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ANOMALIES OF NUCLEAR CRITICALITY

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ABSTRACT

Definitions of anomaly commonly include: A deviation from a common or accepted rule, something that may be out of keeping with regard to accepted notions of fitness and order. There is a scientific way to understand every phenomena and a valid reason for the occurrence of any happening. An anomaly, once disclosed, is therefore amenable to explanation.

During the development of nuclear energy, a number of apparent "anomalies" have become evident in nuclear criticality. Some of these have appeared in the open literature and some have not. Yet, a naive extrapolation or application of existing data, without knowledge of the "anomalies", could lead to potentially serious consequences. Several of the known "anomalies" include, but are not limited to,

1. relationship between criticality in finite spheres and reflected cubes
2. relationships between critical concentrations of ^{233}U, ^{235}U, and ^{239}Pu in infinite cylinders
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21. insertion of a neutron absorbing control rod into a Pu solution sphere wherein the effect of this was to cause the assembly's reactivity to initially increase as the rod entered the solution
22. appearance of critically unbounded regions (of infinite masses) for slightly enriched uranium
23. criticality in the earth
24. criticality in the universe
25. an apparent chain reaction which took place in the Republique of Gabonaise, West Africa, in primeval times with low (3 wt% ^{235}U) enriched uranium
26. criticality of even-n nuclides beginning with the naurally occurring element, $^{231}_{91}Pa$
27. beyond Californium projection of the "micro" critical mass for the doubly-closed shell, super-heavy magic nuclei of the future
28. neutron multiplication and the power reactor (four billion watts and subcritical)

There can be as many as three different fuel concentrations with the same critical volume, and perhaps four different fuel concentrations may result in the same critical mass. Contrary to the usual expectation, the sphere, after all, may not be the configuration of least mass; the reflected cube may be somewhat less under certain circumstances. In some cases, the effect of added scatterers can significantly reduce the critical dimension; whereas, in others the result can be precisely the opposite.

Reducing core density can, under some circumstances, actually decrease the critical mass, contrary to the usual expectation that the mass will be increased. Surprising as it may seem, a system with k_∞ < unity might be made critical by reducing the core size and adding a finite reflector of D_2O, etc., (in the latter case $k_{eff} > k_\infty$!). In some cases, the effect of moderation results in the smallest critical mass; whereas, in others (depending on the evenness or oddness of the nuclide), the effect is again precisely the opposite.

It is noted that a homogeneous aqueous mixture of ^{235}U and ^{238}U could have a smaller ^{235}U critical mass (over a limited concentration range) with low enriched uranium than if the uranium were fully enriched (93.5 wt% ^{235}U), and that an unmoderated interacting array of metal units (spheres of 30 wt% ^{235}U) might have lower critical density than an array of fully enriched uranium spheres, of identical volume, and thus a smaller critical ^{235}U mass in the array.

A number of peculiarities are manifest in the criticality of interacting arrays of subcritical units, that relate to the unit shape, its density, the isotopic fuel composition, the lattice density within the array, and the degree of internal and external moderation and reflection involved. There is an example wherein the effect of inserting a neutron absorbing rod into a Pu-solution-bearing sphere is to cause the reactivity to initially increase rather than decrease.

It is to be noted that in the case of certain heavy, even-n nuclides, which possess fission thresholds, the "worth of the dollar" can become effectively zero because the energy of the delayed neutrons will, for the most part, be less than the fission thresholds. In these cases, ^{238}Pu for example, no state of "delayed criticality" could prevail, but only "prompt criticality."

For very dilute homogeneous PuO_2-$U_{NAT}O_2$-H_2O mixtures, calculations show a maximum in k_∞ to occur at low Pu concentrations; i.e., a higher value of k_∞ can be achieved, or a lower critical concentration of Pu is possible, with

3 wt% PuO_2 than with 8 wt% PuO_2. Based on the data and calculations pertaining to ^{239}Pu-^{240}Pu metal mixtures, there appears to be an inconsistency in the effect of ^{240}Pu on the criticality of the mixture, which is thought to be due to a change in spectrum brought about by the addition of the ^{240}Pu. Calculations now indicate the critical mass for ^{240}Pu metal to be significantly less than that of ^{235}U metal.

It is shown that a processing plant that is "safe by shape" for ^{239}Pu should not be considered safe, on a apriori basis, for processing either ^{233}U or ^{235}U solutions at equal concentrations. This is true even though the ^{239}Pu has the smallest limiting critical concentration, the smallest minimum mass in the form of an aqueous solution, and the smallest mass when in the form of metal.

Pressures up to 10^{12} atm, comparable with the pressure in the center of the sun, are now believed achievable with advanced giant lasers or electron beams which could change the density of a small pellet of fissionable material under irradiation by a factor of some 250, thus making it possible to achieve a supercritical event in a small pellet of Pu containing as little as 10^{-2} g Pu.

One of the more interesting events in the annals of criticality is the discovery of an apparent prehistoric chain reaction (Nature's Anomaly) that took place in the Republique of Gabonaise in primeval times with $\sim$ 3 wt% ^{235}U enriched uranium.

Finally, it is interesting to recall that, in the presence of inherent neutron sources, even the power reactor will be technically subcritical ($k_{eff} < 1$) when operating in a constant power mode at any power level.

Several of the anomalies cited herein would constitute "autocatalytic reactions," for in the event of criticality, the reactivity would be increasingly enhanced as a consequence of the reaction.

The list continues and there are doubtless many other seemingly apparent anomalies that are interesting (albeit they be perhaps hypothetical) that can be cited in the field of criticality. For example, cadmium, gadolinium, samarium, Boron, etc. can be highly effective when used in control rods for reducing the reactivity, or "shut down" of a reactor, if these materials are positioned on the interior of the reactor's core. The same materials could, however, increase the reactivity, or enhance criticality, if they were utilized as reflectors on the exterior or surface of the reactor's core - a fact not always fully appreciated. The reason is that any nuclear material irrespective of the magnitude of its absorption cross section can never have a scattering cross section for return of neutrons that is zero.

ANOMALIES OF NUCLEAR CRITICALITY

A. INTRODUCTION

A large body of knowledge has been accumulated on criticality, and the factors affecting criticality, since inception of the first chain reaction in 1942.[1-5] Criticality, however, is fraught with complexities, and lest we become too complacent in the era of advancement, it may be well, at this time, to recount a few of the apparent anomalies - some of which have heretofore remained unpublished in the open literature. Most of the examples to be cited are not well known. Yet, without knowledge of these "anomalies" an unwise application of existing data could lead to potentially serious consequences. As an introduction to the apparent anomalies that follow, our discussion begins with a qualitative description of the concepts and complexities of nuclear criticality.

B. CONCEPTS AND COMPLEXITY OF CRITICALITY

The phenomenon of criticality depends on the interaction of neutrons with matter, and is characterized by a self-sustaining fission chain reaction. Consider the conditions for achieving criticality. The exact configuration, or spatial density, must be known for each kind of atom present in the system. Criticality, then, depends not only on the quantity of fissile material present, but on the size, shape, and material of any containment vessel which may be used, on the nature of any solvents or diluents, and on the presence of any adjacent material which may possibly return neutrons through scattering back into the fissile material.

The state of criticality can be further expressed in terms of the multiplication factor, which may be defined as the ratio of the number of neutrons in one generation to the number of corresponding neutrons in the immediately preceding generation. Obviously, for each neutron in the

first generation, there must result in at least one neutron in the second generation, etc., if a self-sustaining chain reaction is to continue. The reproduction factor will be unity when a precise balance exists between the production of neutrons through fissions and the subsequent losses. From the neutron balance point of view, the critical condition is defined when:

> the average number of neutrons produced per unit time = average number absorbed per unit time + average number escaping per unit time.

The reproduction factor is the ratio of neutron production to losses, or

$$k_{eff} = \frac{\text{Production}}{\text{Leakage + Absorption}}$$

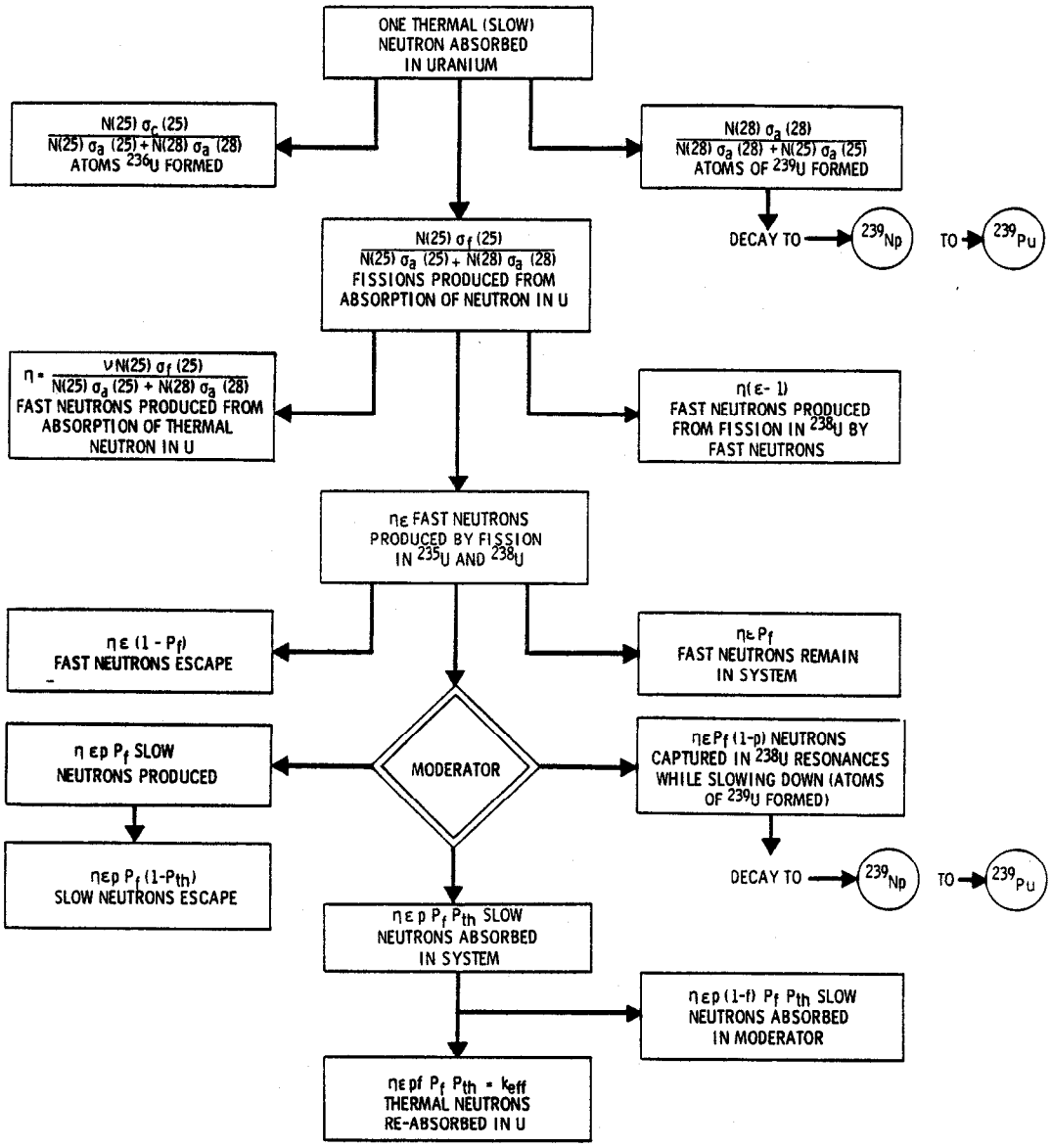
The fate of a fission neutron is to either be absorbed in the fissile material, diluent, or structural material of the containment system, or to escape through leakage.

1. Neutron Economy and Criticality in Uranium

To illustrate, the neutron economy for a homogeneous mixture of U and diluent is presented in abbreviated form in Figure 1.

If the system is of infinite dimension, the fraction of neutrons escaping through leakage (1 - P) becomes zero since the non-leakage fraction P becomes unity. In this case k becomes = $\varepsilon p f \eta$, which is the multiplication constant for an infinite system, k_∞. The reproduction factor for the finite system (k_{eff}) can then be expressed as the simple product

$$k_{eff} = k_\infty P, \text{ where } P = P_{th} P_f$$



SYSTEM IN STATE OF CRITICALITY IF k_{eff} = UNITY

σ_a (ABSORPTION) = σ_c (CAPTURE) + σ_f (FISSION).

ν = NUMBER OF NEUTRONS / FISSION

η = NUMBER OF NEUTRONS PRODUCED PER NEUTRON ABSORBED IN FISSIONABLE MATERIAL

p = RESONANCE ESCAPE PROBABILITY; FRACTION OF NEUTRONS THAT ESCAPE CAPTURE IN ^{238}U

f = FRACTION OF NEUTRONS THAT ARE ABSORBED IN URANIUM AT THERMAL ENERGY

ε = FAST FISSION FACTOR: RATIO OF THE NUMBER OF FAST NEUTRONS RESULTING FROM FISSIONS CAUSED BY NEUTRONS OF ALL ENERGIES (FAST AND SLOW) RELATIVE TO THE NUMBER PRODUCED BY THERMAL (SLOW) NEUTRON FISSION

P_f = FRACTION OF FAST NEUTRONS PRODUCED THAT DO NOT ESCAPE THROUGH LEAKAGE WHILE SLOWING DOWN; FAST NON-LEAKAGE PROBABILITY

P_{th} = FRACTION OF NEUTRONS THAT DO NOT ESCAPE THAT REACH SLOW OR THERMAL NEUTRON ENERGY; THERMAL NON-LEAKAGE PROBABILITY

Figure 1. Neutron Economy in Natural Uranium Reactor System

In order to compute criticality it is required to calculate the interaction of the neutrons with the materials composing the reactor system. The probability of neutron interaction is given by the nuclear cross sections for the various reactions. The problem is complicated, since the cross sections are energy-dependent. The microscopic cross section, σ, for high energy neutrons is of the same order of magnitude as the actual cross sectional area presented by the target nucleus. The average energy of the neutrons released in fission is about 2 MeV. The cross section for absorption of a neutron in ^{235}U at this energy is only about 1.3 barns, whereas, for thermal neutrons (0.025 e.v.) the cross section becomes 681 barns, or some 500 times larger. At 2 MeV the most likely occurrence on collison of a neutorn with a U atom is that the neutron will simply scatter or be deflected. To accurately compute criticality, the various neutron interactions must be determined over the entire energy spectrum of neutrons in the system The neutron spectrum is, in turn, determined by the amount of diluent (especially hydrogeneous materials) that can moderate or slow down the neutrons. Fast neutrons lose energy through collison processes by inelastic and elastic scattering.

