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# IDAHO CHEMICAL PROCESSING PLANT

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CHEMICAL TECHNOLOGY DIVISION

PROCESS DESIGN SECTION

# PRELIMINARY CRITICALITY STUDY IDAHO CHEMICAL PROCESSING PLANT

R. J. Klotzbach

DATE ISSUED

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## TABLE OF CONTENTS


| | | Page |
|---|---|---|
| 0.0 | Abstract | 4 |
| 1.0 | Introduction | 5 |
| 2.0 | Summary | 6 |
| 3.0 | "Always Safe" Tanks | 7 |
| 4.0 | "Provisionally Safe" Equipment | 9 |
| 5.0 | Interaction | 15 |
| 6.0 | Preliminary Equipment Layout | 16 |
| | Bibliography | 18 |
| | Appendix | 19 |

## 0.0 ABSTRACT

The processing of kilogram quantities of $U^{235}$ in the Idaho Chemical Processing Plant necessitates the use of "limit control" for batch operations and "always safe" vessels for continuous column operations to prevent a chain reaction from starting in the process equipment.

## 1.0 Introduction

The Idaho Chemical Processing plant will be designed to process an average of four kilograms of $U^{235}$ per day as 93.4% enriched material, through one line of processing equipment as described in ORNL-779 by H. E. Goeller. Using this report as a basis, a preliminary study of the effects of criticality limitations on processing equipment design and arrangement has been made.

Economically and functionally it is not feasible to design the $U^{235}$ recovery process in its entirety as an "always safe" system because certain phases of the process such as feed solution and preparation and product handling are impossible in vessels of the small size required by always safe conditions. A more reasonable approach is to design the continuous components of the process always safe and the batch components "provisionally safe", maintaining an adequate safety factor and rigorous limit control. The provisionally safe components are justified by the good material balance possible in the batch operations, precise analytical methods, the immobility and remote operations of the equipment in the processing area, and a design safety factor to absorb at least three major human errors. The only inherent disadvantage of a provisionally safe system is that it cannot preclude the possibility of deliberate sabotage. This can be controlled by adequate security measures without additional operating costs and consequently is not a major objection to the system.

In this preliminary survey general design criteria are formulated from available empirical data; however, a detailed criticality study must be made for each equipment piece in the final process layout.

## 2.0 Summary

To prevent a chain reaction from occurring while processing kilogram quantities of $U^{235}$, the following general conditions of criticality control have been established on the basis of uranium concentration, vessel geometry and interaction between uranium containing vessels:

1. All tanks in the continuous component of the process will be "always safe", i.e., with a maximum diameter of 5 inch IPS.
2. Makeup tanks in the batch component of the process and laboratory finishing operations equipment will be mass limited "provisionally safe". The maximum $U^{235}$ charge to each shall not exceed 500 gms.
3. The EBR dissolver, an always safe vessel, will be charged with 1500 grams of $U^{235}$, of which 990 grams will be dissolved. Subsequent charges of 990 grams of $U^{235}$ metal will be made.
4. Tanks at least once removed from the continuous process stream will be buildup mass limited "provisionally safe" and can have standard dimensions.
5. Interaction among "always safe" cylinders will be controlled by limiting the maximum solid angle, $\Omega$, subtended at any cylinder by any array of other cylinders, to one steradian. In addition, no two parallel cylinders will be placed closer than two feet, edge to edge.
6. Provisions must be made so that in the event of a major equipment leak the cell floor area will not be covered with solution of a depth greater than $1\frac{1}{2}$ inches.
7. There will be no direct connection between 25 Processing Areas and waste or collection drains.

Summary (continued)

8. The size of each batch in the laboratory finishing operations shall not exceed 500 grams of $U^{235}$.

3.0 "Always Safe" Tanks

"Always safe" cylindrical tanks are cylinders with a diameter equal to or less than the limiting diameter which would be sub-critical at infinite length. For a given uranium enrichment, the critical mass of a cylindrical configuration filled with uranium-water solution varies with the hydrogen to uranium ratio. There is for each type geometry an optimum water concentration at which the mass of $U^{235}$ is a minimum; this concentration does not correspond to the minimum volume in which some mass of $U^{235}$ can make the tank critical. Selecting this optimum concentration (moderation), the minimum critical mass in water reflected aluminum reactors is plotted against reactor diameter in Fig. 1 and shows that the limiting or always safe diameter is five and one-half inches. Empirical data[1] indicate that the maximum diameter for stainless steel always safe cylinders is six inches with an infinite water reflector and eight inches without the reflector. The thickness of a slab of infinite size which would always be sub-critical appears to be at most five cm. thick.

