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NUCLEAR SAFETY CONSIDERATIONS IN REACTOR FUELS PROCESSING PLANT DESIGN

by

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June 11, 1956

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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NUCLEAR SAFETY CONSIDERATIONS IN REACTOR FUELS PROCESSING PLANT DESIGN

The unit operations principles that apply to non-radioactive chemical separations apply equally as well to reactor fuels processing plants. The designer and operator of radiochemical plants have another factor with which to contend; that is, the raw material feed is radioactive and fissionable. This presents two new types of problems: 1) protection of personnel from radiation, and 2) prevention of conditions that can lead to self-sustaining chain reactions. This paper will be concerned with the latter; namely, some of the design considerations to ensure that no such criticality condition occurs in the chemical separations plants.

With the critical mass information that exists, problems arising under many specific conditions can be solved realistically. However, it is difficult to write in a single paper of this size a set of nuclear safety rules that apply to all plant processing conditions. For spent reactor fuels processing, where a great variety of techniques and conditions may be represented, overall regulations are likely to be so cumbersome as to be easily misapplied. Blind applications of general rules can work in two directions. Misapplication in one direction can lead to criticality accidents that are dangerous to both life and property. Misapplication in the opposite direction with insufficient attention to extenuating conditions may lead to unnecessary expense in both the construction and operation of large-volume processing plants. Therefore, it is necessary to understand the principles of reactor physics as well as the plant processing conditions involved to insure the proper application of these rules as well as determine criticality conditions for process steps that can not be covered by general rules.

Some of the nuclear safety aspects of the design and operation of reactor fuel separations plants will be presented. It is not within the scope of this paper to cover the field of nuclear safety as it pertains to separations plants. The primary purpose is to introduce those desirous of entering the spent reactor fuel processing business with some of the problems that must be considered in the design and operation of such a plant.

Nuclear safety is accomplished by one of several methods: 1) making the process safe by geometry, 2) establishing batch limits on each process vessel, and 3) setting operating limits on solution composition. The method used depends on the plant capacity desired and on operational feasibility. Within a single plant all three methods may be used, each method being applied for most efficient plant operation of each process step. The application of these methods will be illustrated to show how they apply to the processing of spent reactor fuels. For this purpose, the fuel processing steps will be divided as follows:

1) Storage and cooling of reactor fuels.
2) Dissolution of the fuel elements.
3) Separation of fissionable materials from fission products.
4) Separation of plutonium from uranium.
5) Preparation of plutonium and uranium for reuse.

Storage and Cooling of Reactor Fuels

In the storage and handling of natural uranium fuel elements, both unirradiated and irradiated, there are generally no criticality problems involved. In the handling of enriched fuel elements, however, storage of the elements is generally accomplished by limiting the number of elements per storage container and controlling the spacing between containers. For purposes of illustration, a 1.0% enriched U-235 fuel element will be considered. These fuel elements will be solid cylinders having right circular cross-sectional areas. The following is based on theoretical calculations, that have been verified by experiment,(2),(3) made for unirradiated elements of 1.0% enrichment as described above. A conservative correction was made for the fuel element jacket that encased the experimental fuel elements. Figure 1 is a plot of the minimum critical mass for these fuel elements vs water-to-uranium volume ratio for fuel elements of different diameters. It is seen that for each diameter there is an optimum water-to-uranium ratio for minimum critical mass. This optimum ratio is different for each diameter element. Therefore, if the spacing between elements can not be controlled, a safe number per storage container must be set whose mass is below the minimum critical mass for the specific diameter element under consideration. These curves, being based on unirradiated 1.0% enriched elements should not be used for fuel elements of this enrichment after irradiation. To determine the minimum critical mass of these elements after irradiation, the reactor power level to which these elements were subjected must be considered. In dealing with irradiated fuel elements, the plutonium isotopic content build-up as well as the uranium isotopic content must be considered.

Dissolution of the Fuel Elements

In the dissolving of reactor fuel elements, an additional problem must be considered that does not occur in the safe storage of fuel elements. In the dissolution step, the fuel element diameter is continuously being reduced as dissolution takes place. The effect that this has is illustrated in Figure 2. This figure has also been based on unirradiated fuel elements of 1.0% U-235 enrichment. Figure 2 is a plot of the minimum critical mass as a function of fuel element diameter. Therefore, if the diameter of the original fuel element is greater than that for minimum critical mass, the minimum critical mass for the particular enrichment under consideration must be used in establishing safe batch limits. Figure 2, as well as Figure 1, was based on uranium water systems. If nitric acid is being used as the dissolving agent and a minimum nitric acid acidity can always be guaranteed, greater minimum critical masses would result. The presence of nitric acid is equivalent to adding a neutron poison as well as decreasing the hydrogen concentration of the moderator in the dissolver system. However, if another dissolving agent such as hydrofluoric acid is used, the minimum critical mass may not be much different than that in the water system. This is due to the very small neutron absorption cross section of the fluorine in the hydrofluoric acid compared to the nitrogen in the nitric acid system. As dissolution takes place and some of the uranium goes into solution, the minimum critical mass of the system is increased. This is primarily due to the decrease in resonance escape probability for U-238.

In the dissolution of irradiated natural uranium there are generally no criticality problems involved as was found to be the case in the storage and handling of these fuel elements. Where criticality is not a problem, the pros and cons of continuous dissolvers vs batch-type dissolvers will be the same as for other types of dissolution processes. For the processing of enriched fuel elements, however, there are additional factors in favor of continuous dissolvers. Before a new charge can be made in a batch dissolver, the dissolver must be empty, otherwise it is difficult to determine an allowable batch size without knowing the size of the heel remaining in the dissolver from the previous dissolution. Therefore, it would be necessary to lengthen the batch dissolution time until complete dissolution is attained. This greatly limits plant capacity because the rate of dissolution is a function of both acid and metal concentrations. Higher dissolution rates are attained with larger masses of metal for the same acid concentration. If it is desired to process both natural and enriched uranium simultaneously and in the same dissolver, the batch size for the enriched fuel elements may be reduced by a factor greater than two in the presence of an arbitrary amount of natural uranium. Moreover, mixing enriched uranium with natural uranium may reduce the resale value of the recovered uranium.

For a continuous dissolver processing a 1.0% enriched uranium (based on unirradiated fuel elements), the safe diameter is about 22 inches. This is based on a single water-reflected dissolver for a uranium-water system. Of course, if a minimum nitric acid acidity can be guaranteed at all times, this diameter could be increased. For larger enrichments, smaller diameters are required. Since continuous dissolver capacity is a function of dissolver cross section, which in turn is a function of enrichment, one must consider the range of enrichments to be processed and determine whether it is more economical to process the entire range of enrichments in one dissolver or have several sized dissolvers, each one to process a single enrichment or range of enrichments.

