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AUTHOR
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## PLUTONIUM-ALUMINUM ALLOY CRITICALITY

Norman Ketzlach

Critical Mass Physics
PHYSICS AND INSTRUMENT RESEARCH
AND DEVELOPMENT OPERATION

February 28, 1958

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## PLUTONIUM-ALUMINUM ALLOY CRITICALITY

### Introduction

Nuclear safety criteria for the storage and handling of unmoderated systems of plutonium metal,(1,3) oxide, and fluoride(1) as well as criteria for storage, handling and processing of plutonium solutions have already been established. These criteria were based on experimental data. The present study deals with criticality conditions for water-moderated and unmoderated systems of plutonium oxides and fluorides as well as those of plutonium-aluminum alloy materials. Such data is required by those interested in the manufacture of plutonium fuel elements for power reactors. Since there are no experimental data available on light water moderated systems of Pu-Al alloys, the present study is a theoretical one. Therefore, the results presented here should be considered as estimates only until the validity of the theory used is verified by experiment.

### Unmoderated Systems

Critically safe parameters for unmoderated homogeneous systems of plutonium metal, oxide, fluoride, and aluminum alloys will first be considered. These values were based on reported values.(1) The unmoderated systems do not allow for the presence of any water or any other moderator in the system. In fact, the only allowable materials in the system other than plutonium are those in the periodic table from sodium (Atomic No. 11) through bismuth (Atomic No. 83). Elements of lower atomic number such as beryllium, deuterium, or hydrogen must be absent. The reflectors outside these systems may be any of the common materials such as water, steel, magnesium oxide, graphite, etc.

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For the case of plutonium alloys in which the alloying material is $11 \leq Z \leq 83$ (and no oxide or fluoride present), the maximum safe mass of plutonium in the alloy is 27.0 kg when the plutonium concentration in the alloy is less than 20 percent plutonium by weight. For alloys in which the plutonium content is greater than 20% plutonium by weight the maximum safe mass decreases and approaches 4.5 kilograms plutonium for the case of the pure unalloyed plutonium (density = 19.6 gms Pu/cc). The larger safe masses for the alloys of lower plutonium content are allowed only for homogeneous mixtures of plutonium and alloying metal. During the process of making the alloy (i.e. adding 100% Pu to aluminum) the system is not a homogeneous one and, therefore, 4.5 kg plutonium is the maximum safe amount in any unmoderated step of the process before a homogeneous alloy is formed. These maximum safe masses are considered to be about 10 percent below the minimum critical masses. Therefore, where there exists the possibility of errors in metal analysis, double batching, or any other process variable that can lead to the possible accumulation of plutonium in quantities larger than those mentioned above, the maximum allowable mass should be reduced so that a safe system is assured at all times. Where double batching is a possibility, the above safe masses should be reduced by a factor of two.

For process steps in which dry oxides or fluorides are handled, the allowable masses are not as great as those in the plutonium alloy systems. For unmoderated systems of these compounds in which there is at least one atom of plutonium per seven of oxygen or fluorine and the maximum density of plutonium is less than 15% of its full density of 19.6 grams Pu/cc, the maximum safe mass is 13.5 kg plutonium.

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The maximum workable mass limit should be reduced below this value for the same reasons stated above for the plutonium alloy and in comparable percentages. These safe masses of oxides and fluorides also allow for the additions of elements in which $11 \leq Z \leq 83$.

It is conceivable that plutonium fuel elements will be required in which the plutonium is incorporated with natural or depleted uranium. The critical masses for unmoderated systems of plutonium and natural uranium will depend on the uniformity of mixture, plutonium density, and ratio of plutonium to natural uranium. When homogeneously mixed, the nuclear properties of these systems behave in a similar manner as do homogeneously enriched uranium systems for which data has already been reported.(2) Since plutonium is more reactive than U-235, each percentage of Pu corresponds to more than 1% U-235. It was shown there that for moderated systems of uranium of different enrichments the critical mass of U-235 does not change appreciably for U-235 enrichments greater than 20% U-235. For completely unmoderated systems, the minimum critical mass changes much more rapidly with U-235 enrichment.(3) This is shown in Figure 1. Figure 1 indicates that for unmoderated systems no U-235 enrichment below 5% can be made critical.

Since there is no data available for plutonium-natural uranium systems, a more conservative approach must be taken. For such mixtures let us, for the present, assume that the quantity of U-235 in the homogeneous mixture has the same reactivity as plutonium and then use the maximum safe masses as those for the corresponding plutonium metal, oxide, or fluoride systems. The allowable masses using this method should be both operationally practicable and conservative.