The Idaho chemical process always safe tanks will be cylindrical because of the severe limitations imposed by a slab geometry on sampling, mixing, and instrumentation services. The maximum diameter of the always safe cylinders will be five inches IPS, which eliminates the possibility of a chain reaction occurring as a result of accidental or natural flooding of the process cells and allows for a practical degree of interaction between unreflected cylinders. The following tanks will be always safe:

"Always Safe" Tanks (cont.)

1. EBR feed adjustment tank.
2. EBR condenser.
3. EBR feed storage tank.
4. EBR condensate catch tank.
5. MTR and NP feed filters.
6. MTR and NP feed storage tanks.
7. 1st, 2nd, 3rd cycle extraction columns.
8. 1st, 2nd, 3rd cycle strip columns.
9. 1st, 2nd, 3rd cycle aqueous raffinate collection tanks.
10. 1st, 2nd, 3rd cycle solvent raffinate collection tanks.
11. 1st, 2nd, 3rd cycle product evaporators.
12. 1st, 2nd, 3rd cycle column feed, sample and control pots.
13. All condensate separators.
14. Aqueous raffinate recycle storage tanks.
15. Product storage tanks.
16. All cycle product evaporation condensate re-evaporators.
17. MTR and NP acid filter wash catch tank.
18. MTR and NP water filter wash catch tank.
19. All cycle product evaporation concentrate recycle storage.

The processing area floor must be designed as an always safe slab, i.e., any equipment leak should not cover the floor with solution of a depth greater than one and one-half inches. There must be no direct connections between floors and drains to prevent a critical accumulation in the drain lines or waste tanks; this provision is also necessitated by source and fissionable material accountability.

## 4.0 "Provisionally Safe" Equipment

"Provisionally safe" equipment does not have an always safe configuration but is safe because of operational control factors with an allowance for human error. Provisionally safe equipment in the Idaho Chemical Processing Plant falls into two classifications: (1) batch mass limited and (2) buildup mass limited.

### 4.1 Batch Mass Limited

#### 4.11 MTR and NP Makeup Tanks

Batch mass limited safe equipment is a component of the batch type feed preparation operations where the amount of $U^{235}$ charged to the process can be physically limited to a charge safely below that required for a critical accumulation. Limit control is a commonly accepted operational procedure at the K-25 and Y-12 operations in Oak Ridge. At both of these plants the batch size is limited to 500 gms of $U^{235}$ and because 880 grams of $U^{235}$ is critical at optimum moderation in an optimum cylindrical configuration (Fig. 1) any accumulation greater than 500 gms is considered as violating a reasonable safety factor. This limit is highly justifiable for processes in which there is manual handling of uranium bearing solutions in portable laboratory equipment. To maintain an adequate safety factor which operational experience has proven, the MTR and NP makeup tank batches will be limited to 500 gms for normal operation. However, because the equipment is fixed and remotely operated with no manual handling, it is probable that in case of national emergency the batch limit could be safely raised to increase plant through-put without additional equipment.

MTR and NP Makeup Tanks (cont.)

The MTR and NP dissolvers are vertical, cylindrical, stainless steel tanks with a 30 inch or greater diameter. Minimum critical mass for optimum concentration is plotted against reactor diameter in Figure 1. The empirical data are available for reactors up to 16 inches in diameter. A linear extrapolation[2] of the plot to reactors of greater diameter shows that the minimum critical mass for a homogeneous mixture at optimum moderation in a 30 inch diameter reactor is approximately 2.8 kg. Therefore it would be possible to increase the batch size to 800 gms maintaining a safety factor greater than three for a 30 inch or greater diameter dissolver. An 800 gm batch is less than the minimum critical mass of 880 gms which went critical at optimum concentration and optimum geometry. This eliminates the highly improbable possibility of 880 gms settling in a 30 inch reactor so that it passes through the optimum moderation range and has a spatial distribution exactly like the ten inch diameter reactor which went critical (Fig. 1).

Although a 500 gm batch limit will be observed, the MTR and NP makeup tanks will be designed large enough to accomodate an 800 gm batch. Normally the fuel charge for an MTR dissolving will be four assemblies, after 12% burn-up containing 490 gms of $U^{235}$. The NP fuel charge will be 15 slugs after 15% burn up containing 490 gms of $U^{235}$. In time of national emergency it might be possible

MTR and NP Makeup Tanks (cont.)

to increase the dissolving capacity to process six MTR assemblies containing 735 gms of $U^{235}$ or 24 NP slugs containing 796 gms of $U^{235}$.