Separation of Fissionable Material from Fission Products

After the dissolution step, the separations plant is concerned with solution handling. The solution now contains fission products, uranium, and plutonium. With this type solution, safety may be insured by composition, batch size, or by geometry. If precipitation can be precluded, the concentration of fissionable material (U-235 plus Pu-239) can be limited by the more reactive constituent, plutonium. In this case, 6 grams per liter is a safe concentration for the total fissionable material. Safety factors inherent in such a system are acid concentration and fission product concentration. If a precipitate can not be precluded, nuclear safety can be established by maintaining a maximum ratio between fissionable material (U-235 plus Pu-239) and U-238. The safe ratio of this system in water is a function of the enrichment of the original fuel elements as well as the pile exposure conditions. Safe diameters for single vessels containing this solution will be greater than that of the dissolver for fuel elements of the same enrichment. It is a function of enrichment and for the case considered above may be as high as 30 inches compared to 22 inches for the safe dissolver. After the fission products have been removed from the fissionable material, the same methods can be used to guarantee plant nuclear safety. Now, however, the safety factor of having fission products (neutron poisons) present no longer exists.

Separation of Plutonium from Uranium

When the separation of plutonium from uranium is attained, safety of the plutonium, due to the presence of U-238, no longer exists. The two streams to be considered are now a plutonium solution and a uranium solution. The nuclear safety of the uranium solution is a function of the U-235 to U-238 ratio. Experimental results on U-235 solutions have already been reported in the unclassified literature.(1) Experimental data with uranium systems of different enrichments have also been performed at the Oak Ridge National Laboratories. The nuclear safety of plutonium solutions will be examined here in more detail. The study will be based on experiments performed at the Hanford Atomic Products Operation.(4) A safe design can be based on the same considerations as above; safety by batch size, composition, and geometry. One other factor will also be considered; namely, the effect of neutron reflectors on criticality. In the previous processing steps, completely water-tamped systems were assumed. Before the separation of fission products, the plant structure and neutron reflectors (vessel walls, structural supports, concrete walls, etc.) are generally so great as to be equivalent to complete water reflection.

Figures will now be presented to illustrate how these variables affect the nuclear safety of separations plants. These curves are based on all the plutonium being present as Pu-239. Figure 3 is a plot of minimum critical mass of plutonium vs cylinder diameter as a function of equivalent reflector (vessel walls, structural supports, framework, concrete walls, presence of personnel, etc.) thicknesses. These calculated values for minimum critical mass are based on plutonium-water systems. As stated above, if the minimum acidity could be guaranteed, the critical parameters would not be so restrictive. However, in a number of process steps neither a minium acidity nor no precipitation can be guaranteed. It is seen that reflector thickness is more important for small diameter vessels than for those of larger diameters.

Figure 4 gives the estimated minimum critical mass for effectively water-tamped cylinders of smaller diameters. The curve is not extended to diameters smaller than six inches because of the larger uncertainties in the calculations for vessels of smaller diameter. The very rapid rise in minimum critical mass for vessels of smaller diameter is evident from this curve. The minimum critical mass approaches infinity as the vessel diameter approaches that for a safe diameter for plutonium solutions.

Figure 5 is a plot of safe diameter for plutonium solutions as a function of nitric acid acidity. The curve is broken for acidities below about 1.3 M nitric acid as plutonium nitrate in high concentrations begins to decompose below this acidity. Under these conditions the safe diameter is about 5.4 inches for plutonium concentrations below 500 grams per liter "solution" and may be as low as 5 inches for somewhat higher concentrations. This uncertainty is due to lack of experimental information on plutonium "solutions" of this type.

When plutonium nitrate in solution begins to decompose, what has been referred to as a plutonium polymer, is believed to be formed. This has been assumed to have the composition, $PuO_2 \cdot (H_2O)_{7\text{-}1/2}$. Figure 6 is a plot of plutonium concentration vs nitric acid acidity showing the region in which this polymer is formed.(5) It appears that there is no polymer formation for acidities greater than 1.3 M nitric acid.

Figure 7 is a plot of safe diameter as a function of equivalent reflector thickness. For "bare" vessels (no reflector) the safe diameter approaches 8 inches. For very thick equivalent reflectors, the safe diameter approaches 5.4 inches. The same restrictions on plutonium concentration as described above should be applied to these safe diameters.

Figure 8 is a plot of critical solution height as a function of vessel diameter for effectively water-reflected vessels. From this curve the critical volume as a function of vessel diameter can readily be calculated. The same uncertainties exist, as that described above, for plutonium solutions whose concentration is greater than about 500 grams per liter.

Figure 9 is a plot of critical concentration of plutonium as a function of vessel diameter for water-tamped vessels. Calculations, upon which this curve is based, did not give credit for nitrate ion that may be present. It is therefore conservative for actual solutions of plutonium nitrate in the presence of excess nitric acid.

Application of Nuclear Safety Rules

In applying criticality data to the different steps of a separations plant, the following must be considered. Whether safety is determined by batch limitations, geometry, or by solution concentration, the effect of reflectors must be carefully considered. Where no contact maintenance problems are involved, reflectors may be taken into account more closely in vessel design. However, where contact maintenance may be involved, a more liberal consideration of neutron reflectors is necessary. Allowance must be made for maintenance personnel approaching plutonium solution-bearing vessels as they add more neutron reflection to the vessel the closer they approach the vessel. One very important factor which has not been considered above is the effect of interaction between vessels. In the above discussion, vessels were considered as isolated vessels in which there are no vessel inlets into the side. Side inlets have the effect of increasing the effective vessel diameter in the same way that interaction with other vessels does. In actual practice, it is generally not feasible to build a plant in which there is no interaction between vessels. This is too demanding on space requirements. Among the factors to be considered for interaction between vessels are vessel sizes, separation between vessels, neutron reflectors between vessels, and solution composition. This subject is beyond the scope of this paper. However, the following generalities will be made: 1) For a given vessel diameter interaction varies inversely as a function of separation; 2) For a given vessel diameter and given spacing interaction varies inversely as a function of the amount of neutron reflectors between vessels; and 3) For a given spacing, interaction varies directly as a function of diameter.

In operating process steps by batch limitations, the following should be considered: accuracy of analyses, efficiency of process control, and the possibility of double batching. By double batching is meant the possibility of making a single operating error and adding a solution batch to a vessel already containing one. Where double batching is a possibility, the batch size should be less than half the estimated minimum critical mass after the above operating and analytical uncertainties are considered. In the control of process steps by concentration, where precipitation can be precluded, 75% of the critical concentration may be used as a safe concentration.

For safety by geometry, the data presented above for infinitely tall vessels may be considered safe for plutonium concentrations less than 500 grams per liter. For aqueous solutions of somewhat higher concentration, the minimum safe diameter may be about 10% less.