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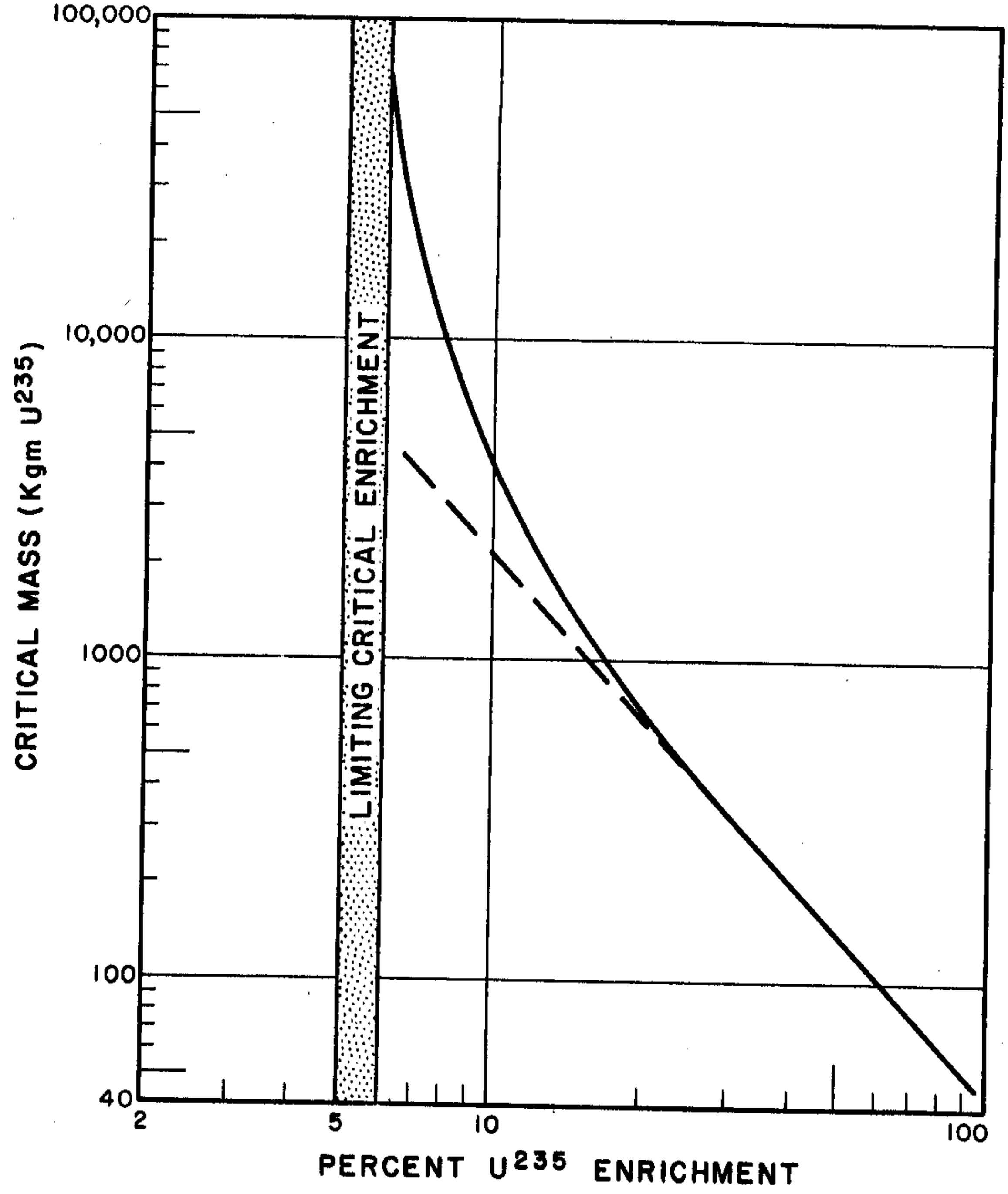

FIGURE - 1
CRITICAL MASS $U^{235}$ vs PERCENT $U^{235}$ ENRICHMENT
(BARE AND UNMODERATED SYSTEMS)

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## Moderated Systems

Theoretical calculations of critical bucklings (the buckling is a measure of the curvature of the neutron flux in a critical reactor; a small critical size implies high reactivity and neutron curvature) were made using an approximation to the age theory formulation outlined previously(4) for slightly enriched uranium-water systems.(8) The only deviation from this formulation was that used in the calculation of neutron age. The method used for the calculation of neutron age will be discussed in more detail here.

The study of alloy fuel elements containing uranium or plutonium indicated that they differ quite markedly in their characteristics from those of slightly enriched uranium fuel elements. The high aluminum content of aluminum alloy fuel elements very markedly affects the neutron age of systems containing these fuel elements immersed in water. It has already been shown(5,6), both in theory and by experiment, that the neutron age of homogeneous systems of aluminum and water increases with increase in aluminum to water volume ratio. One of the curves of Figure 2 shows the relationship(3) between the neutron age in the homogeneous mixture to that in pure water as a function of water to uranium ratio. The curve has been extended (dotted portion) to zero aluminum to water ratio (pure water). It is not expected that the neutron age will increase as rapidly with aluminum to water ratio for heterogeneous systems of aluminum and water.

Since no criticality data are available for heterogeneous lattices of aluminum alloys in light water other than those for J-slugs in water,(7) these data were used to estimate the age of aluminum water mixtures for heterogeneous systems. It was assumed that both the fast effect and resonance escape probability were unity.