The dissolver feed is filtered and adjusted before passing to the always safe column feed tanks. The following tanks, intermediate between the dissolver and column feed tanks, which will never contain more than one MTR or NP dissolving charge, will have the same physical dimensions as the dissolver:

(1) MTR and NP coarse feed adjustment tanks.

(2) MTR and NP filtrate catch tanks.

(3) MTR and NP fine feed adjustment tanks.

4.12 EBR Dissolver

This dissolver, will have an always safe geometry, i.e., a diameter equal to or less than 5 inches. The flowsheet shows a charge of 1.5 kg of 25 metal to the dissolver, of which 990 grams will be dissolved in each batch. The empirical criticality data apply to moderated assemblies with a maximum density of 4.8 $gm/cm^3$(3). The problem involved in the EBR dissolver is to determine whether a charge of 1.5 kg of pure metal (no moderation) with a density of 19 $gm/cm^3$, completely tamped with water(dissolving solution) is safe.

Linear extrapolation of experimental data(3) indicates that the critical mass for unmoderated assemblies with a maximum density

EBR Dissolver (cont.)

of 4.8 gm/cm$^3$ is at least 125 kg and may be as high as 160 kg. Theoretically, we would expect that the critical mass should vary approximately inversely with the density squared[4]. With actual assemblies it would be expected that the greatest deviation from this inverse square rule would occur with small unmoderated assemblies that are greatly affected by the surrounding reflector. In this case it would be expected that the numerical value of the negative exponent of density sould be somewhat less than two. Empirical data (4) show that critical mass for low moderation varies as the density$^{-1.7}$.

Applying the inverse square rule and using the empirical data available, the critical mass for unmoderated reflected metal is indicated to be at least 8 kg and can be as high as 15.5 kg. Therefore, it is possible to charge the EBR always safe dissolver with 1.5 kg of fuel and still maintain a safety factor of greater than five.

4.2 Buildup Mass Limited

Equipment which receives process wastes and condensates will be buildup mass limited provisionally safe and can have standard physical dimensions because of the small quantity of $U^{235}$ contained in these solutions.

4.21 Column Waste Solution Tanks

The aqueous and solvent raffinate streams are collected in always safe tanks which have an eight hour capacity. These solutions are sampled, analyzed, and, according to the analysis, are either recycled to always safe equipment in the case of excessive losses or transferred to waste processing tanks. The analytical accuracy is of the order of 3% for solutions containing 10γ of 25/cc, so that any loss can be easily detected.

In the worst case, when processing 8 kg of EBR fuel per day, the maximum amount of $U^{235}$ lost in any particular waste stream is 0.8 gm/day at a maximum concentration of 0.004 gm/liter. The hydrogen to $U^{235}$ atomic ratio for these solutions is of the order of $10^7$. In the limiting case, the hydrogen to $U^{235}$ atomic ratio above which a $U^{235}$ solution cannot be made critical in any configuration is $\sim 2.3 \times 10^3$ as shown in Fig. 2.

Since it is possible to maintain good control because of the low concentration of the uranium solution (large H: $U^{235}$ ratio) and because the waste tanks are at least once removed from the continuous process streams, the following vessels can have standard physical dimensions:

(1) 1st cycle aqueous holdup tank and evaporator.

(2) All cycle solvent holdup tanks.

(3) 2nd and 3rd cycle aqueous raffinate evaporators.

[redacted]

4.22 Filter Wash Waste Tank

The process "crud" filter is backwashed twice with nitric acid and water solution which is recycled to the next dissolving via always safe tanks. The third wash, a water wash of approximately 20 gallons, is collected in a waste tank which can be provisionally safe. The maximum 25 loss in the wash waste is in the order of 0.05% and in the case of the NP flowsheet would mean a loss of 0.4 gm of 25 in a solution with a concentration of 0.005 gm 25/l ($H:U^{235} \sim 10^7$).

Because of the low concentration of $U^{235}$ possible in the wash, the filter wash waste tank can be provisionally safe with standard dimensions.

4.23 Condensate Catch Tanks

The process solutions are concentrated at various steps in the process by evaporation. The expected maximum $U^{235}$ entrainment loss to any condensate collection tank is approximately 0.2%. In the case of the EBR flowsheet the $U^{235}$ loss will be 16 grams per day in any one evaporation and the $U^{235}$ concentration in the condensate will be 0.07 gm/l ($H:U^{235} > 10^5$). Therefore, because of the low concentration of $U^{235}$, the following tanks can be provisionally safe with standard dimensions:

(1) NP and MTR condensate catch tank.

