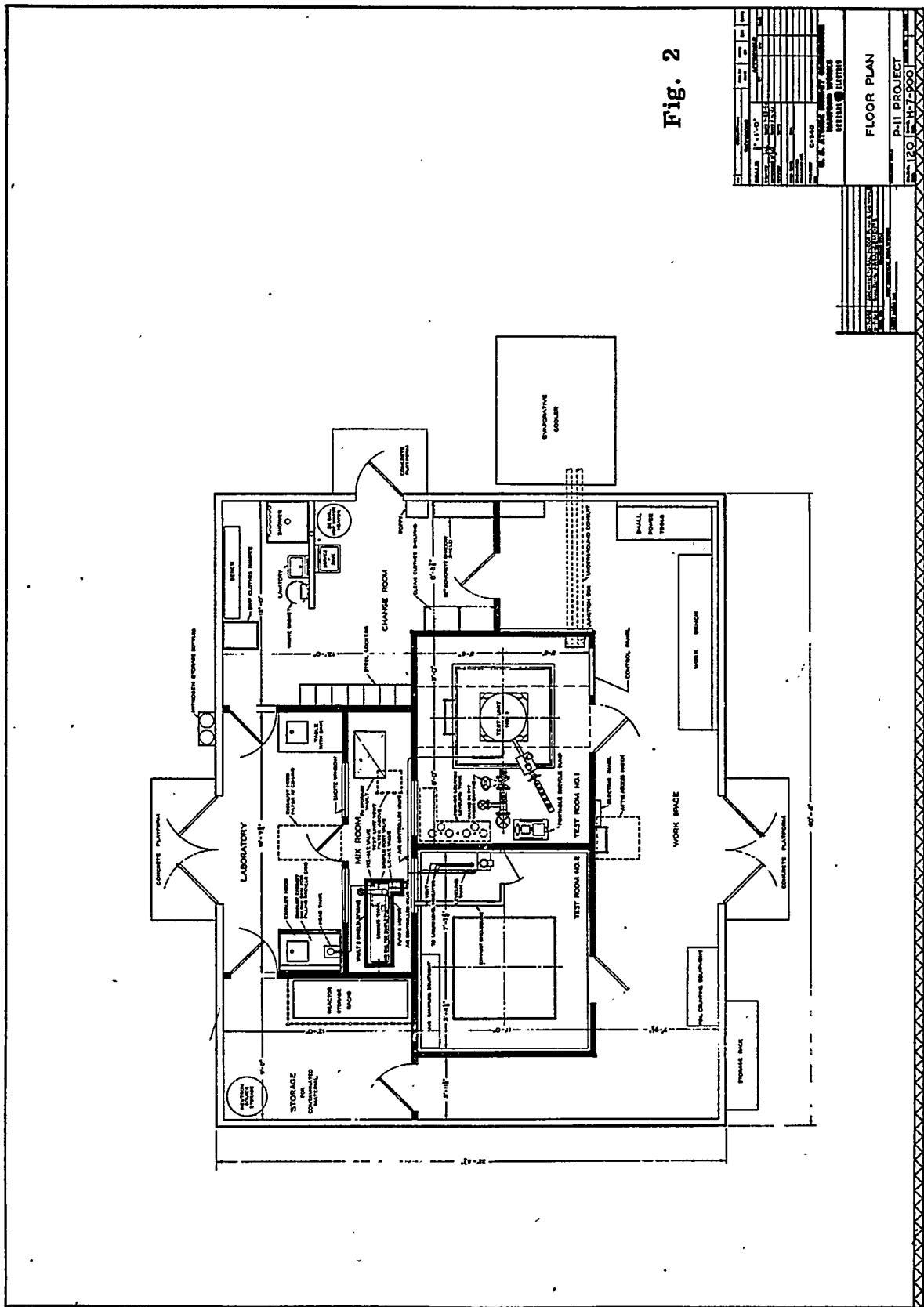
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Fig. 2



Plutonium solutions were prepared and introduced into the fluid system in an exhaust hood located in the chemical laboratory. One hundred fifty linear feet/minute of air were provided across the open face of the hood. The output of the hood was filtered through six parallel CWS Type 197-54-303C filters. The air sample activity of the filtered exhaust air varied directly with the activity of chemical processing. General levels in the chemical laboratory were about 10^{-11} microgram Pu/cc while individual samples ran as high as 10^{-9} microgram Pu/cc.

The chief components of the fluid system were the head tank, mixing tanks and agitator, leveling tanks and reactors. Solution was cycled through the system after passing through appropriate valves in the following manner:

- a) A vacuum pump isolated from gross contamination by a CWS gas mask filter evacuated the head tank.
- b) A quantity of solution not exceeding 2.5 liters was drawn through the decant tube into the tangential head tank opening and the vacuum released. (A sight glass of polythene tubing paralleled the head tank for volumetric measurement purposes.)
- c) This solution was released to the mix tanks and agitated with previous fuel additions to attain homogeniety. The mix tanks with a 35 liter capacity consisted of two paralleled cylinders five inches in diameter and five feet in length, spaced 23 inches between centers. An agitator or recirculating pump drew solution from the outlet of both tanks and exhausted to the inlet of the tanks. A sampling port was provided at the outlet of the recycling pump.
- d) This homogeneous fuel mixture drained to the leveling tanks as they were lowered. The leveling tanks (35 liter capacity) consisted of a single vertical bank of six paralleled cylinders which were raised or lowered with respect to the reactor and mix tanks to provide the required fluid heads. Each tank was five inches in diameter, 18 inches

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in height, and separated from adjacent tanks by a minimum of ten inches between centers. Fluid level within the tanks was indicated by a Republic liquid level bubbler to within $\pm 1/16$ inch after correcting for specific gravity.

- e) The reactor vessel was filled as outlined in the procedures section of this report by raising the leveling tanks from the control room to provide the desired flow rate. A remotely operated valve (solenoid operated air cylinder actuating diaphragm type valve) controlled fluid flow.
- f) Fluid was returned to the mix tanks or head tank for further processing or withdrawal by reversing this process.

All piping and fluid tanks were fabricated from type 347 stainless steel. Flexible portions of the system were fabricated from 3/8 inch O.D. polythene tubing which was protected by a one inch I.D. tygon tubing. A compression type squeeze clamp was used with excellent results to join the plastic and stainless portions of the fuel lines. Polythene diaphragm type valves were used throughout the system to eliminate valve stem contamination or the need for bellows. Air seals prevented contamination spread where removable components were introduced into the fluid system.

Upon completion of the greater part of the experimental program discussed in this report modifications were made to the fluid system because of fluid capacity limitations and corrosion failure at welded joints. Sufficient criticality data on plutonium had been gained to allow a relaxation in vessel geometry limitations. The mix tanks described above were replaced by a single tank having 6 x 14 x 60 inch dimensions with three internal equally spaced cadmium absorbing curtains paralleling the long dimension of the tank. The leveling tanks were replaced with a single 10 x 15 x 30 inch tank containing cadmium absorbing curtains spaced 2-1/2 inches apart. The capacity of the system was raised to 80 liters in this manner.

Figure 3 shows a schematic (diagram) of the fluid handling system.

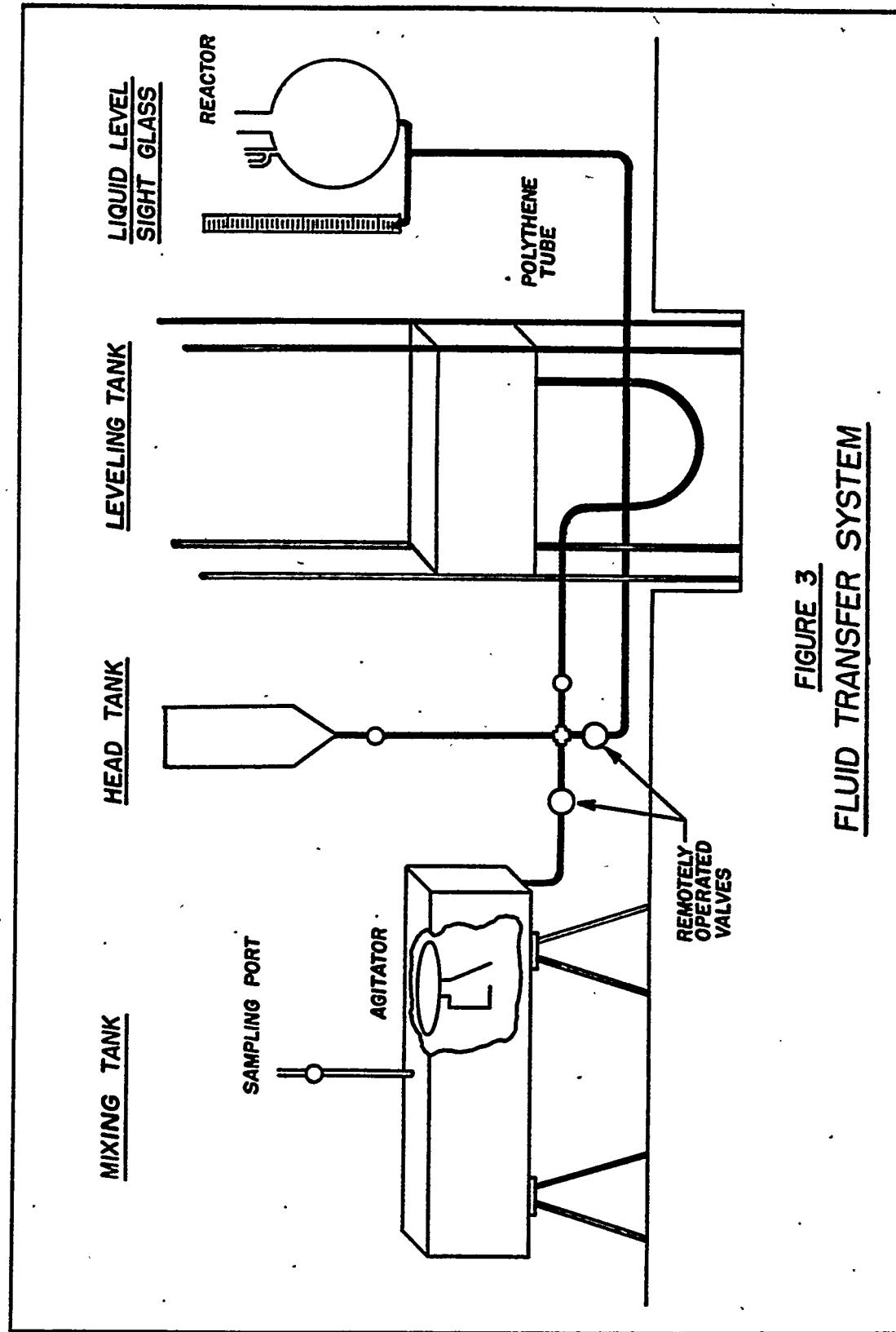


FIGURE 3
FLUID TRANSFER SYSTEM

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~~REF ID: A65181~~~~REF ID: A65181~~Fuel

Plutonium nitrate was utilized as the basic fuel material in view of its 1) availability as normal isolation plant output, 2) chemical stability, 3) solubility at required concentrations, 4) ease of reconcentration and recovery in the Hanford process, and 5) its prevalence in the bismuth phosphate extraction process. Subsequent alterations in chemical form and solution constituents were made by the critical mass experimental group as necessary. All fuel was returned to normal production channels for recovery and reconcentration. Fuel was prepared for critical mass experimentation in the following manner.

- a) Plutonium was received in 160 gram batches either as a 1000 gram Pu/liter, viscous concentrate or as a 350 gram Pu/liter solution depending upon extraction process considerations. The viscous concentrate was converted to 320 gram Pu/liter solution by the addition of weak nitric acid with about two hours of continuous agitation.
- b) This solution was introduced to the fluid system either by decanting into the head tank when bulk additions were required or by pipetting measured volumes directly to the mix tank through the sample port for small concentration adjustments.
- c) Distilled water and 70 per cent nitric acid were added as in (b) to produce the required fuel concentrations. Precautions were taken to maintain a fuel acidity of at least 1 M HNO_3 to prevent hydrolysis and subsequent precipitation of basic plutonium compounds.
- d) This fuel mixture was thoroughly agitated after each concentration adjustment for a period of about 15 minutes to insure complete homogeneity. The entire fluid content of the system was drained to the mix tanks prior to agitation.

e) Analytical samples representative of fuels reaching criticality were taken through the sampling port by lowering a clean dry glass pipette into contact with the solution and withdrawing the required sample. (Usually 10 ml.)

Fuel analyses were obtained as outlined in the analytical section of this report. Concentration estimates based on known fuel additions were not better than \pm 5.0 per cent owing to volume changes through displacement, assay inaccuracies (\pm 2.0 per cent) in arriving plutonium material, and small solution hold-ups in various parts of the system.

Fuel was returned to the extraction process in 300 grams of plutonium batches in standard recycle cans. The plutonium concentration was adjusted to 10 to 20 grams of plutonium per liter range at 1.5 to 2.0 M HNO₃, where possible, for recovery purposes. The frequency of recycling depended upon required concentrations, impurities, fluid system capacity limitations, and production commitments.

Criticality considerations required an investigation of the effects of various fuel impurities. These studies included bismuth, phosphate, lithium, sulfate, and the isotopic content of the fuel.

Bismuth

Bi(NO₃)₃ · 5 H₂O crystals were dissolved in plutonium nitrate solution in concentrations ranging up to 300 grams Bismuth ion/liter. It was necessary to maintain an acidity of 1.5 M HNO₃ to prevent hydrolysis and subsequent precipitation of basic bismuth nitrate compounds.

Phosphate

Eighty-five per cent phosphoric acid was added to plutonium nitrate in the plus six oxidation state with resulting concentrations ranging up to 280 grams of PO₄ ion per liter. Potassium permanganate was used as an oxidant to prevent the formation of insoluble Pu(IV) phosphate. Difficulties were encountered in preventing precipitation and in removal of the MnO₂ reduction product.

Lithium

Weighed quantities of lithium nitrate were dissolved in distilled water and added step wise to the fuel in concentrations ranging up to 1.5 grams lithium ion/liter.

Plutonium Isotopic Content

Effects of the relative isotopic contents of Pu^{239} and Pu^{240} on criticality were studied by experimenting with fuels of different pile exposure histories as expressed in megawatt-days per ton (MWD/T) exposure. Fuels of 55, 212, 420, 572, and 605 MWD/T exposure were utilized. The entire fuel content was removed and the system flushed with 2.0 M HNO_3 prior to the introduction of each new fuel.

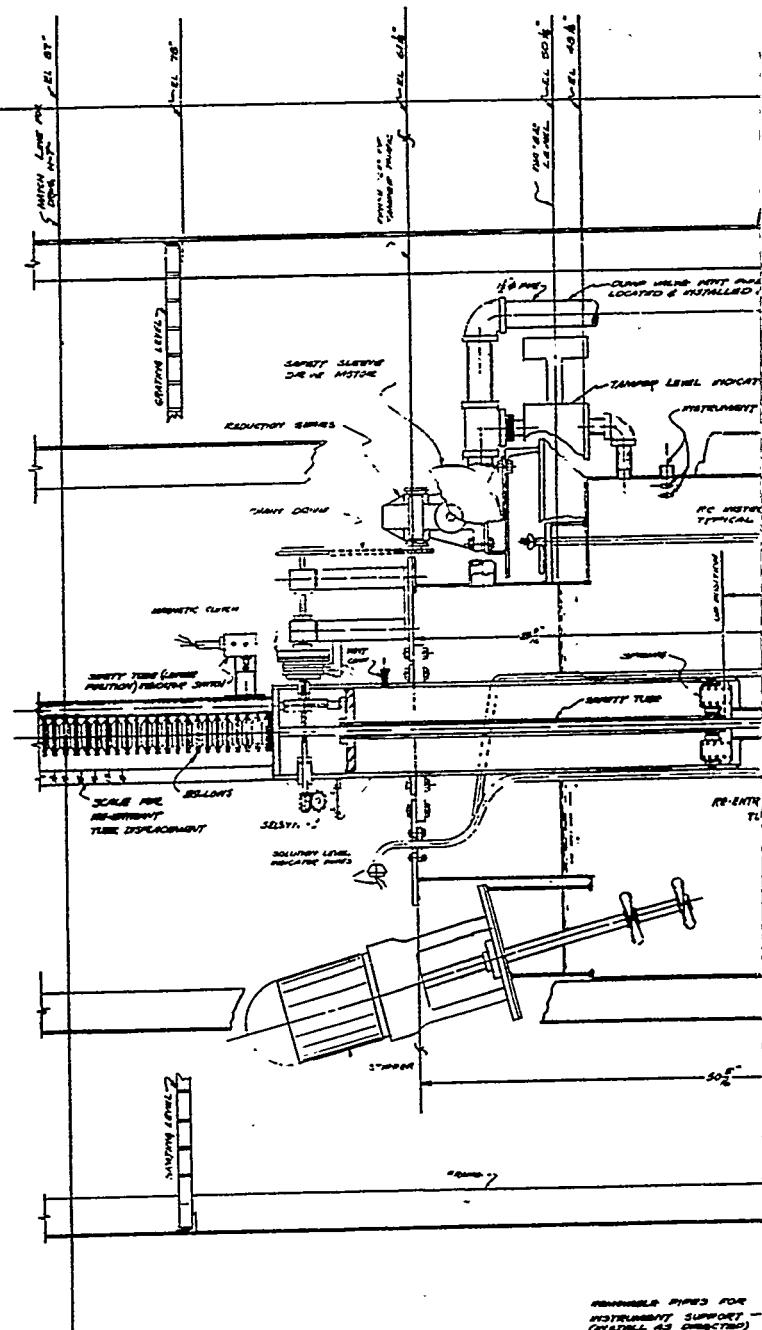
A total of about 32 kilograms of plutonium was processed during the course of the experimentation. An inventory of about three kilograms was required for experimental purposes. An accountability loss of less than one per cent was apparent upon completion of the experimental program.

REACTOR ASSEMBLY

The reactor assembly used in these experiments was designed for criticality measurements in spherical and cylindrical geometries under conditions of effectively infinite water tamping. Principles of nuclear safety, experimental parameters, precision requirements, and the elimination or reduction of perturbing effects were given primary consideration in determining equipment specifications.

Fundamentally, the reactor assembly consisted of a reactor, immersed in a water tamper, fed from the leveling tanks, and controlled by cadmium absorbers. The reactor was driven by an artificial neutron source inserted through a reentrant tube until such time as background multiplication met instrument sensitivity requirements. Auxiliary apparatus was provided for nuclear safety requirements and the control of experimental parameters. Figures 4 and 5 are assembly drawings for a tamped

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NOTE

1. THIS DRAWING IS INTENDED ONLY TO SHOW THE VARIOUS PARTS OF THE REACTOR. IT DOES NOT SHOW THE PARTS IN THEIR ORIENTATION FOR DETAILS OF ASSEMBLY, SEE THE INDIVIDUAL ASSEMBLY DRAWINGS AS LISTED ON H-7-681.

2. ALL ELEVATIONS ARE IN INCHES TAKEN ABOVE CATCH BASIN TOP (FLOOR LINE).

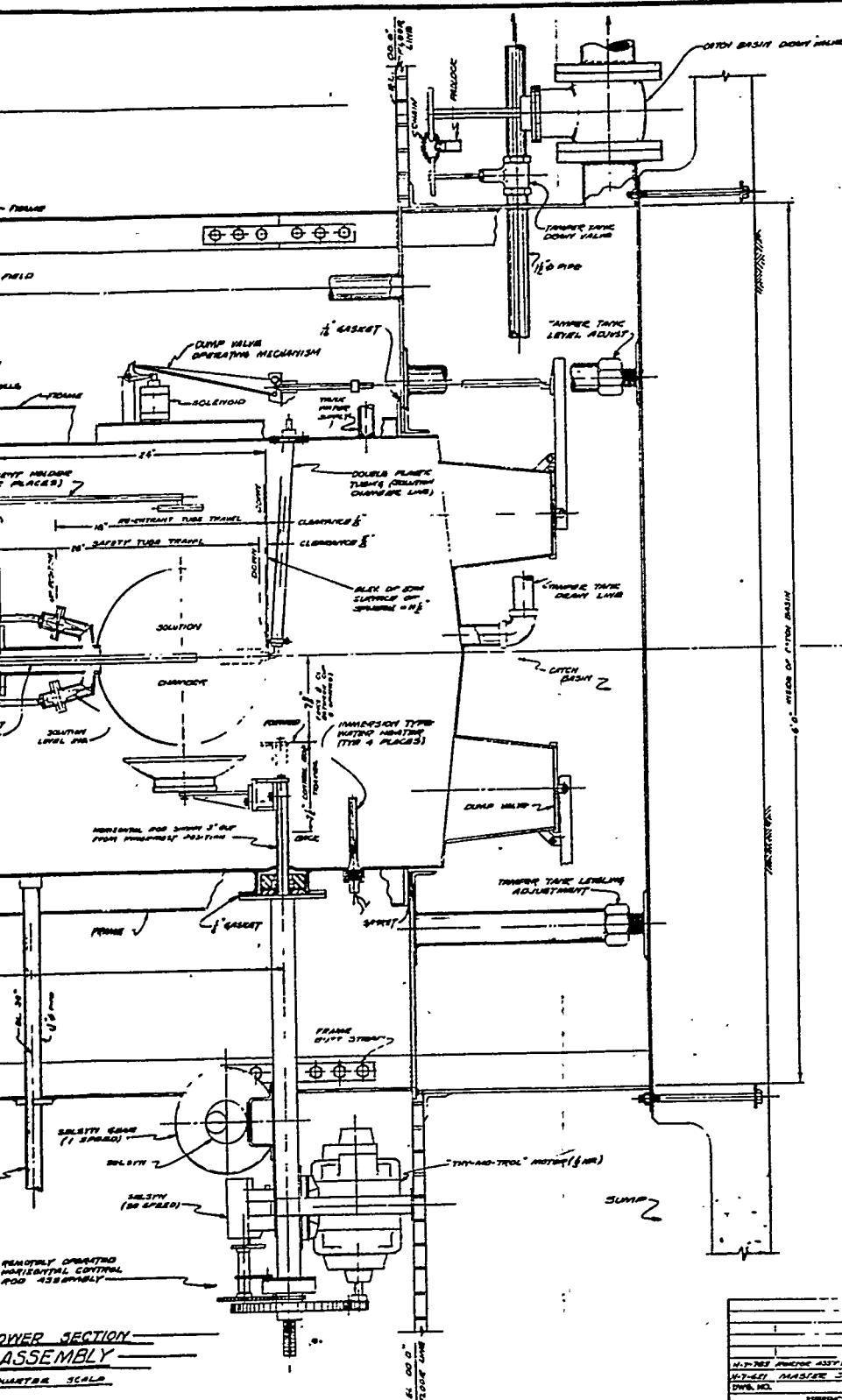
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GENERAL NOTES

2. TO MAKE A COMPLETE DRAWING, THIS SHEET
SHOWING THE SPHERICAL ASSEMBLY MUST
BE USED WITH DWS. H-T-703 SHOWING THE
UPPER, HALF OF THIS ASSEMBLY.