Summary

Process variables that affect nuclear safety in the design and operation of aqueous separations plants of spent reactor fuels have been presented. Calculations based on experimental and extrapolated experimental data have been made and presented in curves to indicate how criticality is affected by these variables. Examples of how these are applied to plant processes have been given as well as a few general rules in their application. The importance of understanding the process variables, as well as the reactor physics principles involved in applying either experimental data or theoretical calculations, can not be overemphasized.

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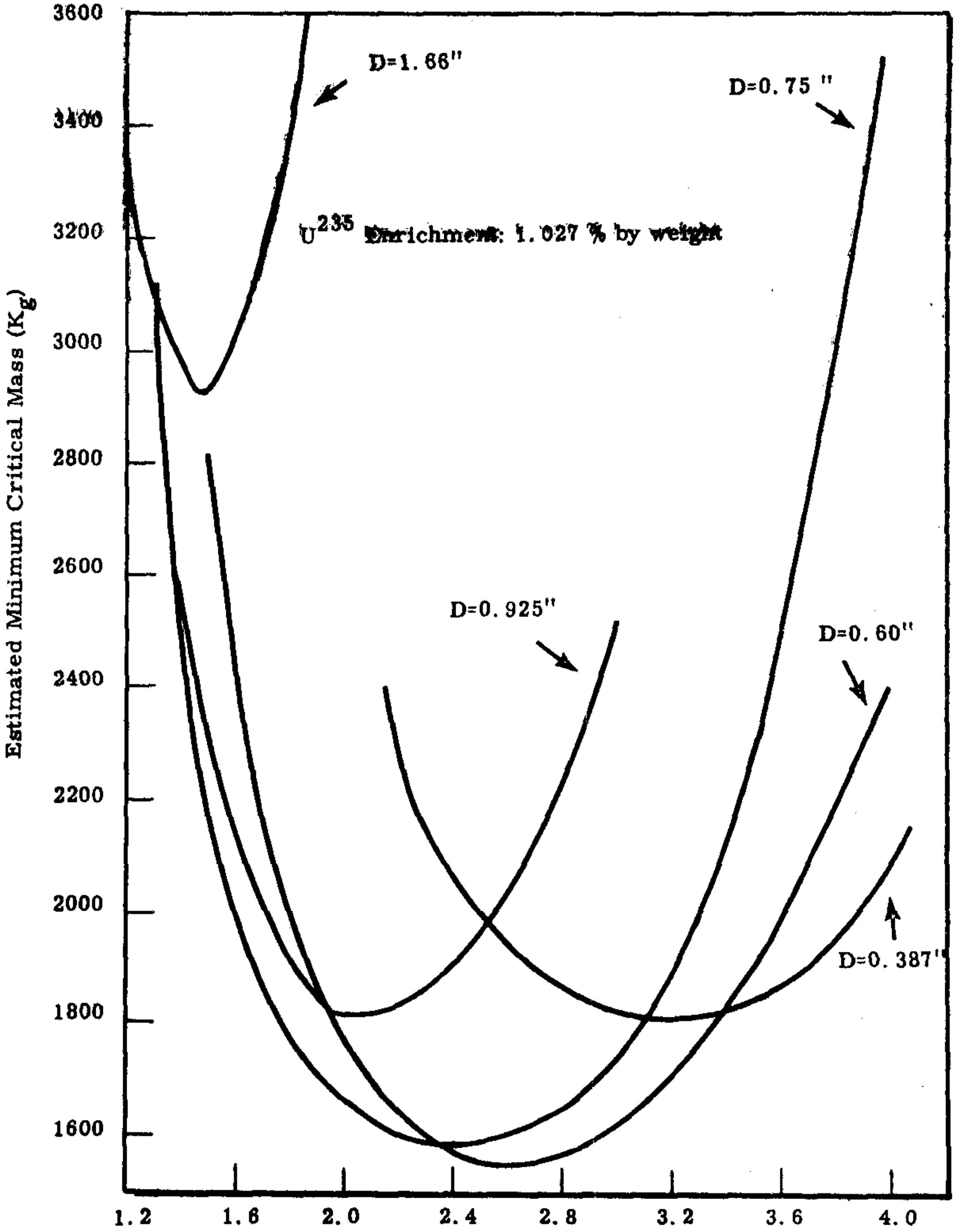