(2) 1st, 2nd, and 3rd cycle product evaporation condensers.

Condensate Catch Tanks (cont.)

(3) 1st cycle aqueous raffinate condenser.

(4) 2nd and 3rd cycle aqueous raffinate condenser.

(5) Product condensate re-evaporation condensate holdup.

## 5.0 Interaction

The possibility of a particular assembly going critical is enhanced by the proximity of other vessels. Neutrons that leak from the walls of one container enter the other and increase the multiplication. The effect of interaction in terms of critical mass, dimensions, and concentrations has not been completely formulated and a conservative assumption has been made[5] that the neutron interaction depends on the magnitude of the solid angle "seen" from points on the surface of one container to the surface of adjacent containers. In the light of empirical data, limiting the solid angle subtended at any five inch cylinder by all other cylinders of five inch diameter or less to one steradian appears to insure a favorable margin of safety provided that no two cylinders are closer than 2 feet[6]. The restriction that no two parallel 5 inch cylinders be spaced closer than 2 feet takes into account the possibility that the cylinders may be surrounded by reflector on all sides except the mutually adjacent sides.

Applying these restrictions the following separations will be observed for 5 inch cylinders arranged in air:

(1) Two parallel 5 inch diameter cylinders of any length, will be separated by two feet or more.

(2) A system of any number of parallel 5 inch diameter cylinders, of any length, may be placed two feet apart, edge to edge, provided that their axes lie in the same plane.

Interaction (cont.)

(3) Plane arrangements (see 2) of parallel banks of 5 inch cylinders must be separated by at least 10 feet.

In the final plant design each item of equipment will have to be reviewed in the light of the one steradian restriction.

## 6.0 Preliminary Equipment Layout

A preliminary equipment layout has been tentatively approved on a criticality basis for tank sizes, spacing and uranium solution concentrations. Tanks were spaced so that the total solid angle subtended on any one was less than one steradian. In all parts of the process the maximum uranium solution concentrations were assumed.

In the case of the extraction columns and aqueous raffinate catch tanks, it was assumed that scrub and solvent solutions were cut off and the equipment filled with feed solution. For the stripping columns and solvent raffinate catch tanks, it was assumed that the strip solution was cut off and the equipment filled with hexone-uranium solution.

Table I shows the preliminary concept of the process and equipment variables for the major process operations.

[redacted]

Table I

| PROCESS OPERATION | TYPE OPERATION | TANK DIAM in. | TANK CLASS | PROCESS SOLUTION | ATOMIC RATIOS | COMMENT |
|---|---|---|---|---|---|---|
| EBR Feed Makeup | 1000 gm. Batch | 5 | Always safe | 1.5 M UNH 0.2 N acid def. | $H:U^{235} = 64$ | 1500 gm. of metal charged to the dissolver per batch |
| EBR Feed Storage | Continuous | 5 | " | " | " | |
| MTR and NP Feed Makeup | 500 gm. Batch | >30 | Batch Mass Limited | 1.6 M $Al(NO_3)_3$ 0.114 M UNH | $H:U^{235} = 1000$ $Al:U^{235} = 16$ | Solution 0.2 N acid deficient, 0.66 M $NH_4OH$ |
| MTR and NP Feed Storage | Continuous | 5 | Always safe | " | " | " |
| 1 and 2 Cycle Extraction and Aqueous Raff. Stor. | Continuous | Column 4.5 Tanks 5 | Always safe | 1.5 M UNH 0.2 N acid def. | $H:U^{235} = 64$ | Ordinarily there is no $U^{235}$ in Raffinate Storage |
| 3rd Cycle Extraction and Aq. Raff. Storage | Continuous | Column 2 Tanks 5 | Always safe | 2.0 M UNH | $H:U^{235} = 47$ | " |
| 1st Cycle Stripping and Solvent Raff. Storage | Continuous | Column 4.5 Tanks 5 | Always safe | Hexone-UNT sol. 18.6 gm.$U^{235}$/L | $H:U^{235} = 1200$ $C:U^{235} = 588$ | " |
| 2nd Cycle Stripping and Solvent Raff. Storage | Continuous | Column 2 Tanks 5 | Always safe | Hexone-UNT sol. 36.9 gm. $U^{235}$/L | $H:U^{235} = 603$ $C:U^{235} = 293$ | " |
| 3rd Cycle Stripping and Solvent Raff. Storage | Continuous | Column 1.25 Tanks 5 | Always safe | Hexone-UNT sol. 104 gm. $U^{235}$/L | $H:U^{235} = 201$ $C:U^{235} = 100$ | " |
| 1 and 2 Cycle Product Evaporation | Continuous | 5 | Always safe | 1.5 M UNH | $H:U^{235} = 64$ | |
| 3rd Cycle Product Evaporation | Continuous | 5 | Always safe | 2.0 M UNH | $H:U^{235} = 47$ | |
| Condensate Catch Tanks | Continuous | Standard size | Buildup Limited | 0.07 gm. $U^{235}$/L | $H:U^{235} > 10^5$ | |
| Filter Wash Catch Tank | Batch | Standard size | Buildup Limited | 0.005 gm.$U^{235}$/L | $H:U^{235} \sim 10^7$ | |