3. TO CONVERT THE ORIGINAL REACTOR
USED WITH CYLINDRICAL GEOMETRY TO
USE WITH SPHERICAL GEOMETRY AS SHOWN
HEREON, THE FOLLOWING ALTERNATES
MUST BE MADE:

- (a) REMOVE THE CYLINDER & PISTON
ASSEMBLIES.
- (b) REMOVE SAFETY SHELL, DRIVE, CATCHES,
& ETC. ATTACHED.
- (c) REMOVE TAMPER TANK TOP COVER
PLATE.
- (d) REMOVE PISTON DRIVE SHAFT &
COPING.

3. ITEMS 1 THRU 5 MAY BE RE-USED.

4. REMOVE THE HORIZONTAL ROD DRIVE
ASSEMBLY & HOISTS AS SHOWN ON
DWS. H-T-700. RE-INSTALL IN PRESENT
LOCATION.

5. ONE NEW (2 1/2 INCH) RE-ENTRANT TUBE
IS REQUIRED AS SHOWN ON DWS. H-T-791.

6. INSTALL NEW, DESIGNATED SAFETY TUBE
DRIVE, DWS. H-T-707, WITH SPHERICAL
ASSEMBLY OF SELECTED SIZE, DWS. H-T-705,
IN TAMPER TANK.

7. PLATED COUPLINGS ARE RE-USED IN
OTT AS SHOWN ON DWS. H-T-702.

8. FIELD WILL RE-INSTALL RE-ENTRANT
TUBE DISPLACEMENT SCALE & PISTON

10. THE PISTON DRIVE MOTOR & GEAR REDUCER
WILL BE USED ON ITS PRESENT
FOUNDATION.

11. THERE ARE NO PISTONS, THE PISTON DIS-
PLACEMENT SCALE & PISTON SUPPORTS
WILL BE RE-USED. THE JELLY
CATCHERS (MOTORS) IN THE COTTERLE
SACHING PISTON LOCATION SHALL BE
REMOVED, BUT NOT RE-ARMED.
E-STOP JELLY CATCHERS (MOTORS) (COTTERLE)
ARE TO BE REMOVED & DISPOSED.

12. SAFETY TUBE WILL BE REMOVED FROM
EXTERIOR PISTON MOTORS & REDUCERS.

13. PROVIDE NEW, LARGE PLATED AT COPROX
PIPE, WHICH PROVIDE SUITABLE CAVITIES
FOR HOLDING OF REACTOR CENTER ASSEMBLY
WHEN CHARGING SPHERES SIZED WITH THE
END OF THE CHAIN HOIST.

15. HANDLING TRUCK DWS. H-T-702 WILL BE
RE-ARMED AS SHOWN ON DWS. H-T-703
IN ORDER TO HANDLE THESE SPHERES.

16. WHEN DISMANTLING THE SPHERICAL
ASSEMBLY, THE SPHERES SHALL BE TAKEN TO PRESENT
CATCHERS. ALL SPHERES SHALL BE HANDLED &
STORED SO THAT THEY MAY BE RE-INSTALLED
& REARMED AT ANY TIME IN THE FUTURE.

17. SUITABLE PROTECTION IS TO BE PROVIDED
FOR THE SPHERICAL CHARGING SPHERES
DWS. H-T-705.

Fig. 4

Fig. 4

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SCALE: 3'-0" X 10"		APPROVALS	
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PROJECT NO. <i>C-340</i>	1	PROJECT NO. <i>C-340</i>	1
GENERAL ELECTRIC CO. HANFORD WORKS			
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- SPHERE PHASE, SHEET 1 -			
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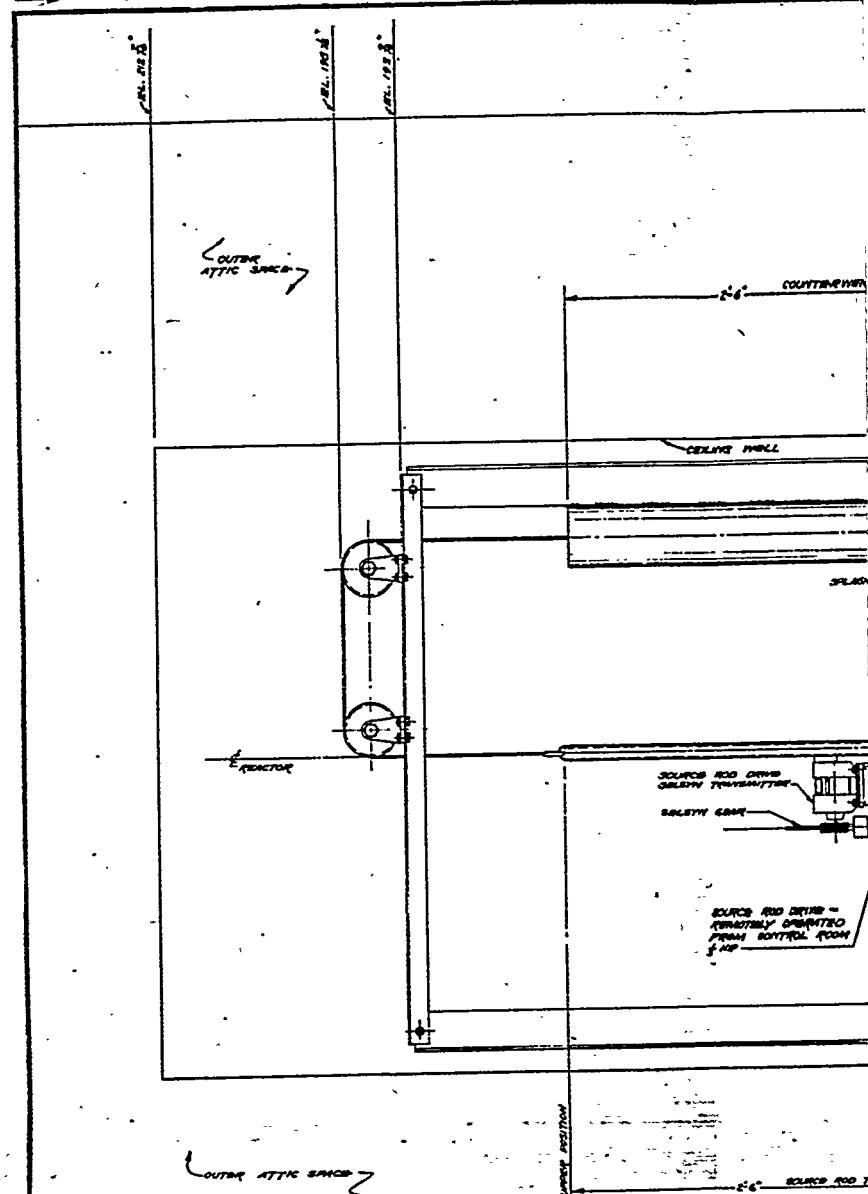
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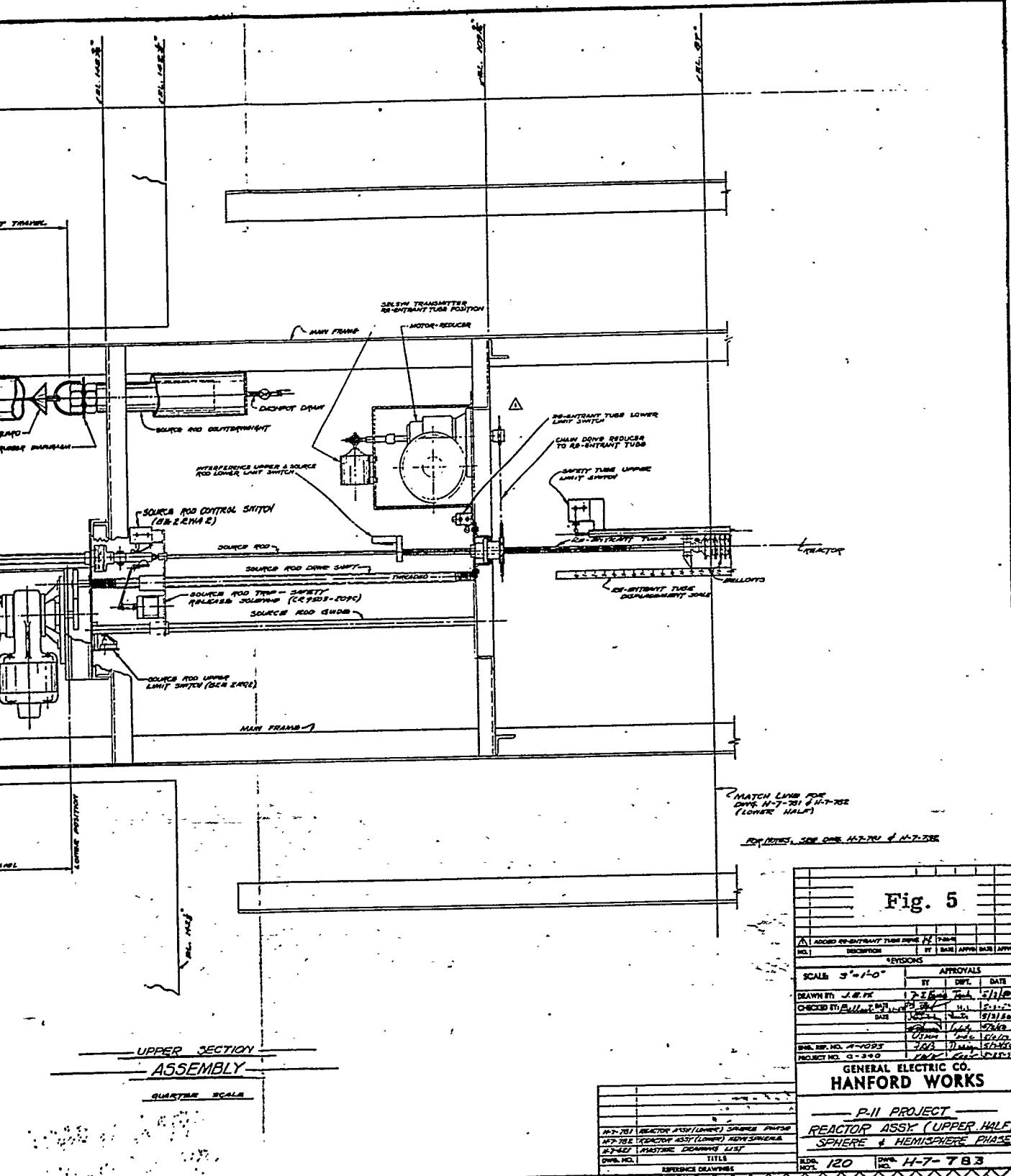
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spherical reactor. The following is a detailed description of the different components of the reactor assembly.

Reactors

Spherical reactors were fabricated by the pressing and spinning of 0.050 inch, type 347, stainless steel sheet with the exception of one reactor fabricated from 20 gauge 2S aluminum. Volumes of tamped spheres ranged from 11.37 to 29.00 liters corresponding to nominal diameters of 11 and 15 inches. Radial sphericity aberation did not exceed 1/8 inch. A fuel feed pipe of 3/8 inch O.D. stainless tubing was attached to the base of each sphere. A supporting neck, 2-1/8 inch O.D., 1/8 inch wall, suspended the reactor in the water tamper. The coaxial safety, reentrant, and source tubes passed through the supporting neck into the reactor. Two electrical fluid contactor housings of one inch pipe projected from the supporting neck.

Cylindrical reactor units with a twelve inch water filled piston tamper were fabricated from 0.062 inch stainless steel sheet. The reactor volume was determined by the piston height which was adjustable to a maximum of 19 inches either by manual or remote operation to a precision of ± 0.010 inches. The cylinder wall - piston annulus did not exceed 0.050 inches. Four cylindrical reactors ranged from 8.0 to 12.0 inches inside diameter. Two electrodes insulated by a one inch diameter polythene body recessed into the piston tamper served as fluid contactors. The reentrant tube and source rod were inserted into the reactor through a 3/4 inch column centered in the piston tamper. A 1/4 inch spiral vent tube passed displaced air through the tamper piston. Grease seals and air bellows prevented the escape of contamination where adjustable mechanisms passed through the top of the cylinders.

Three untamped spherical reactors were fabricated from 0.050 inch stainless steel with capacities of 34.15, 49.00, and 67.61 liters corresponding to nominal diameters of 16, 18, and 20 inches. A control rod was

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introduced into the reactor core parallel to the vertical axis of the sphere and displaced four inches from it.

Tamper

Reactor units were surrounded by a minimum of 12 inches of water tamper contained in a 300 gallon cylindrical tamper tank, the main body of which was three feet in diameter and four feet in height. The work of Beck, et.al. ⁽¹⁾ demonstrates that this is an effectively infinite tamper. The temperature of the tamper and subsequently that of the reactor was normally held to 80° F. which was 5.0° F. above ambient room temperature and was controlled to within $\pm 0.5^{\circ}$ F. by electrical immersion heaters, agitator, and thermostatic regulators. The bottom of the tamper tank was dished to improve water circulation. Four quick opening dump valves (4 inch diameter) in the tank bottom released the tamper to a catch tank in about four seconds upon interruption of the secondary safety circuit. The catch tank had sufficient capacity for two tamper scrams and could be either drained to the crib or recycled to the tamper tank to reduce the time required to obtain temperature equilibrium.

Control Rod

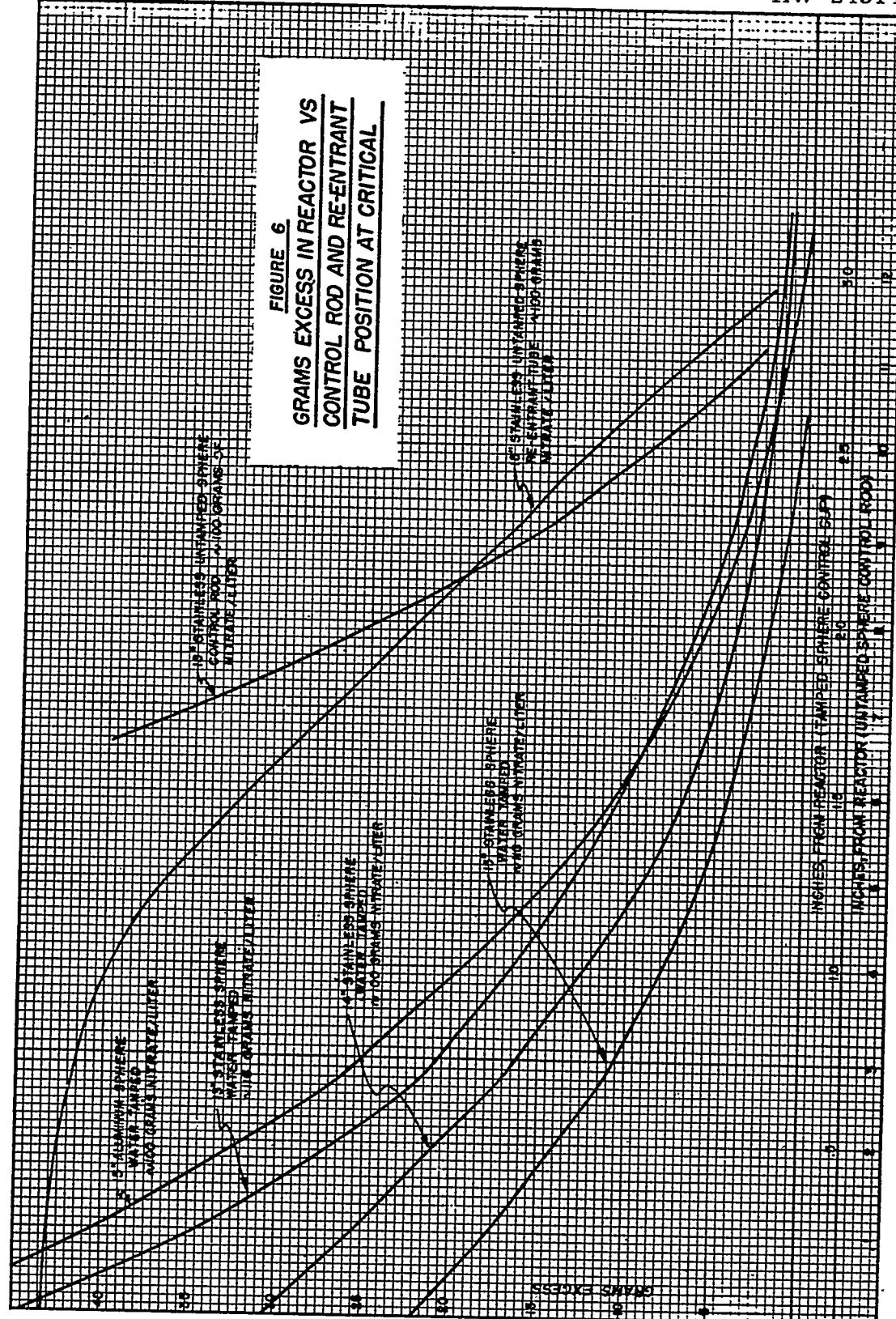
The reactivity of tamped spherical reactor assemblies was controlled by a cadmium cup in the tamper with a surface area approximately ten per cent of the reactor surface area. Bare spheres were controlled by a two inch wide cadmium blade sandwiched by stainless steel that was introduced into the reactor core. A 36 sq. inch cadmium sheet passed horizontally 7/8 inch below the bottom of the cylindrical reactors affording the necessary control. The control cup or blade approached or receded from the reactor with a variable speed between the limits of 1.5 and 25 inches per minute and could be positioned to a precision within ± 0.003 inches as indicated by a coarse and vernier selsyn combination.

Figure 6 shows experimental control rod calibrations in terms of equivalent grams of plutonium.

1. C. K. Beck, A. D. Callihan, J. W. Morfitt, R. L. Murray, K-343, April 19, 1949.

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Safety Control

The safety control used in conjunction with the spherical reactors consisted of a 1.75 inch diameter cadmium tube, sandwiched by stainless steel, suspended 12 inches above the top of the reactor by a magnetically held rack and pinion. Upon release, the 18 inch length of active absorber passed through the supporting neck and came to rest along a sphere diameter. The time lapse between release of the magnetic clutch and completion of the rod motion was about one second. The safety tube was controlled remotely, its position being indicated by a selsyn receiver. Graphite filled fluorthene was used for bearing surfaces in the corrosive atmosphere above the reactor.

The safety control used in conjunction with the cylindrical reactor was a cadmium sheath external to and coaxial with the reactor. This sheath was 12 inches in height and of such diameter as to clear the reactor wall by 1/4 inch on a diameter. The sheath dropped to a position surrounding the reactor in about one second after being released.

Source Rod and Re-entrant Tube

A Polonium-Beryllium neutron source of approximately 10^6 neutrons/second attached to a 3/8 inch diameter aluminum rod was remotely positioned within the reactor or removed from it as required. The rate of travel of the source was about 30 inches per minute when motor driven. A release and counter-weight mechanism permitted a rapid source withdrawal for "source jerk" criticality tests. The total source travel was 30 inches as measured from the reactor base and indicated on a selsyn receiver.

A 5/16 inch I.D. stainless steel re-entrant tube isolated the source from the reactor fuel for contamination reasons. This tube was either manually or remotely positioned over its 20 inches of travel.