In the case of inelastic scattering, part of the energy of the incoming neutron goes into internal excitation of the target nucleus with subsequent release by gamma emission; a portion of the kinetic energy of the neutron has been converted into gamma emission, leaving the neutron with less energy. Inelastic scattering is important chiefly in heavy nuclei such as uranium. The threshold energy below which the reaction cannot occur is about 0.1 MeV. The loss of energy by elastic scattering is determined by the mechanics of the interaction and is thus greater for the lightest nuclei, such as from the hydrogen contained in water. Through the above two processes, the fast fission neutrons can be moderated to thermal energies ($\sim$ 0.025 e.v.)

The state of criticality for the system may further be defined in terms of the value k:

State of Criticality

k ≡ Unity	Delayed Critical
k > Unity	Supercritical
	i Delayed
	ii Prompt
k < Unity	Subcritical

Delayed critical defines a condition of precise balance between production and losses of neutrons whereby all of the neutrons released in fission (including those that are delayed) are required to obtain a reproduction factor of unity. Two supercritical conditions are defined: Delayed and Prompt. In the delayed supercritical state, k exceeds unity, but only by an amount that is less than the total contribution possible from delayed neutrons. In the prompt supercritical state, k exceeds unity by an amount that is equal to, or greater than, the contribution from delayed neutrons.

Changes in k above unity will cause exponential changes in the neutron population at a rate dependent on the average neutron lifetime. If the system is delayed critical, this lifetime is determined principally by the mean life of the delayed neutron emitters, whereas, if the system is prompt critical the lifetime becomes essentially the time from birth to death of a neutron emitted promptly in fission. Since the latter lifetime is extremely short, $\sim 10^{-4} - 10^{-8}$ sec., the neutron population will increase at a rapid and uncontrollable rate, resulting in a "criticality accident." The system becomes prompt critical when $\frac{k_{eff} - 1}{k_{eff}} = \beta_{eff}$ where β_{eff} is the effective delayed neutron

fraction. In the case of uranium, this would mean the system would become prompt critical with $k_{eff} \simeq 1.007$ or with k only slightly above unity.

2. K_{eff} As An Index Of Criticality

As criticality is approached, or as the size is increased, for any given concentration, k_{eff} will increase and approach unity; k_{eff} is, therefore, an index of criticality. A pertinent question, applicable to any system, concerns the value of k_{eff} for any given fraction of critical mass. In criticality safety analysis, it is common to evaluate safety in terms of a given value of k_{eff}; i. e., the system is safe, providing k_{eff} does not exceed 0.9 or 0.95, etc. A problem arises because there is no general consistency between k_{eff} and fraction of critical mass except at the point of criticality (where k_{eff} = unity). Two different systems which have the same fraction of critical mass may have different values of k_{eff}. Stating it another way, for a specified value of k_{eff} on two systems (with different fuel compositions), one system may have a higher fraction of critical mass and be "less safe" than the other. This fact has perhaps not always been fully appreciated by those in the field.

The weird-complex variation in k_{eff} of fraction of critical mass vs. critical mass is shown in Figure 2, where k_{eff} has been computed by R. D. Carter, et. al., for two cases; 50% of a critical mass and 75%, spanning the range of concentrations from Pu metal (19.6 g Pu/cc) to dilute aqueous solutions (0.01 g Pu/cc) for both unreflected and water-reflected systems.[1]

Figure 3 gives k_{eff} of the fractional critical cylinder diameter vs. the critical cylinder diameter.[1] In the case of the unreflected cylinders, where the diameters are 85% of the critical values, k_{eff} is seen to vary from about 0.96 to 0.86 throughout the range covered by the calculations. Since the

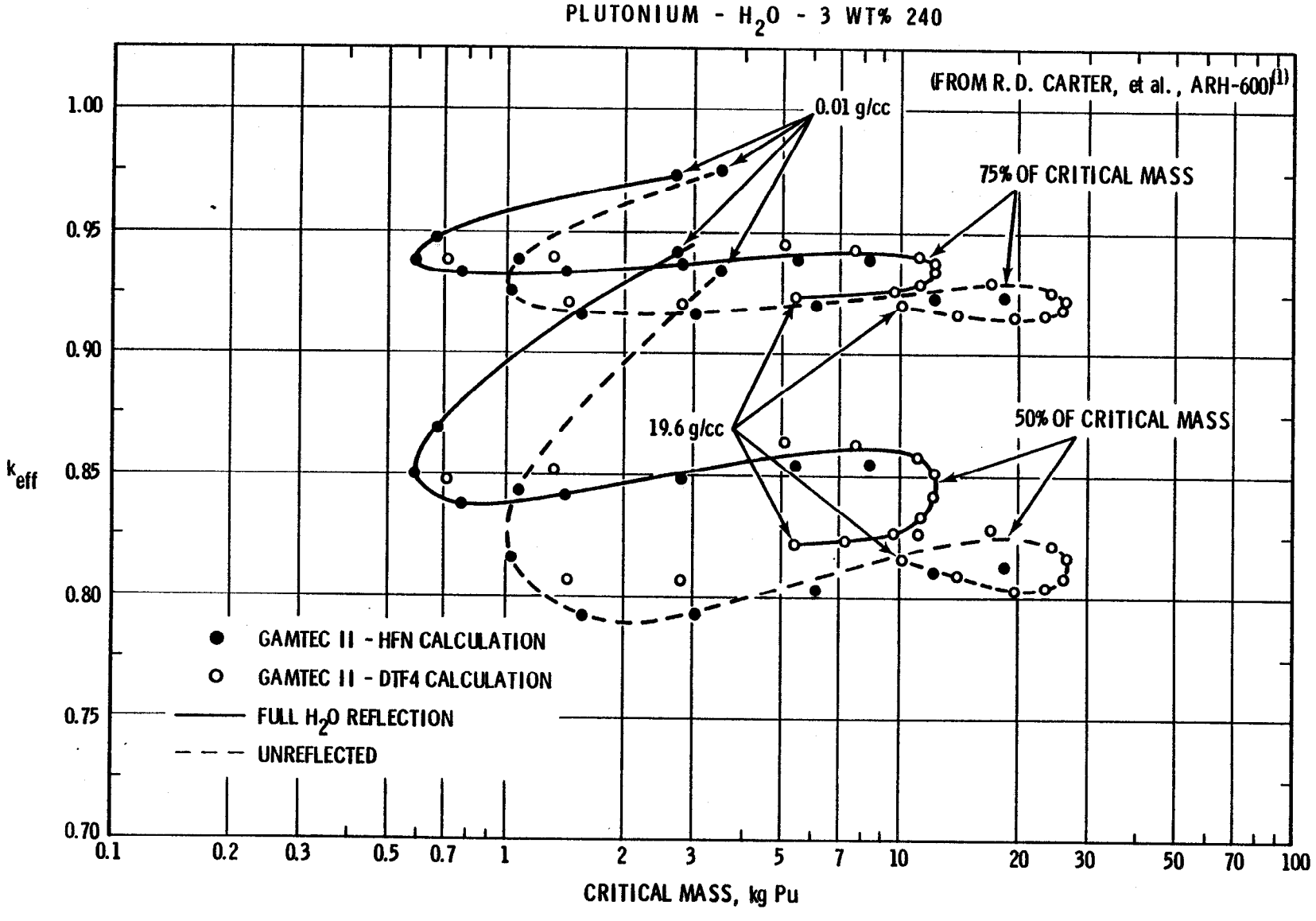


Figure 2. K_{eff} of Fractional Critical Mass vs. Critical Mass

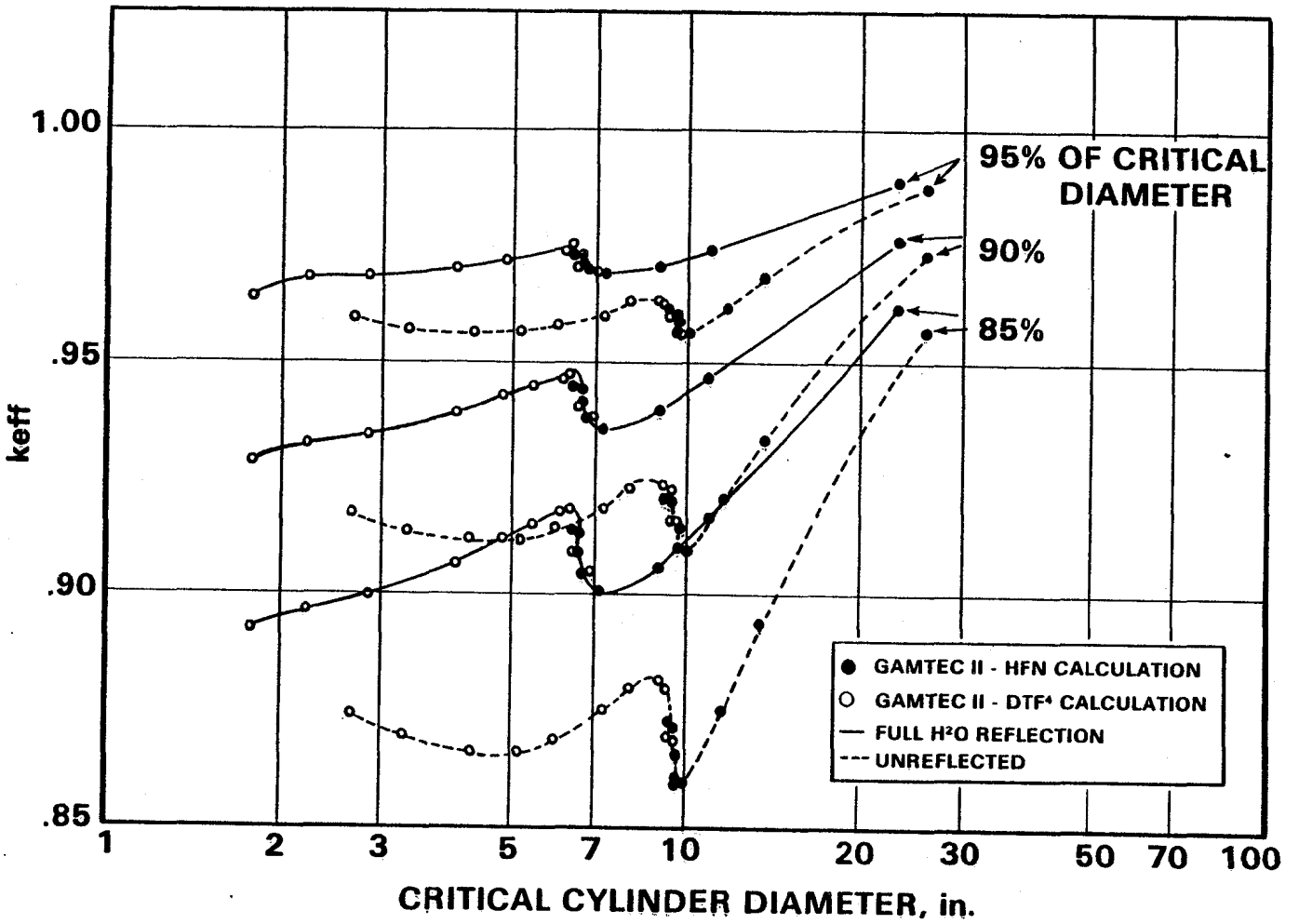


Figure 3. K_{eff} of Fractional Critical Cylinder Diameter vs. Critical Cylinder Diameter

critical cylinder diameter depends on the Pu concentration, the k_{eff} of cylinders with the same fraction of critical diameter also varies with concentration - but in a highly non linear fashion.

3. Successive Generations and Source Multiplication

If N_1 be the number of neutrons in the first generation, then the number in the nth generation will given by: $N_n = N_1 k^{n-1}$ where k is the effective multiplication constant.

The count rate observed during the construction of a critical assembly is the sum of the source neutrons, plus those arising from fissions caused by the source neutrons and by the progeny of neutrons born in earlier fissions. If Co is the count rate in the absence of any fissionable material, then in simplest terms:

$$Ct = Co + Co\ k + Co\ k^2 \quad \ldots Co\ k^{n-1}.$$

When the value of k is less than unity, the preceding may be written as,

$$\frac{Ct}{Co} = \frac{1}{1 - k} = \text{Source Multiplication.}$$

As k approaches unity the source multiplication becomes infinite.

During an approach to criticality, the reciprocal of the observed multiplication, $\frac{Co}{Ct}$, may be plotted against one of the controlling variables. Extrapolation of the plot to zero intercept yields the critical value of the variable, or the point at which k becomes unity. Although conceptually simple, a precise measurement of M is difficult to carry out, in practice, on a subcritical system. The observed multiplication depends on the location of the source and its distribution. It is,

therefore, generally not practical to evaluate k by means of the observed multiplication. The following table is presented for purposes of illustration.(6)

TABLE I

k_{eff} and Computed Multiplication with ^{252}Cf Point Source

k_{eff} Without Source	Concentration g/ℓ	Core Radius cm	Multiplication With Source
	$^{233}U\text{-}H_2O$		
0.98	20	23.76	100.1
	30	17.47	94.8
	40	15.06	91.3
	50	13.74	86.0
	60	12.90	84.7
	80	11.88	82.8
	100	11.27	81.3
	200	10.00	75.5
0.99	20	24.33	240.6
	30	17.76	216.1
	40	15.28	203.0
	50	13.93	186.8
	60	13.07	174.4
	80	12.03	171.1
	100	11.41	165.9
	200	10.12	146.9
	$^{235}U\text{-}H_2O$		
0.98	13.3	55.37	98.5
	35.1	17.75	88.2
	50.7	15.24	84.8
	85.8	13.15	82.7

The calculations of Table I were made for homogeneous uranium-water mixtures over a range of uranium concentrations corresponding to systems with k_{eff} = 0.98 and 0.99. In all cases, the multiplying core (a subcritical sphere) was surrounded by a water reflector of at least 4 cm thickness.

The neutron multiplication (ratio of total source to fixed source) is considerably higher than would be calculated from the expression,

$$M = \frac{1}{1 - k_{eff}} ,$$

where k_{eff} is the effective multiplication constant. The multiplication is higher, because with the $^{252}_{98}Cf$ fission neutron source positioned at the center of the core, the neutron flux is more sharply peaked in the center of the assembly, with the result that the leakage is smaller.

In practice, the approach to criticality utilizing an external source will involve a flat source on which is imposed the point source. The flat source is the result of α,n and γ,n reactions, and of spontaneous fission, which has nothing to do with the process of criticality itself. (For example, the neutron emission from spontaneous fission in ^{240}Pu is at the rate of about one million neutrons/kg/sec. - and all Pu contains varying amounts of ^{240}Pu.)

In principle, the value of k_{eff} may be determined from the ratio of the number of neutrons in successive generations, but this, also, is difficult to accomplish.