[redacted]

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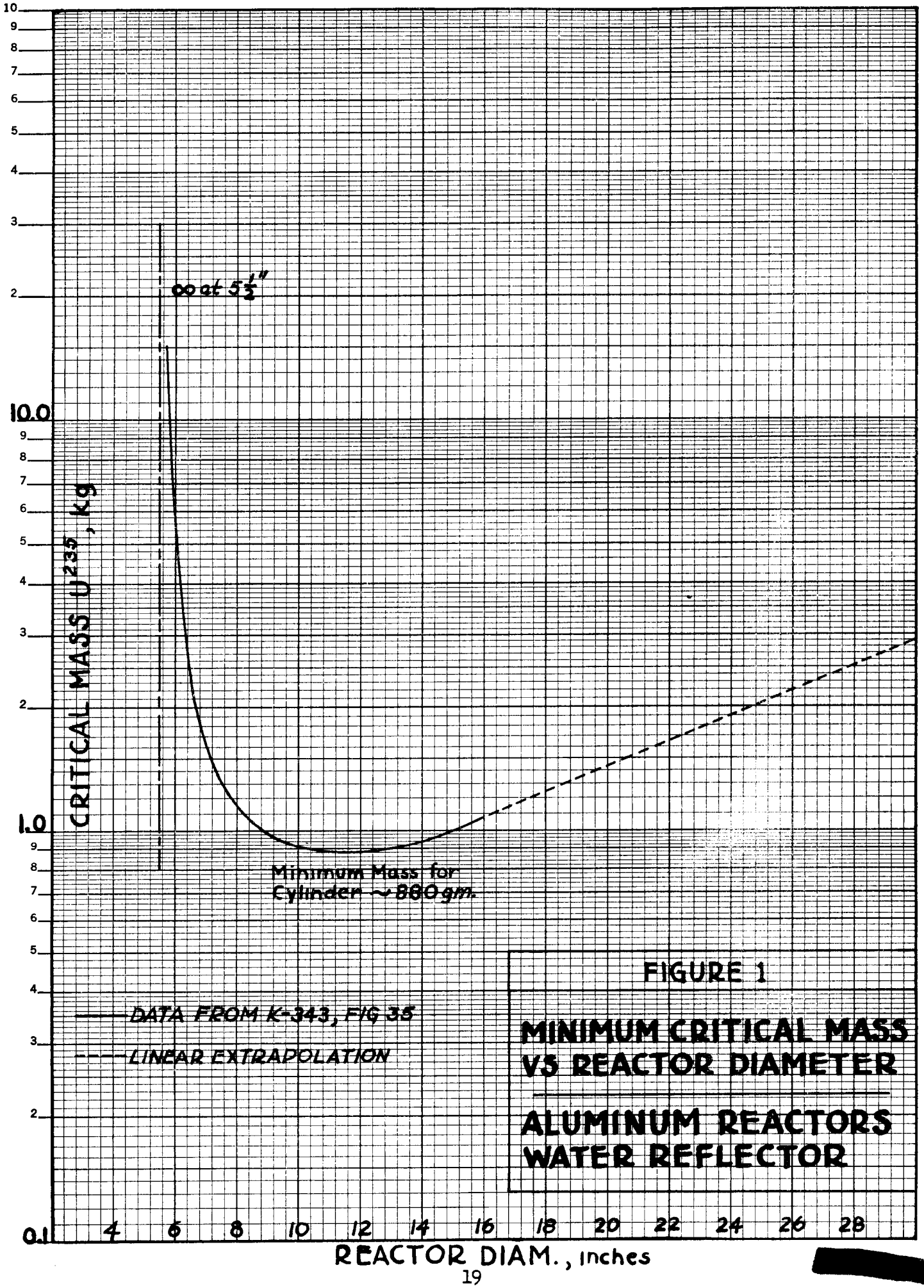