FIGURE 1

ESTIMATED MINIMUM CRITICAL MASS vs WATER TO URANIUM RATIO

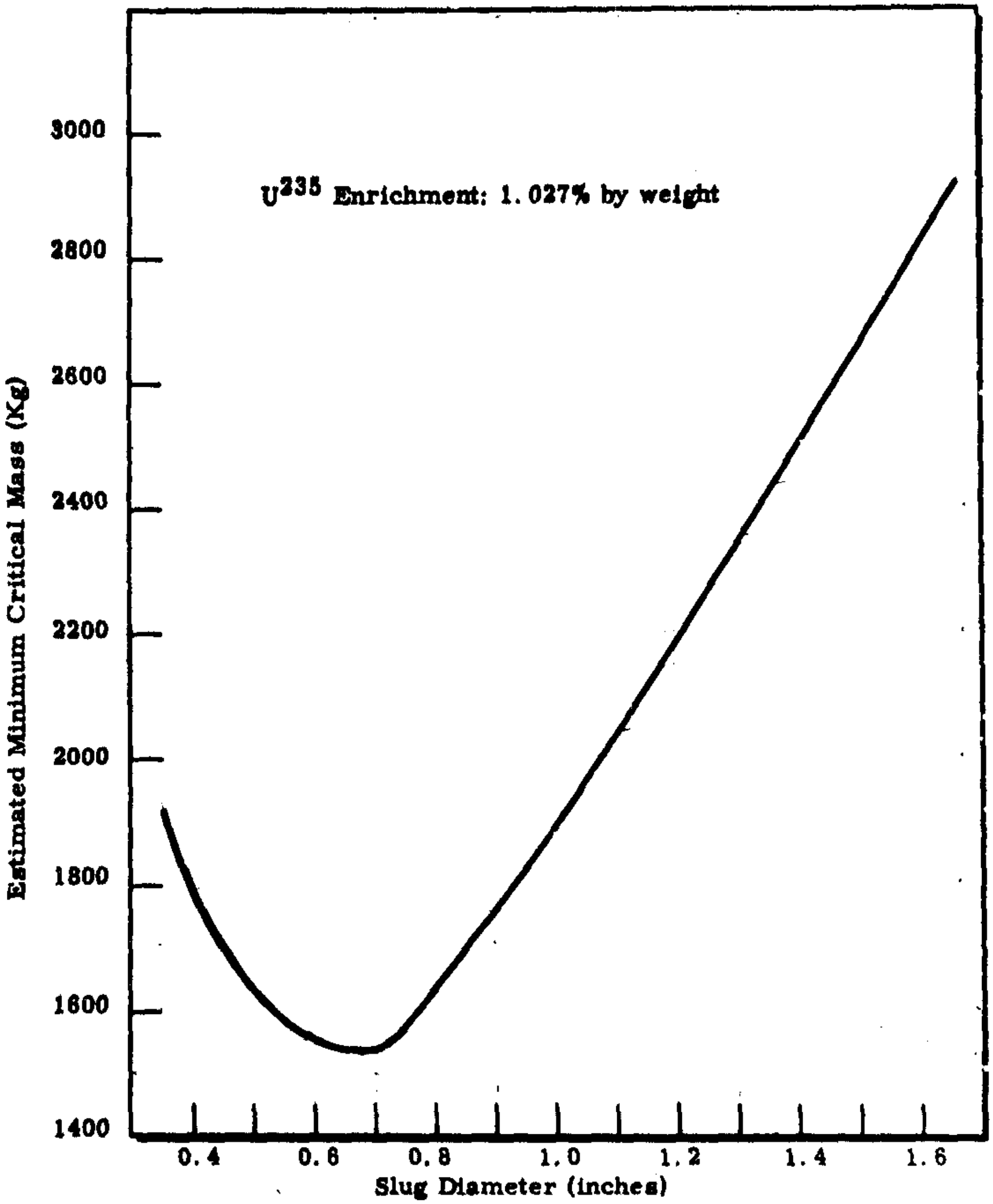

FIGURE 2

ESTIMATED MINIMUM CRITICAL MASS vs SLUG DIAMETER

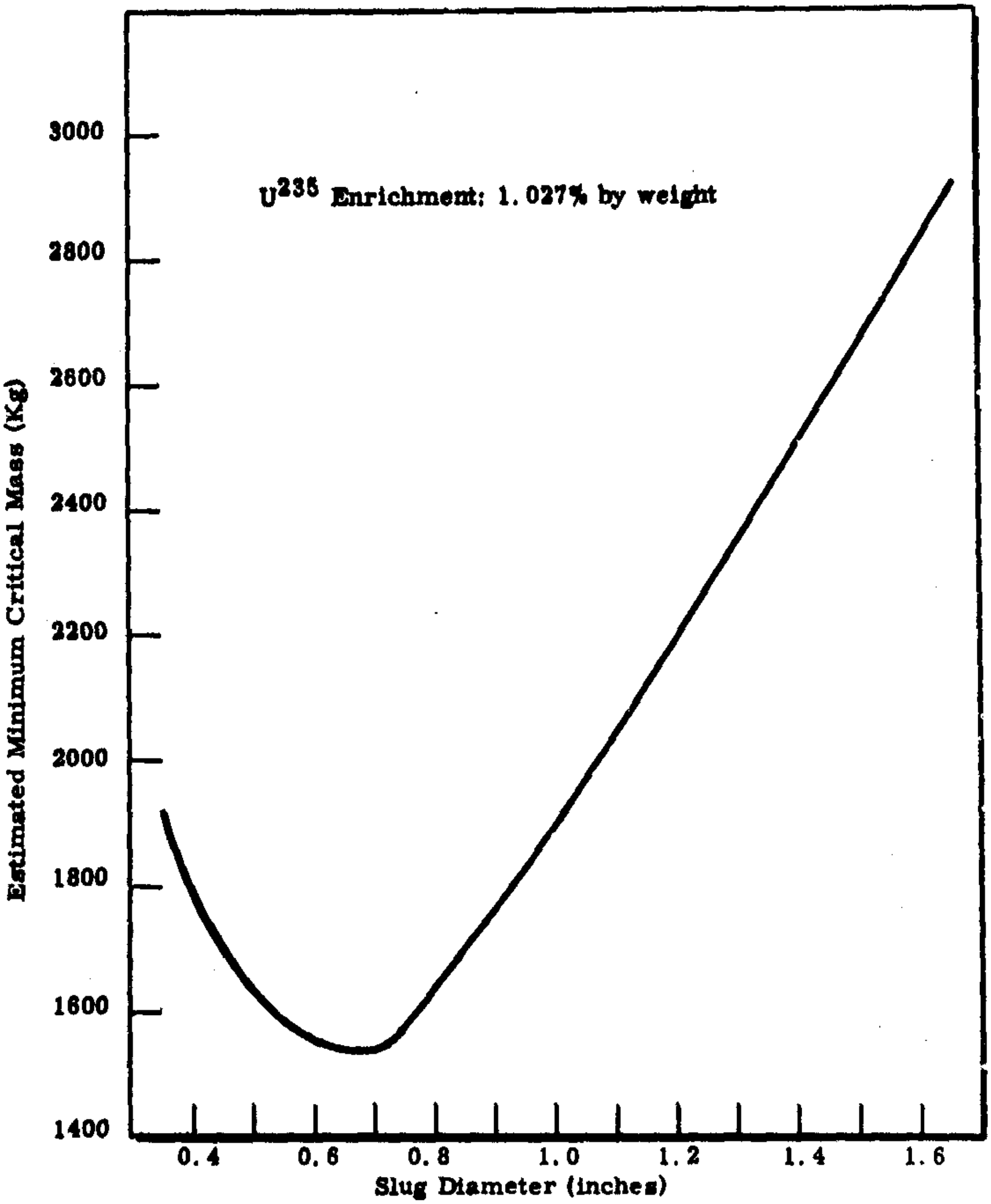

FIGURE 2

ESTIMATED MINIMUM CRITICAL MASS vs SLUG DIAMETER