4. Variation of Critical Mass With Sphere Radius for Homogeneous ^{239}Pu - Water Mixtures

All factors which influence the interaction of neutrons with matter affect criticality. The following curves (Figure 4), show the complex variation of critical mass with sphere radius for homogeneous ^{239}Pu-water mixtures, and serve to illustrate several effects.[7] The curves show critical radii of spheres, and critical masses of plutonium contained therein, as a function of water dilution. The upper curve is for bare, homogeneous plutonium-water spheres and the lower one for plutonium-water spheres immersed in water. Striking changes are seen to occur in the critical mass as the plutonium is diluted with water. Beginning with alpha-phase plutonium metal (ρ = 19.6 g/cm^3), the critical mass and radius both increase on dilution with water. The mass passes through a maximum value at an H/Pu ratio of about 4 (Pu density $\sim$ 5 g/cm^3).

The effect of partially moderating (or slowing down) the fission neutrons causes a significant reduction in the value of η (number of neutrons produced per neutron absorbed in Pu), due to the change in the ratio of the neutron capture and fission cross sections with neutron energy. In addition, the dilution of the metal with water also decreases the density of Pu and increases the neutron leakage. The system must then be made larger to maintain a balance between production and losses of neutrons. On further dilution, moderation by the hydrogen in water becomes increasingly more effective and the probability for fissioning with slow neutrons is enhanced. The effect of adding water is seen to cause a further increase in leakage (and critical size), but the net overall result is a decrease in mass, due to the reduction in Pu density. Finally, on further dilution, or moderation, an optimum condition for production and leakage is obtained, such that the combination results in the smallest critical mass. At this point, the Pu concentration has been reduced to 32 g Pu/ℓ (H/Pu

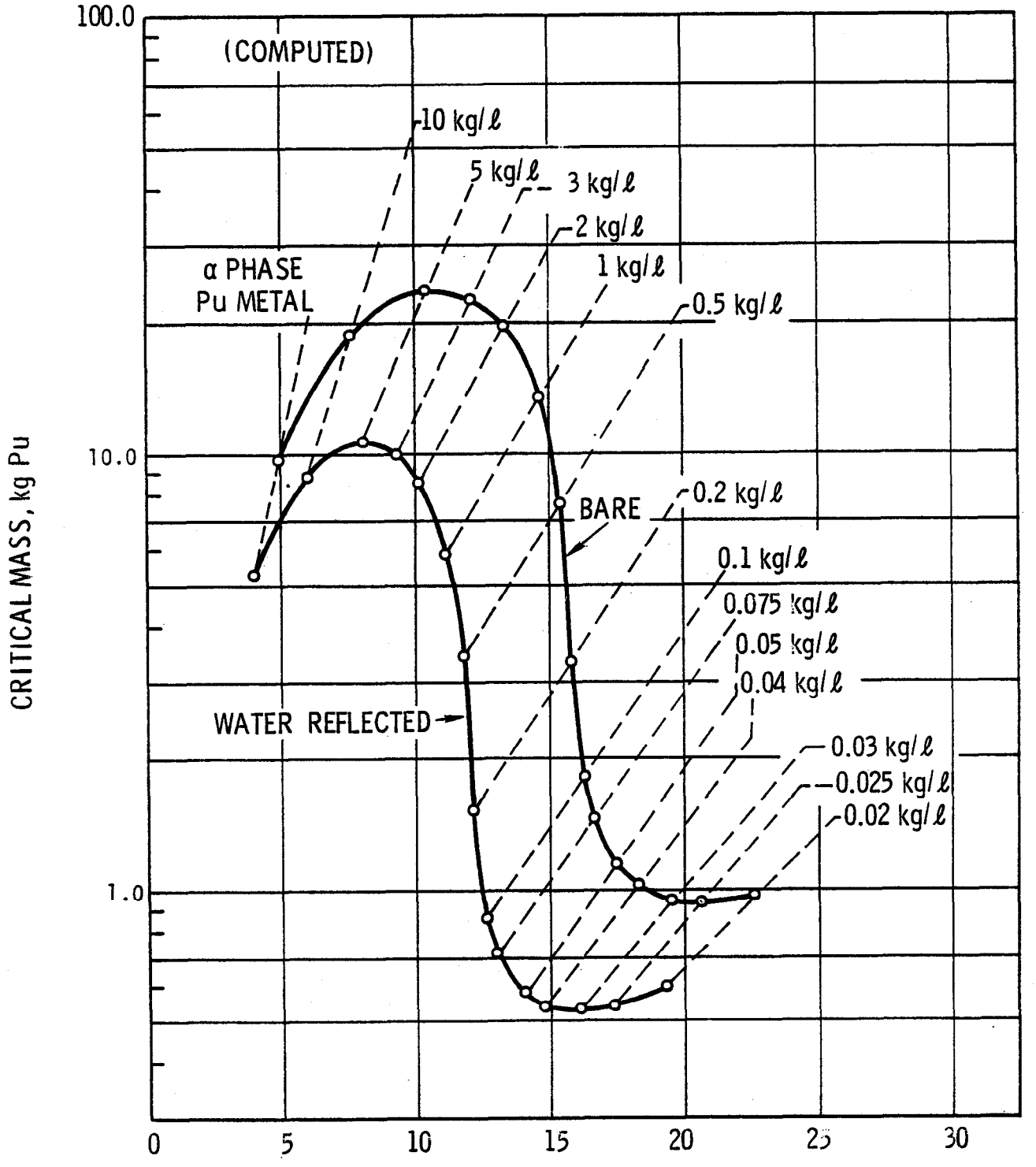


Figure 4. Estimated Mass and Radius of Critical Plutonium-Water Spheres

atomic ratio ∿ 900). The critical solution volume is about 75 times larger than that for undiluted Pu metal, but the quantity of Pu contained in the sphere is only about 1/10 the metal value. This condition of "optimum moderation" gives a minimum critical mass for the water-reflected sphere that is ∿ 530 g Pu.[8] Finally, both the critical radius and mass increase once again on further dilution with water, due to increasing neutron absorption in the water (principally in the hydrogen). Both become infinite at a plutonium concentration of 7.19 ± 0.1 g Pu/ℓ (H/Pu ratio of ∿ 3680).[9] At this point, about half the neutrons released in fission are absorbed in the diluent.

Figure 4 shows, also, the effect of neutron reflection. For the sphere immersed in water, some of the neutrons which would otherwise escape are reflected (scattered) back into the sphere, reducing the leakage. The curves show the difference in critical radii brought about by the water reflector.

Figure 4 illustrates, also, that the same critical mass could be achieved with three different Pu concentrations, but that the critical volume of size would differ in each case.

5. A Triple Point in Criticality (Identical Critical Volume But Three Different Critical Concentrations)

Figure 4 was used to illustrate some of the factors (such as moderation and reflection) that affect the criticality of systems containing a single actinide isotope (for purposes of illustration, ^{239}Pu).

The system becomes inherently more complex for mixtures of isotopes. The second most prevalent isotope of plutonium is ^{240}Pu. The effect of the ^{240}Pu isotope on the criticality of ^{239}Pu-^{240}Pu

mixtures is shown in Figure 5.[10] Calculations indicate that ^{240}Pu could, by itself, be made critical under certain conditions - specifically those under which there would be no moderation by a diluent. The interaction of a thermal neutron with ^{240}Pu results principally in scattering or the formation of ^{241}Pu, since the fission cross section for slow neutrons is negligible. Thus, the effect of ^{240}Pu on the criticality of ^{239}Pu will be largely dependent on the neutron spectrum, which in turn is determined by the concentration and type of diluent present. The curves of Figure 5 show the effect of ^{240}Pu on the critical radius, and clearly indicate the existence of triple points of criticality. The effect is more clearly portrayed (schematically) in Figure 6.

Note that for a given radius (or fixed volume) there may now be as many as three different critical concentrations! The system would then oscillate between regions that are subcritical and supercritical as a function of the fuel concentration. This is brought about by the mere addition or removal of fuel that changes the hydrogen-to-fuel ratio and, consequently, the neutron spectrum.

6. Identical Critical Mass at Four Concentrations

Critical mass calculations are presented in Figure 7 for mixed oxides of U and Pu in which the Pu contains 25 wt% ^{240}Pu.[11] If the peculiar curve shape at the higher concentration range is correct (note curves for 15 and 30 wt% Pu in U) the results imply that it would be possible to achieve the same critical mass at four different concentrations of mixed oxides in water. In this case there would be four different volumes having the same critical mass; whereas, in the previous example (Figure 6) there was one critical volume at three different fuel concentrations.