FIGURE 1

MINIMUM CRITICAL MASS VS REACTOR DIAMETER

ALUMINUM REACTORS WATER REFLECTOR

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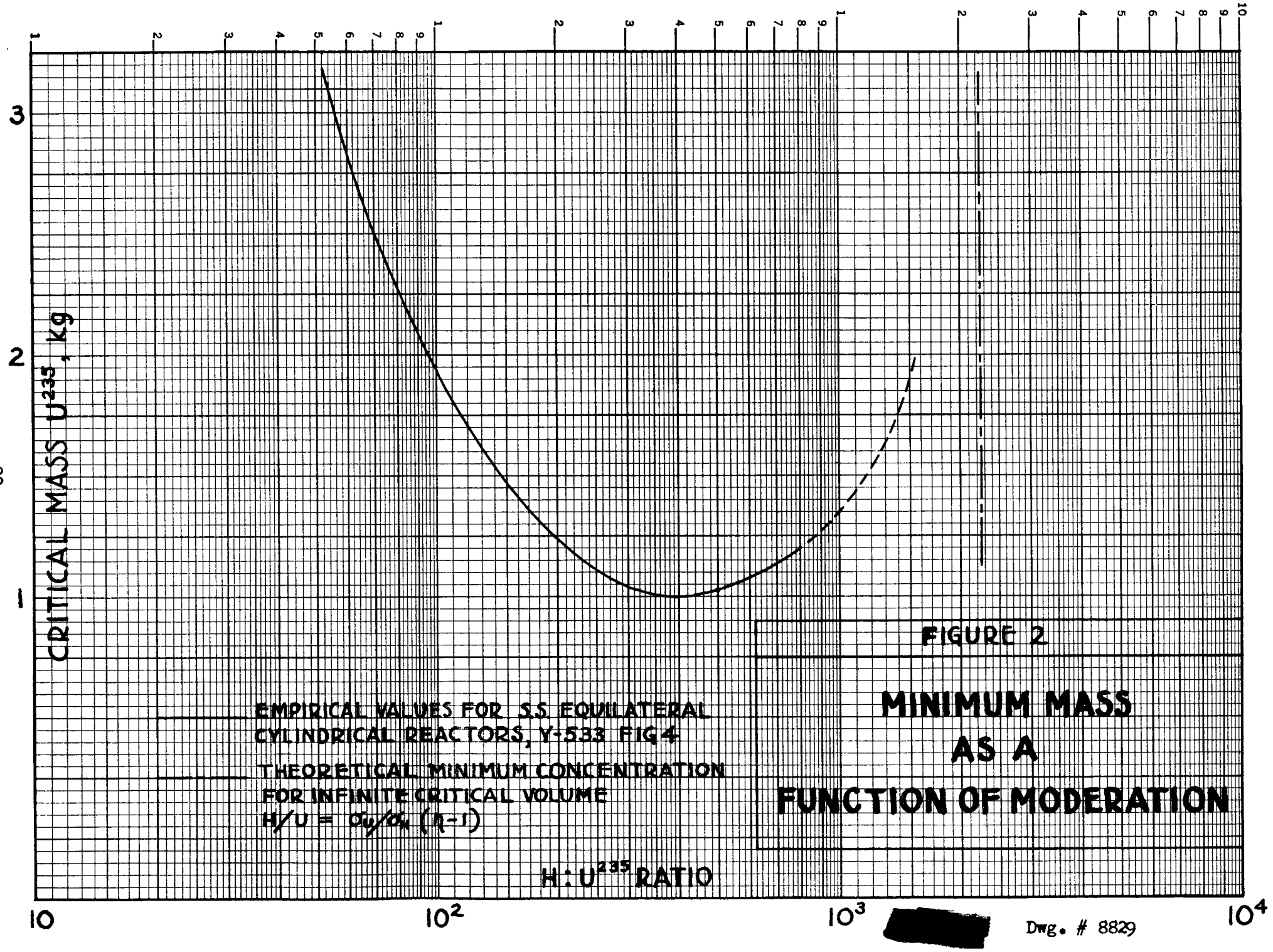


FIGURE 2

**MINIMUM MASS AS A FUNCTION OF MODERATION**