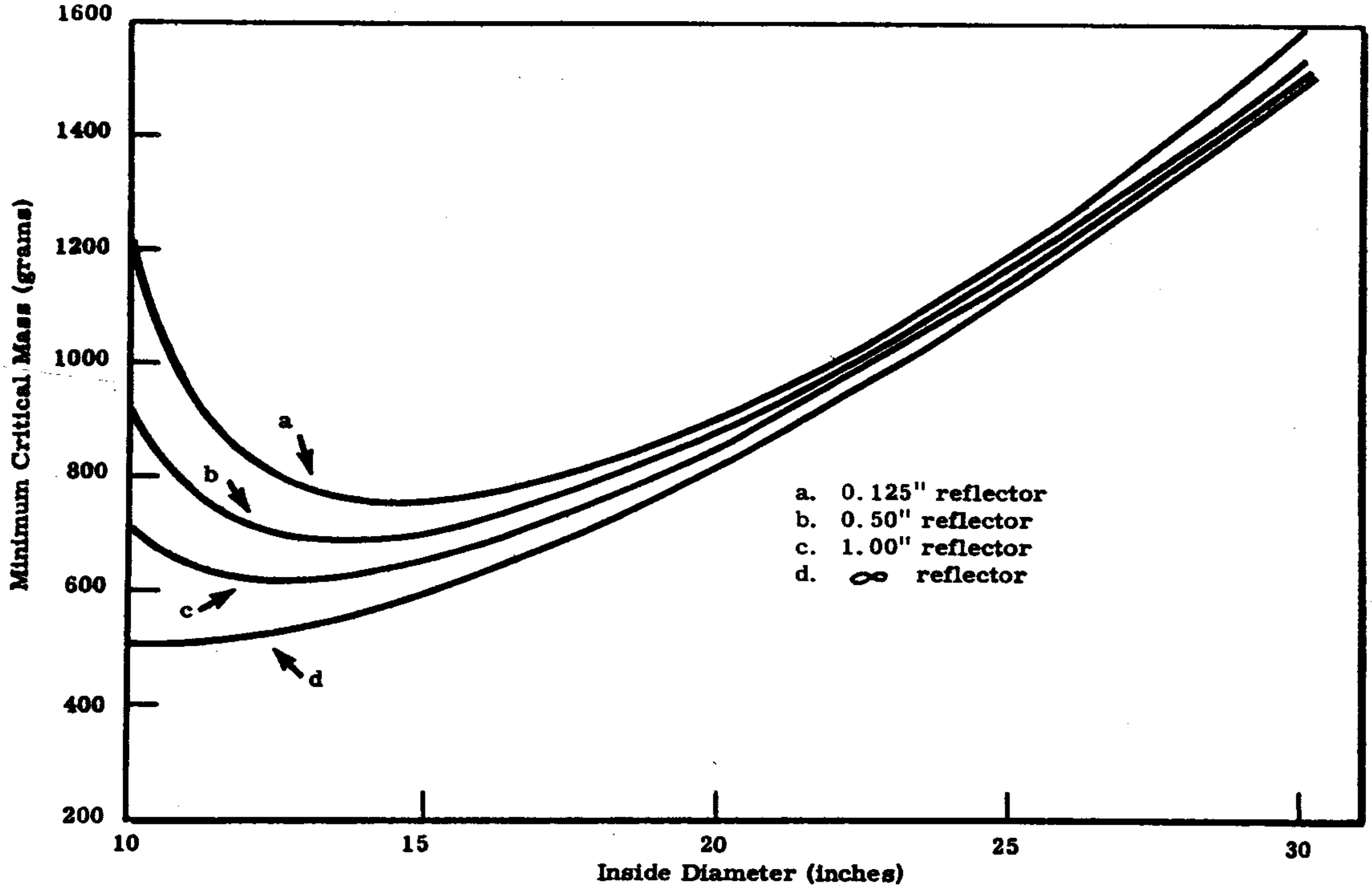

FIGURE 3

MINIMUM CRITICAL MASS vs CYLINDER DIAMETER FOR VARIOUS EQUIVALENT REFLECTOR THICKNESSES

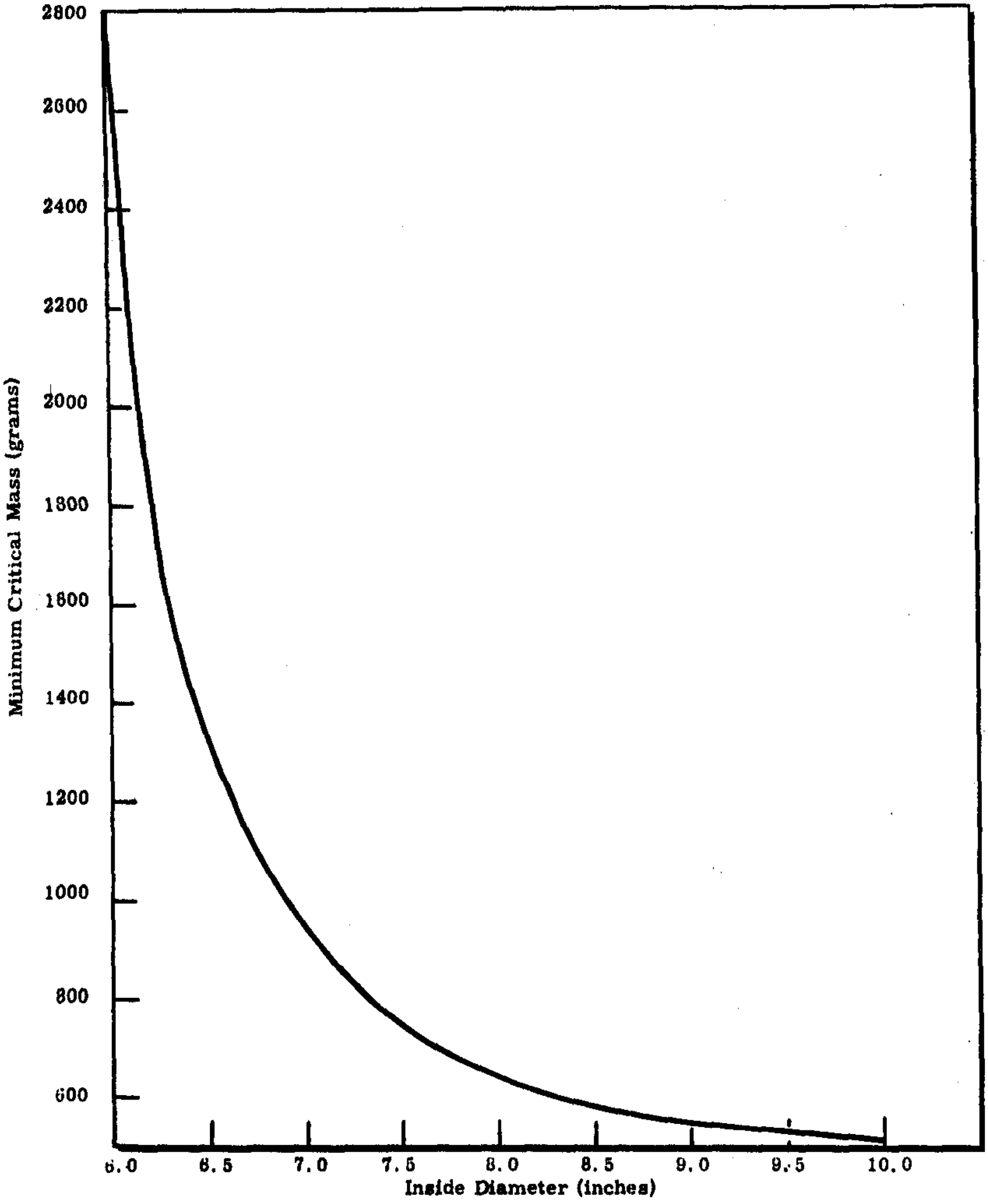

FIGURE 4

MINIMUM CRITICAL MASS vs CYLINDER DIAMETER (FOR WATER-TAMPED CYLINDERS)