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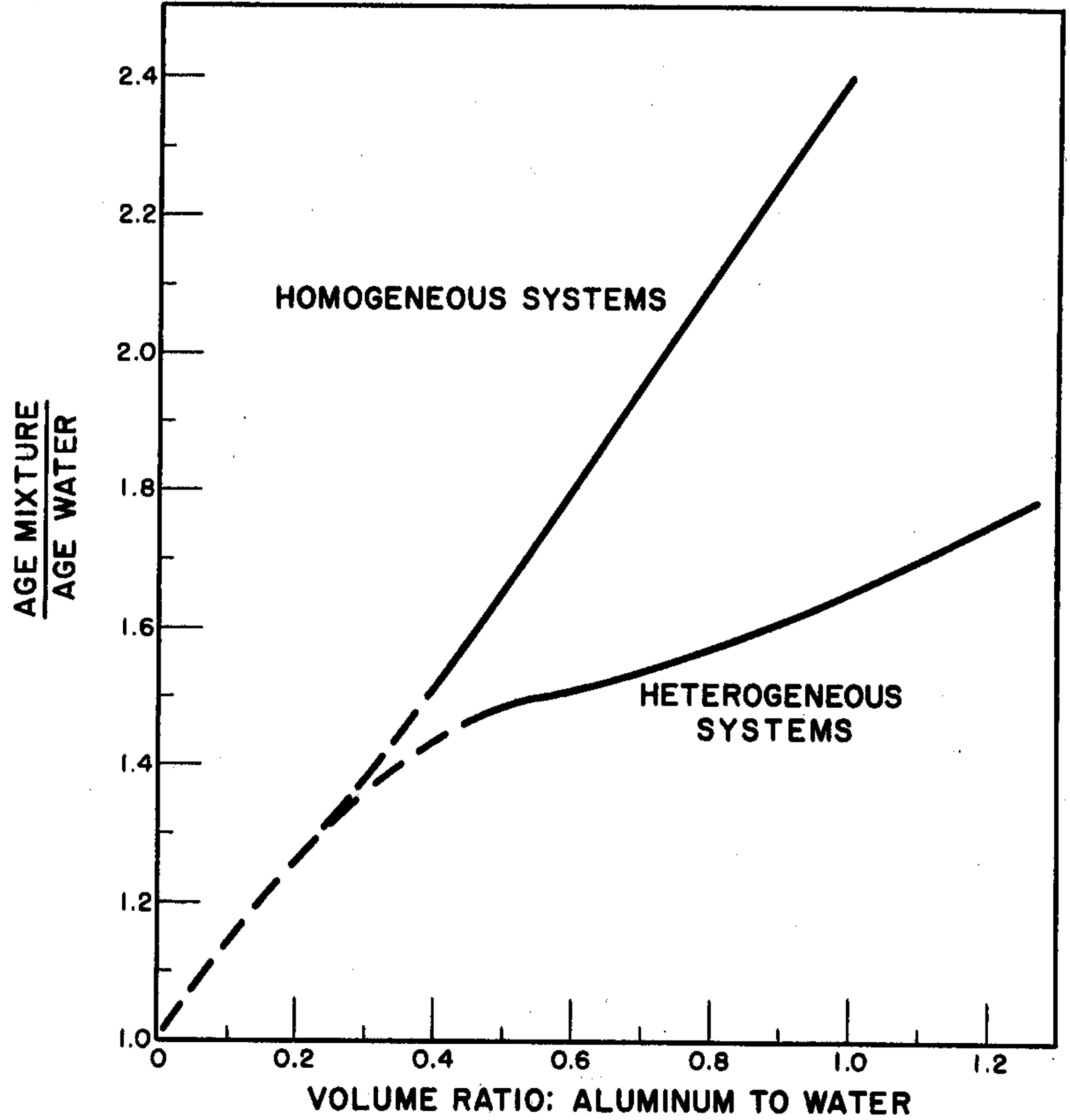

FIGURE - 2
NEUTRON AGE vs ALUMINUM TO WATER VOLUME RATIO

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Since the sizes of the experimental critical systems were known the ages were then calculated from the relation

$$e^{B^2\tau} = \frac{k_\infty}{1 + L^2 B^2}$$

where the terms have their usual meaning in reactor physics. The ratio of these ages to that of pure water (31.5 $cm^2$) are plotted in Figure 2 (heterogeneous systems) as a function of aluminum-to-water ratios. This curve, too, was extended (dotted portion) to pure water.

Using these values for the neutron ages and the formulation previously described,(4) critical bucklings and critical masses as a function of rod size and water-to-fuel ratio were calculated for plutonium-aluminum alloy fuel elements of 5% and 15% plutonium by weight. Figure 3 is a plot of material buckling as a function of moderating ratio for 5% Pu alloys for several rod sizes. It is seen that the maximum buckling increases with decreasing rod size. Figure 4 is a similar plot for aluminum alloys containing 15% plutonium by weight. Figure 5 is a plot of the maximum buckling for each rod size as a function of rod diameter for plutonium-aluminum alloys of 5% and 15% plutonium content. It is seen from Figure 5 that the maximum buckling for Pu-Al alloys of the indicated enrichments increases with decreasing rod size. A mixture of water and alloy of zero rod diameter should correspond to a homogeneous mixture. From the maximum bucklings of Figure 5, the minimum critical diameters for 5% and 15% plutonium-aluminum alloys are 10.6 inches and 8.7 inches respectively for water-reflected cylinders. In other words, an assembly of fuel elements of the above enrichments cannot be made critical in infinitely tall vessels whose diameters are smaller than those indicated above. The corresponding maximum safe diameters are 5.4 inches for plutonium solutions (no aluminum present) and 1.4 inches for plutonium-239 metal.