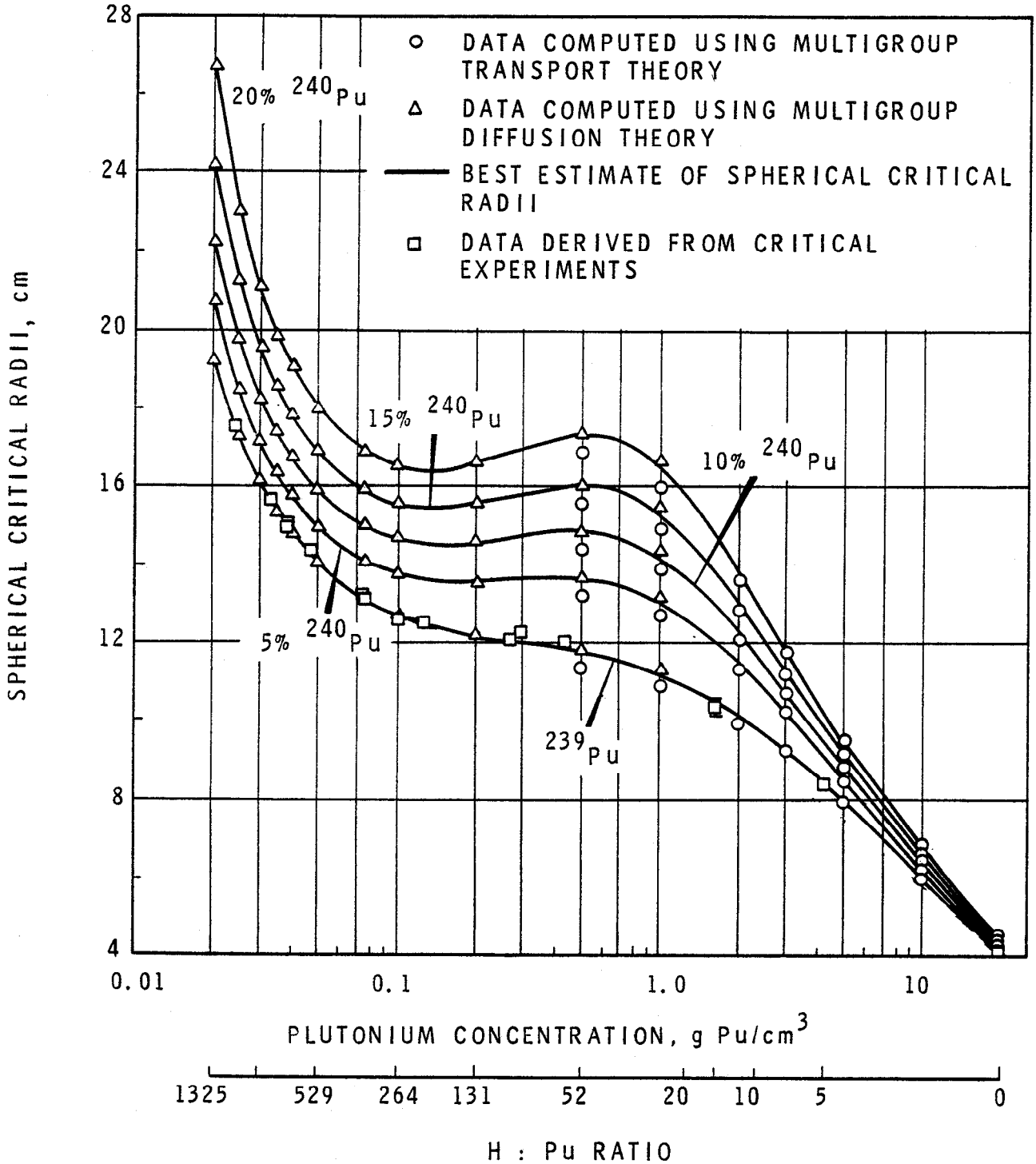


Figure 5. Water Reflected Spherical Critical Radii of Pu(Metal)-Water Mixtures

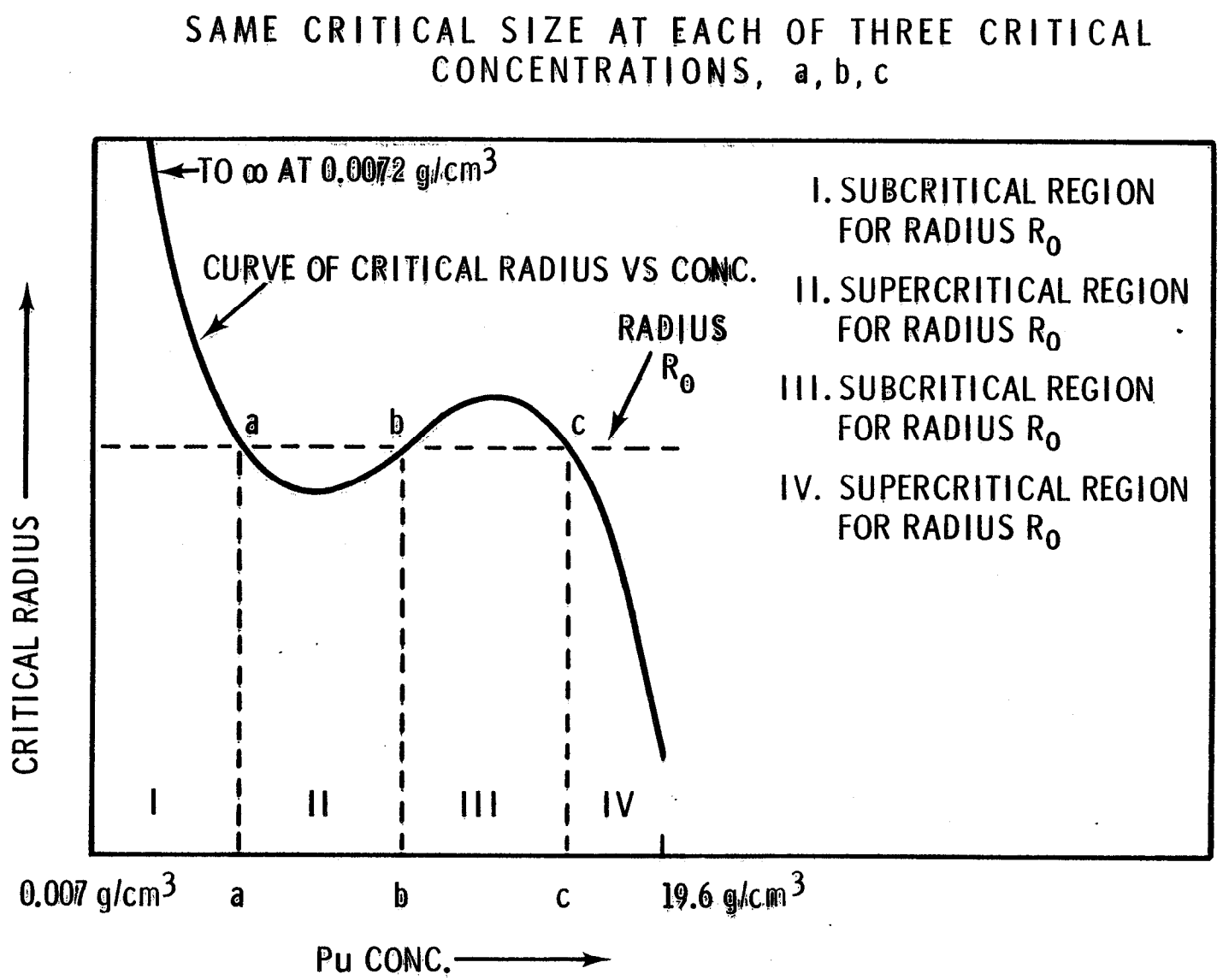


Figure 6. Illustration of Triple Point of Criticality

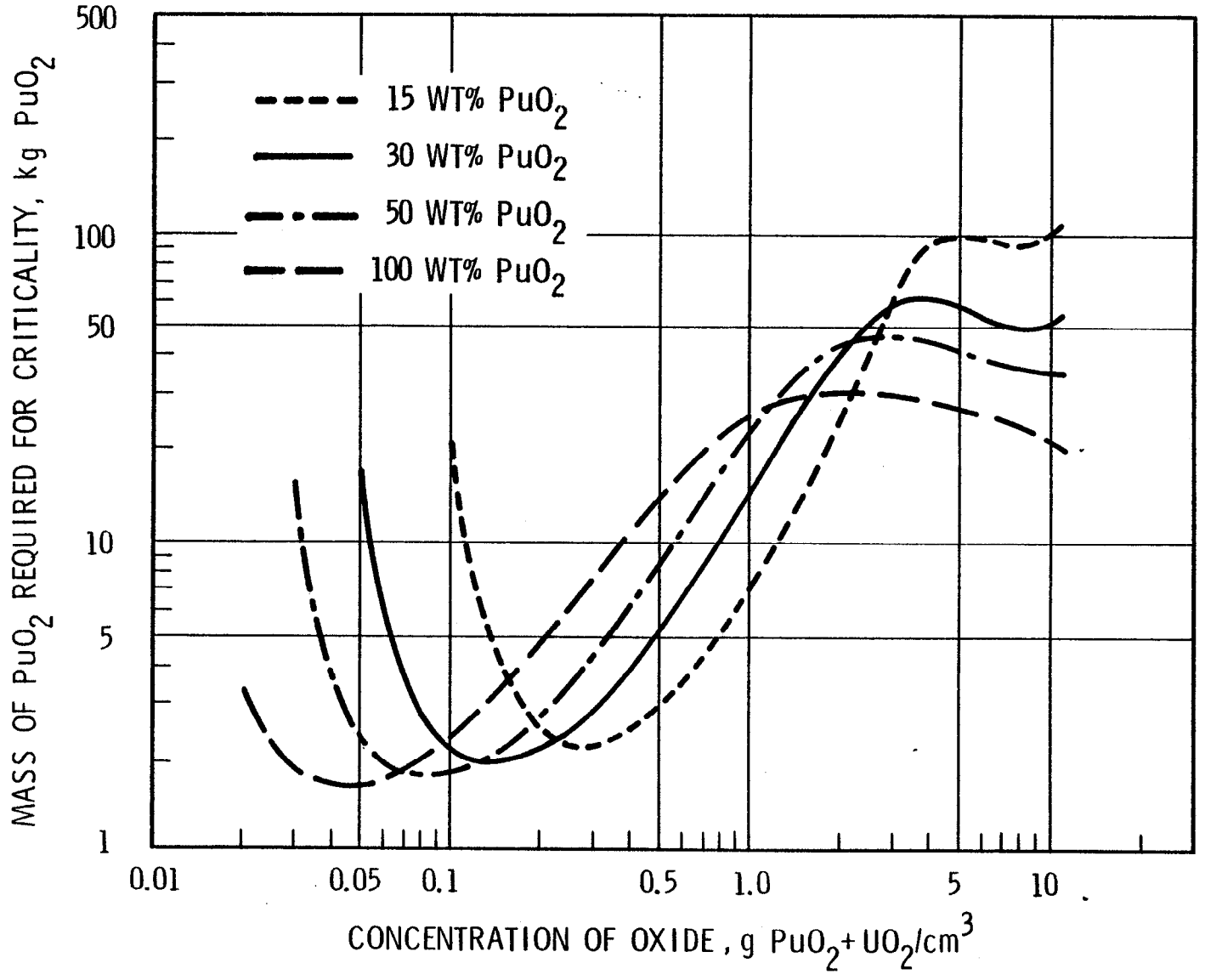


Figure 7. Computed Reflected Spherical Critical Mass of PuO_2-UO_2-Water Mixtures With 25 Isotopic Percent ^{240}Pu

7. Limiting Critical Enrichment of Uranium for Aqueous Homogeneous Solutions

A series of calculations by B. M. Durst of Battelle - Pacific Northwest Laboratories, are presented in Figure 8 on the values of k_∞ for UO_3 - water mixtures beginning with natural uranium extending through various ^{235}U enrichments up to highly enriched uranium. The curves illustrate the increase in k_∞ with ^{235}U content, or enrichment, and show the range of uranium concentrations, or $H/^{235}U$ atom ratios, over which criticality would be possible in homogeneous UO_3 water mixtures.

In the case of 30 wt% ^{235}U enriched uranium and higher enrichments, it is evident from the figure there can be as many as three different H/X atom ratios, or uranium concentrations, that yield the same value of k_∞.

For homogeneous uranium-water solutions, there is one enrichment for which criticality is possible with only one hydrogen - ^{235}U atom ratio; this is the limiting enrichment for criticality. From k_∞ measurement data, the enrichment was found to be 1.035 ± 0.010% ^{235}U.[12] At this particular enrichment, the largest value that can be obtained for the reproduction factor, k_∞, for an infinite system, under optimum conditions of moderation is unity.

8. Thirty-Six Years and the Criticality of Intermediate Uranium Enrichments (What is Known)

Data on the criticality of uranium in the intermediate enrichment range ($\sim$ 6 wt% to $< \sim$ 93 wt% ^{235}U) from which to deduce subcritical limits for criticality control are limited.[i] It is indeed surprising (perhaps tantamount to an anomaly) that in the intervening period of some 36 years since the first man made nuclear chain reaction (Dec. 2, 1942) that few data have become available, nor is this intermediate

enrichment range adequately covered in the "American National Standard for Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors," N16.1-1975 (Revision of N16.1-1969), American Nuclear Society, (April 1975).[13] The data on uranium are for uranium enriched to no more than 5% ^{235}U, and then for high enriched uranium. It should be noted that the critical mass for uranium enriched in ^{235}U to 6 wt% or less is lower for a heterogeneous system than a homgeneous system; the critical volume also is smaller for the heterogeneous system. For enrichments above 6 wt%, however, the minimum critical mass for uranium in an aqueous solution will be less than the minimum achieveable for a lattice of rods immersed in water. In this case the smallest critical mass is found to occur with rods of vanishingly small diameter, or of zero diameter. On the other hand, although the minimum critical mass may be less for the homogeneous case, (for enrichments above 6 wt%), the minimum critical volume will not be. It is then possible to achieve criticality in a smaller volume with a heterogeneous system, for example with fuel rods in water, but the critical mass will be greater than the minimum value for uranium in a homogeneous aqueous solution at the same uranium enrichment. The type of system giving rise to the smallest critical volume and mass including intermediate enrichments is summarized in Table II.

(i) Subcritical limit is defined basically as the limiting value assigned to controlled parameter that results in a system known to be subcritical . . . See Reference No. 13.

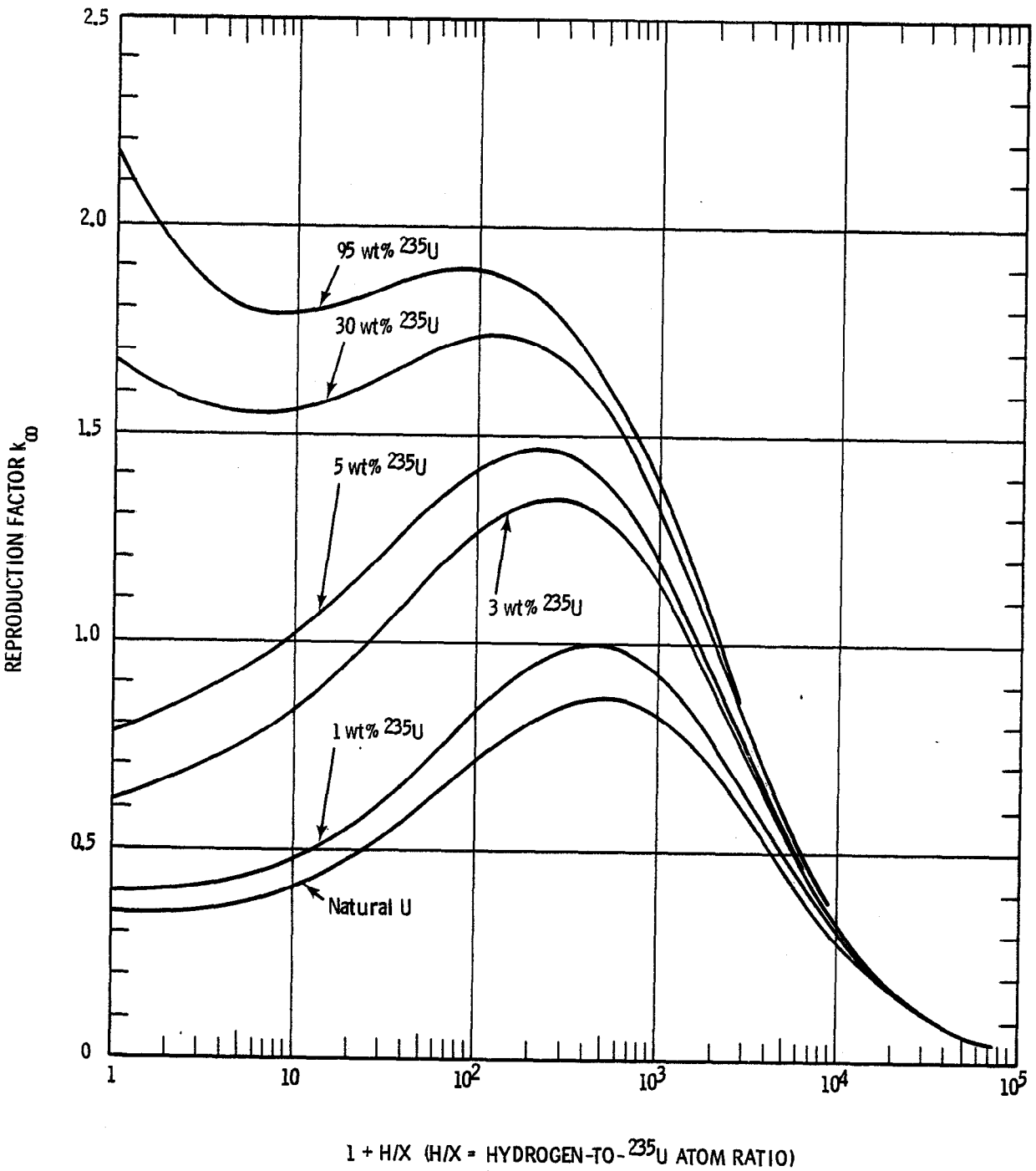


Figure 8. Computed Values of Reproduction Factors for Homogeneous UO_3 Water Mixtures at Various ^{235}U Enrichments

TABLE II

Type of Water-Reflected Uranium System That Gives The Smallest Critical Volume and The Smallest Critical Mass Including Uranium of Intermediate Enrichments

Uranium Enrichment	Type System Giving Smallest Critical Volume	Type System Giving Smallest Critical Mass
0.71 wt% to ∿ 6 wt%	Hetrogeneous[(a)]	Heterogeneous[(a)]
> ∿ 6 wt% to ∿ 34 wt%	Heterogeneous[(a)]	Homgeneous[(b)]
> ∿ 34 wt% to 100 wt%	Single Metal Unit[(c)]	Homogeneous[(b)]

(a) Such as an array of fuel elements (of optimum diameter) positioned in water (at optimum spacing) and reflected with water; note that minimum volumes and minimum masses will occur at different spacings.

(b) Uniform aqueous mixture of uranium and water at that concentration giving the minimum mass, and reflected with water.

(c) Single units of metal at theoretical density (18.9 g/cc) reflected with water.

In view of the lack of appropriate critical experiment data, calculations have been made by R. A. Libby of Battelle - Pacific Northwest Laboratories to provide an estimate of minimum critical volumes for uranium in the intermediate enrichment range (in the range from $\sim$ 6% to $< \sim$ 93%) as shown in Figure 9. These are calculations of the minimum critical volumes applicable to uranium systems regardless of the size and shape of the uranium as reflected by an unlimited thickness of water. The region of the curve beyond about 6 wt% ^{235}U is the area wherein lack of data prevails, which actually includes most of the possible enrichment range beyond natural uranium. Based on these calculations, a possible subcritical limit curve, covering the intermediate enrichment range, would appear somewhat as indicated on the figure in the lower dashed curve.