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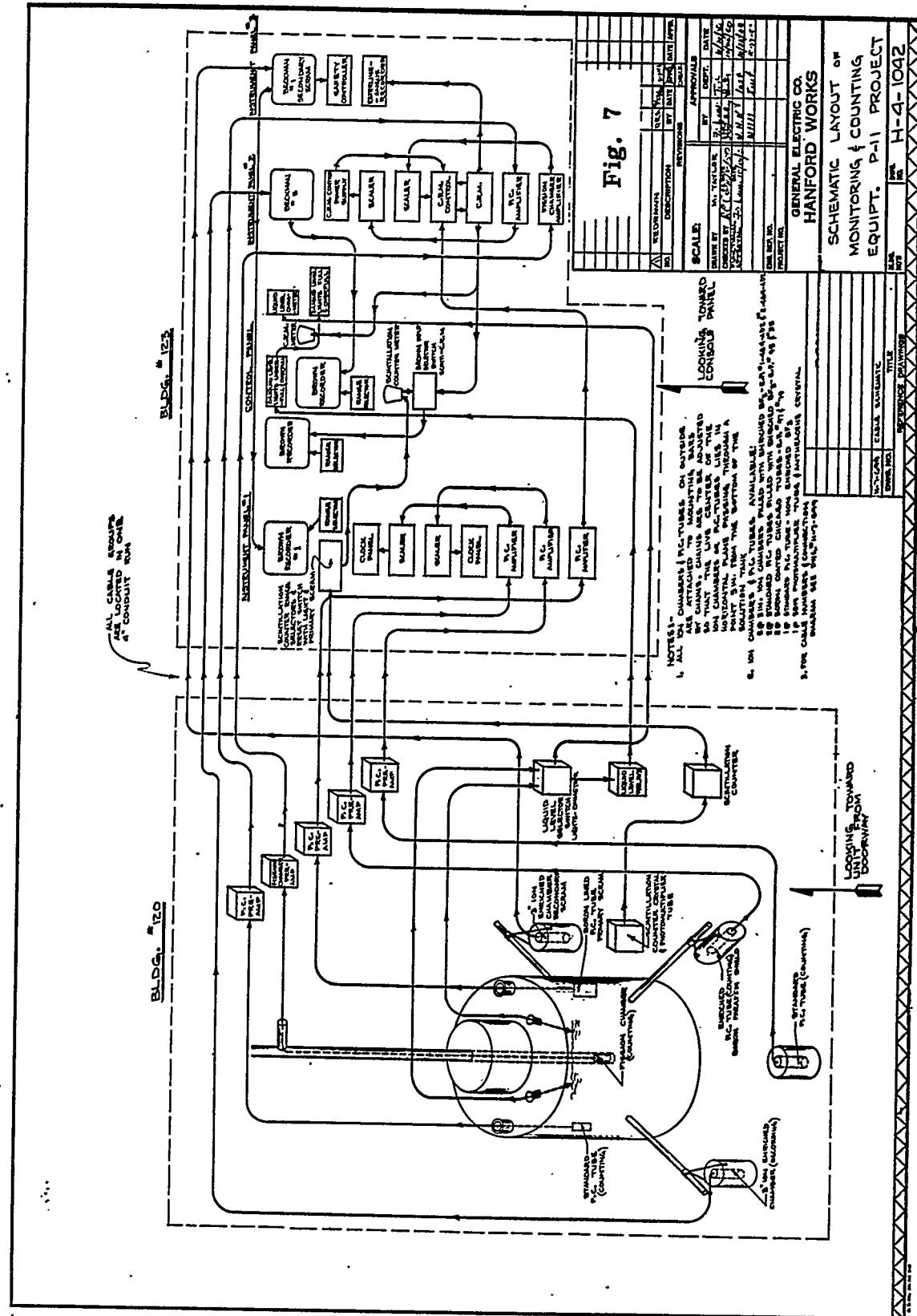
INSTRUMENTATION AND SAFETY CONTROL CIRCUITS

The instrumentation required for monitoring and recording the reactivity variations of the critical assemblies consisted of neutron detectors driving scalers, a counting rate meter, and recording potentiometers. Boron trifluoride and boron lined proportional counters and boron trifluoride filled ionization chambers monitored the neutron flux external to the reactor. A uranium ²³⁵ cylindrical fission counter of 3/8 inch O.D. measured the neutron flux within the reactor depending upon the position of the source rod to which it was attached. Neutron detectors external to the tamper tank were filled with enriched BF_3 and were surrounded by paraffin moderators to increase sensitivity. Gamma activity was followed by a scintillation counter with an anthracene phosphor (3 x 3 x 1 cm) and 5819 photomultiplier. A logarithmic amplifier and period meter having a range of six decades and capable of measuring periods as short as three seconds was provided. A standard five inch gamma chamber was located in the control room for personnel monitoring. The placement and use of these detectors is discussed in the Operating Procedures section of this report and shown schematically in Figure 7.

The signals from the proportional counters and fission counter were fed into preamplifiers located in the reactor assembly room and were further amplified after transmission (200 feet) to the control room before being fed into scalers (usually four including the fission counter) and a General Radio counting rate meter. Scale of 256 scalers were used. The current from the enriched BF_3 ionization chambers was amplified by Beckman micro-micro ammeters which in turn actuated Brown strip recorders. The output from the scintillation counter amplifier also actuated a Brown recorder. A modification to this recorder circuit permitted either 3 x or 10 x normal sensitivity for observation of small reactivity changes.

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Primary and secondary scram circuits were provided to rapidly deactivate the reactor assembly. These scram circuits were activated by a rise of neutron or gamma flux above a predetermined level, a period of the system shorter than a predetermined value, an electrical power or circuit failure, or manual operation of a push button.

A primary scram occurred if the neutron or gamma levels exceeded nine tenths of full scale on the counting rate meter or scintillation counter meter on any sensitivity range, or by a period shorter than one second as recorded by the counting rate meter. The primary scram circuit was designed to allow a reasonably rapid reactivation upon clearance of trouble without requiring personnel to leave the control room except to correct circuit or equipment failure.

The secondary scram circuit was provided to more completely deactivate the unit in the event the primary scram circuits failed. A trip level exceeding nine tenths full scale of a Beckman amplifier and controller driven by a BF_3 ionization chamber automatically activated this scram circuit.

In the case of the tamped spheres and cylinders, the primary scram caused the safety tube (or sleeve) to drop to the all-in position. This also prevented the raising of the fuel leveling tanks, however, the leveling tanks could be lowered at any time. The secondary scram initiated the events of a primary scram and in addition opened the dump valves in the tamper tank releasing the water from the tank.

For the bare spheres, a primary scram dropped the control rod to an all-in position while a secondary scram dropped both the control rod and the safety tube into the reactor.

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PLUTONIUM CONTAMINATION PROBLEMS

A significant part of the total effort required to accomplish these experiments was spent on special problems associated with the handling of plutonium contamination. Experience with plutonium nitrate solutions showed that all sources of contamination on open lines or equipment had to be eliminated upon detection as it was very difficult to prevent the spread of contamination once the material became dry. A loose contamination source of the order of 10^{-7} grams of plutonium was found to produce a significant rise in air sample activity in the reactor room. Routine exposure without respiratory protection was permissible where air levels were known to be less than 4×10^{-12} micrograms plutonium per cubic centimeter. Assault masks were required with CWS red cannisters for all work in zones exceeding this level. No equipment was released from the laboratory building unless a negative "poppy" survey or smear resulted. Potentially contaminated equipment not lending itself to surveying was not released except under controlled conditions. Equipment and tools probing less than 500 d/m were released from contaminated work zones for general use within the building. Routine "poppy" surveys and air and tamper water sampling were required throughout the experimental program.

The laboratory building designed for these experiments was divided into a number of small rooms, previously described, in order to localize potential contamination hazards. Normal activity was possible in most parts of the laboratory even though one or more rooms were contaminated. Various techniques were used in removing contamination from working areas and equipment. Floors, walls, and some large equipment in rooms containing plutonium were covered with G. E. cocoon, a strippable coating. In addition, 10 ply absorbent paper or heavy sisal paper was taped to the floor in these rooms. Grossly contaminated areas where decontamination was not feasible were covered over with cloth backed industrial adhesive tape and coated over with G. E. cocoon. Polished metallic or glass surfaces were easily cleaned with dilute nitric acid liberally applied with glass

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wool followed by paper tissue wipes to absorb the acid and plutonium. Dry contamination could be removed by contact with tacky masking tape or by wiping down a surface with a damp rag.

Plutonium solution leaks through valve castings, flanged joints, and particularly at welded joints and seams, were experienced during the course of the experiments. Several techniques of prevention and repair that proved reliable for this type of application are discussed below:

- a) Flange leaks were avoided by turning flange facings to leave a slightly raised ring to cut into the gasket material. Rings on opposing flanges were made of slightly different radii. Polythene and teflon were used as gasket material.
- b) Piping leaks frequently occurred where connections were directly welded. These failures were eliminated by welding an overlapping sleeve with an inside diameter slightly greater than the outside diameter of the tubing to one tube slipping the sleeve over the second tube, and welding. No attempts were made in repairing a leak by direct welding on exposed lines as contamination considerations prohibited this procedure. Piping leaks were successfully arrested by tightly wrapping the faulty joint with #33 Scotch Electrician Tape. This tape adhered well to a clean dry surface and was found to be resistant to all solutions used in these studies and gave no signs of deterioration after a year of service.
- c) No successful techniques were found to repair leaks that would not lend themselves to the above treatment. Temporary repair was effected by the use of industrial adhesive or absorbent dressings other than glass wool. A number of acid resisting cements were tried with poor results.
- d) A highly successful preventative technique for tubing and equipment not previously contaminated was that of flame spraying polythene on

stainless steel and other surfaces. This plastic coating is impervious to most chemical reagents, is flexible and tough, and is strongly bonded to the sprayed surface when applied correctly.

- e) Faulty equipment components could be removed with a minimum of contamination difficulty by first wrapping the tube section to be cut with masking tape and then cutting through the tape with a hack saw. The replacement equipment was welded onto the existing line using the sleeve technique outlined above.

PROCEDURES AND TECHNIQUES

Approach to Critical Height

Establishment of the critical height in the various cylinders at different fuel concentrations was accomplished by the inverse multiplication extrapolation method. Stepwise addition of fuel to the reactor in increments limited in magnitude by the observed source multiplication at preceding cylinder heights is the basis of this method. The initial height to which the vessels were filled was arbitrarily chosen in the early experiments as one-third the predicted critical height. Accurate extrapolation from multiplication measurements taken at less than one-half critical height, however, was difficult to achieve since the neutron distribution undergoes marked changes as the cylinder height increases. After several experiments had been completed and due confidence gained in both the equipment and accuracy of predictions all initial counting was done at one-half predicted critical height. This alteration of procedure allowed larger increments of fuel to be added to the cylinder as a result of the more accurate extrapolations.

The inverse multiplication curves are most frequently found to extrapolate to a premature critical height if the detectors from which they arise are located outside the reactor and some distance from the background neutron source. The foregoing conditions were both necessary in

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these experiments in order to avoid perturbation effects. There was determined only one detector location that consistently yielded a near straight line on the reciprocal multiplication versus height plot. All other locations, both inside and outside the tamper, produced concave curves. This most useful position was eight to ten inches directly below the center of the reactor with the longitudinal axis of the counter in a horizontal plane. All monitors, however, produced extremely accurate predictions, i.e. ± 0.05 inch, of the critical height after the fuel height had been increased to within about two inches of criticality.

The actual operating procedure followed in a routine criticality determination was as follows:

- (1) A thorough checklist was completed to insure that all the equipment was functioning properly.
- (2) All personnel were removed to the control building and the area was put on an operating basis which prohibited the entry of visiting personnel.
- (3) The neutron source was positioned in the reactor.
- (4) Safety devices were made ready and automatic trip levels set.
- (5) The height of the cylinder was adjusted remotely to one-half the predicted critical height.
- (6) Fuel was introduced continuously into the reactor until its level reached the moveable piston in the cylinder, at which time flow was interrupted by the closing of the remotely operated feed line valve.
- (7) A negative hydrostatic head was placed on the remote valve line by repositioning the leveling tanks.
- (8) The neutron activity, as detected by four individual monitors, was recorded and assigned the value of unity for the multiplication plot.

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- (9) The control rod was returned to the "all in" position.
- (10) The cylinder height was then adjusted to allow sufficient fuel to enter the reactor to give approximately twice the initial source multiplication as based on the predicated critical height.
- (11) Steps six through ten were repeated in the order given and a second multiplication measurement taken which, along with the previously acquired data determined the third piston height setting. This process was repeated until at some final height the system was determined to be critical, as indicated by a linear rise of flux, with the control rod mostly withdrawn.
- (12) The fuel level in the reactor was lowered approximately one-half inch and the source and re-entrant tube withdrawn from the reactor.
- (13) Steps six through ten were again repeated, this time without the re-entrant tube present and with the source at the top of the reactor.
- (14) After two new multiplication measurements were taken the cylinder was made critical at the height indicated by the extrapolation of these two measurements.

The increment of cylinder height used in the final repositioning of the piston was dictated by the extrapolated prediction of control rod strength. Because the control rod was beneath the cylinder its effectiveness was quite sensitive to changes in cylinder height. In the initial experiments the final multiplication measurements were taken in duplicate, one set with the rod "all-in", the other with it "all-out". The height difference between these two infinite multiplication extrapolations was assigned to the strength of the control rod. This procedure was dispensed with as soon as it was evident that the control rod had been evaluated under all significantly different conditions.

A range of values for the poisoning effect of the reentrant tube has resulted from habitually determining two critical heights, one with the tube in place, the other with it withdrawn. This range indicates that the fuel must be over concentrated so as to add 15 to 35 grams in cylindrical reactors 18 inches tall and 12 to 9 inches in diameter respectively in order to compensate for the poisoning effect of such a tube.

Approach to Critical Concentration

The data taken during the investigation of criticality conditions for cylinders contributed significantly to the accuracy of predictions of critical concentration for the range of spherical reactors used. In those cases where comparatively unpredictable poisons were introduced in the fuel, criticality was reached by successive concentrations of the fuel. A plot involving reciprocal multiplication and concentration normally led to a reliable estimate of the critical concentration if the full reactor was twenty-five to fifty grams subcritical. The one exception to be noted here is the case involving very concentrated fuel, as illustrated by the attempt to make the eleven inch sphere critical. The fuel additions to this sphere eventually resulted in such extensive leakage because of poor moderation that the ineffectiveness of further concentration was clearly reflected in strongly concave reciprocal multiplication plots. The control cups used with the spheres facilitated large stepwise fuel concentrations by merit of their large control strength. This is in view of the fact that the fuel was never concentrated in any one operation by an amount which resulted in the addition of more grams of plutonium to the reactor than the control cup was able to compensate. The nominal range of poisoning by these cadmium coated cups was of the order of twenty-five to fifty grams, with the largest sphere having the weakest control cup.

After the initial sphere experiments the operating procedure was revised to exclude the necessity of the preliminary critical concentration determination with the reentrant tube in place. For each sphere, however,

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two fuel concentrations were sampled and analyzed, one of which had resulted in a slightly supercritical sphere without control rod, the other just subcritical.

The spontaneous fission source of neutrons within the fuel itself accompanied by the artificial source at the edge of the reactor was deemed sufficient neutron background to meet the sensitivity requirements of the monitoring and safety instruments. This precluded the preliminary determination with the reentrant tube down and all later tests were made with it withdrawn into the neck of the sphere. Criticality determinations were frequently conducted with fuels of greater than 400 MWD/T exposure in the absence of an artificial source since the number of spontaneous fission neutrons is of a magnitude suitable for detection through several inches of water.

Periods

Period measurements were taken in several spheres to establish the amount of excess plutonium required to produce periods between five and 90 seconds. A typical plot of the data taken appears in Figure 8. This type of plot indicated reliably, in the case of a slightly overconcentrated sphere, the amount of plutonium to be removed from the reactor in order to make it just subcritical without control rod.

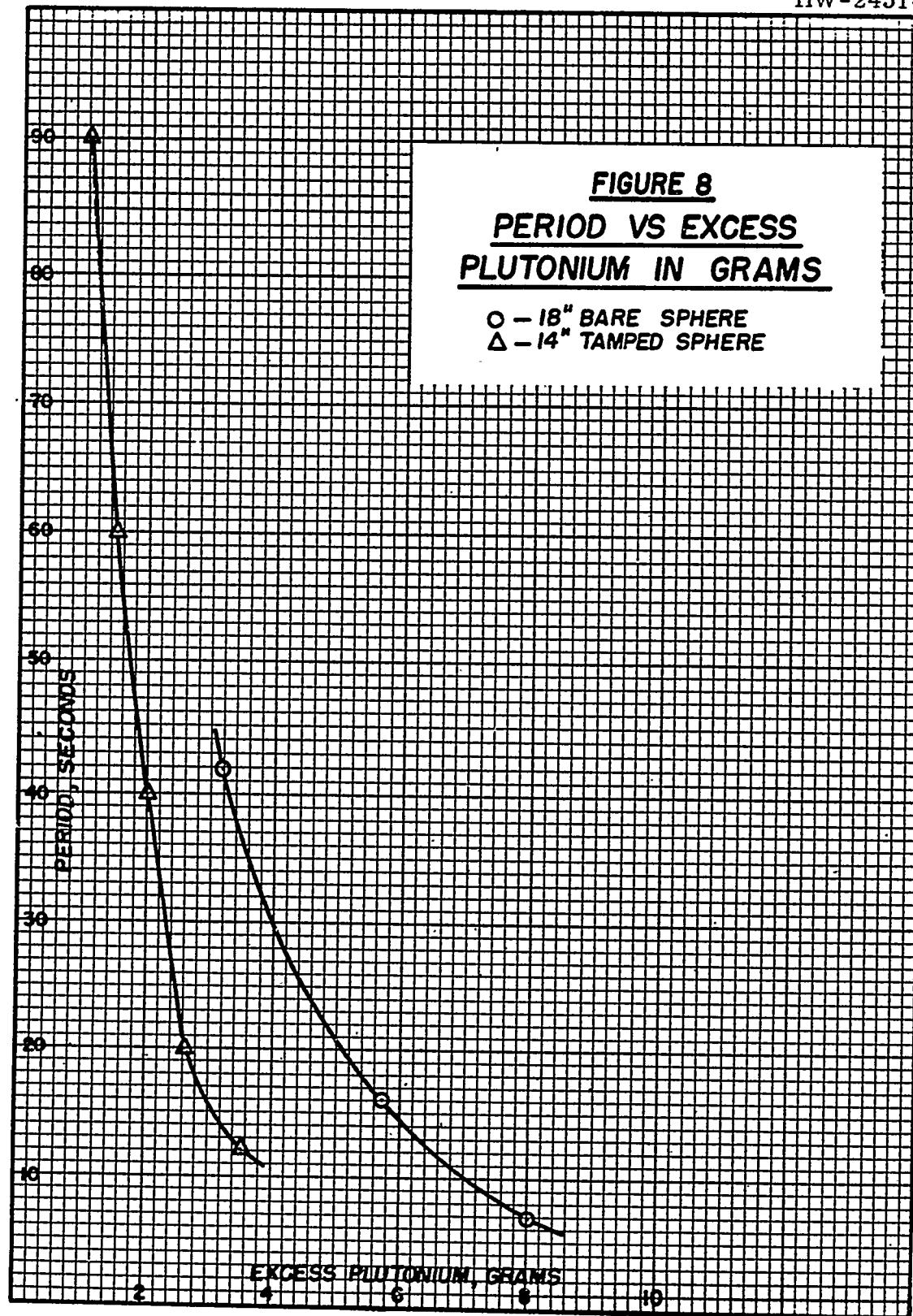
Power Levels

Nominal power levels at critical were of the order of twenty milliwatts. The highest exposure experienced by personnel in the control room was approximately 25 milliroentgens per hour, this being an unusually high level experiment. Personnel exposure seldom exceeded one-quarter mr/hr during normal runs. Operating power levels were purposely limited to as low values as possible to avoid overexposure of personnel from residual fuel activity during fuel alterations.

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Correction for Sphere Neck

A mock-up of the supporting neck assembly for the sphere was installed on the lower half of the reactor. It was mounted against the side of the sphere which opposed the control rod. The critical position of the rod was noted, the mock-up removed, and a new critical rod position determined. The difference, in grams, represented by the two rod positions was attributed to the mock-up and subsequently assigned to the sphere neck. This value is a maximum of five grams for all spheres made critical.

Cross Section Measurements

Estimates of the thermal neutron absorption cross section for two isotopes were arrived at from criticality studies. A comparison of the neutron absorption by a known amount of boron with that of the isotopes studied is the basis of these estimates.

Gas samples of these isotopes were placed in the reentrant tube inserted in a spherical reactor which was several grams supercritical with the control rod completely withdrawn. The unknown samples were introduced into the reactor alternately with boron (BF_3) standards in identical containers. Comparison of the relative periods resulting from the same control rod setting produced the unknown cross section values. The sensitivity of the system to internal poisoning of the reactor was set at 10^{19} atoms of boron as a result of these experiments.

Analytical Procedures

Consistent biases in the analyses of P-11 samples were detected by analyzing standard samples, and gross discrepancies were checked by the use of multiple analyses. The precisions reported indicate the magnitude of the variation from the true value which should be exceeded only about once in one hundred analyses.

Plutonium

Plutonium is reduced to the +3 state with titanous chloride and then titrated (reoxidation to the +4 state) with standard ceric sulfate. The titration is carried out in 3 N sulfuric acid under an inert atmosphere of nitrogen and is followed potentiometrically. Nitrates are removed by evaporation with sulfuric acid and iron corrections are applied, if iron is present. The precision of this method is \pm 1.0 per cent.

Hydrogen Ion

Total acid or hydrogen ion concentration in plutonium solutions is obtained by oxalate complexing of plutonium and direct titration of the acid with standard base. Methyl red is used to indicate the end point. The precision of this method is \pm 1.4 per cent.

Iron

Iron is reduced to the ferrous state with hydroxylamine hydrochloride and reacted with ortho-phenanthroline in slightly acidic solutions to form an intense reddish-purple complex which is measured spectrophotometrically. The acidity of the solution is maintained between 3.5 - 4.5 pH by using a buffer. The precision of this method is \pm 1.4 per cent.

Density

This method is based on weighing a known volume of the liquid at 25°C. The value reported is density in grams per cubic centimeter. The precision of the method is \pm 0.03 per cent.

Phosphate

Phosphate ion is precipitated as ammonium phosphomolybdate. The precipitate is collected on a suitable filter, washed, and dissolved in an excess of standard sodium hydroxide. The excess sodium hydroxide is titrated with standard acid using phenol red-bromothymol blue as the indicator. The precision of this method is \pm 2 per cent.

Nitrate

Nitric acid formed from the action of sulfuric acid on a nitrate compound is distilled under vacuum and collected in a condenser coil which is immersed in a dry ice-isopropyl alcohol mixture. The acid distillate is titrated with standard sodium hydroxide. The precision of the method is ± 0.6 per cent.

Water

The Carl Fisher method for determination of water was used. In this application, the sample is added to a known amount of Carl Fisher reagent and the excess reagent back titrated with a methanol-water mixture to a dead stop end point. A tungsten electrode is used. The precision of the method is ± 0.9 per cent.