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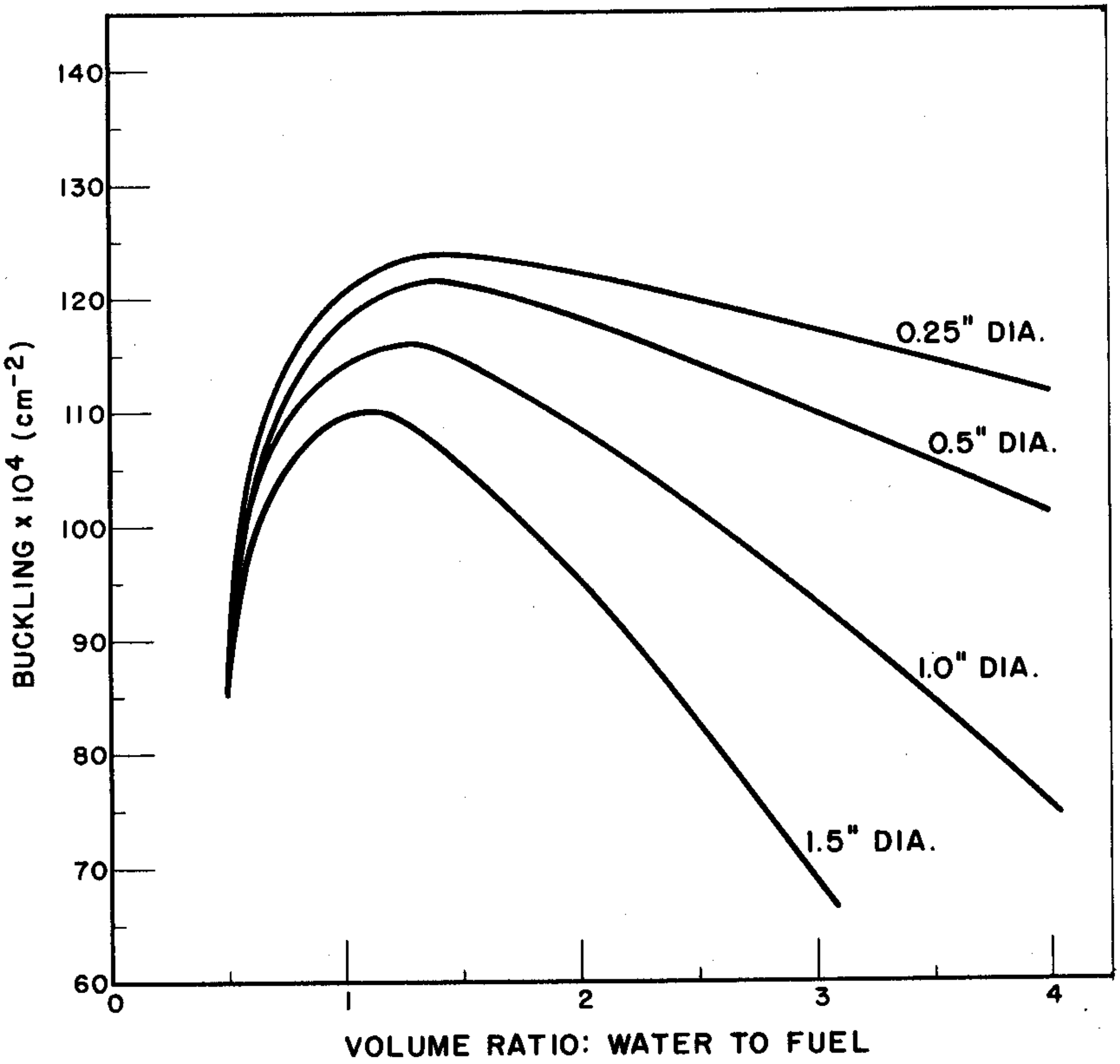

FIGURE - 3
MATERIAL BUCKLING vs VOLUME RATIO
WATER TO FUEL ( 5% Pu )