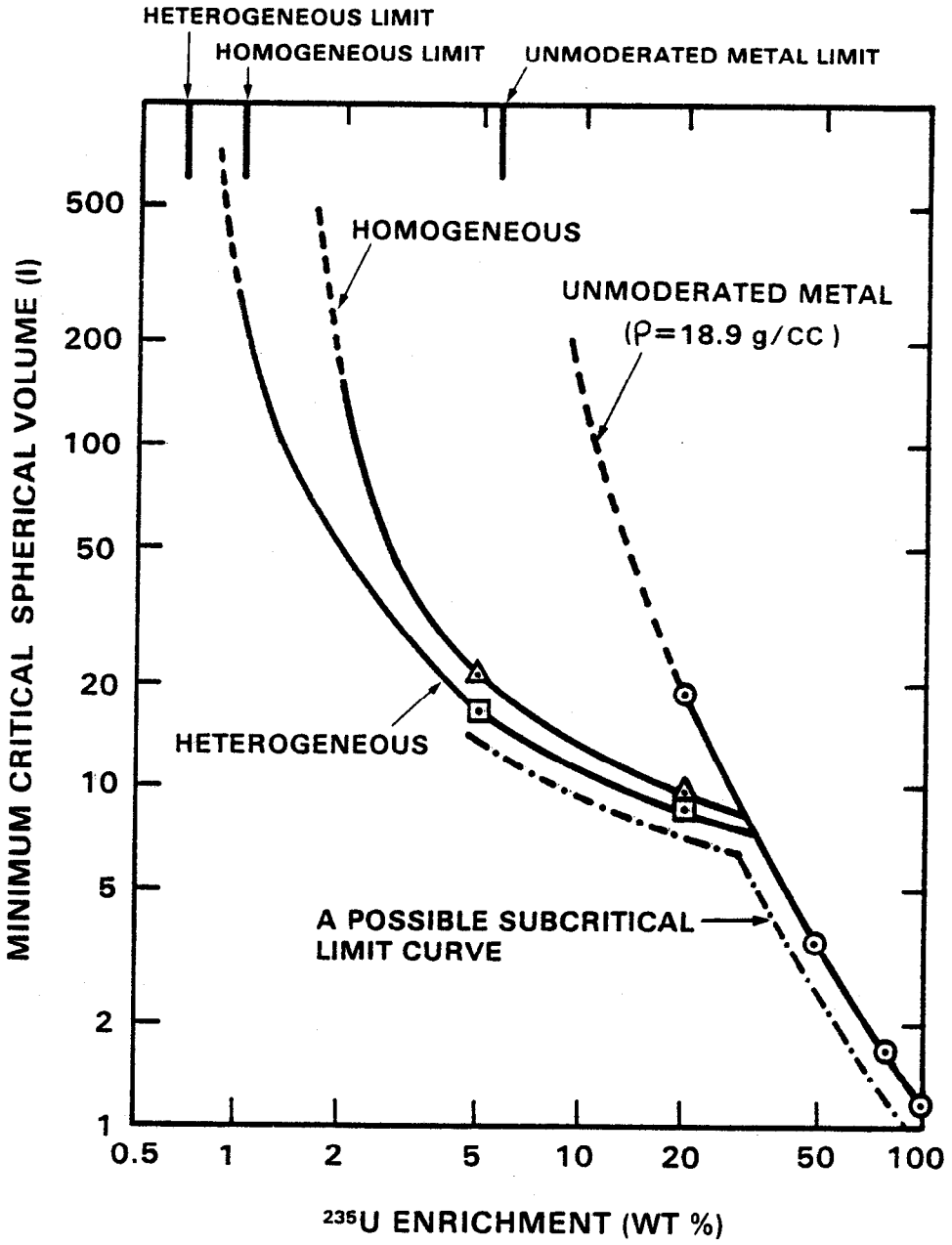


Figure 9. Minimum Critical Volume vs. Uranium Enrichment

C. COMMENT ON CRITICAL CONCENTRATIONS FOR ^{233}U, ^{235}U AND ^{239}Pu (CAN THE LIMIT OF ANY ONE BE SAFE FOR ALL THE OTHERS?)

Of the above three isotopes, ^{239}Pu has the smallest "infinte sea" or limiting critical concentration in water (that concentration for which k_∞ becomes unity in an infinite sea of water). The value is 7.19 ± 0.15 g/ℓ (H/Pu atomic ratio $\simeq$ 3680).[9] Fuel processing operations involve cylindrical vessels, for the most part, which are of such diameter as to preclude criticality for the concentrations of nuclear materials contained therein. These "safe by geometry" vessels may contain many times the minimum quantity of U or Pu that could potentially be made critical in some other geometry; for example, a water-reflected sphere of the proper diameter.

Estimated critical concentrations are presented in Table III as a function of cylinder diameter. On an a priori basis, could a plant that is "safe by geometry" for one of these three isotopes be considered inherently safe for either of the others? The anser is "no"! Note that Pu has the smallest limiting critical concentration of the three. It also has the smallest minimum critical mass in an aqueous solution and the smallest mass when in the form of the metal (reference Table VIII). The critical ^{235}U concentrations are, however, smaller than those of ^{239}Pu, by up to a factor of $\sim$ 3, for cylinders in the 5 - 1/2 to 6 inch diameter range; but below about 5 - 1/2 inch the critical concentrations exceed those for ^{239}Pu. For cylinders about 7 inch diameter and smaller, the critical concentration for ^{233}U is significantly less than either that of ^{235}U or ^{239}Pu, but the critical diameter for ^{239}Pu metal will be less than that of ^{233}U.

TABLE III

Estimated Critical Concentrations of Fissile Isotopes in Infinite Length, Water-Reflected Cylinders[1, 4]

Cylinder Diameter (in.)	^{233}U (g/ℓ)	$^{235}U_{93.2\%}$ (g/ℓ)	^{239}Pu (g/ℓ)
∞	11.3	11.8	7.19
8.0	42	58	37
7.0	57	90	65
6.5	70	125	100
6.0	90	200	450
5.75	110	290	850
5.5	140	1000	1100
5.0	230	3000	1900
4.5	780	6000	2900

In early days it was sometimes suggested (usually by persons somewhat unfamiliar with the complexities of neutron interaction) that scaling factors might be developed from the more extensive data that was available on ^{235}U solutions, that could be used to provide critical concentrations, or safe subcritical limits, for Pu. The idea was to perform several critical experiments on a vessel with ^{235}U solutions and then repeat the process with Pu solutions. From the results "scaling factors" might then be developed. It is a good thing that this procedure was not attempted, as the data of the Table show no consistent scaling factor to exist. The differences in variation of critical concentrations are due to the variations in eta, and in the cross sections, with changing spectrum which also depends on the concentration or H/X ratio of the fissile isotopes in the aqueous solutions.

D. THE CUBE AND THE SPHERE

Since the ratio of surface area to volume is a minimum in the case of a sphere, and since neutron production depends on volume and neutron leakage on surface area, the sphere can be expected to have the smallest critical volume of any shape. There are data, however, that indicate that a reflected cube might, under some circumstances, have a smaller critical volume (and mass) than if the fissile material were in the form of a sphere. This somewhat surprising result stems from experiments performed with PuO_2-plastic compacts arranged in cubic geometry and reflected with Plexiglas.[14] [15]

For PuO_2 at an H/Pu ratio of 0.04 (essentially unmoderated) the analysis indicates that a reflected cube would have a critical volume about 14% less than that for the reflected sphere. However, the phenomenon is not so pronounced that the apparent anomaly could not result from inaccuracies in the measurements. Monte Carlo calculations have been made by S. R. Bierman of Battelle - Pacific Northwest Laboratories, utilizing the KENO code[16] on a reflected cube and a reflected

sphere of unmoderated PuO_2 having precisely the same volumes; the results show the cube, in this case, to have a higher k_{eff} (about 1%) and tend to support the above, but the statistical uncertaintly in the Monte Carlo calculations rules out firm conclusions. However, in examining data from a number of other experiments involving cubes of Pu-bearing fuels, it is to be noted that the effect (ratio of critical sphere volume to critical cube volume) is uniformly dependent on the H/Pu ratio, or degree of moderation, as is evident from Figure 10. In the case of well moderated (and larger systems) the reflected sphere does, as expected, have a critical volume or mass about 20% less than that of the reflected cube, and the Monte Carlo calculations are in support of these results. It also has been concluded that for some undermoderated mixtures of U(93.5% enriched) and water, a right circular cylinder with height-to-diameter ratio (h/d) of about 0.9 may have a slightly smaller water-reflected critical volume than a sphere.[4] [17] This same effect also was reported to have been seen in the results of some two dimensional neutron transport calculations made by G. E. Hansen.[17] These conclusions lend additional support to our conclusions regarding the cube and the sphere. In passing, it may be of interest to note that, in practice, materials are more likely to be encountered in the form of rectangular parallelepipeds or cylinders than in the form of spheres.

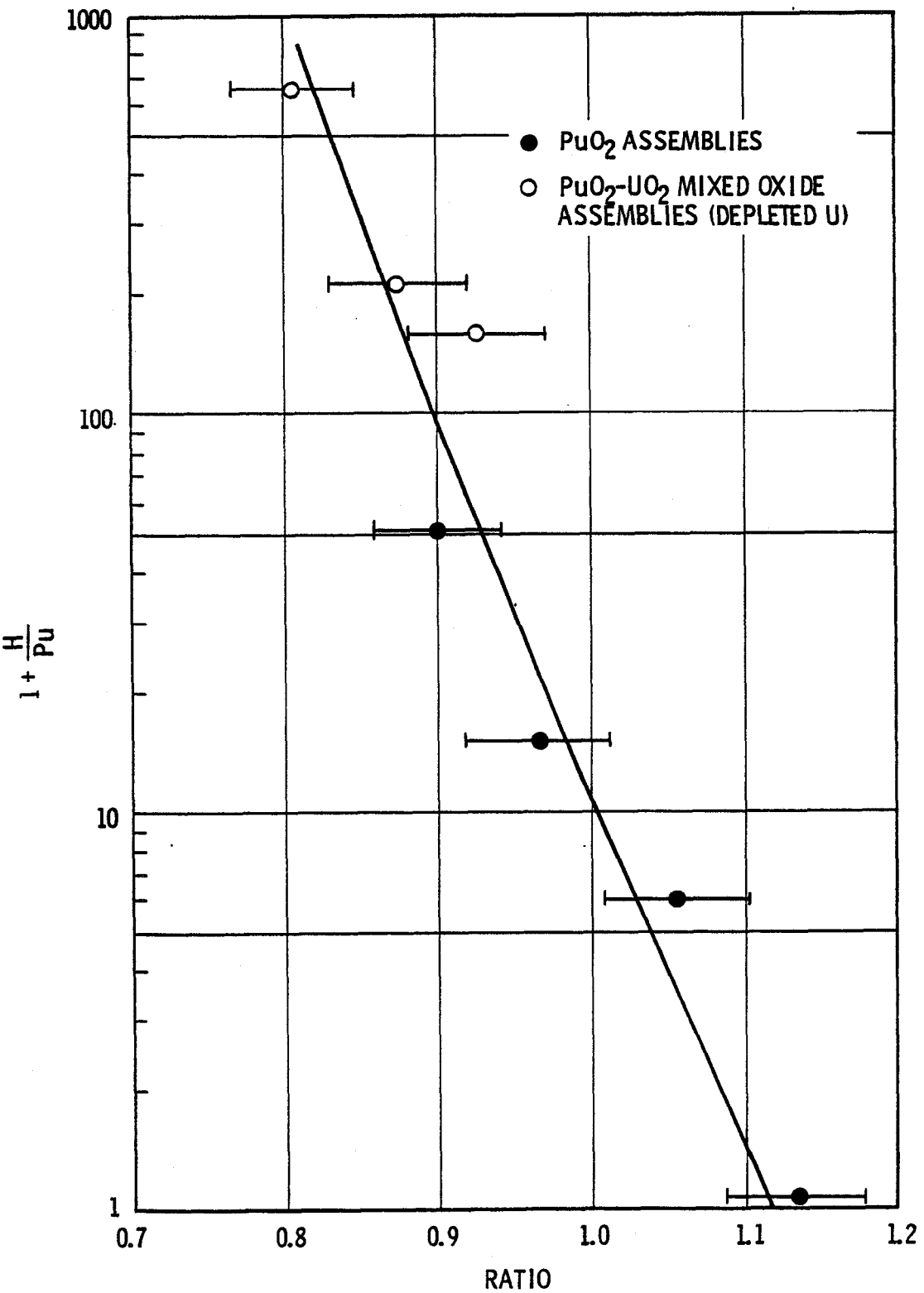


Figure 10. Ratio of Critical Sphere Volume to Cube Volume - Plexiglas Reflected Assemblies

E. THE CRITICALITY OF LARGE BILLETS VS. SMALL RODS - CONDITIONS FOR MINIMUM MASS (Trangular vs. Square Lattice)

Data from critical experiments have been reported on large uranium metal cylindrical rods and annuli immersed in water wherein both triangular and square lattice patterns were used.[18] [19] See Figure 11. The experiments were performed at two uranium enrichments, 1.95% and 3.85% ^{235}U. Data were obtained with the 1.95% enriched uranium in the form of cylindrical annuli, 7.2 in. O. D., 2.6 in. I. D. In the case of the 3.85% enriched uranium the outside diameters of the annuli were 7.2, 6.2, and 5.2 in., with inside diameters of 2.6 in. Solid rods slightly less than 2.6 in. in diameter could be inserted into these annuli to produce effectively solid rods of each of the three outside diameters. All rods and annuli were 30 inches long.

It was noted that arrangement of the units of both U(1.95) and U(3.85) in both triangular and square lattice patterns resulted in significantly different quantities of uranium required for criticality. The number of rods required for criticality in a square pattern of the U(1.95) cylindrical annuli, 7.2-in. O.D. and 2.6-in. I.D., was a factor of 2 greater (at optimum moderation) than that for these annuli arranged in triangular patterns. In the case of the U(3.85), the same effect was observed but the magnitude was reduced to a factor of about 1.3 for the larger diameter annuli. However, for the same outside diameter (7.2 in.), the minimum critical number of solid rods was a factor of 1.7 greater when arranged in a square pattern than when in a triangular pattern.