Bismuth

Bismuth is titrated (reduced to metallic state) with standard chromous sulfate. The titration is carried out in a 0.5 M H_2SO_4 - 2.5 per cent NaCl solution buffered with tartaric acid. The titration is done under an inner atmosphere of CO_2 and followed potentiometrically. Ferric iron is added to obtain the first potentiometric end point since the small amounts of plutonium present did not reveal this end point. Nitrates are removed by heating with formic acid. The precision of the method is ± 1.7 per cent.

Manganese

Permanganate formed by periodate oxidation of manganese in an acid solution is determined spectrophotometrically. A 1 cm. light path length is used and the optical density determined at wave lengths of 525 and 545 milli microns. The precision of this method is $\pm 1-2$ per cent.

Lithium

Lithium is determined spectrochemically by the copper spark method using barium as an internal standard. Lithium line 6707 and

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barium line 6694 are photometered and intensity ratios are determined by means of a Hurter-Driffield curve. The precision of the method is \pm 3.0 per cent.

Megawatt Days/Ton

This quantity is reported to have a precision of \pm 10 per cent, according to HW-23487. (2)

Grams Plutonium/Ton

Determinations of this quantity have a precision of \pm 3 per cent. A brief discussion of the method used in this determination is given in HW-23487. (2)

Per Cent Plutonium²⁴⁰ by Weight

As reported in HW-23487, (2) the precision of these determinations is \pm 7 per cent. Where this quantity is not reported in the data tables, reference may be made to Figure 13.

DESIGN AND DISCUSSION OF EXPERIMENTS

A review of the critical mass problems pertaining to the Hanford separations processing had indicated the desirability of determining the minimum critical mass of a plutonium solution. Such a measurement cannot be accomplished in practice because of features of plutonium chemistry, the presence of the Pu²⁴⁰ isotope, the necessity of providing a reactor shell, and the dependence of critical mass on temperature. In these experiments, it was necessary to provide as favorable a condition for criticality as was feasible and to evaluate the magnitude of each of the above factors by additional experimentation.

2. G. J. Alkire, H. R. Schmidt, and E. M. Kinderman, HW-23487, 2-25-52.

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Nitrate Ion Effect

The basic fuel selected was plutonium nitrate dissolved in dilute nitric acid. The presence of the nitrate ion associated with the plutonium and nitric acid has an important influence on the critical mass due to its ability to displace moderating media and its parasitic absorption of thermal neutrons. The nuclear properties of the fuel are determined essentially by the plutonium, nitrate, and hydrogen concentrations. The effect of an increase in nitrate concentration is to cause a displacement of hydrogen and an increase in parasitic macroscopic absorption and therefore an increase in critical mass. Figures 9 and 10 show the dependence of critical mass on nitrate concentration for tamped and bare spherical reactors respectively. Within the range of data taken and experimental error imposed it is found that a linear relationship exists between the logarithm of the critical mass and the nitrate concentration, as is shown in Figures 11 and 12.

Isotopic Content of Plutonium

Plutonium produced by reactor conversion is principally a mixture of the Pu^{239} and Pu^{240} isotopes. As the nuclear properties of these isotopes are quite different, it is necessary to specify the isotopic content of a fuel under consideration. This is customarily expressed by stating the pile exposure of the fuel in MWD/T. Figure 13 shows the percentage of Pu^{240} in fuel of a given pile exposure as reported in HW-23487. (2) The critical mass of fuel solutions of several different exposure levels was measured as a means of determining the dependence of critical mass on isotopic concentration. Figure 14 shows this effect in the tamped 14 inch diameter sphere for fuels ranging between 55 and 605 MWD/T, Figure 15 for 212 and 420 MWD/T in the 13 inch sphere. Presumably, the difference in critical mass between fuels differing only in pile exposure is due to the difference in the Pu^{240} content of the fuel. Such data indicates a large total absorption cross section for Pu^{240} of 925 ± 200 barns. Figure 16 expresses the variation in critical mass with MWD/T in the 14 inch sphere at a nitrate concentration of 110 grams/liter.

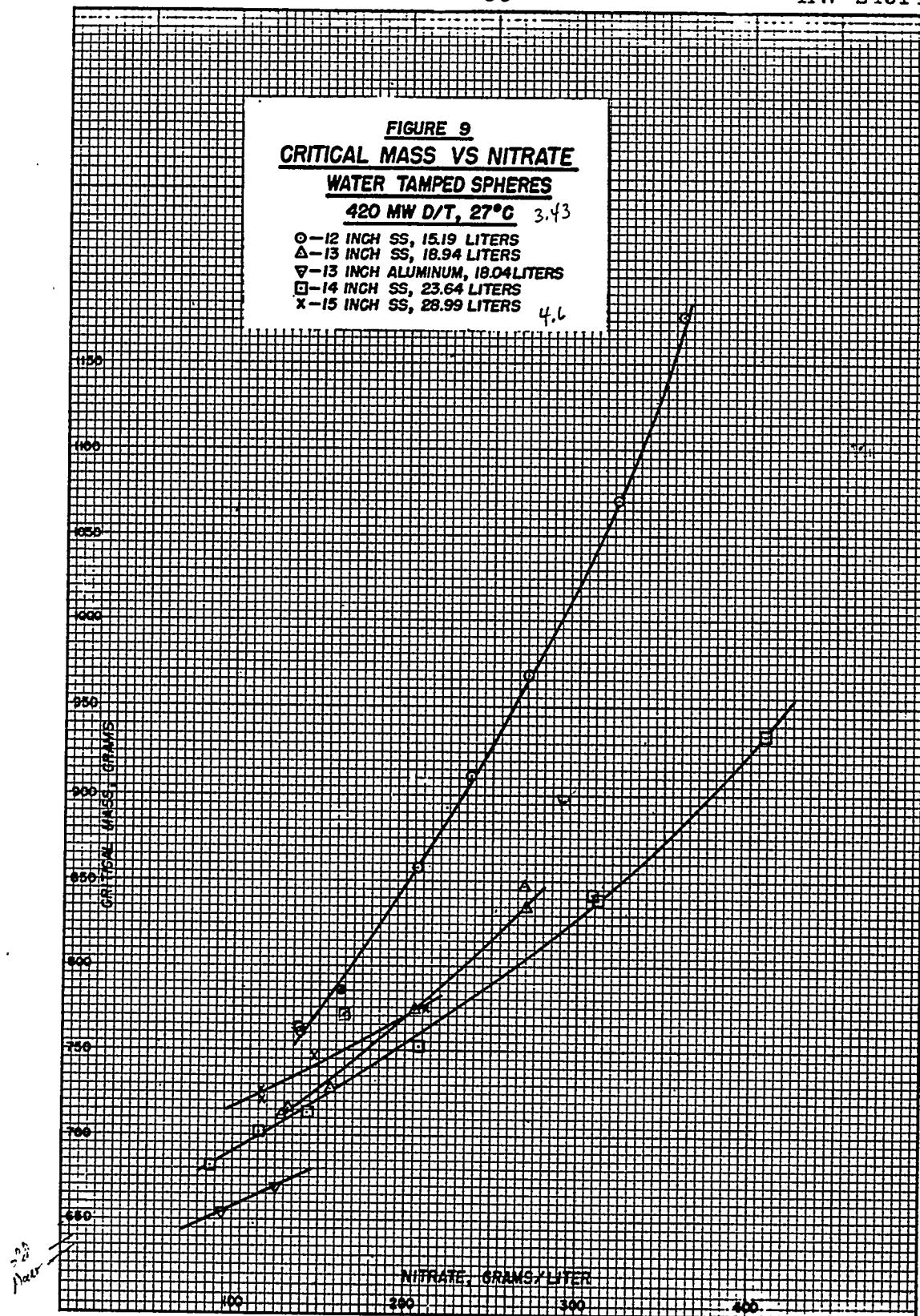


FIGURE 10
CRITICAL MASS VS NITRATE
BARE SS SPHERES
572 MW D/T

- - 16 INCH, 34.15 LITERS
- - 18 INCH, 49.0 LITERS
- △ - 20 INCH, (ESTIMATED)

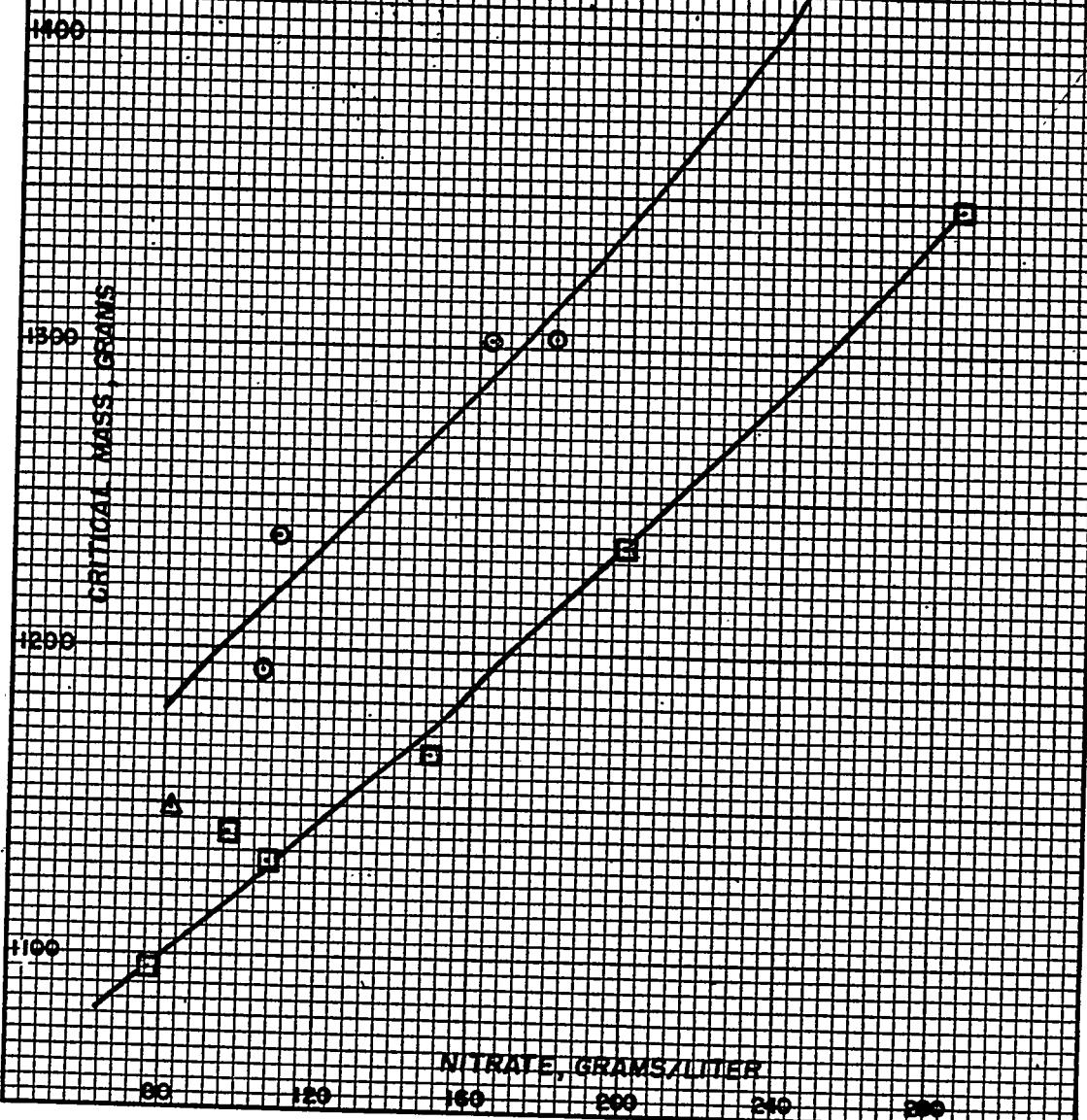


FIGURE II
LOG₁₀ CRITICAL MASS VS NITRATE
WATER TAMPED SS SPHERES
420 MW D/T, 27°C

○ - 12 INCH
▽ - 13 INCH
△ - 14 INCH
□ - 15 INCH

3.95

3.90

LOG₁₀ CRITICAL MASS
3.85
3.80
3.75

3.80

3.75

100

200

400

600

800

1000

1200

1400

1600

1800

2000

2200

2400

2600

2800

3000

3200

3400

3600

3800

4000

4200

4400

4600

4800

5000

5200

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43200

43400

43600

43800

44000

44200

44400

44600

44800

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45800

46000

46200

46400

46600

46800

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54600

54800

55000

55200

55400

55600

55800

56000

56200

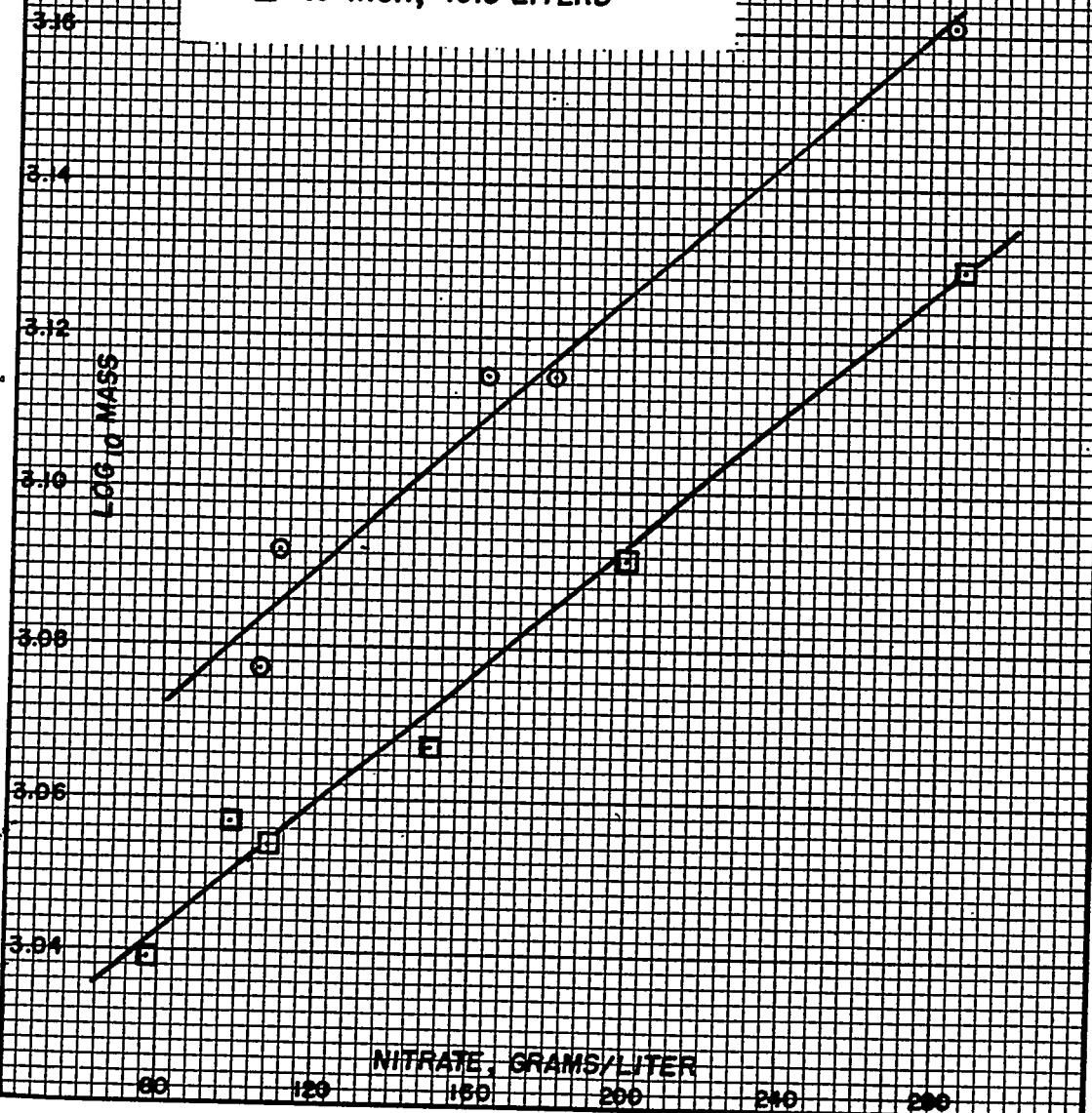
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FIGURE 12
LOG₁₀ MASS VS NITRATE
BARE SS SPHERES
572 MW D/T

○ - 16 INCH, 34.15 LITERS
□ - 18 INCH, 49.0 LITERS



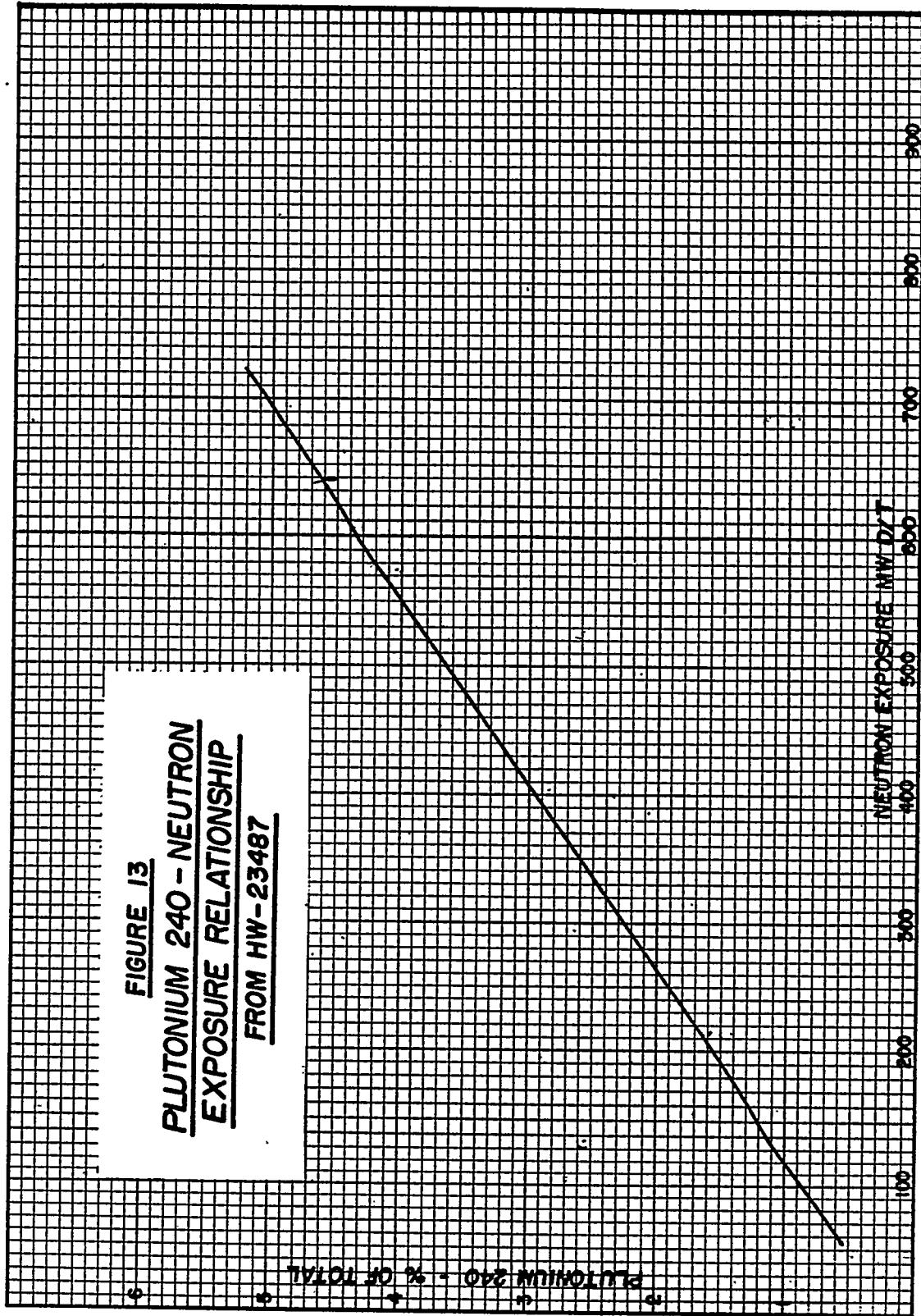
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FIGURE 13
PLUTONIUM 240 - NEUTRON
EXPOSURE RELATIONSHIP
FROM HW-23487