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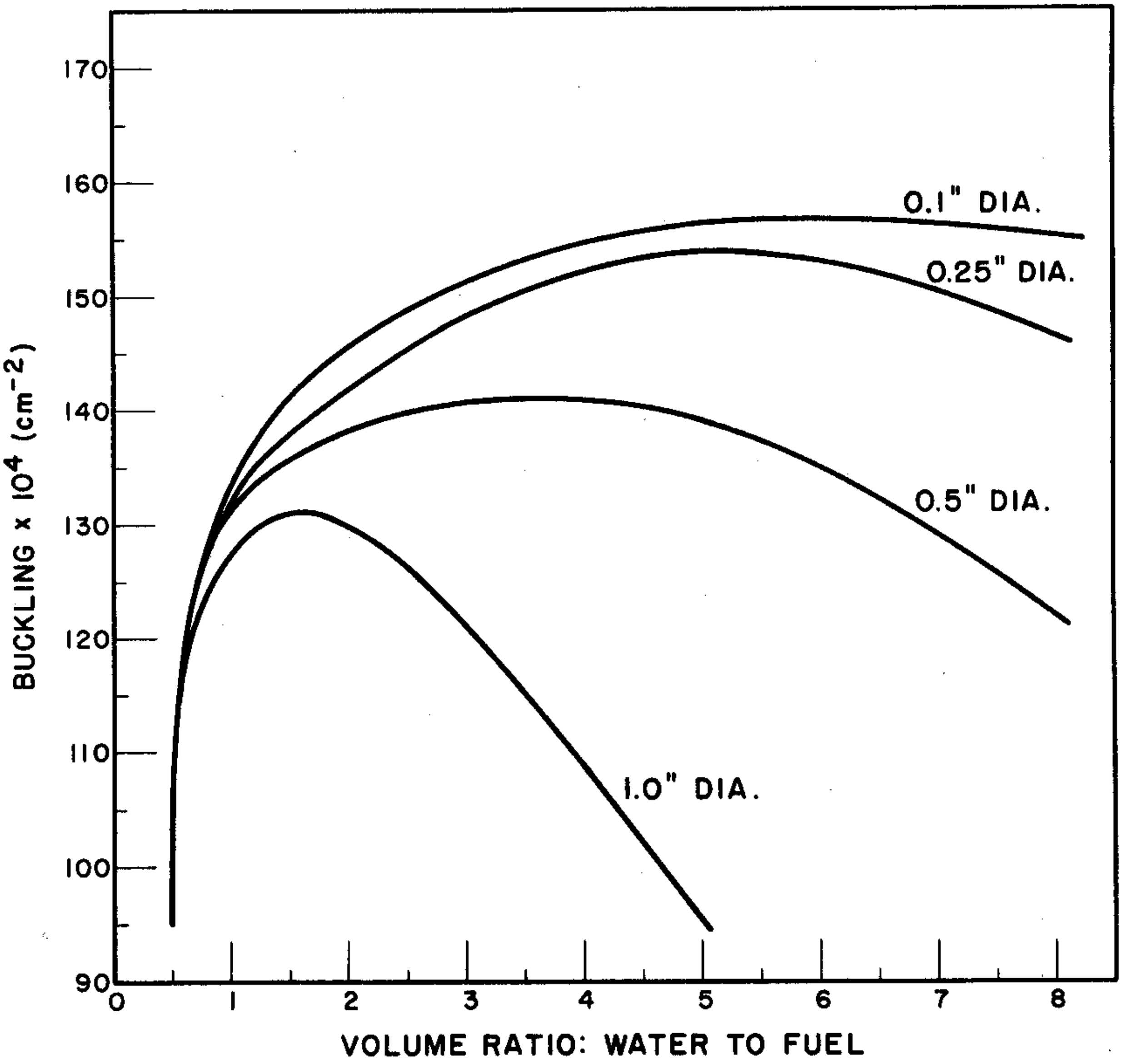

FIGURE - 4
MATERIAL BUCKLING vs VOLUME RATIO
WATER TO FUEL (15% Pu)

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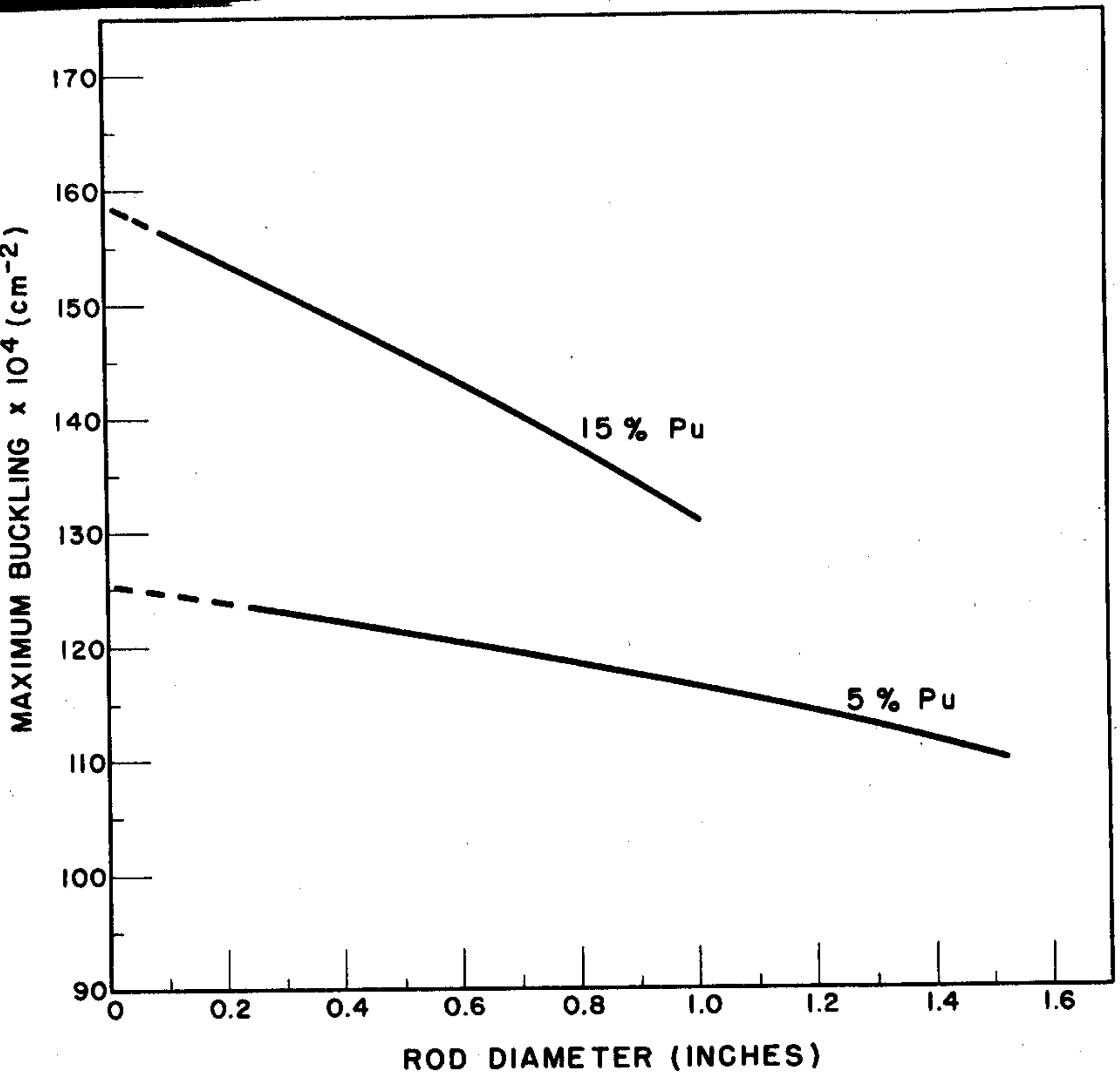