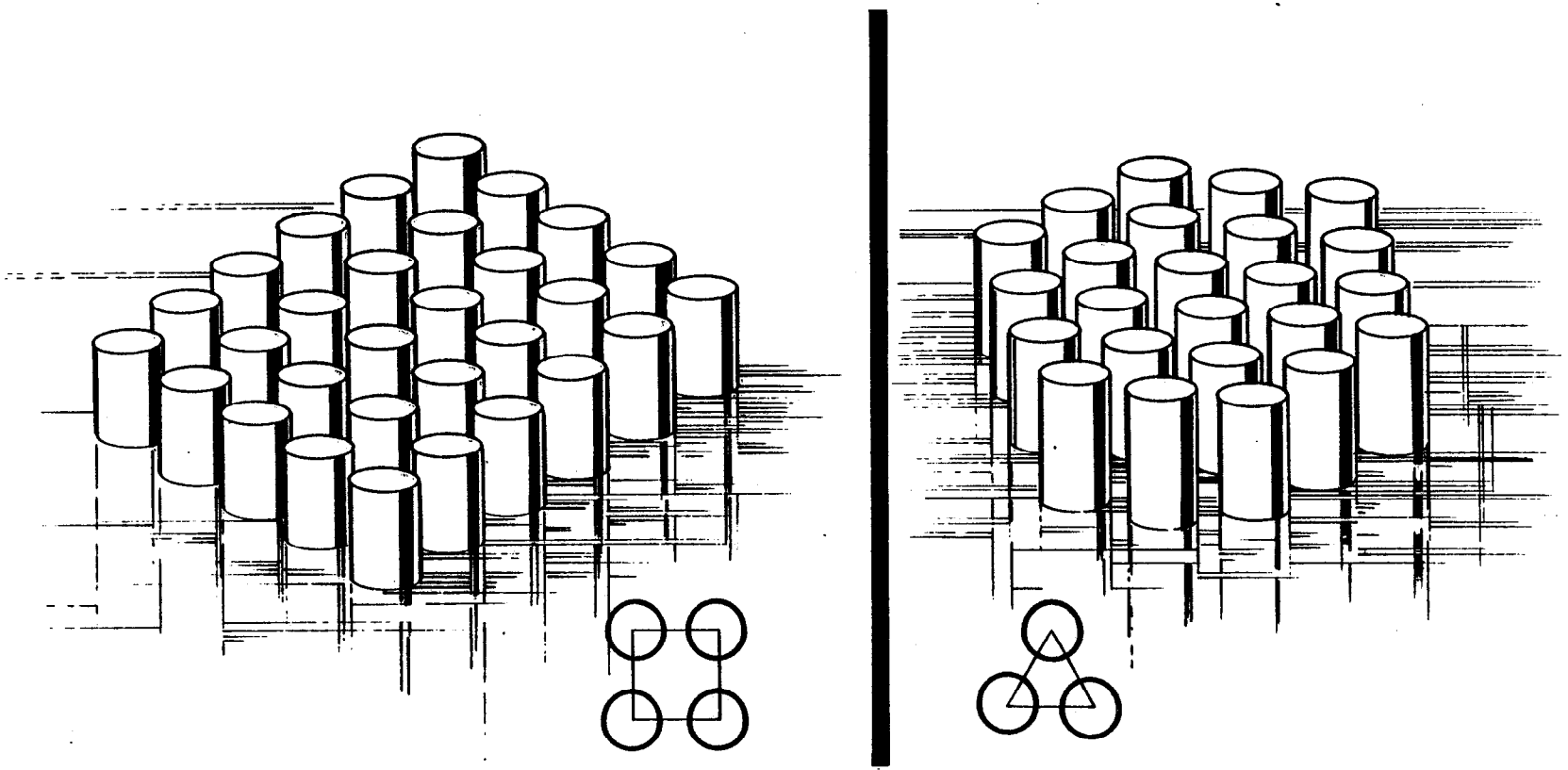


Figure 11. Square Lattice vs. Triangular Lattice Pattern

Data are presented below which show the percent difference in the minima observed for the U(3.85) both as annuli and as rods, when arranged in each pattern.

TABLE IV

Comparison of Minima for Lattices of U(3.85)
Arranged in Square and Triangular Patterns

Outside Diameter of Unit (in.)	Change Between Patterns %	
	Annuli	Rods
7.2	34	74
6.2	23	60
5.2	25	33
2.5	--	2

As the rod size decreases the difference becomes insignificant - but prior to these experiments on large billets there was no evidence to indicate that such a large difference in minima would prevail between the square and triangular pattern at the larger rod size. Attempts to calculate the critical lattices with such large units have proved marginal.

Then, in the case of large billets, and in the interest of criticality prevention, the triangular lattice, with its smaller mass, should be avoided.

F. LIMITING CRITICAL FUEL ROD CONCEPT. (WHEN LARGER IS BETTER - SAFER)

Questions concerning the criticality of large rods, slabs, and billets frequently arise in connection with fuel element fabrication, such as in the extrusion process in which fuel tubes are extruded from large billets.

In heterogeneous assemblies of uranium and water, lumping the fuel effects three of the factors, p, f and ε, entering in k. Lumping the fuel will cause k to increase on three counts and decrease on one. By lumping the fuel the fast neutrons will have a better chance of slowing down in the moderator and thus of passing through the resonance energy region before encountering ^{238}U than in the case of a homogeneous mixture of uranium and moderator. The most important effect of lumping, however, is due to the self shielding effect. Because of the large values of the absorption cross section for neutrons in the resonance energy region, the flux will be strongly self depressed in the uranium. Since this depression, or minima in the flux, is caused by the absorption itself, the total absorption in ^{238}U resonances will be greatly reduced if the uranium and moderator are separated; as under these conditions, the resonance flux will be at a minimum in the presence of the uranium.

Lumping the fuel also increases the probability of causing fission in ^{238}U before the neutron energy is degraded below the fast fission threshold by collision in the moderator; thus, ε (fast fission factor) will be increased. Lumping the fuel will, on the other hand, cause f (thermal utilization factor) to decrease. Neutrons that become thermal in the moderator, apart from the uranium, will have less chance of being absorbed in the uranium than if the uranium and moderator were intimately mixed. Also, self shielding of thermal neutrons takes place in the fuel lump, so that the uranium on the interior is in a lower thermal flux than would be the case for a homogeneous uranium-moderator mixture.

Criticality data are generally lacking for slightly enriched uranium fuel rods greater than about 2 in. diameter. The data available on large rods or billets consists of a series of exponential experiments with 3 in. diameter rods of 3.0 wt% U-235 enrichment made at the Savannah River Laboratory[20] and critical experiments performed at ORNL with 1.95 and 3.85 wt% ^{235}U large annual cylinders and solid rods.[18], [19] The outside diameter of the annual cylinders was 7.2, 6.2 and 5.2 in. and the inside diameter 2.6 in. In the case of the 3.85 wt% ^{235}U, experiments also were completed with solid rods of 2.6, 5.2, 6.2 and 7.2 in. diameter. The calculation of these large rods and billets proved only marginal. Both triangular and square lattice patterns were used in the experiments.

The Savannah River Measurements correlate reasonably well with the Hanford measurements at 3.06% U-235 at the smaller rod diameters of 0.175, 0.60, and 0.925 in.

By comparing these data, it is apparent that the maximum buckling for a given enrichment is a slowly varying function of rod diameter. For example, in the case of the 3% enriched uranium, the buckling for a 0.6 in. diameter rod is about $15,400 \times 10^{-6}$ cm.$^{-2}$, whereas for a rod diameter five times larger (a 3-in. diameter rod), the optimum buckling is still approximately $10,000 \times 10^{-6}$ cm.$^{-2}$. As the rod size further increases, the buckling is finally reduced to zero, and the critical mass becomes infinite.

An attempt has been made to estimate the largest diameter rods which can be made critical in a water lattice as a function of enrichment. The results are shown in Figure 12.[21] For 3% enriched uranium, the diameter is about 15 in.; for 1% the diameter appears to be about 4 in. For about 6% enriched uranium, the rod diameter for zero bucklings would be infinite. In the case of natural uranium, the rod diameter would appear to be about 1 in. for zero buckling.

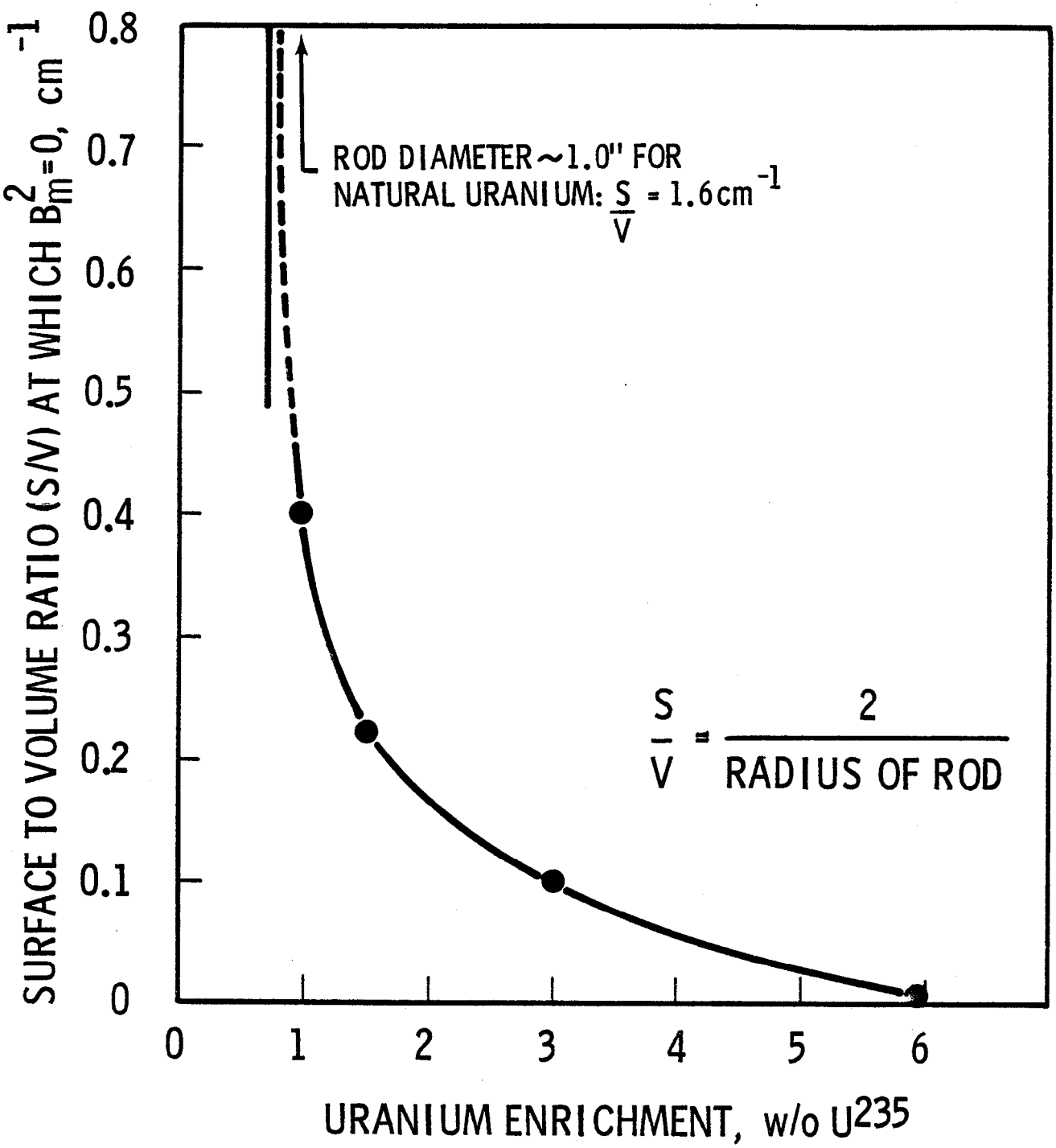


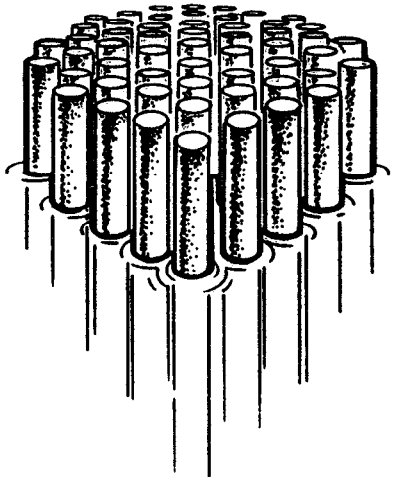
Figure 12. Estimated Surface-to-Volume Ratios of Large Rods Which Result in Zero Bucklings (Infinite Critical Masses)

Figure 12 illustrates the necessity for making nuclear safety reviews in operations involving large billets. The limit at approximately 6% represents an entirely fast system, whereas the limit for natural uranium would be a thermal system.

An interesting point of the limiting fuel rod concept is that criticality of slightly enriched uranium could be prevented (under water immersion) if only the enriched uranium fuel rods were large enough. An illustration of this is provided in Figure 13 wherein a finite number of slightly enriched uranium fuel rods are depicted as being critical if spaced properly in water, but if these same fuel rods were bundled tightly together (so as to effectively preclude water moderation on the interior of the fuel bundle, or if water were excluded from the bundle) and if each fuel bundle were of sufficient size (diameter), an infinite number of fuel bundles, (that would contain an infinite number of individual rods), could conceivably be subcritical in any arrangement whatsoever in water, for example, a storage pool.

This is somewhat contrary to the usual way of thinking on matters of criticality prevention, but the uranium enrichment must be low, and must certainly be less than that required for criticality in a fast or unmoderated metal system.

FINITE NUMBER OF RODS CRITICAL IF POSITIONED IN WATER



INFINITE NUMBER OF TIGHTLY PACKED FUEL BUNDLES MAY BE SUBCRITICAL IN ANY CONFIGURATION IN WATER

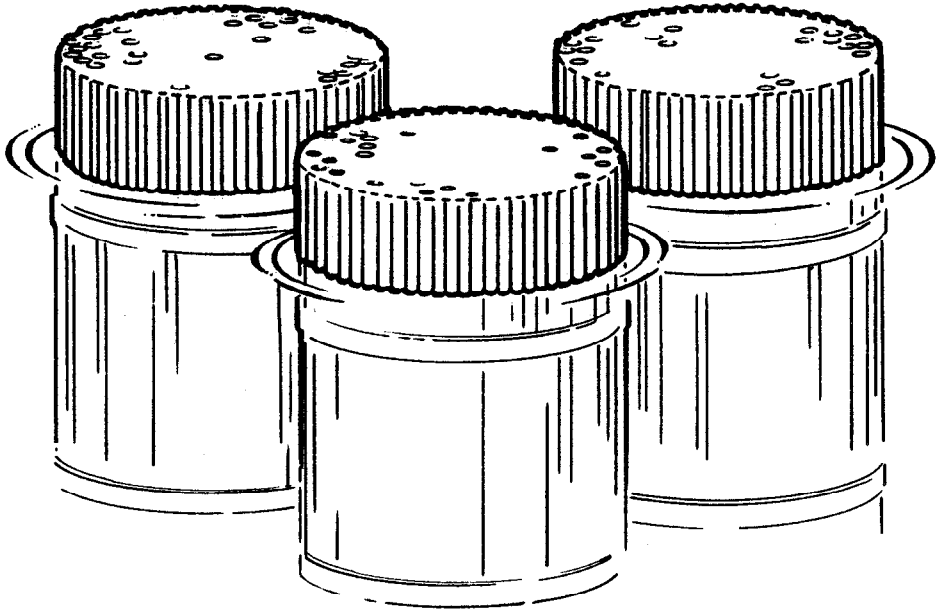


Figure 13. The Criticality of Tightly Packed Low Enriched ^{235}U Fuel Bundles in Water

G. ADDED SCATTERERS AND MODERATION

In Figures 4 and 5, the critical mass and radius for various plutonium concentrations were seen to vary continuously, in a smooth but somewhat complex manner. We shall now consider the effect of adding water to the fissile core without at first changing the density of the fissile isotope.