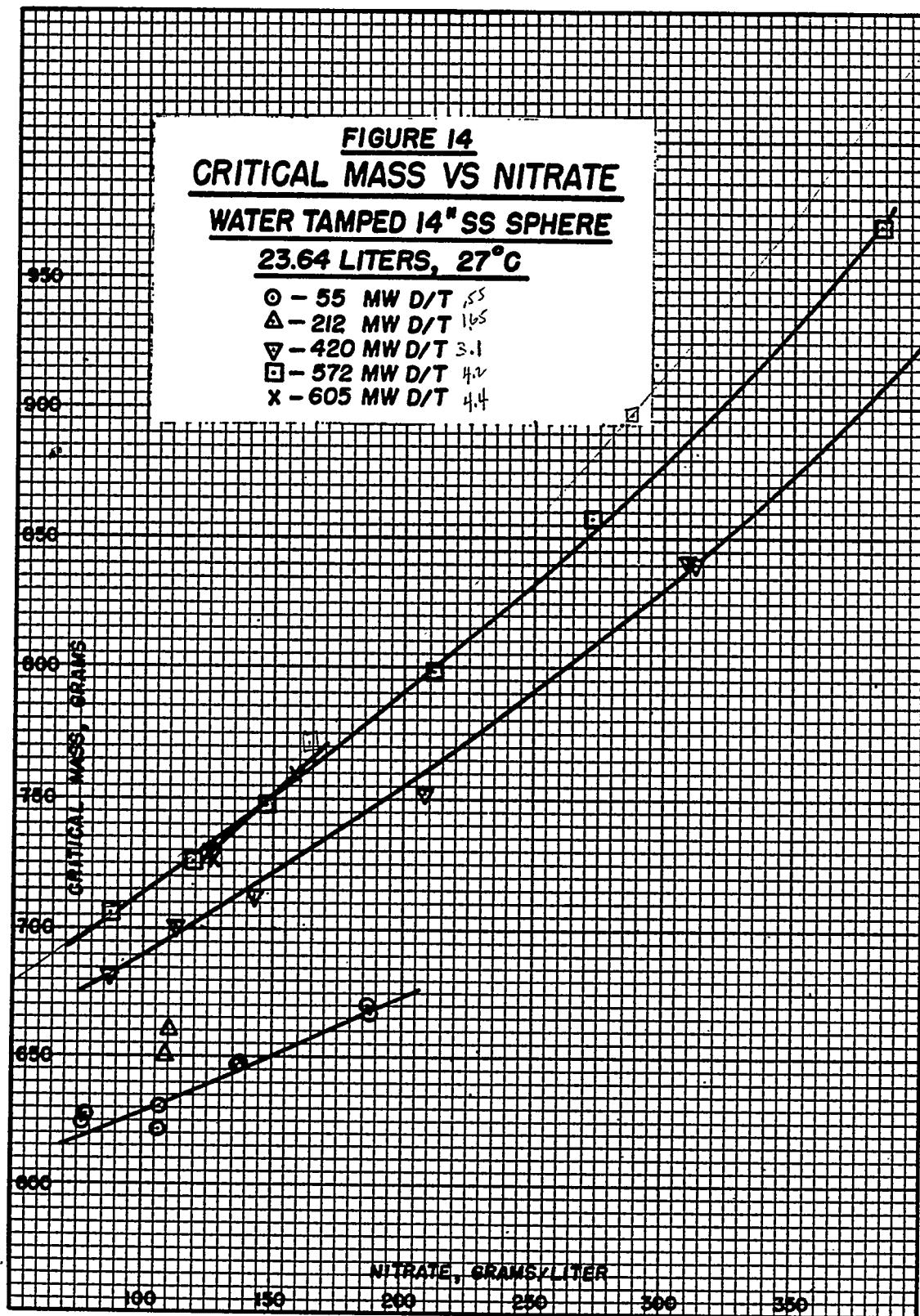


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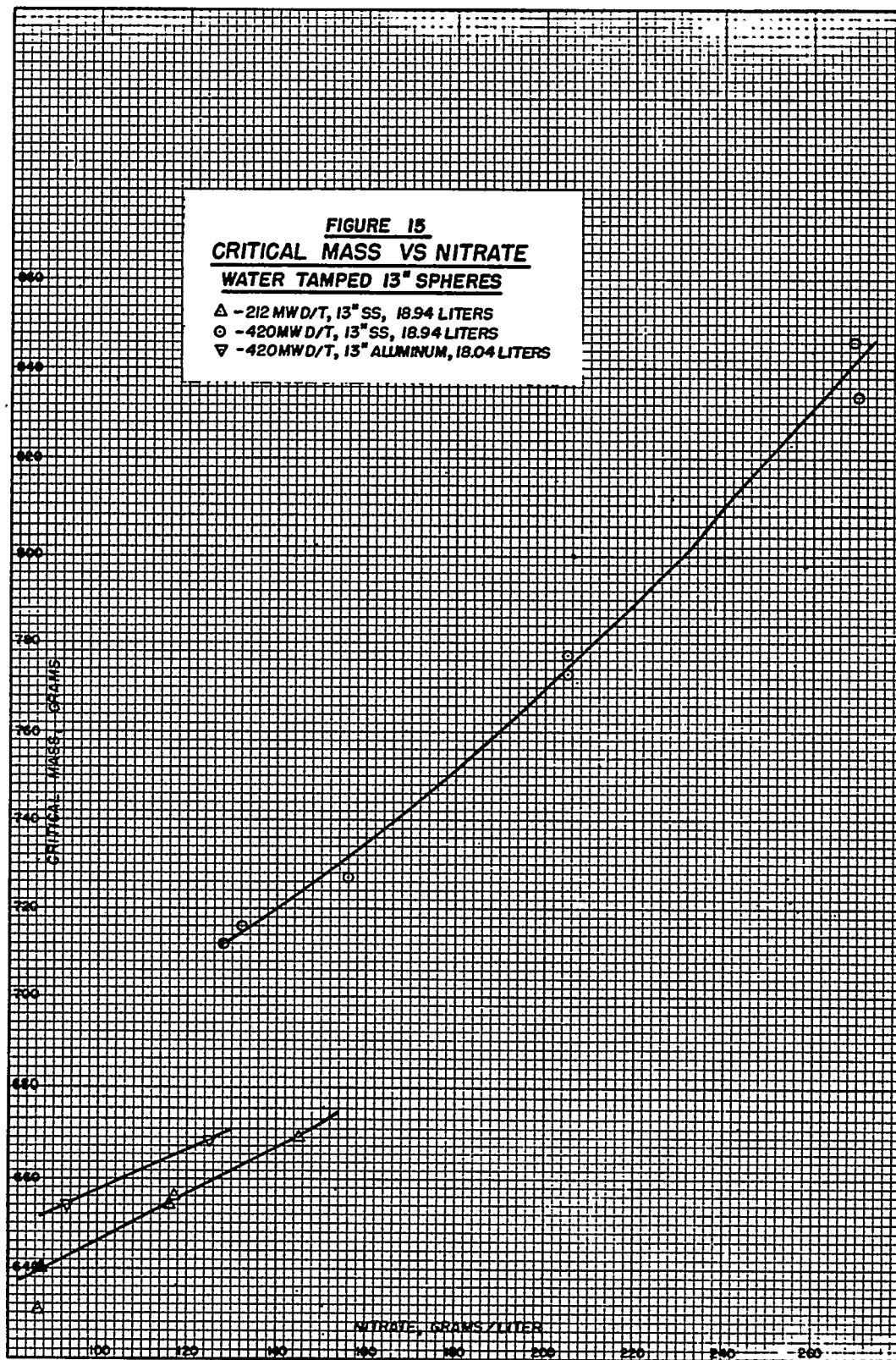


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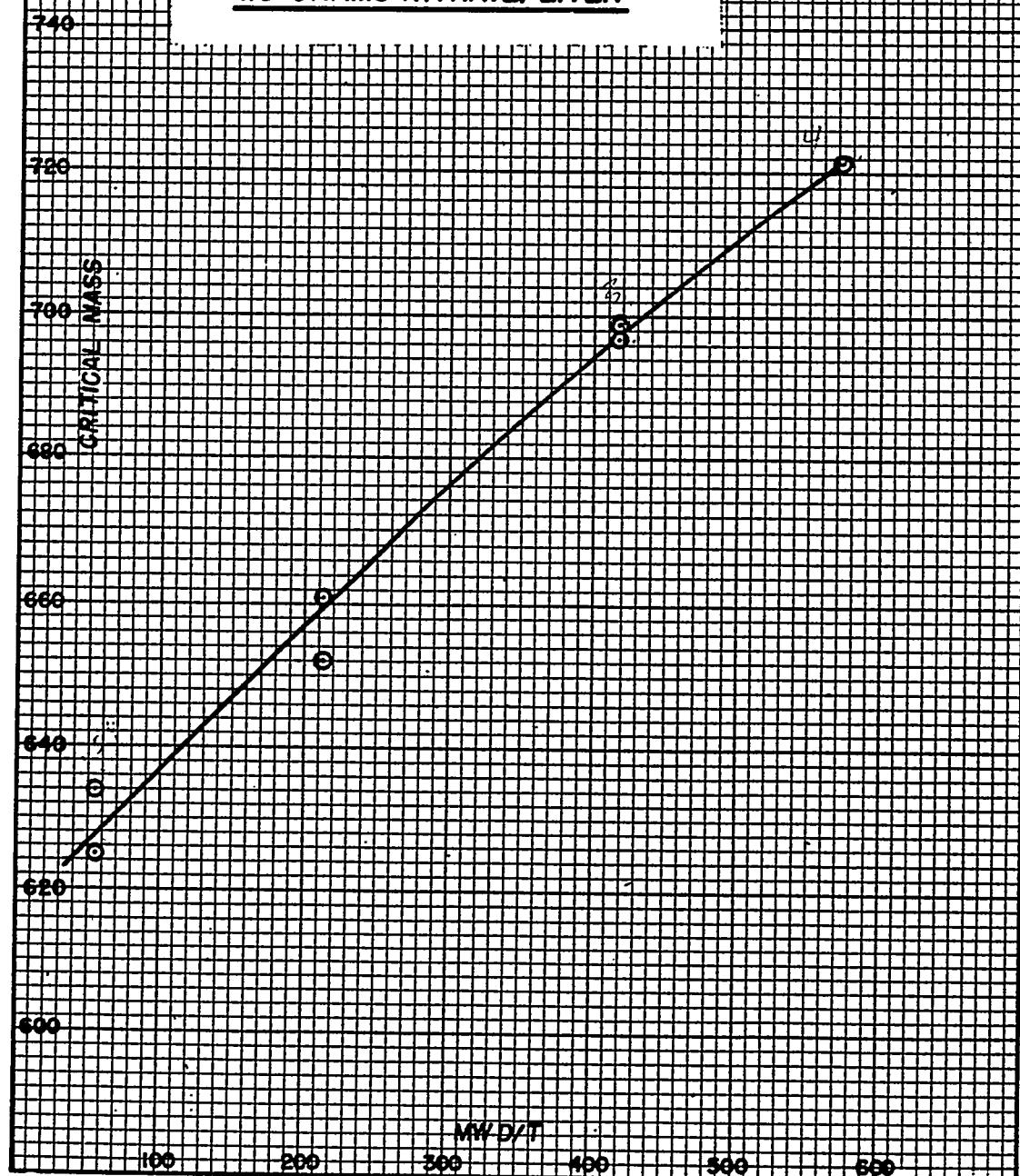
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FIGURE 16
CRITICAL MASS VS MW D/T
14" WATER TAMPED SPHERE
110 GRAMS NITRATE/LITER



620

640

660

740

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Optimum Concentration

It is necessary to establish experimentally the optimum concentration of plutonium in order to find a minimum critical mass. Experiments were made in a series of four spheres of sufficient volume range to bracket the experimentally determined minimum mass. Figure 17 shows the dependence of critical mass on reactor volume for fuels having identical nitrate and isotopic content. These curves show that the optimum concentration of plutonium in aqueous solution lies at about 33 grams of plutonium per liter.

These curves also show the dependence of critical mass on volume for the range studied.

The Effect of Other Non-fissionable Elements

The effect of the addition of compounds of non-fissionable elements other than those of the original fuel on criticality was investigated. These impurities were introduced in order to predict their behavior when used as process reagents in the separation plant processes. These tests were directed toward an independent evaluation of effect of poisoning and displacement in the reactor core.

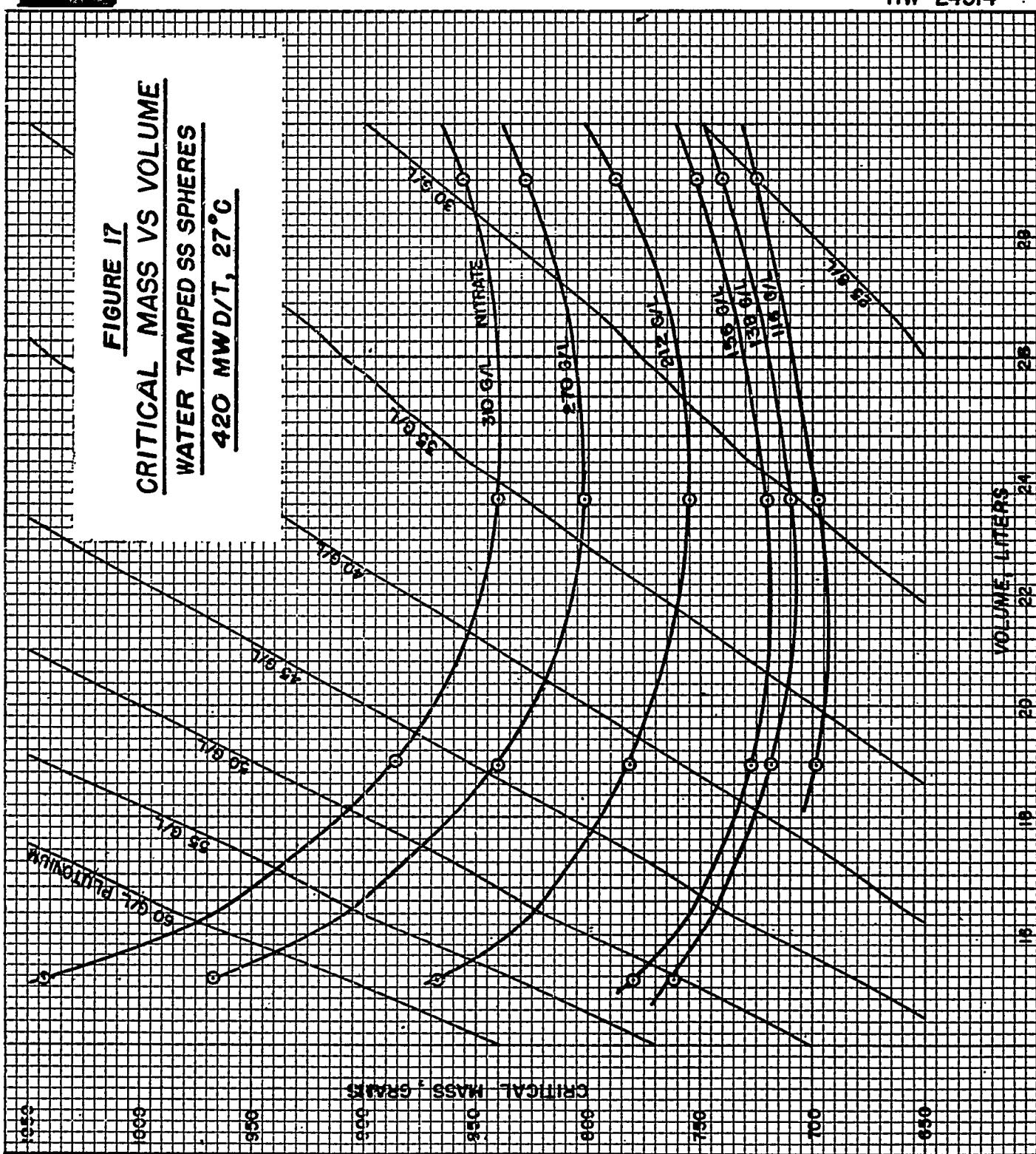
Lithium nitrate was added in increments to obtain a poison calibration in a tamped and bare reactor. It was selected for this purpose because of its large absorption cross section with a \sqrt{E} energy dependence. At the concentrations required here, about 1 gram of lithium per liter, displacement effects are negligible. The change in critical mass with lithium for the tamped 14 inch reactor is shown in Figure 18. The rather wide variation in experimental points is attributed to difficulties encountered in the analytical determination of fuel constituents. The short section of graph represents the variation of mass with lithium at a nitrate concentration of about 207 grams $\text{NO}_3^-/1$. The effect of the progressive increase in nitrate concentration associated with the lithium and fuel additions, was corrected by the adjustment of the data to a nitrate concentration of 110 grams per liter.

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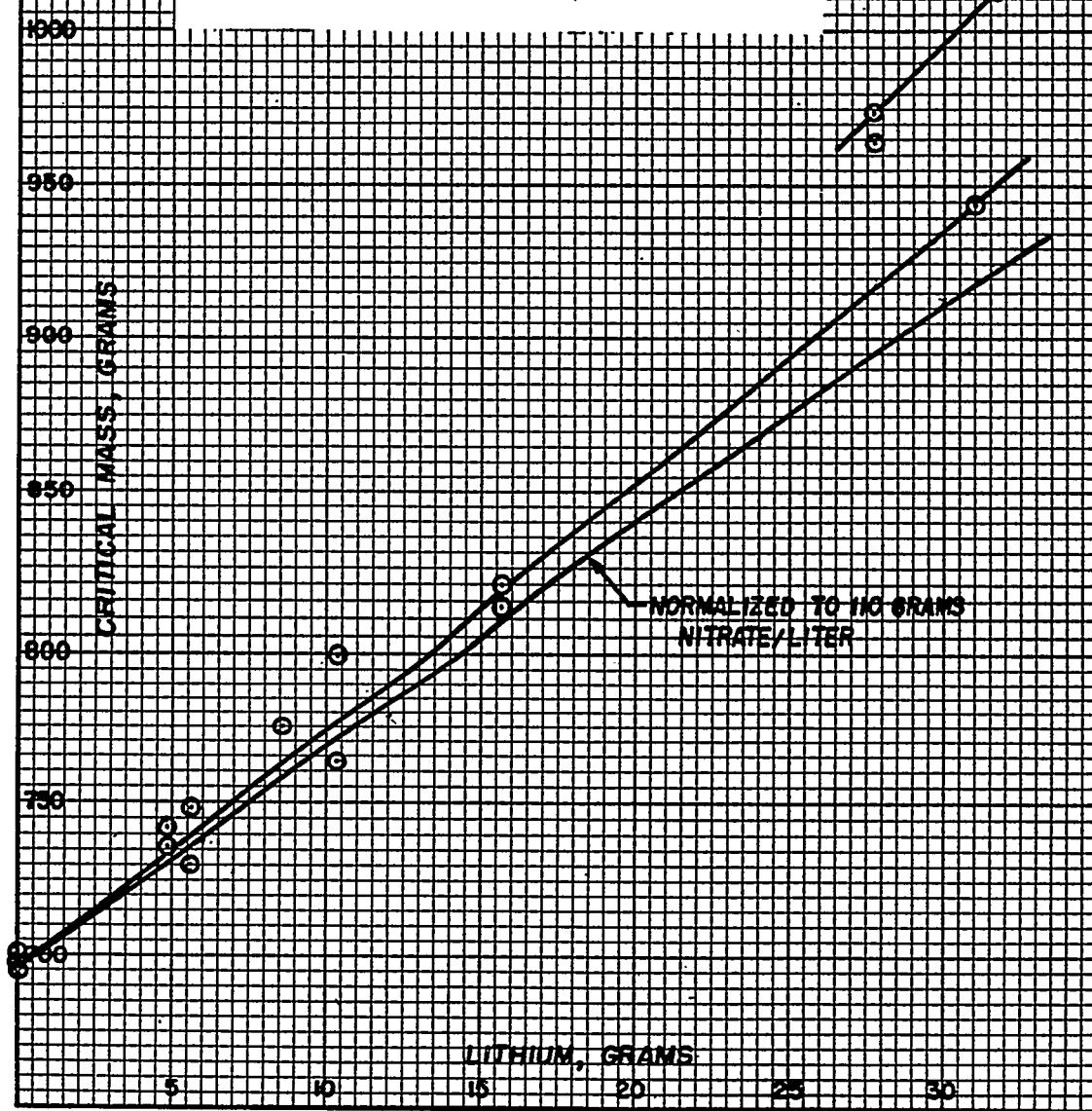
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FIGURE 17
CRITICAL MASS VS VOLUME
WATER TAMPED SS SPHERES
420 MW D/T, 27°C



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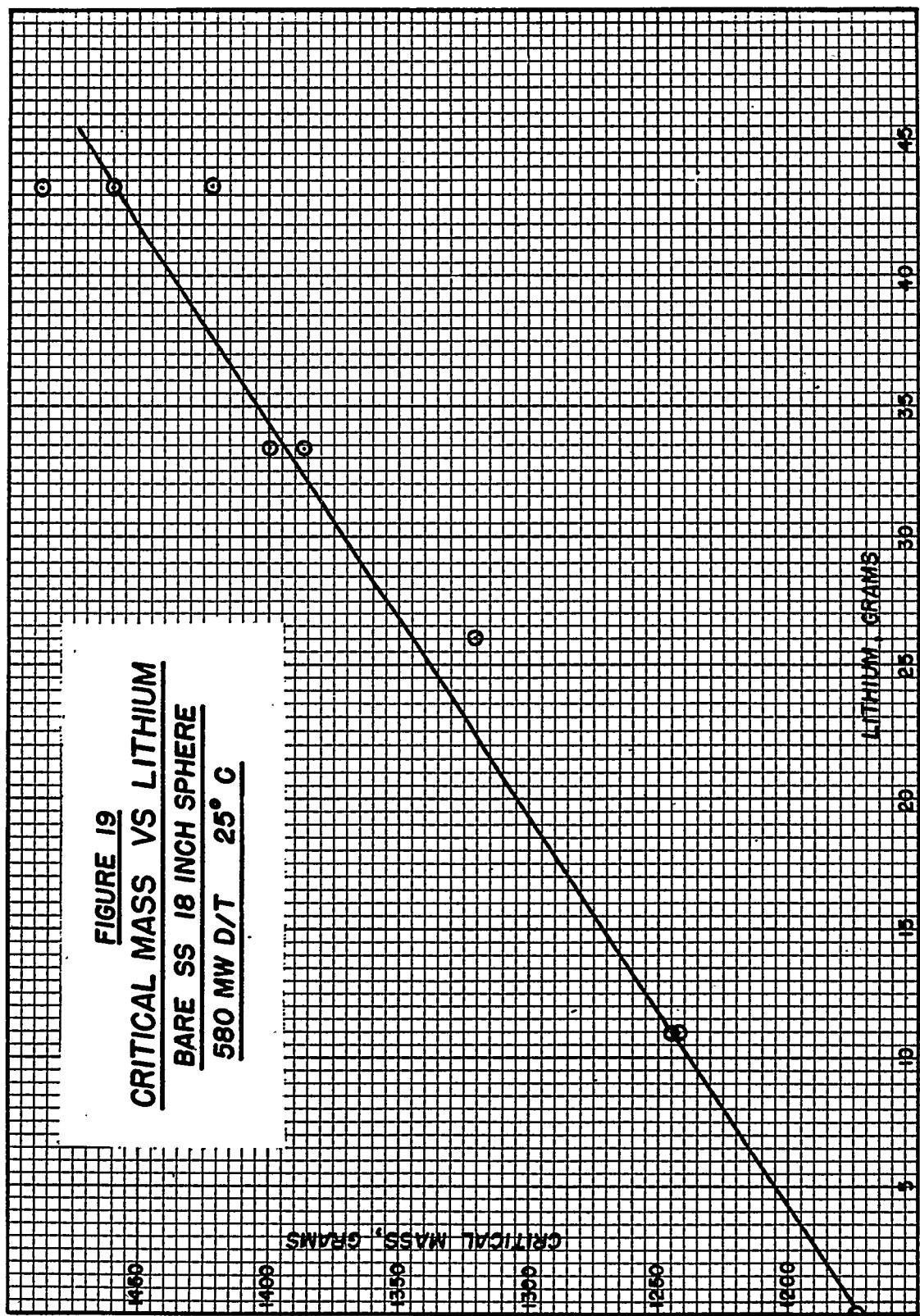
FIGURE 18
CRITICAL MASS VS LITHIUM
WATER TAMPED 14 INCH SS SPHERE
23.64 LITERS, 27°C
~420 MW D/T, 3.43 % Pu²⁴⁰