FIGURE - 5
MAXIMUM MATERIAL BUCKLING vs ROD DIAMETER
(Pu - Al ALLOYS)

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From the buckling data, the minimum critical masses as a function of rod size and moderation have been calculated for the above plutonium enrichments. Figure 6 is a plot of minimum critical mass as a function of moderation for several rod sizes for plutonium-aluminum alloys containing 5% plutonium by weight. Figure 7 is a plot of the same parameters for the 15% plutonium alloy. Figure 8 is the plot of the minimum critical mass as a function of rod size for both the 5% and 15% plutonium alloys. The curves in Figure 8 also indicate that the minimum critical masses for plutonium-aluminum alloys of the indicated enrichment decrease with decreasing rod sizes. Here again a system in which the rod size is zero should correspond to a homogeneous system. It is seen that the zero rod diameter critical masses get closer and closer to the minimum critical mass of a homogeneous solution of Pu-239 in water (500 grams plutonium) as the plutonium enrichment of the alloy increases; this is to be expected. It is also not surprising that the minimum critical mass of the 15% alloy is only about 15% greater than that for 100% plutonium-water systems. This relatively small change in critical mass was also found(2) between homogeneous solutions of about 15% enriched U-235 solutions (remainder U-238) and 100% enriched U-235 solutions. This approach to the minimum critical mass of 500 grams for Pu-239 water systems is an indication that the theory used in this study for plutonium-aluminum alloy rods is a fair approach to criticality conditions of such systems.

The above critical masses and vessel geometries are based on individual vessels or containers. Where a number of individually safe units are assembled, it must be ascertained that they are far enough from other units so that the interaction between them is not enough to affect their criticality. Since the critical masses of plutonium

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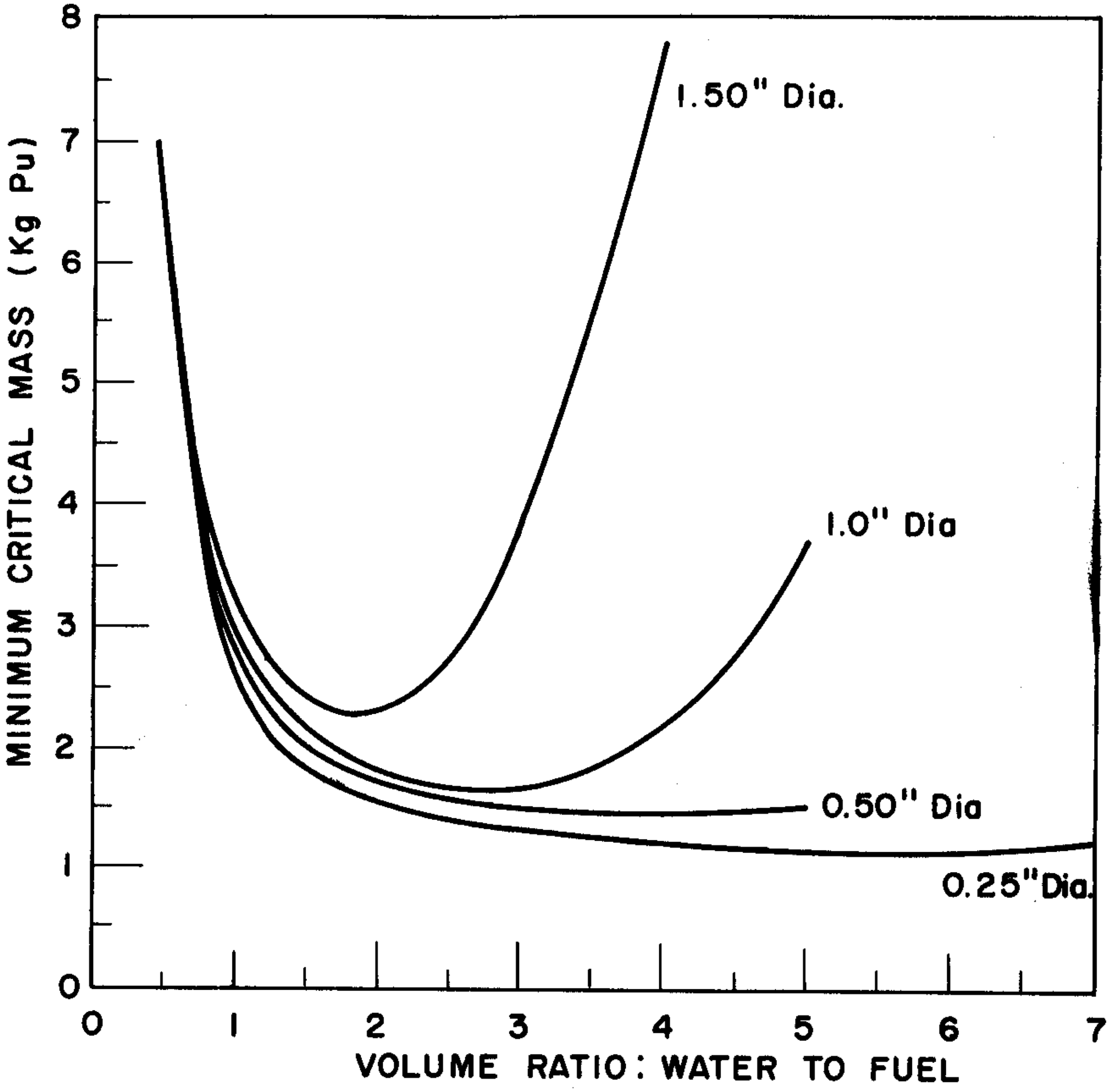