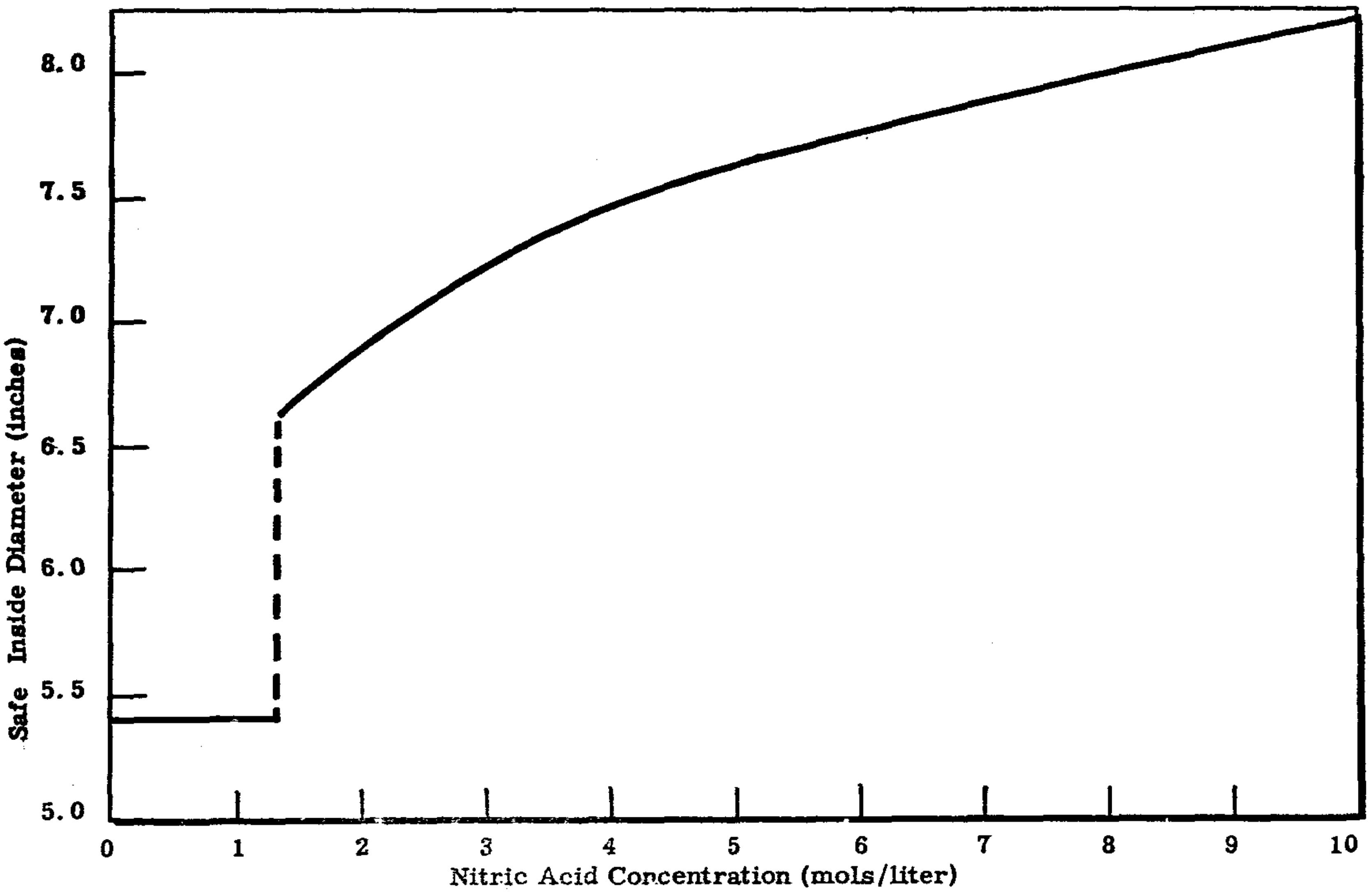

FIGURE 5

SAFE DIAMETER vs NITRIC ACID CONCENTRATION (PLUTONIUM NITRATE SOLUTIONS)