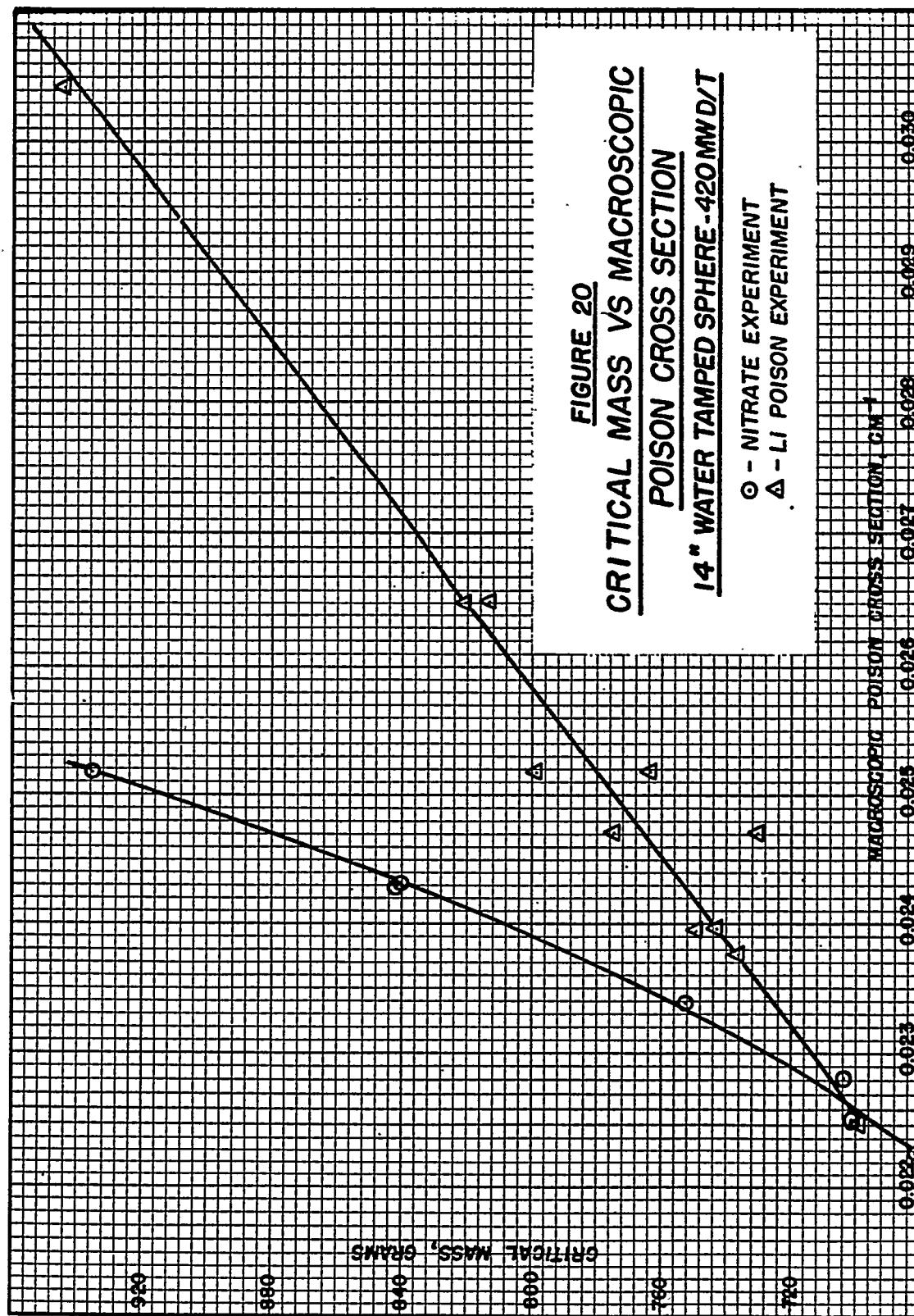


A similar calibration, Figure 19, was made in the 18 inch bare sphere at a nitrate concentration ranging from 116 to 132 grams $\text{NO}_3/1$.

Two attempts were made to investigate the effect of displacement by impurities. Additions of phosphoric acid were made, after first oxidizing the fuel to the Pu(VI) oxidation state. Difficulties were encountered because of the fuel precipitation and because of the relatively large quantities of potassium permanganate required. A second attempt was made using sulfuric acid for displacement purposes. The absorption cross section of phosphorous and sulfur are sufficiently low that either acid could have produced the desired effect. Unfortunately, difficulties encountered in the analytical determination of the fuel constituents prevent a quantitative measurement of the increase in mass with displacement. It is readily apparent, however, that a decrease in core hydrogen density as the result of the addition of impurities causes an increase in critical mass even though the total parasitic absorption in the core is reduced by this process. Figure 20 shows the effect of displacement of hydrogen in the case of additions of nitrate as compared with lithium additions.

Bismuth was added to a fuel solution to determine whether its inelastic scattering properties would have an appreciable effect on criticality at the concentrations prevalent in the Hanford BiPO_4 separations process. Figure 21 shows the increase in mass with bismuth concentration. However, this curve must be corrected for the nitrate which was added with the $\text{Bi}(\text{NO}_3)_3 \cdot 5 \text{H}_2\text{O}$ crystals and fuel. The necessary correction is not fully understood. If the increase in nitrate is corrected for by subtracting the incremental change in mass for the equivalent nitrate increase, the resulting curve indicates an increase in critical mass with increasing bismuth concentration, followed by a decrease to just less than the original bismuth free mass at the highest bismuth concentration reached. It does not seem reasonable that this should be the case as inelastic properties should manifest themselves on an atomic concentration basis.

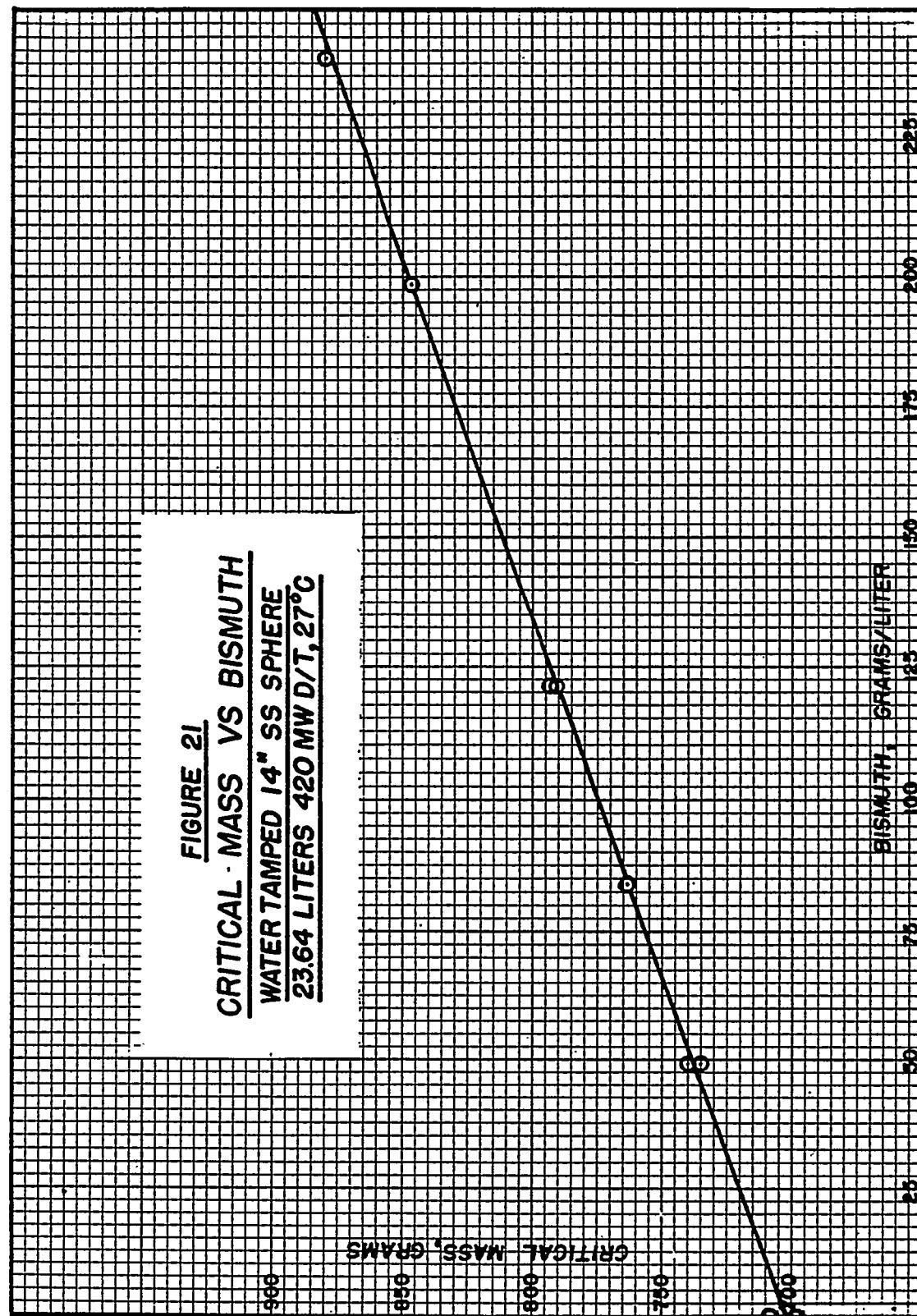




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Reactor Shell Effect

An attempt was made to correct the measured critical mass values for the perturbation caused by the thin stainless steel reactor shells. The presence of such a wall between the fuel and reflector increases the necessary critical concentration, or for a given concentration, the critical volume. This effect is due to the large capture cross section of iron for thermal neutrons reflected back into the core. An evaluation of the magnitude of this effect was made by comparing the critical radius of a tamped aluminum sphere with negligible wall absorption, and a tamped sphere with a thin stainless steel wall using the same fuel. A single reactor made of 20 gauge 2S aluminum was used for this measurement. A second test was made by surrounding a cylindrical reactor with an additional thickness of stainless steel and noting the increase in critical height required to attain criticality.

It was apparent from these limited experiments that the effect of the wall increased as the hydrogen content or moderation in the core was decreased. This observation was qualitatively made by noting an increase in the control cup strength under these conditions.

It was found that the addition of a 0.020 inch layer of cadmium to a bare spherical reactor effected a reduction in critical mass by reflection of neutrons, of 20 grams at low nitrate concentrations. This indicated that a correction must also be made for the reflection of the 0.050 inch wall of the bare reactors.

The extent and reliability of the data is not sufficient to correct for the effect of the reactor shell under all conditions. Comparison of the bare and tamped stainless steel spheres gives an extrapolation length of about 3.2 cm. at a hydrogen concentration of about 108 grams/liter, which may be increased to about 5 cm. after correcting for reflection and absorption. Only the absorption effect of the steel shell was measured. It was

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found that the presence of the steel shell required an increase in the critical radius of about 0.22 cm. or about 30 grams in the critical mass.

Temperature Effect

The temperature dependence of critical mass as measured in a reactor is due to the change in density of the moderator with temperature and to a change in the average neutron energy. Measurements were made in a range of 60 to 120° F. It is not possible on the basis of the data to fix the values of these effects. A rise in temperature in tamped reactors produced a negative reactivity coefficient of not more than -1.0 gram Pu/°C. over the range of these experiments. The temperature at criticality was held at 27°C. for the greater part of this work.

PREDICTION FORMULA

Derivation of Formula

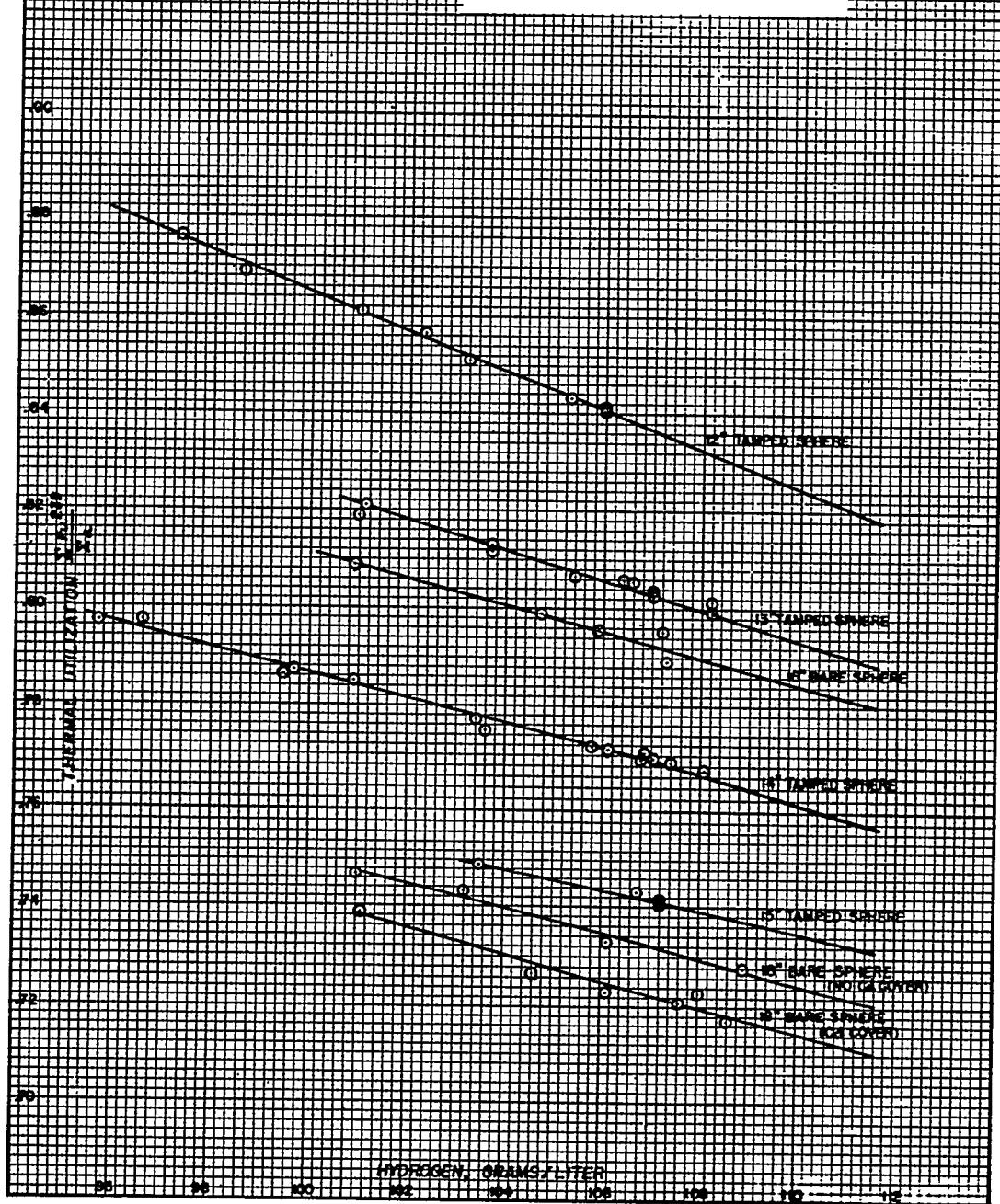
It has not been possible on the basis of simple diffusion theory to predict the critical mass of plutonium solutions quantitatively. The data obtained in the course of these studies has been analyzed in an attempt to arrive at an expression which will predict the critical mass over the entire range of the work done with plutonium nitrate solutions.

The thermal utilization, f , of the fuel is found to vary linearly with hydrogen concentration as shown in Figure 22 using cross section values as given below. Since the change in thermal utilization is relatively insensitive to small poison changes, the increase in thermal utilization with decreasing hydrogen concentration must be due to a change in moderation. The change in f is also a function of the sphere volume, or the buckling (B), as given by $B = \frac{\pi^2}{(R + 1)^2}$ in diffusion theory where R and l are the reactor radius and extrapolation length respectively. Using an extrapolation length of 4 cm. for tamped steel spheres as approximately indicated by Figure 23, the thermal utilization for plutonium nitrate fuels is given by:

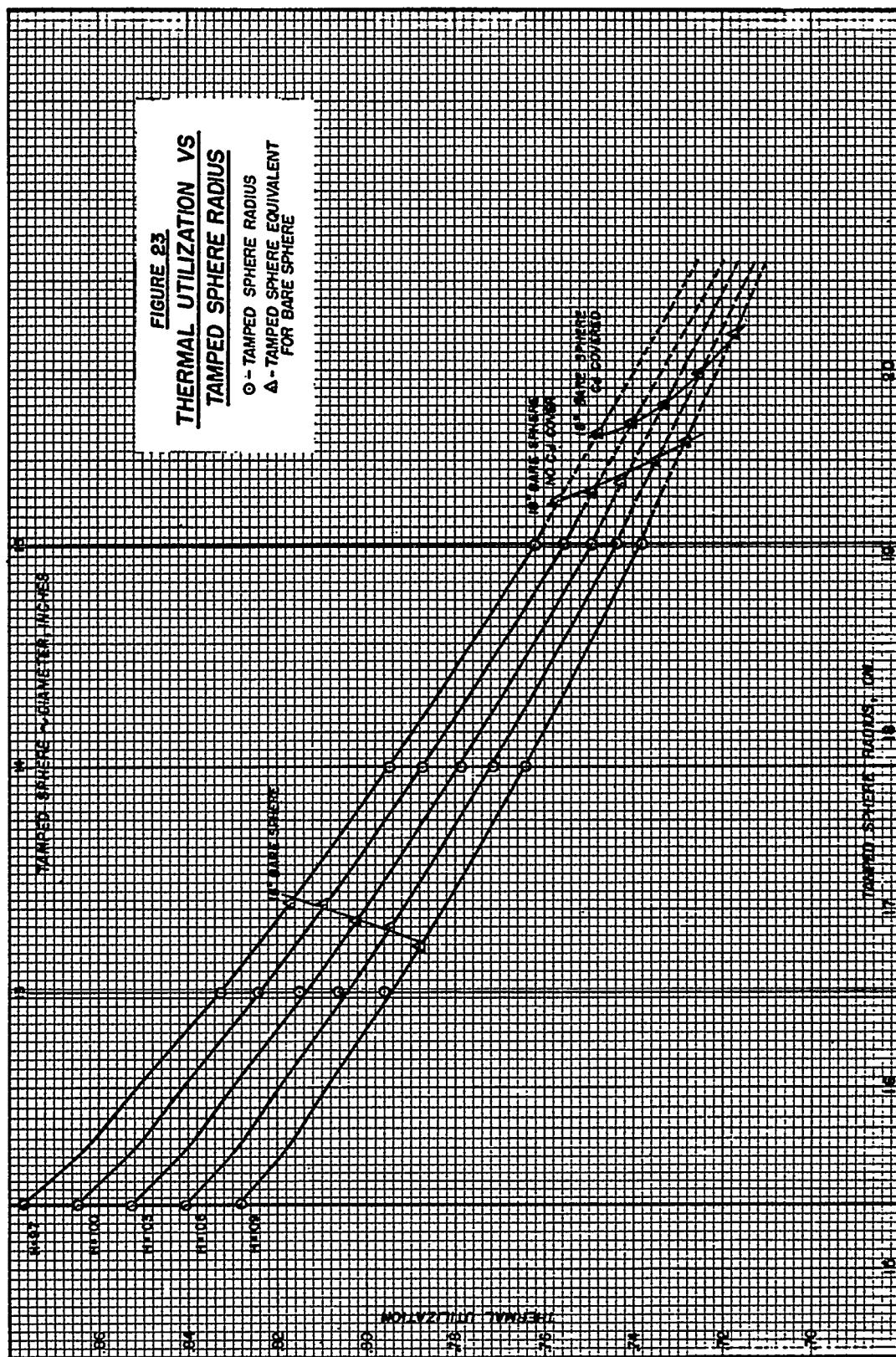
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FIGURE 22
THERMAL UTILIZATION
VERSUS HYDROGEN



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$$f = (38.90 - 0.2494 H)B + 2.743 \times 10^{-3} H + 0.2221$$

where H is the hydrogen concentration in grams/liter.

The critical mass is then given by:

$$M = \frac{V \cdot 239 \times 10^4}{\sigma_{239} (6.02)(1-w)} \left[\frac{\sum \text{poison}}{1/f - 1 - \left(\frac{w}{1-w} \right) \frac{239}{240} \frac{\sigma_{240}}{\sigma_{239}}} \right]$$

where

V = volume in liters

σ = cross section in barns

w = Pu^{240} fuel fraction

σ_{240} = 1000

σ_{239} = 1150

σ_H = 0.32

σ_N = 1.7

This formula predicts the critical mass over the range of these experiments of tamped spheres within experimental error and tamped cylinders to within ± 3 per cent.