FIGURE 6
MINIMUM CRITICAL MASS vs VOLUME RATIO WATER TO FUEL (5% Pu)

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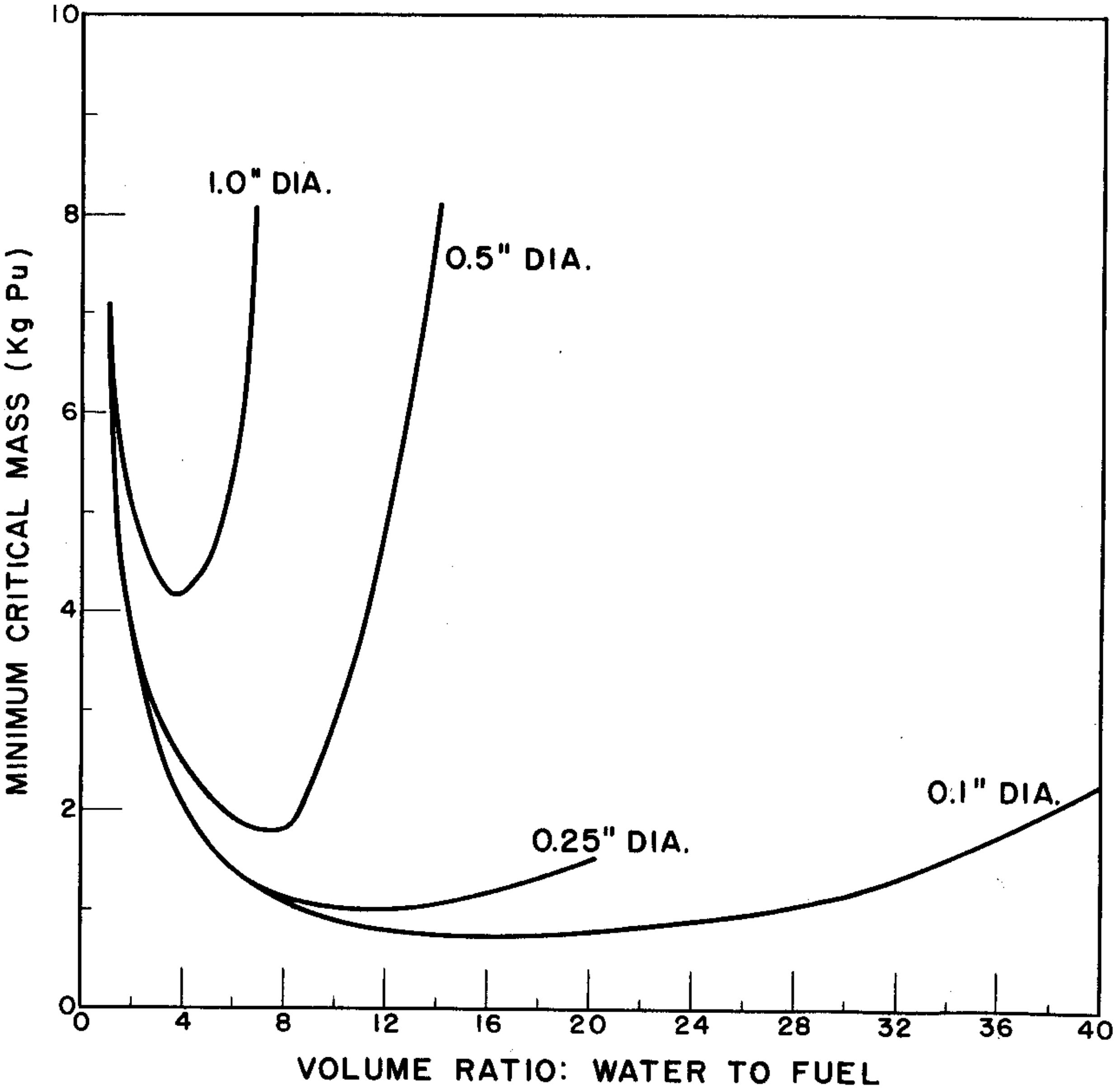