1. A Point of Discontinuity

Figure 14 shows the effect of adding water to mixed oxides of Pu and U beginning at 7 g/cm^3 in water plotted as a function of the fractional weight of water added.[22] The sphere volume is seen to decrease initially as the water fills the void space in the oxides. A point of discontinuity occurs at saturation in the example given. Beyond this point the further addition of water reduces the density of the mixed oxides and the critical volume is seen to increase. The result is that the critical volume changes abruptly from a decreasing to an increasing function.

The curve shape is the result of four effects: added scatterers which initially reduce neutron leakage, moderation by hydrogen, the change in density of mixed oxides, and, finally, excess neutron absorption in hydrogen becomes predominant.

Also note in Figure 17 (Section H.2), the occurrence of points of discontinuity in the case of ^{235}U enrichments below about 12 wt% ^{235}U. As the metal is diluted with carbon, the curves of critical mass vs. ^{235}U density undergo sharp changes in curvature, to exhibit a cusp (the curves appear concave upward from both right and left of the point) for ^{235}U concentrations in carbon near 0.1 g $^{235}U/cm^3$.

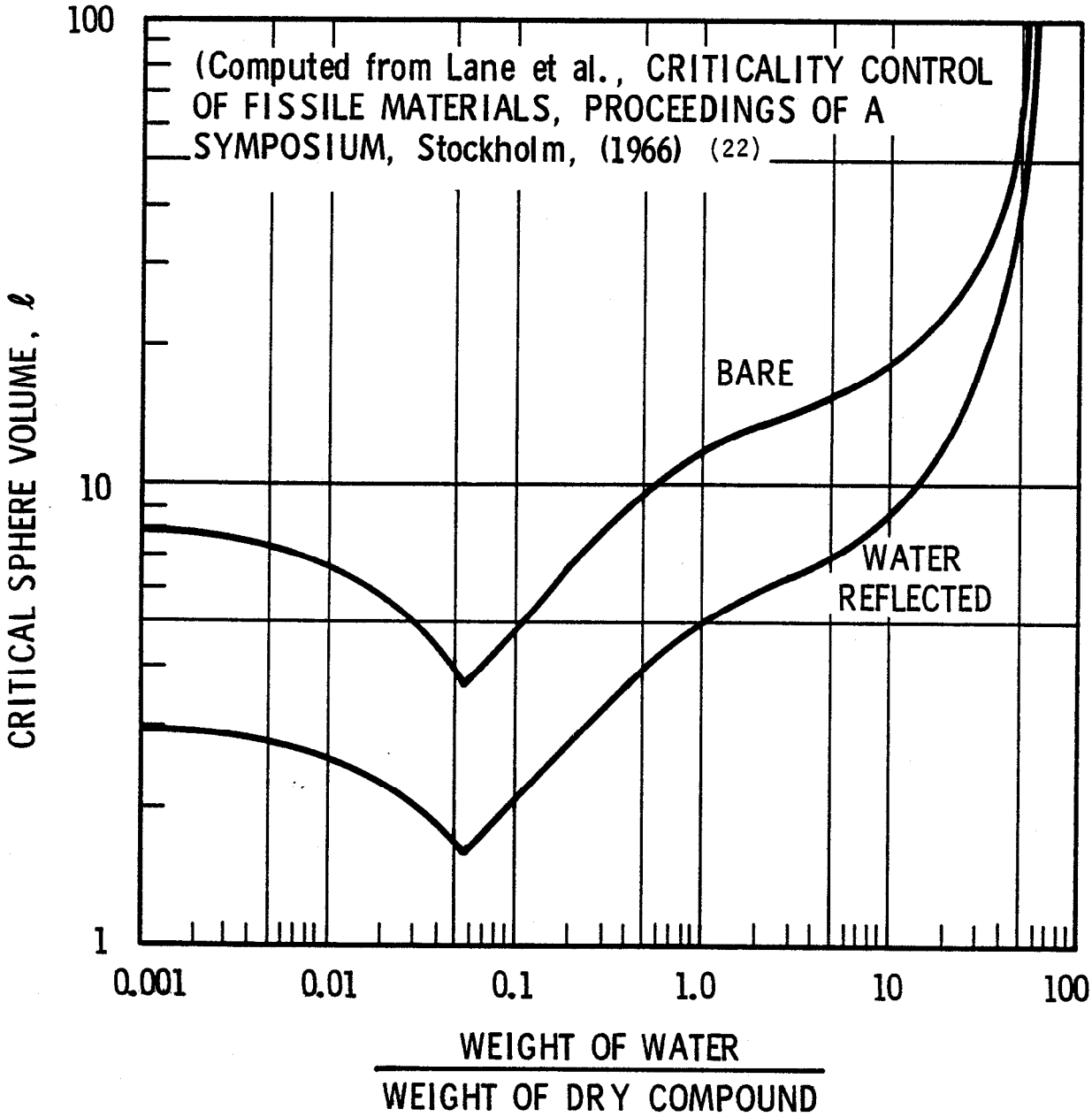


Figure 14. Computed Critical Volume as Water is Added to 30/70 PuO_2/ $^{235}UO_2$ at 7 g/cm^3

2. The Reduction in Mass of the Sphere

The next example (Figure 15) serves to illustrate the large reduction in critical mass that can be brought about by the mere addition of water to oxide at reduced density.[22] The straight lines show the increase in mass as a result of reducing the density of the mixed oxides. The bottom curves give the critical masses for saturated oxides. Note that critical mass reductions of about 200 are theoretically possible on simple saturation of the reduced density oxides with water.

3. The Paradox of the Infinite Slab

The following example is interesting because it demonstrates that, under some circumstances, the effects will be directly opposite to those illustrated in the previous examples; not only will there be no reduction in critical mass with added scatterers, but the critical size can actually be increased.

The effect of added scatterers on the criticality of slabs was first reported by E. R. Woodcock in 1961[23] and later studied in detail by Makoto Iwai.[24]

E. R. Woodcock had reported that, if the core were in the form of a thin disc or slab, a reverse effect could occur in which the additional scattering centers would now tend to scatter neutrons out of the core and the critical size would increase.

Mokota Iwai performed a study on the effect of added scatterers (O, C, N) on criticality by means of transport theory calculations utilizing the DTF-IV code.[25] His study pertained to plutonium compounds likely to be encountered through the nuclear industry -

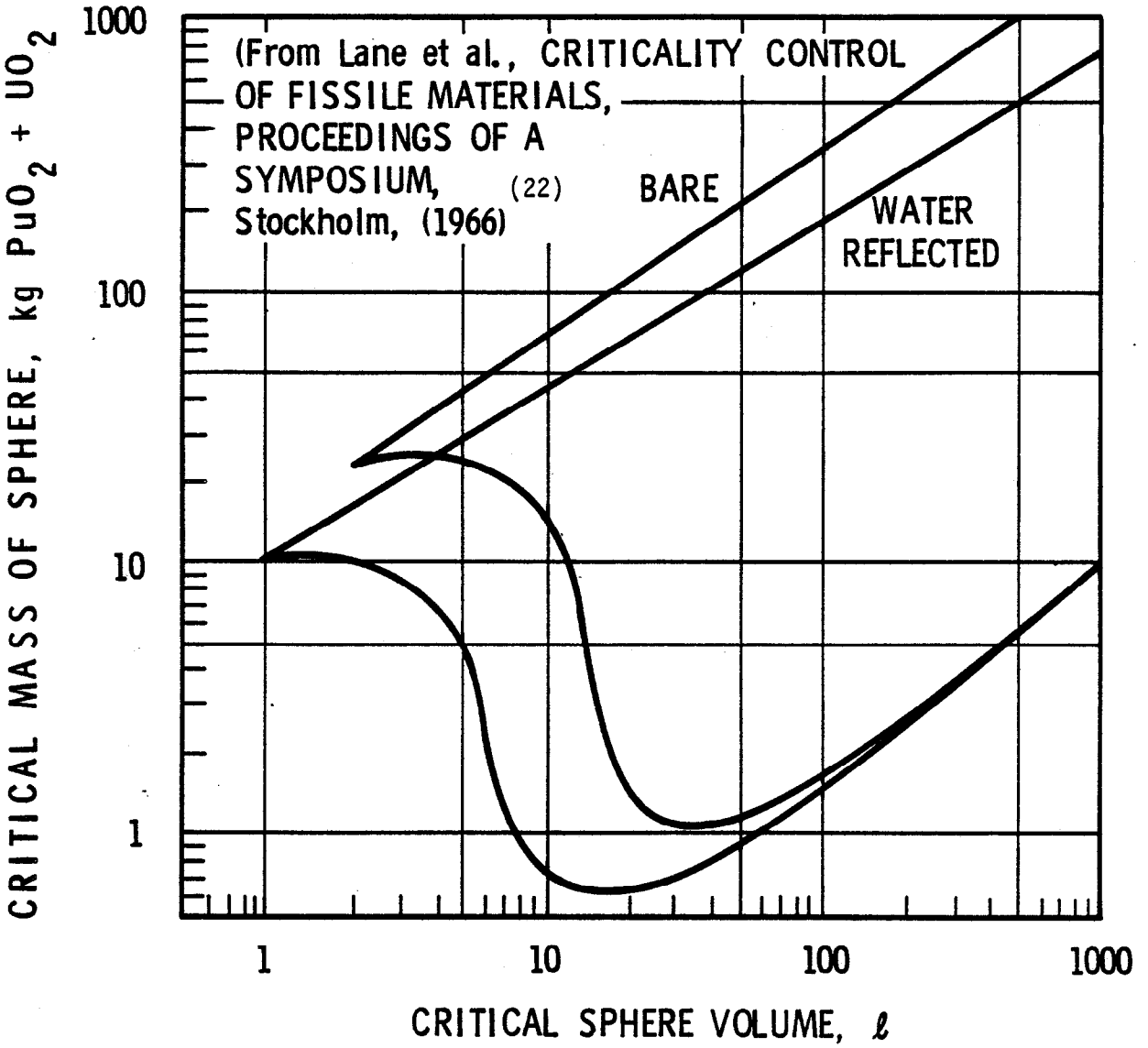


Figure 15. Computed Mass/Volume Curves for 30/70 $PuO_2/^{235}UO_2$

in fuel processing and fabrication processes. His results do, indeed, confirm that, in some cases of unmoderated thin slabs with hydrogeneous reflectors, the effect of added scatterers can cause an increase in the critical dimension contrary to the usual expectation that the size should be reduced in such cases. The dominant factor causing the increase in slab thickness was the decrease of neutron leakage into the moderating reflector.

H. DENSITY EFFECTS

The variation of critical size and mass with changes in density is of special interest. For a bare system to remain critical, while the density is changed uniformly, all the linear dimensions must be scaled inversely as the density. To maintain the same nonleakage probability or the same number of mean free paths in the system, the dimensions and density must be inversely proportional. It follows that the critical mass of an unreflected sphere will vary inversely as the square of the density, $M_c \sim \rho^{-2}$. For infinitely long cylinders, the critical mass per unit length will vary inversely with density, $M_c \sim \rho^{-1}$. In the case of infinite slabs, the mass per unit area, $M_c = \rho^0$ = constant, and remains unchanged. An unreflected infinite slab which is subcritical remains so, irrespective of the density; criticality in this case could be achieved only by adding more material to the slab so as to increase the mass per unit area. For reflected systems, in which the core and reflector density are varied independently, the variation in the critical mass for finite geometries is given by

$$M_c \propto (\text{core density})^{-m} \, (\text{reflector density})^{-n},$$

with the provision that $m + n = 2$.[26]

We shall now cite an example contrary to the usual expectation that the critical mass should be increased as the core density is reduced.

1. External Moderation

Surrounding the fissile material with thick moderating and weakly absorbing reflectors such as graphite, heavy water, or beryllium, can cause striking and unexpected changes to occur with core density change.[17] [27] The effect is illustrated in Figure 16, where the critical mass of U(93.5) metal reflected by graphite and beryllium has been plotted against density of ^{235}U metal in the core. The critical mass is, at first, seen to increase, and then, contrary to the usual expectation, the change reverses itself and the critical mass decreases with decreasing core density. It should be borne in mind that the core is not being diluted with any material, but merely reduced in density.

The region of core density throughout which a decrease causes a smaller critical mass would be critically unstable with respect to an increase in temperature. In the event of criticality, the heat from fission would reduce the core density and cause a further increase in reactivity. This autocatalytic process would then continue until the core density was sufficiently low that the critical mass was again increasing as the core density was reduced, or until the reflector was reduced in density sufficiently to reduce its effectiveness.

G. Safonov has also studied externally moderated reactors.[28] Externally moderated reactors wherein the interior consists of very-low-density fissile material cores have been referred to as "cavity reactors." The critical particle densities of the fissile atoms correspond to molecular densities of gases at less than atmospheric pressures. Thus the term "cavity reactor" has been used to describe such systems with extremly low density interiors.