Minimum Critical Mass

This formula enables us to predict the hypothetical minimum critical mass of plutonium²³⁹ and water. A mass of 509 grams is predicted on this basis at a concentration of about 32.6 grams/liter in a 12.2 inch diameter thin steel sphere at a temperature of 27°C. A somewhat smaller mass would result in the absence of the spherical shell.

The predictions arrived at by this formula are in good agreement with values obtained from other data analyses and graph extrapolation.

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A maximum safe tamped cylindrical diameter of 6 inches is indicated. The maximum safe thickness of 2.8 inches is found for an infinite tamped slab. The maximum safe volume of a moderated fuel solution is 5.8 liters, containable in a 8.8 inch diameter thin steel sphere.

CYLINDRICAL REACTORS

The initial critical mass studies at Hanford were undertaken in cylindrical reactors due to 1) the relative ease of changing reactor volume as compared with concentration, 2) the desire to make early comparisons with similar data on U^{235} reported by Beck, et al. (1) and 3) the large uncertainty in theoretical prediction of the range of concentration required to obtain a minimum mass. These experiments provided a means of scoping the work requiring investigation. It was not recognized until the completion of a number of these experiments that the nitrate concentration of the fuel would have such a significant bearing on critical mass. Unfortunately, no attempt was made to hold nitrate concentration constant although the free acid content was held between 1.5 and 2.0 $N\ HNO_3$. These data should be corrected for nitrate variation which is difficult to accomplish in the geometries encountered. Figure 24 shows the relationship between critical concentration and height in each of four cylinders made critical. Figure 25 gives the critical mass in terms of plutonium concentration.

An estimate of the nitrogen and hydrogen concentrations in each of the reactors is given in Figure 26 as a function of reactor height. This information is provided to indicate that the shape of the above curves will be altered somewhat after the nitrate correction is applied due to an increase in nitrate concentrations as the fuel is concentrated.

Acknowledgements

The authors wish to acknowledge the contributions to the P-11 Project of G. M. Muller and J. W. Flora.

F. E. Kruesi / by J.O.E.
F. E. Kruesi

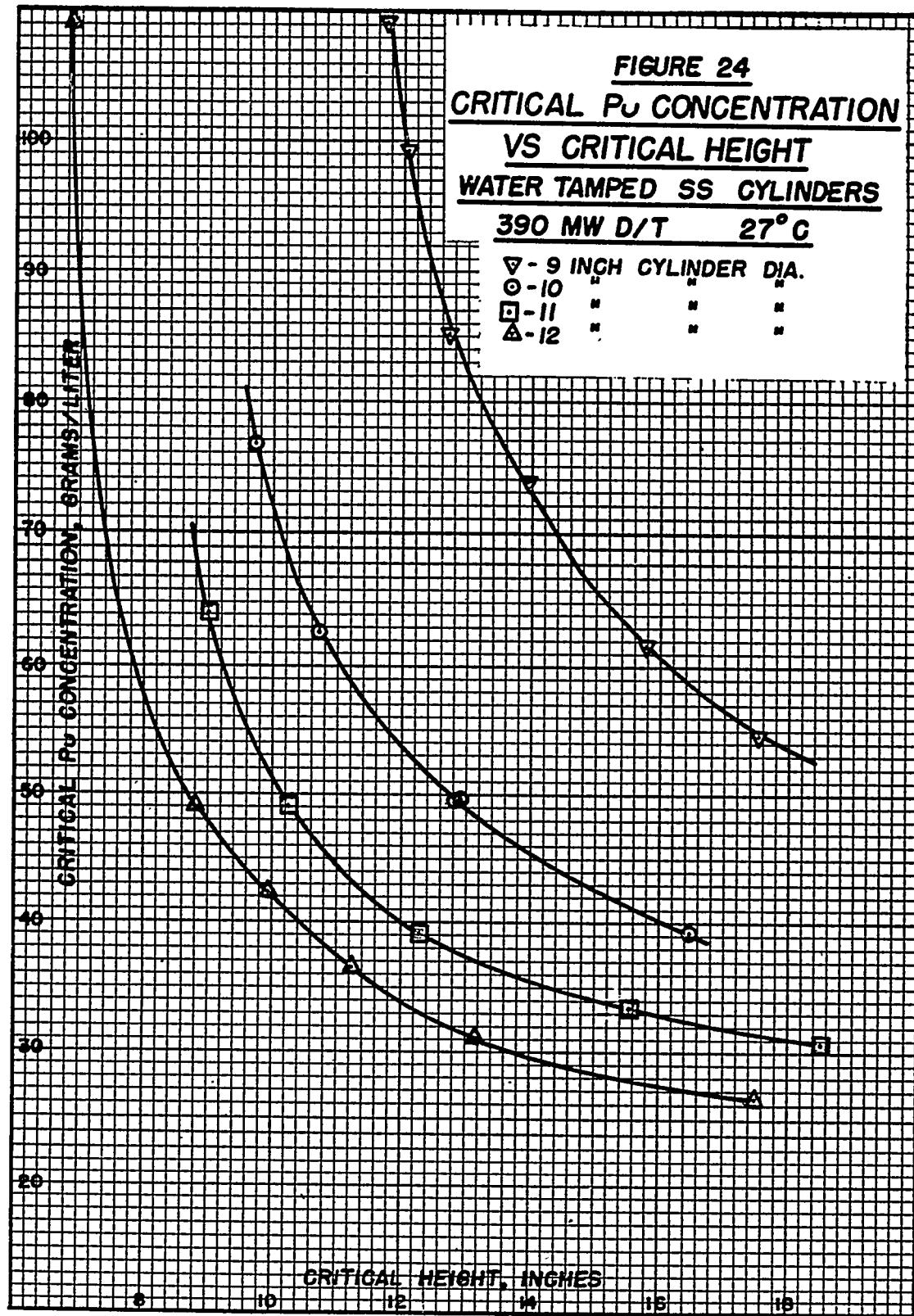
J.O. Erkman
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D. D. Lanning
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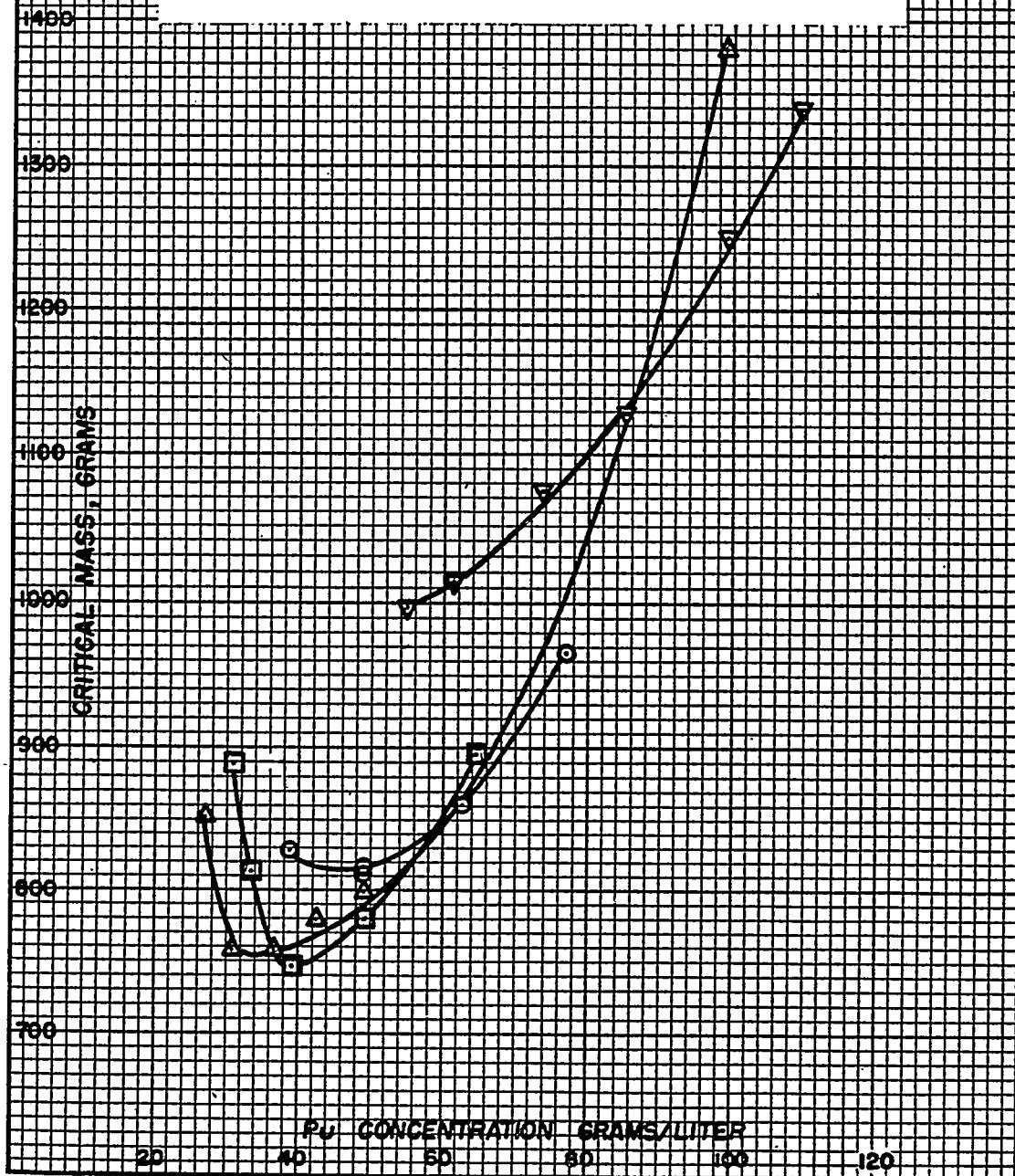
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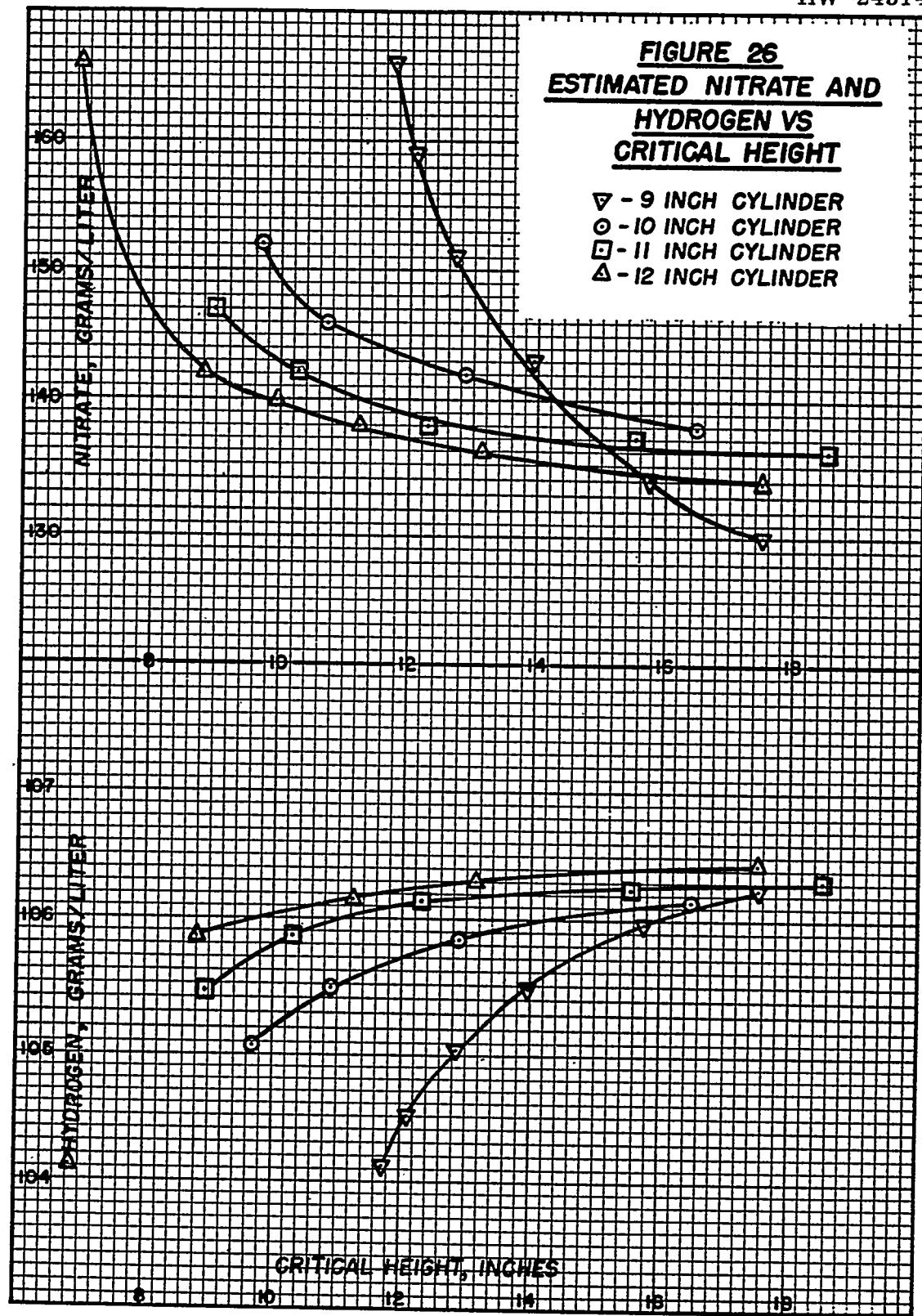


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FIGURE 25
CRITICAL MASS VS Pu CONCENTRATION

▽ - 9" CYLINDER DIAMETER
○ - 10" CYLINDER DIAMETER
□ - 11" CYLINDER DIAMETER
△ - 12" CYLINDER DIAMETER





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DATA

Explanation of Data Tables

In the tables which follow, the results of the P-11 Criticality Experiments are summarized. A description of the chemical analysis of the fuels, and an indication of the accuracy of the results was given on page 33. A blank space in these tables indicates that no analysis was made in that particular case, as when a duplicate sample was submitted for plutonium and iron assays only. In some cases where analytical results were not obtained, estimated values have been used which are based on recorded fuel additions. These values may be in error as much as ± 5 per cent, as indicated on page 14.

Entries in the data tables not covered by the reference above are described below.

Reactor Diameter:- For cylinders, the reactor diameter is reported to within ± 0.02 inches. The nominal diameter is given for the spheres.

Reactor Height:- The height of the cylinders was measured to ± 0.01 inches.

Reactor Volume:- Cylinder volume was calculated to be uncertain to the extent of ± 0.6 per cent. The volume of the spheres was measured to ± 0.3 per cent.

Temperature:- The temperature of the tamper was controlled to within $\pm 0.25^{\circ}\text{F}$. The temperature of the fuel used in the untamped reactors is reported to within $\pm 1.0^{\circ}\text{F}$.

Total Hydrogen:- This quantity may be calculated in one of three ways for most of the experiments reported.

1. By calculations based on the chemical analysis for water described under analytical procedure.

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2. The water content may be calculated by subtracting the plutonium, iron, nitrate, hydrogen ion and any other fuel constituent from the density. Total hydrogen is then found as in (1).
3. An equation, based on nitric acid tables, was developed giving total hydrogen in terms of nitrate and hydrogen ion concentrations only i. e. $\text{Hydrogen} = 111.8 + \text{hydrogen ion concentration} - 0.0535 \times \text{nitrate ion concentration}$.

Analyses for water were not made for the cylinder and bare sphere experiments, so that the first method is not generally applicable. The second method was considered to be reliable, and was used for experiments other than nitrate experiments and lithium experiments. The cumulative error is a disadvantage of this method. The third method was used for the nitrate and lithium experiments. Over the range of these experiments (up to 80 grams plutonium per liter), this method gives good results when compared to method number two, even though the displacement of the plutonium has been neglected. The smaller cumulative error and the ease of computation are the advantages of this method when used for the fuels mentioned above. The error in this quantity is estimated to be less than 1.0 per cent.

Critical Mass:- The critical mass, as reported, is the product of the volume of the reactor and the plutonium concentration, with a correction being made for the control rod, if necessary. These critical masses are estimated to have an uncertainty of 1.5 per cent.

Spectrographic Analysis:- Samples of the fuel were subjected to spectrographic analysis to determine if any impurities were present in significant amounts. The tamper water was also analyzed, and was found to contain no significant amount of impurities. Results of the tamper water analysis are not included in this report.

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WATER TAMPED CYLINDER DATA

Reactor Diameter (inches)	8.0 ^a	9.0	9.0	9.0	9.0	9.0	9.0	10.0	10.0	10.0	10.0
Plutonium Assay (grams/liter)	<u>77.40</u> ^a	109.16	99.09	85.14	73.92	61.49	54.53	77.40	76.93	62.47	
Acid Normality	<u>1.41</u> ^b	1.68	1.37	1.29 ^b	1.78	1.34 ^b	1.36	1.41 ^b	1.41 ^b	1.52 ^b	
Megawatt Days/Ton	<u>385</u>	383	383	383	383	383	383	385	385	385	
Tamper Temperature (°F)	81.0	81.5	80.0	80.5	81.0	81.0	79.5	83.5	83.0	83.0	
Density (grams/cc.)	<u>1.175</u>	1.2110	1.1997	1.1859	1.1592	1.1441	1.1329	1.175	1.165	1.146	
Total Nitrate (grams/liter)	<u>152</u> ^b	<u>166</u> ^b	136.5	<u>151</u> ^b	125.6	134 ^b	119.8	152 ^b	152 ^b	146 ^b	
Iron Assay (grams/liter)	0.644	0.395	0.378	0.321	0.231	0.303	0.257	0.644	0.327	0.269	
Total Hydrogen (grams/liter)	<u>105.1</u> ^b _{94.7}	104.6 ^b	105.9	<u>105</u> ^b	106.8	106.0 ^b	106.7	105.1 ^b	105.1 ^b	105.5 ^b	
Critical Height (inches)	(19.01)	11.76	12.09	12.73	13.94	15.78	17.55	9.93	9.74	10.73	
Critical Volume (liters)	(15.66)	12.25	12.60	13.26	14.52	16.44	18.29	12.78	12.53	13.79	
Critical Mass (grams)	(1212)	1337	1249	1128	1073	1011	997	989	964	860	

a. This cylinder was subcritical by an unknown amount.

b. These numbers are estimates based on log book entries.

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WATER TAMPED CYLINDER DATA (CONTD.)

Reactor Diameter (inches)	10.0	10.0	10.0	10.0	10.0	11.0	11.0 ^d	11.0	11.0
Plutonium Assay (grams/liter)	49.26	49.26	39.10	39.10 ^c	39.10 ^c	39.10	54.53	47.21	41.73
Acid Normality	1.63 ^b	1.63 ^b	1.70 ^b	1.70 ^b	1.70 ^b	1.70 ^b	1.36	1.38	2.77
Megawatt Days/Ton	385	385	385	385	385	385	383	383	383
Tamper Temperature (°F)	84.0	84.0	83.0	64.0	122	85.4	80.5	80.5	81.5
Density (grams/cc.)	1.130	1.130	1.110	1.110	1.110	1.110	1.1329	1.1202	1.1604
Total Nitrate (grams/liter)	142 ^b	142 ^b	138 ^b	138 ^b	138 ^b	138 ^b	120	117	215
Iron Assay (grams/liter)	0.260	0.260	0.172	0.172	0.172	0.172	0.257	0.275	0.255
Total Hydrogen (grams/liter)	105.8 ^b	105.8 ^b	106.1 ^b	106.1 ^b	106.1 ^b	106.1 ^b	106.7	106.9	103
Critical Height (inches)	12.87	12.82	16.44	16.24	17.49	12.28	10.08	11.09	10.66
Critical Volume (liters)	16.56	16.50	21.16	20.90	22.51	19.12	15.69	17.27	16.60
Critical Mass (grams)	816	813	827	817	879	747	856	815	784

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c. Assays for fuel used in temperature coefficient experiments have not been corrected to the temperature at which the experiment was made.

d. In this experiment, a 0.065 inch layer of stainless steel was placed around the cylinder. The ends of the cylinder were not changed.

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WATER TAMPED CYLINDER DATA (CONT'D.)

Reactor Diameter (inches)	11.0	11.0	11.0 ^e	11.0	11.0	11.0	11.0	11.0	12.0	12.0	12.0
Plutonium Assay (grams/liter)	36.90	33.54	30.81	63.99	48.98	48.75 ^c	48.75 ^c	48.75	42.29	36.52	
Acid Normality	4.28	1.76 ^b	1.78 ^b	1.17	1.38	1.27	1.27	1.27	1.36	1.39	
Megawatt Days/Ton	383	385	385	391	391	391	391	391	391	391	391
Tamper Temperature (°F)	81.5	86.0	83.5	80.5	80.5	69.5	69.5	114.0	84.5	84.0	81.0
Density (grams/cc.)	1.2022	1.099	1.098	1.1483	1.1360	1.1260	1.1260	1.1260	1.1152	1.1077	
Total Nitrate (grams/liter)	300.0	137 ^b	136 ^b	121.1	139.0	116.3	116.3	116.3	126.6	107.1	
Iron Assay (grams/liter)	0.263	0.193	0.173	0.298	0.238	0.223	0.223	0.223	0.174	0.161	
Total Hydrogen (grams/liter)	100.6	106.2 ^b	106.3 ^b	106.5	105.7	106.8	106.8	106.8	106.4	107.4	
Critical Height (inches)	16.93	15.57	18.55	8.98	10.22	10.18	10.52	8.80	9.94	11.21	
Critical Volume (liters)	26.36	24.24	28.88	13.98	15.91	15.85	16.38	16.31	18.42	20.77	
Critical Mass (grams)	973	813	889	895	780	773	799	799	779	758	

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e. This experiment was subcritical. A reliable extrapolation to the critical height was made.