FIGURE - 7
MINIMUM CRITICAL MASS vs VOLUME RATIO WATER TO FUEL (15% Pu)

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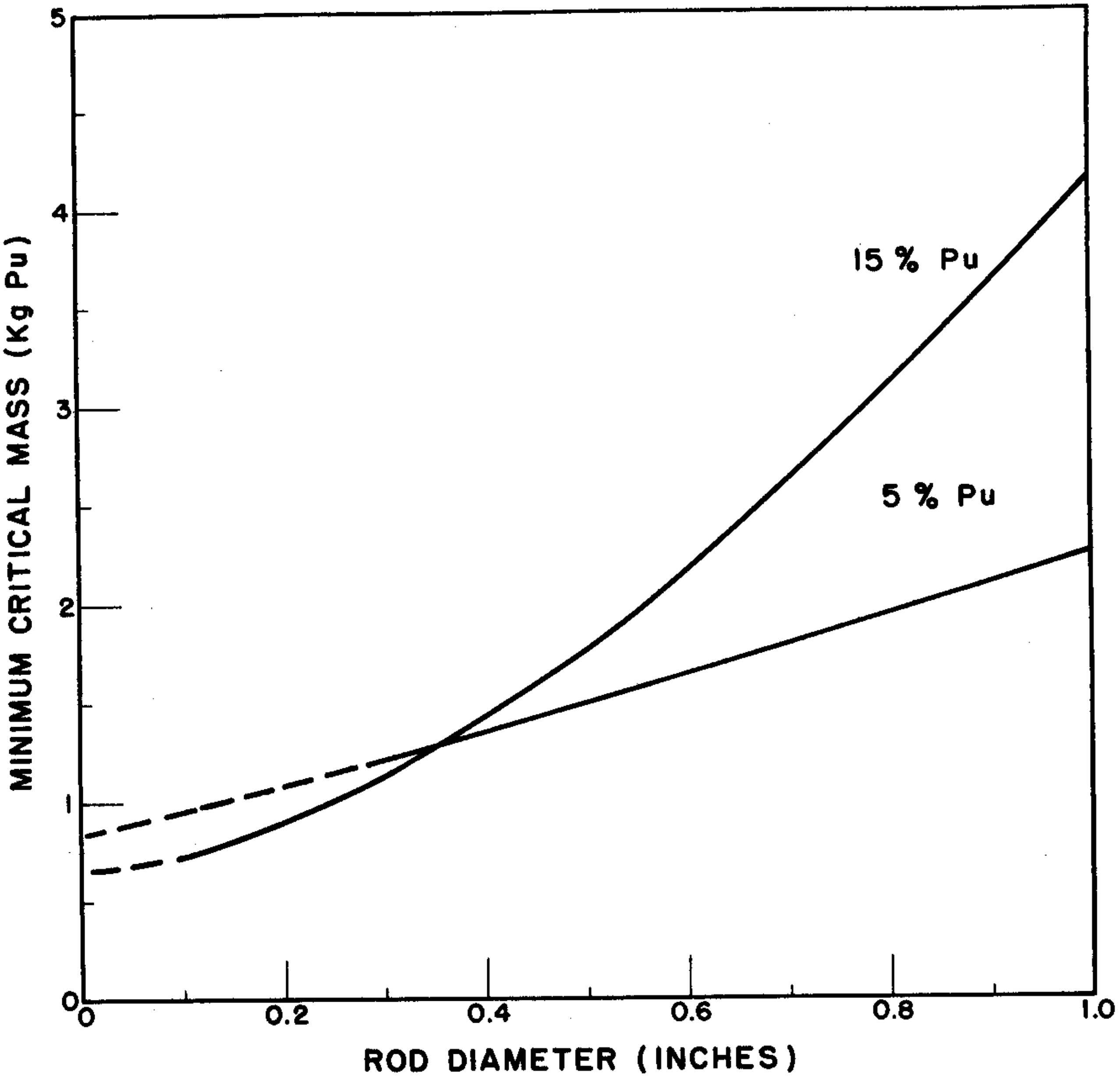

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FIGURE – 8
MINIMUM CRITICAL MASS vs ROD DIAMETER
(Pu-Al ALLOYS)

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for Pu-Al alloys approach those for homogeneous systems as the fuel rod diameter approaches zero and the percentage Pu in the alloy increases, a safe array may be conservatively based on the maximum safe concentration of plutonium in solution; namely 6 grams Pu per liter (170 grams Pu/ft$^3$). Therefore, if each unit in the array contains a safe mass of plutonium (less than 500 grams), and the units of the array are separated so that the maximum spatial density is less than 170 grams Pu/ft$^3$, the array will be safe.

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