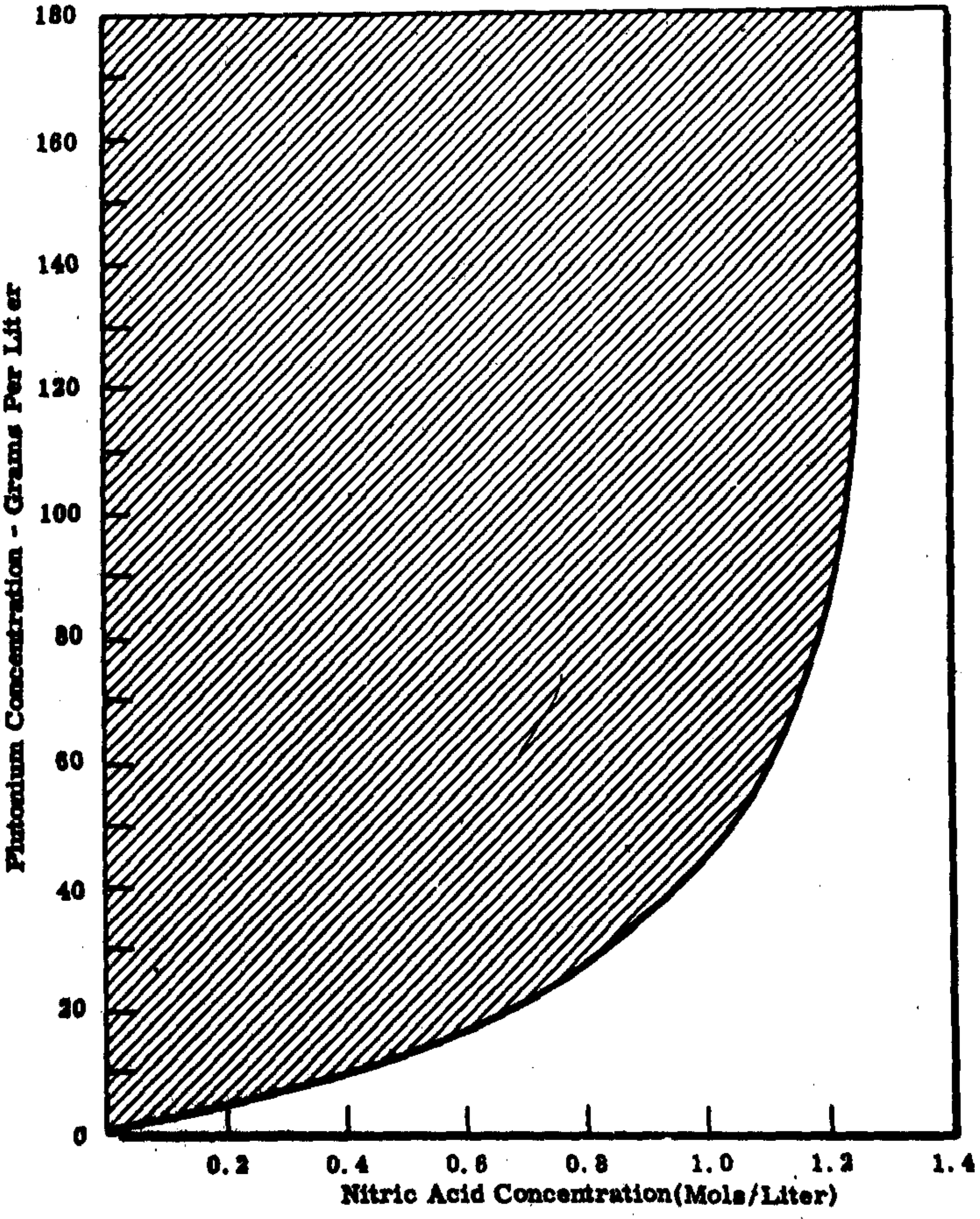

FIGURE 6

REGION OF PLUTONIUM POLYMER FORMATION

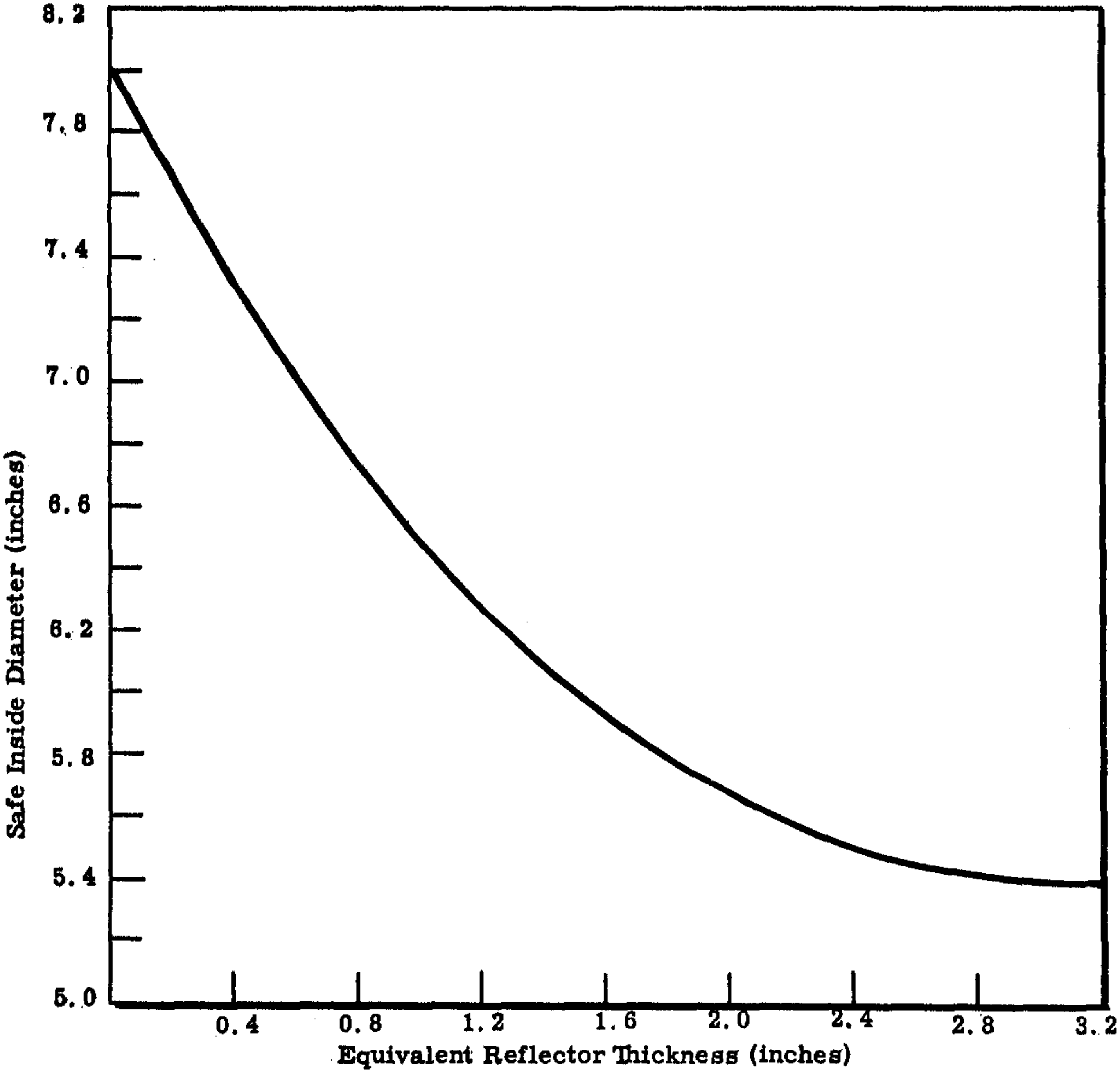

FIGURE 7

SAFE DIAMETER vs REFLECTOR THICKNESS