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WATER TAMPED CYLINDER DATA (CONTD.)

Reactor Diameter (inches)	12.0	12.0	12.0	12.0	12.0	12.0 ^e
Plutonium Assay (grams/liter)	31.14	31.14 ^c	31.14 ^c	26.45	109.16	
Acid Normality	1.44	1.44	1.44	1.09	1.09	1.68
Megawatt Days/Ton	391	391	391	391	391	383
Tamper Temperature (°F)	82.5	67.0	116.6	78.5	82.0	
Density (grams/cc.)	1.1028	1.1028	1.1028	-	-	1.2110
Total Nitrate (grams/liter)	114.0	114.0	114.0	134 ^b	166 ^b	
Iron Assay (grams/liter)	0.153	0.153	0.153	0.154	0.154	0.395
Total Hydrogen (grams/liter)	107.1	107.1	107.1	105.7 ^b	105.7 ^b	104.6 ^b
Critical Height (inches)	13.16	13.03	13.59	17.50	17.50	6.82
Critical Volume (liters)	24.39	24.14	25.18	32.43	32.43	12.64
Critical Mass (grams)	759	752	784	858	858	1380

NITRATE EXPERIMENTS, TAMPED SPHERES

Nominal Reactor Diameter (inches)	12.0	12.0	12.0	12.0	12.0	12.0	12.0	12.0
Reactor V volume (liters)	15.19	15.19	15.19	15.19	15.19	15.19	15.19	15.19
Megawatt Days/Ton	420	420	420	420	420	420	420	420
Grams Plutonium/Ton	-	-	-	-	-	-	-	-
% Plutonium ²⁴⁰ By Weight	-	-	-	-	-	-	-	-
Plutonium Assay (grams/liter)	50.39	50.25	51.80	56.20	56.75 ^f	59.93	60.35 ^f	63.75
Total Nitrate (grams/liter)	138	139	163	207	-	237	-	270
Density (grams/cc.)	1.1318	1.1306	1.1482	1.1682	-	1.1898	-	1.2152
Iron Assay (grams/liter)	0.178	0.199	0.272	0.203	0.286	0.199	0.184	0.204
Acid Normality	1.57	1.61	2.20	2.49	-	3.20	-	3.62
Water Assay (grams/liter)	922.8	941.3	920.0	899.2	-	879.8	-	860.9
Total Hydrogen (grams/liter)	106.0	106.0	105.3	103.2	-	102.3	-	101.0
Temperature (°F)	80.5	81.0	80.5	80.5	80.5	80.5	80.5	80.5
Critical Mass (grams)	763	761	786	854	860	911	911	973

f. The sample represented by data in this column was taken from the same fuel as the sample represented by the immediately preceding column.

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NITRATE EXPERIMENTS, TAMPED SPHERES (CONT'D.)

Nominal Reactor Diameter (inches)	12.0	13.0	13.0	13.0	13.0	13.0	13.0	13.0	13.0	13.0
Reactor Volume (liters)	15.19	18.94	18.94	18.94	18.94	18.94	18.94	18.94	18.94	18.94
Megawatt Days/Ton	420	212	212	212	212	212	212	212	212	212
Grams Plutonium/Ton	-	222	222	222	222	222	222	222	222	222
% Plutonium ²⁴⁰ By Weight	-	1.76	1.76	1.76	1.76	1.76	1.76	1.76	1.76	1.76
Plutonium Assays (grams/liters)	77.22	34.06	33.17	34.59	34.81	35.65 ^b	38.11	37.99	38.83	40.69
Total Nitrate (grams/liter)	359	87.1	86.2	117.0	116.0	145.0	128	132	156	205
Density (grams/cc.)	1.2695	1.0858	1.0848	1.1012	1.1006	1.1161	1.1107	1.1106	1.1279	1.1547
Iron Assay (grams/liter)	0.237	0.123	0.124	0.113	0.121	0.124	0.110	0.095	0.141	0.248
Acid Normality	4.72	1.04	1.02	1.44	1.46	1.88	1.67	1.64	2.01	2.90
Water Assay (grams/liter)	822.7	955.6	962.4	939.8	940.1	929.2	-	-	926.7	906.2
Total Hydrogen (grams/liter)	97.3	108.2	108.2	107.0	107.0	105.9	106.6	106.4	105.5	103.7
Temperature (°F)	80.5	80.5	80.5	80.5	80.5	80.5	81.0	80.5	77.0	80.5
Critical Mass (grams)	1176	640	631	656	654	670	712	716	727	773

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NITRATE EXPERIMENTS, TAMPED SPHERES (CONT'D.)

Nominal Reactor Diameter (inches)	13.0	13.0	13.0	13.0 ^g	13.0 ^g	14.0	14.0	14.0	14.0	14.0
Reactor Volume (liters)	18.94	18.94	18.94	18.04	18.04	23.64	23.64	23.64	23.64	23.64
Megawatt Days/Ton	420	420	420	420	420	55	55	55	55	55
Grams Plutonium/Ton	-	-	-	-	-	56.8	56.8	56.8	56.8	56.8
% Plutonium 240 By Weight	-	-	-	-	-	0.54	0.54	0.54	0.54	0.54
Plutonium Assays (grams/liter)	41.10	44.64	44.12	36.27	37.11	26.33	26.69	26.77	26.23	27.91
Total Nitrate (grams/liter)	205	269	270	93.1	125	77.3	78.3	107	107	138
Density (grams/cc.)	1.1548	1.1905	1.1910	1.0910	1.1106	1.0692	1.0695	1.0869	1.0858	1.1030
Iron Assay (grams/liter)	0.266	0.269	0.262	0.114	0.128	0.143	0.146	0.147	0.150	0.160
Acid Normality	2.90	3.71	3.63	1.15	1.66	0.99	0.97	1.42	1.41	1.85
Water Assay (grams/liter)	908.6	865.2	868.3	955.1	942.5	967.6	967.2	955.5	954.9	936.6
Total Hydrogen (grams/liter)	103.7	101.1	101.0	108.0	106.8	108.6	108.6	107.5	107.5	106.3
Temperature (°F)	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5
Critical Mass	777	846	834	654	669	625	627	631	622	648

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g. Aluminum sphere.

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NITRATE EXPERIMENTS, TAMPED SPHERES (CONT'D.)

Nominal Reactor Diameter (inches)	14.0	14.0	14.0	14.0	14.0	14.0	14.0	14.0	14.0
Reactor Volume (liters)	23.64	23.64	23.64	23.64	23.64	23.64	23.64	23.64	23.64
Megawatt Days/Ton	55	55	55	212	212	420	420	420	420
Grams Plutonium/Ton	56.8	56.8	56.8	222	222	389	-	-	389
% Plutonium 240 By Weight	0.54	0.54	0.54	1.76	1.76	3.12	-	-	3.12
Plutonium Assays (grams/liter)	27.39	28.63	28.50	27.95	27.92	28.78	30.15	29.61	29.74
Total Nitrate (grams/liter)	137	188	187	109	110	87.4	109	110	115
Density (grams/cc.)	1.1040	1.1321	1.1300	1.0885	1.0885	1.0769	1.0918	1.0914	1.0931
Iron Assay (grams/liter)	0.152	0.165	0.163	0.092	0.096	0.110	0.129	0.127	0.113
Acid Normality	1.90	2.72	2.70	1.44	1.42	1.02	1.39	1.40	1.51
Water Assay (grams/liter)	945.0	915.1	914.8	939.4	938.6	957.7	951.8	948.2	941.4
Total Hydrogen (grams/liter)	106.4	104.5	104.5	107.4	107.3	108.1	107.3	107.3	107.1
Temperature (°F)	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5
Critical Mass	647	666	671	652	661	681	708	699	701

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REF ID: A62625

NITRATE EXPERIMENTS, TAMPED SPHERES (CONT'D.)

Nominal Reactor Diameter (inches)	14.0	1.40	14.0	14.0	14.0	14.0	14.0	14.0	14.0
Reactor Volume (liters)	23.64	23.64	23.64	23.64	23.64	23.64	23.64	23.64	23.64
Megawatt Days/Ton	420	420	420	420	572	572	572	572	572
Grams Plutonium/Ton	389	389	389	389	429	429	429	429	429
% Plutonium 240 By Weight	3.12	3.12	3.12	3.12	4.05	4.05	4.05	4.05	4.05
Plutonium Assay (grams/liter)	31.79	35.82	35.53	39.62	29.82	29.94	30.79	30.75 ^f	31.72
Total Nitrate (grams/liter)	208	308	311	408	-	87.49	-	119.3	146.8
Density (grams/cc.)	1.1460	1.1993	1.2001	1.2508	-	1.0777	-	1.0967	1.1117
Iron Assay (grams/liter)	0.120	0.144	0.146	0.147	0.104	0.128	0.083	0.074	0.086
Acid Normality	2.90	4.39	4.32	5.75	-	1.01	-	1.44	1.88
Water Assay (grams/liter)	898.6	844.7	852.4	799.7	-	959.8	-	940.3	-
Total Hydrogen (grams/liter)	103.6	99.7	99.5	95.7	-	108.1	-	106.8	105.8
Temperature (°F)	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5
Critical Mass (grams)	752	841	840	935	706	706	727	726	748

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NITRATE EXPERIMENTS, TAMPED SPHERES (CONT'D.)

Nominal Reactor Diameter (inches)	14.0	14.0	14.0	14.0	14.0	14.0	14.0	14.0	15.0
Reactor Volume (liters)	23.64	23.64	23.64	23.64	23.64	23.64	23.64	23.64	28.99
Megawatt Days/Ton	572	572	572	572	572	572	605	605	420
Grams Plutonium/Ton	429	429	429	429	429	429	480	480	-
% Plutonium 240 By Weight	4.05	4.05	4.05	4.05	4.05	4.05	4.40	4.40	-
Plutonium Assay (grams/liter)	33.81 ^f	36.38	37.84	38.72	41.12	41.19 ^f	31.04	30.79	32.41
Total Nitrate (grams/liter)	-	272.4	334 ^b	335.2	384.9	-	127	126	158
Density (grams/cc.)	-	1.1826	-	1.2172	1.2405	-	1.1021	1.1016	1.0899
Iron Assay (grams/liter)	0.100	0.104	0.115	0.126	0.126	0.126	0.110	0.105	0.102
Acid Normality	-	3.69	-	5.46	5.40	-	1.71	1.63	2.22
Water Assay (grams/liter)	-	865.7	-	833.4	811.9	-	930.8	944.0	926.3
Total Hydrogen (grams/liter)	-	100.9	-	-	96.6	-	106.7	106.7	105.6
Temperature (°F)	-	80.5	-	80.5	80.5	-	80.5	80.5	80.5
Critical Mass (grams)	795	857	892	915	971	973	727	729	758

NITRATE EXPERIMENTS, TAMPED SPHERES (CONT'D.)

Nominal Reactor Diameter (inches)	15.0	15.0	15.0	11.0 ^h
Reactor Volume (liters)	28.99	28.99	28.99	11.37
Megawatt Days/Ton	420	420	420	420
Grams Plutonium/Ton	-	-	-	-
% Plutonium 240 By Weight	-	-	-	-
Plutonium Assays (grams/liter)	25.10	25.83	27.05	135.8
Total Nitrate (grams/liter)	116	147	212	229
Density (grams/cc.)	1.0899	1.1068	1.1409	1.2733
Iron Assay (grams/liter)	0.092	0.094	0.096	0.464
Acid Normality	1.60	2.08	3.07	1.81
Water Assay (grams/liter)	946.5	927.1	900.7	901.7
Total Hydrogen (grams/liter)	107.2	106.0	103.5	101.3
Temperature (°F)	80.5	80.5	80.5	80.5
Critical Mass (grams)	722	746	787	-

h. This reactor was subcritical by an unknown amount.

PHOSPHATE AND BISMUTH EXPERIMENTS

Reactor: 14 inch nominal diameter (23.64 liters), water tamped stainless steel sphere.

Fuel: 420 MWDT/T.

Plutonium Assay (grams/liter)	30.15	29.61	31.21	31.05 ^f	32.22	33.46	33.36 ^f	35.75	37.37	32.14
Total Nitrate (grams/liter)	109	110	153	154	187	220	227	315	360	114.8
Density (grams/cc.)	1.0918	1.0914	1.1698	1.1753	1.2245	1.2854	1.2957	1.4103	1.4795	1.0999
Iron Assay (grams/liter)	0.129	0.127	0.139	0.159	0.141	0.142	0.160	0.169	0.230	0.130
Acid Normality	1.39	1.40	1.36	1.42	1.54	2.37	2.35	2.2 ^b	2.3 ^b	1.85 ^b
Water Assay (grams/liter)	951.8	948.2	932.1	924.7	899.7	894.6	894.4	856.7 ⁱ	838.9 ⁱ	925.7
Total Hydrogen (grams/liter)	107.8	107.7	105.9	106.4	104.5	103.9	104.3	98.1	96.2	104.6
Temperature (°F)	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5
Bismuth (grams/liter)	0.0	0.0	49.8	49.8	83.65	121.6	121.6	196.7	241.0	0.0
Phosphate (grams/liter)	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Manganese (grams/liter)	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	2.66
Critical Mass (grams)	708	699	739	735	763	792	790	846	880	760

i. The results on these two water analyses are questionable due to interference of bismuth in the samples.

PHOSPHATE AND BISMUTH EXPERIMENTS (CONTD.)

Plutonium Assay (grams/liter)	32.67	32.95	.35.85	35.62
Total Nitrate (grams/liter)	112.4	117.3	128.6	113.2
Density (grams/cc.)	1.1339	1.1654	1.2268	1.2603
Iron Assay (grams/liter)	0.149	0.178	--	--
Acid Normality	1.81 ^b	1.89 ^b	2.07 ^b	1.83 ^b
Water Assay (grams/liter)	899.1	861.4	804.3	764.4
Total Hydrogen (grams/liter)	103.3	100.5	99.19	95.57
Temperature (°F)	80.5	80.5	80.5	80.5
Bismuth (grams/liter)	0.0	0.0	0.0	0.0
Phosphate (grams/liter)	50.61	92.92	244.4	280.0
Manganese (grams/liter)	3.0	2.02	0.59	0.303
Critical Mass (grams)	775	781	844	845

LITHIUM EXPERIMENT

Reactor: 14 inch nominal diameter (23.64 liter volume), water tamped stainless steel sphere.

Fuel: 420 MWD/T, 358 grams plutonium/ton, 3.43% Pu 240 by weight.

Plutonium Assay (grams/liter)	29.60	29.75 ^f	29.55 ^f	31.19	31.42 ^f	31.66	30.84 ^f	32.82	33.92	32.40 ^f
Total Nitrate (grams/liter)	111	111	112	118	117.6	119	-	122	125	-
Density (grams/cc.)	1.0925	1.0923	1.0916	1.0968	1.0966	1.0984	-	1.1008	1.1030	-
Iron Assay (grams/liter)	0.142	0.129	0.148	0.157	0.187	0.151	0.165	0.147	0.154	0.165
Acid Normality	1.51	1.46	1.46	1.49	1.50	1.54	-	1.53	1.54	-
Water Assay (grams/liter)	948.3	944.7	947.4	946.1	943.5	944.2	-	943.9	943.8	-
Lithium Assay (grams/liter)	0.0	0.0	0.0	0.206	0.206	0.235	-	0.362	0.441	-
Total Hydrogen (grams/liter)	107.3	107.3	107.2	107.0	-	107.0	-	106.8	106.6	-
Temperature (°F)	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5	80.5
Critical Mass (grams)	695	698	699	736	742	749	730	775	799	763

LITHIUM EXPERIMENT (CONT'D.)

Plutonium Assay (grams/liter)	34.70	34.39 ^f	39.94	41.34	40.99 ^f	42.98	43.31 ^f
Total Nitrate (grams/liter)	131	-	147	206	-	210	-
Density (grams/cc.)	1.1086	-	1.1243	1.1566	-	1.1599	-
Iron Assay (grams/liter)	0.168	0.161	0.162	0.192	0.259	0.188	0.171
Acid Normality	1.38	-	1.84	2.77	-	2.75	-
Water Assay (grams/liter)	927.4	-	921.5	900.7	-	895.2	-
Lithium Assay (grams/liter)	0.661	-	1.31 ^b	1.17	-	1.34	-
Total Hydrogen (grams/liter)	106.4	-	105.7	103.5	-	103.3	-
Temperature (°F)	80.5	80.5	80.5	80.5	80.5	80.5	80.5
Critical Mass (grams)	820	813	944	973	964	1012	1020

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NITRATE EXPERIMENTS, UNTAMPED STAINLESS STEEL SPHERES

Nominal Reactor Diameter (inches)	18	18	18	18	18	18	16	16	16	16	16
Reactor Volume (liters)	49.0	49.0	49.0	49.0	49.0	49.0	34.15	34.15	34.15	34.15	34.15
Megawatt Days/Ton	580	580	580	580	580	580	573	573	573	573	573
Plutonium Assay (grams/liter)	22.35	23.40	23.14	23.85	25.18	27.48	34.80	36.22	38.31	38.15	43.20
Total Nitrate (grams/liter)	76.12	97.77	107.78	148.41	199.16	285.75	104.4	109.4	163.0	180.0	281.8
Density (grams/cc.)	1.0662	1.0788	1.0860	1.1020	1.1472	1.1816	1.0961	1.0991	1.1374	1.1396	1.1958
Iron Assay (grams/liter)	0.120	0.126	0.135	0.123	0.137	0.141	0.104	0.120	0.124	0.178	0.179
Acid Normality	0.935	1.431	1.560	2.27	3.49	4.63	11.12	1.31	2.79	2.58	4.18
Total Hydrogen (grams/liter)	108.70	108.0	107.6	106.1	104.6	101.1	107.3	107.2	106.9	104.7	100.9
Temperature (°F)	72	71	68	70	73	69	73	75	75	73	72
Critical Mass (grams)	1095	1140	1132	1167	1235	1347	1194	1237	1302	1303	1483

j. 18 inch reactor was covered with 0.020 inch cadmium.

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LITHIUM EXPERIMENT

Reactor: 18 inch nominal diameter, (49.0 liters volume), untamped stainless steel sphere
(covered with 0.020 inch cadmium).

Fuel: 577 MWDT/T

Plutonium Assay (grams/liter)	23.94	25.43	25.47 ^f	25.98	27.00 ^f	28.28	28.54 ^f	29.83	30.43 ^f	29.06 ^f
Total Nitrate (grams/liter)	125.6	123.3	116.5	132.6	103.0	132.7	132.2	141.4	134.2	131.8
Density (grams/cc.)	1.0894	1.0917	1.0914	1.0964	1.0985	1.1001	1.1006	1.1043	1.1059	1.1050
Iron Assay ^k (grams/liter)	0.0389	0.0407	0.165	0.0361	0.154	0.0392	0.133	0.0435	0.092	0.208
Acid Normality	1.66	1.64	1.45	1.74	1.74	1.64	1.37	1.69	1.49	1.47
Lithium Assay (grams/liter)	0.0	0.223	0.223	0.528	0.528	0.680	0.680	0.88	0.88	0.88
Total Hydrogen (grams/liter)	106.7	106.8	107.0	106.4	108.0	106.3	106.1	105.9	106.1	106.2
Temperature (°F)	75	77	77	73	73	78	78	81	81	81
Critical Mass (grams)	1173	1242	1244	1270	1320	1386	1399	1459	1488	1421

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k. Where duplicate samples occur, the high iron assay is more reliable.

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SULFATE EXPERIMENT

Reactor: 18 inch nominal diameter (49.0 liters volume), untamped stainless steel sphere.

Fuel: 572 MWD/T

Plutonium Assay (grams/liter)	23.87	24.94 ^f	24.76	25.20	24.48 ^f	26.07	25.82 ^f	26.88 ^b
Total Nitrate (grams/liter)	84.3		80.2	84.38	87.3	82.54	89.9	93.36
Density (grams/cc.)	1.0730	1.0725	1.1247	1.1716	1.1725	1.2242	1.2252	1.2567
Iron Assay (grams/liter)	0.099	0.082	0.117	0.136	0.181	0.103	0.121	0.110
Hydrogen Ion (grams/liter)	1.24	1.02	2.79	4.05	4.40	6.05	7.28	7.09
Sulfate Assay (grams/liter)	0.0	0.0	76.3	149.0	-	231.0	-	279.0
Total Hydrogen (grams/liter)	109	108.6	108.0	105.7	105.9	104.3	104.7	102.2
Temperature (°F)	72.5	-	73.0	76.0	-	73.0	-	74.5
Critical Mass (grams)	1166	1227	1219	1231	1206	1282	1270	1317

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FUEL IMPURITIES

By Spectro Chemical Analysis in Parts Per Million

Element	10" Cylinder Experiment	11" Cylinder Experiment	13" Sphere Experiment	18" Sphere Li Experiment
Ag	50	10	L1	--
Al	20	100	L10	50
As	L50	L50	L50	--
B	--	10	5	--
Ba	L20	L200	L20	L30
Be	L0.02	L2	L0.02	L0.1
Bi	500	500	500	--
Ca	1000	200	20	200
Cd	L20	200	10	80
Ce	L200	--	L200	L500
Co	20	L20	L20	--
Cr	1000	1000	2000	30
Cu	10	100	20	--
Fe	G1000	--	2000	--
Hg	L200	--	L200	L1000
In	5	5	L2	L130
K	500	1000	100	--
La	G2000	--	G2000	5000
Li	L1	L5	L2	L5
Mg	50	20	200	30
Mn	200	200	200	50
Na	500	1000	50	500
Ni	1000	400	1000	500
P	1000	G1000	100	--
Pb	50	500	20	L200
Sb	100	500	L50	--
Si	50	50	L50	--
Sn	5	50	5	--
Sr	L4	--	L4	L8
V	L200	L200	L200	--
Zn	L100	200	L100	L500
Tl	L100	--	L100	L300

L Less than

G More than

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