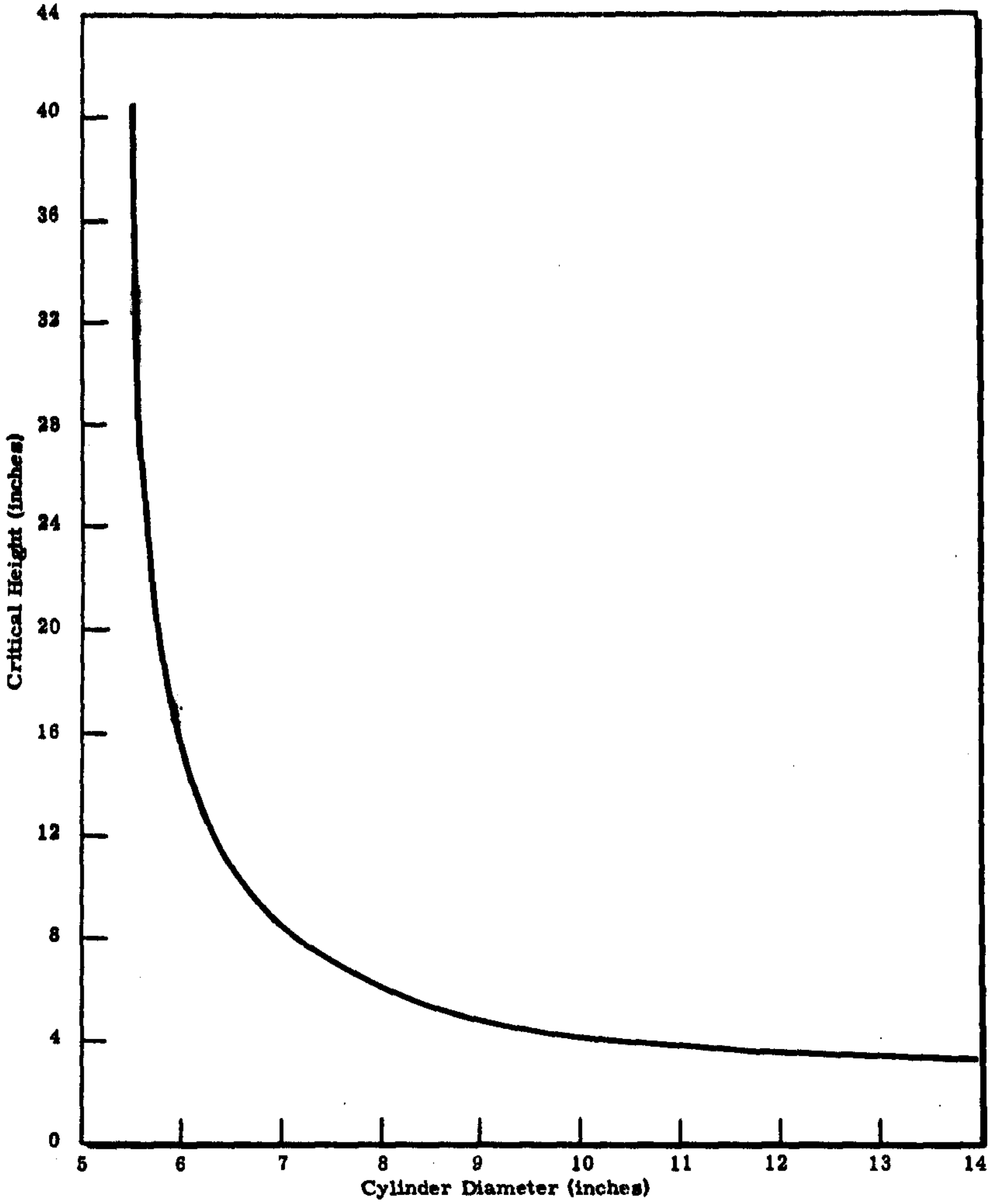

FIGURE 8

CRITICAL SOLUTION HEIGHT FOR WATER-TAMPED CYLINDERS

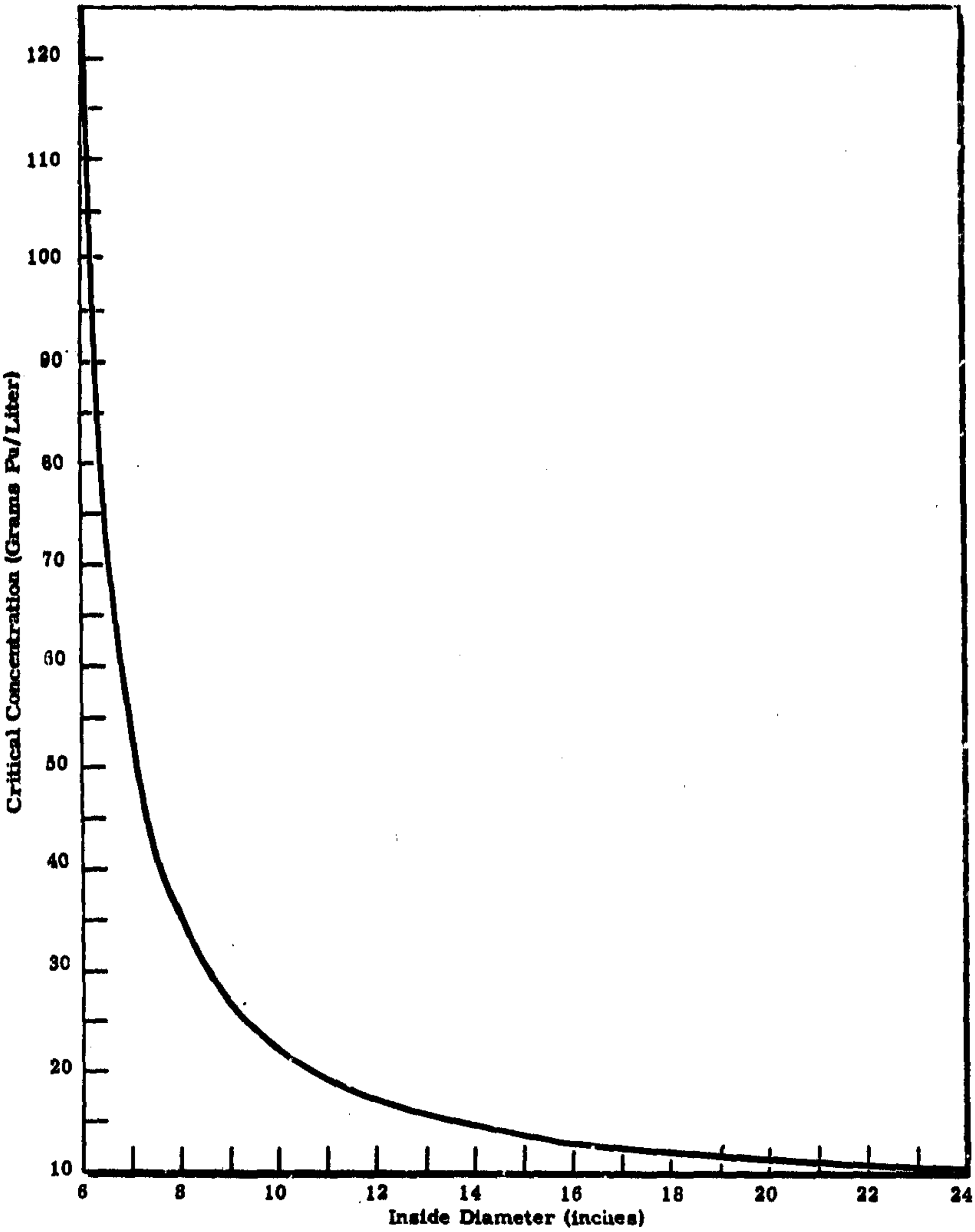

FIGURE 9

CRITICAL CONCENTRATION OF Pu FOR WATER-TAMPED CYLINDERS