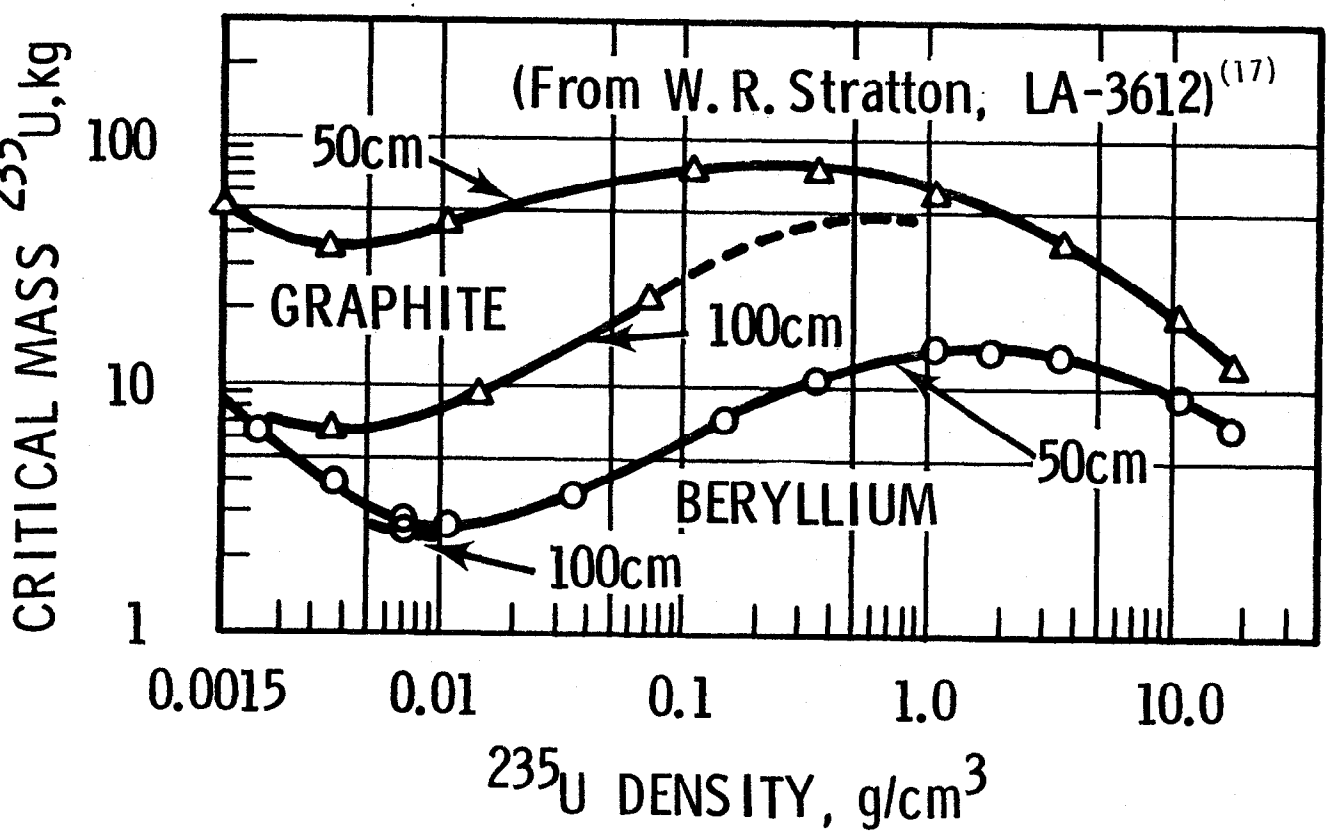


Figure 16. Computed Critical Masses of U(93.5) Metal Reflected by Thick Graphite or Beryllium for a Wide Range of ^{235}U Densities

In his report, G. Safonov calculates the critical mass of cavity type reactors fueled with ^{235}U, ^{233}U, or ^{239}Pu that are externally moderated by D_2O, Be or C. For each fuel and moderator combination, the critical mass is shown as a function of the interior radius by a family of curves for various thicknesses of moderating exteriors.

Safonov's calculations show the critical mass to first decrease with increasing interior radius due to the rapid initial rise in the cavity thermal albedo. With large radii, however, the albedo tends to saturate and criticality is obtained when the cavity radius corresponds to a constant fraction of the interior thermal mean free path. Quoting from his document: "Thus, at large radii, the critical mass varies as radius squared. This is in contrast to the bare, internally moderated cores, where critical mass increases asymptatically with radius cubed once a limiting moderated-to-fuel ratio is obtained."

2. Internal Moderation - Unbounded Regions and Multiple Infinity

As interesting as the preceding example may be, the following anomaly is perhaps even more strange. The variation in critical mass with core density change for a weakly absorbing reflector (such as graphite) was shown in Figure 16. It should be borne in mind that this variation was merely the result of a simple density change within the core. Let us now consider the combined effects of reducing the core density and also filling the void space with graphite (diluting the core with graphite). For this particular illustration there will be no external reflector; the core will be unreflected, or bare, but internally moderated.

A parametric study was made in 1967 by L. B. Engle and W. R. Stratton[29] of bare homogeneous spheres containing ^{235}U, ^{238}U and carbon in various mixtures. Figure 17 shows the unusual results of their calculations. There is nothing unusual about the curve for fully enriched uranium (93 - 1/2%), but note the appearance of critically unbounded regions for ^{235}U densities between about 10^{-2} and 2 g/cc for uranium enrichments less than $\sim$ 11%. It is also true that, for every enrichment, the critical mass will become infinite on the left side of the Figure; i. e., for sufficiently small ^{235}U densities (at large C/^{235}U ratios). The minimum critical enrichment for metal (enrichment for which k_∞ is unity with no dilution) was computed to be 5.694%.[29] Now it is clear that whenever k_∞ becomes unity the critical mass becomes infinite.

The calculations show the critical mass to become infinite at three different ^{235}U densities, providing the enrichment is in the range between 5.7 and 11%. This can be explained as follows. As carbon is added to the metal, the neutron spectrum will be degraded slightly in energy. Eta for ^{235}U will be reduced somewhat, as will fast fission in ^{238}U; to the contrary, resonance capture in ^{238}U will be somewhat enhanced. Over a range of C/U ratios k_∞ will become, and remain, less than unity; but on further moderation, as the neutron spectrum becomes sufficiently well thermalized, resonance capture in ^{238}U will be significantly reduced and k_∞ will now exceed unity. Ultimately, excessive absorption in the graphite (at very low ^{235}U densities) reduces k_∞ to values that are again less than unity. Thus, within the enrichment ranges defined, there can be as many as three different C/^{235}U atom ratios for which k_∞ is unity and the critical masses and dimensions become infinite.

3. Moderation and Density Effects in Dry and Damp Powders

The curves of Figure 4, previously discussed, show a significant increase to occur in the critical mass of Pu as the metal is initially

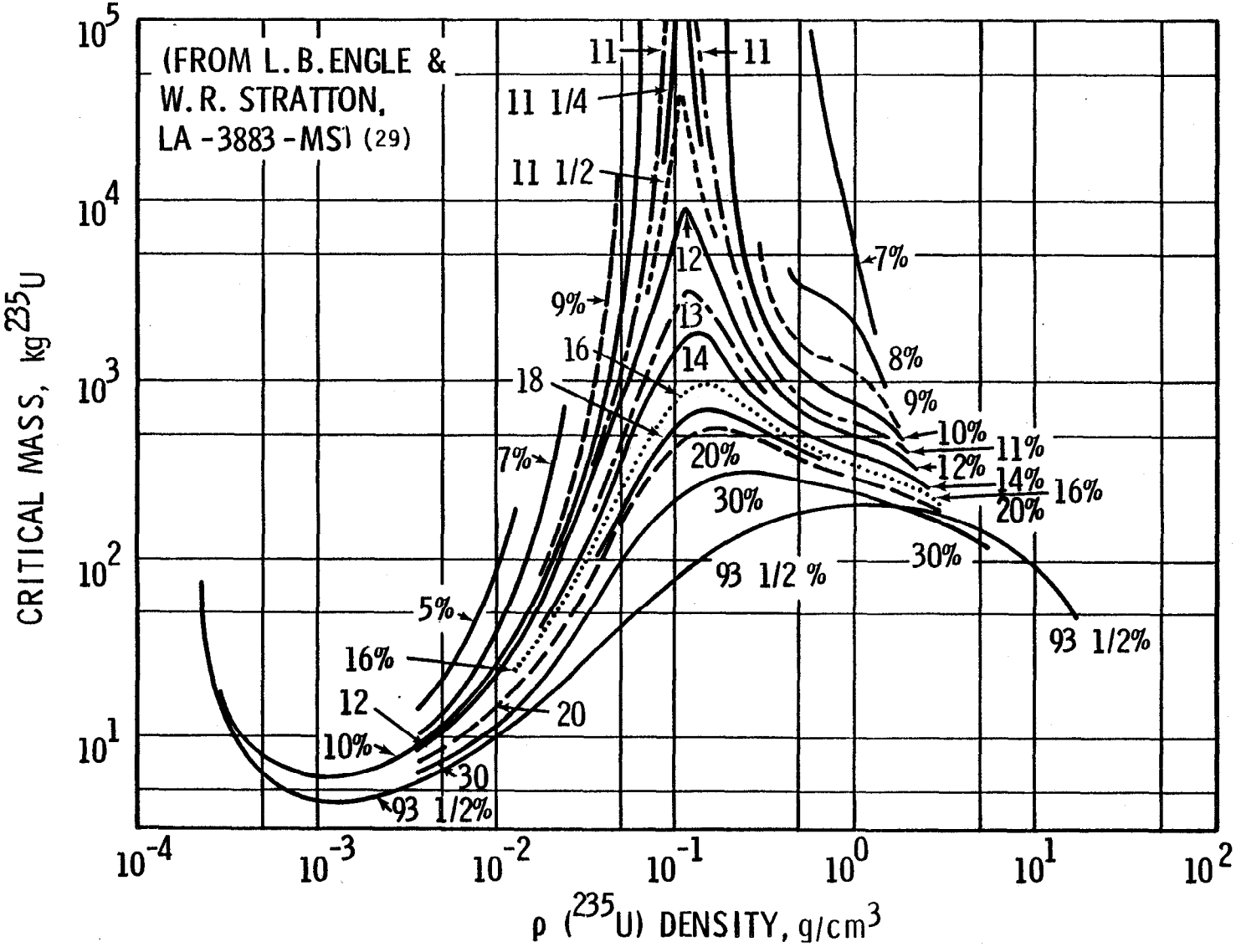


Figure 17. Critical Mass (kg ^{235}U) of Homogeneous Spheres Containing ^{235}U, ^{238}U and Carbon vs. ^{235}U Density for Various ^{235}U Enrichments

diluted with water. There is some evidence to imply that the same effect also occurs, but to a lesser extent, with PuO_2 + $U(NAT)O_2$ mixtures having a Pu content down to as low as 15 wt% or less. Criticality calculations, by J. H. Chalmers, Health and Safety Executive, Nuclear Installations Inspectorate, England, made on dry and damp mixed oxide powders in 1975 bear this out. Data taken from calculations made on mixed oxides containing 15 wt% PuO_2 are presented below:

TABLE V

Calculated Water-Reflected Spheres for $^{239}PuO_2$ - $U(NTAT)O_2$
(Dry and Damp Powders)

Wt% PuO_2	H / (Pu + U) Atom Ratio	H / Pu Atom Ratio	Fraction of Theoretical Density	Critical Radius	Critical Mass kg Pu
15	0	0	0.5	41.5	218.5
15	0.1	0.66	0.5	42.7	233.7
15	0.45	3.00	0.5	37.0	143.0

The explanation for the occurrence of this peak is similar to the explanation of the curves in Figure 4, except that it occurs at a lower H/Pu ratio as a consequence of the uranium present in the mixture. The occurrence of this peak can easily be missed unless the effects of H/Pu ratio changes between zero and unity are explored in detail. It may be concluded from this that a little bit of dampness is a safer situation than complete dryness.

Oxygen itself can cause pronounced changes in criticality irrespective of density effects. For example, the maximum value of k_∞ measured for a 3.04% enriched UO_3 hydrogeneous mixture is 1.35 ± 0.013 which occurs at an H/U ratio of about 7 ($H/^{235}U$ ratio about 240).[30]

Some interesting results were reported on Monte Carlo calculations of k_∞ for unmoderated 3.04% enriched uranium metal and for UO_3.[21] The results are given below:

MONTE CARLO CALCULATIONS OF K_∞ FOR DRY 3.04 WT% U-235 ENRICHED URANIUM

	k_∞
Uranium Metal	0.720 ± 0.012
UO_3	0.584 ± 0.019

The dry UO_3 salt is seen to have a value of k_∞ which is actually less than the value for uranium metal. The smaller k_∞ value for the UO_3 system is primarily due to scattering and moderation by the oxygen. The oxygen degrades the fast neutron spectrum slightly, which reduces the value of eta for the ^{235}U, and fast fission in ^{238}U, and enhances resonance absorption in ^{238}U. It is estimated that for UO_3 the median capture energy shifts from 0.1 to 0.2 MeV down to 0.025 to 0.050 MeV, and the median fission energy shifts from 0.4 to 0.5 MeV down to 0.075 to 0.1 MeV. In the case of uranium metal, the only significant moderating effect the neutrons experience is due to inelastic scattering.

The net effect of the oxygen in dry 3.04 wt% ^{235}U-enriched UO_3 appears to be a reduction in k_∞ of appoximately 135 mk. With the proper amount of hydrogen, however, k_∞ for the 3% oxide is raised from 0.58 to 1.35.

4. The Dilute Fissile Bearing Solution (Criticality and Evaporation)

Apart from reactors, all of the nuclear criticality incidents have involved uranium or plutonium in the form of solutions.(31) Solutions can concentrate, leak, siphon, or be inadvertantly transferred from safe to non-safe geometry vessels - or accumulate in non-safe configurations. In the case of the OKLO mine (See Section T to follow), the processes of nature concentrated the uranium and provided the water for moderation resulting in its criticality. In Section S to follow, a discussion is given on "infinte sea" critical concentrations of fissile nuclides in water such as Pu or ^{235}U; criticality becomes possible when the concentration of the fissile nuclide is high enough such that about one half of the neutrons are absorbed in the fissile material and one half in the water. In the case of Pu, this condition prevails at a concentration of about 7.2 g Pu/ℓ (H/Pu atom ratio $\sim$ 3680).

In a long water reflected vessel of restricted diameter (for example 200 mm diameter) the Pu concentration required for criticality will be greater than the "infinite sea" critical concentration, due to neutron leakage through the sides and ends of the vessel. The critical concentration in a vessel of given diameter depends on its length. However, for vessels taller than about 10 times their diameter, there will be little difference in the critical concentration as the vessel height is increased indefintely. The reason is that since the fraction of neutrons that leak out the ends of a moderately long vessel (for example 6 ft.) is already small, further

increases in length will not have an appreciable effect on the reproduction factor. To exclude criticality the vessel must remain subcritical under all credible solution concentrations, and dilute solutions in long column can be concentrated by evaporation, boiling or precipitation. If the vessel is tall enough it is possible that evaporation could produce a sufficiently concentrated solution to yield criticality. This must be precluded.

For example, in a 200 mm diameter vessel, the solution would be well subcritical in any length if the concentration were only 20 g Pu/ℓ. However, if precipitation were to occur, or evaporation take place, the concentration might well exceed 40 g/ℓ (the critical concentration) over a significant length, resulting in a criticality. (See Figure 18 for artists rendition). Further, in the event that criticality were to occur by this process the reaction might well be autocatalytic depending on the quantity of fuel available. As the fuel was further concentrated, through evaporation, boiling, and radiolytic decomposition of the water the effect could be to further enhance the reactivity.

To prevent criticality in our hypothetical vessel under such an event, the total mass of Pu permitted therein would have to be less than the minimum quantity required for criticality in the vessel. That is, if precipitation, or concentration through evaporation or loss of process control cannot be excluded, the safe concentration must be based on the minimum mass for criticality in the vessel and not on the minimum concentration for criticality in the "infinite" vessel. Then under these circumstances, depending on the vessels dimension, a concentration could be required as low as one or two g Pu/ℓ, being even less than the "infinite sea" critical concentrations. In an infintely long vessel, in the limit, the safe concentration would approach zero - unless the vessel was "safe by geometry" to begin with for all credible concentrations therein.

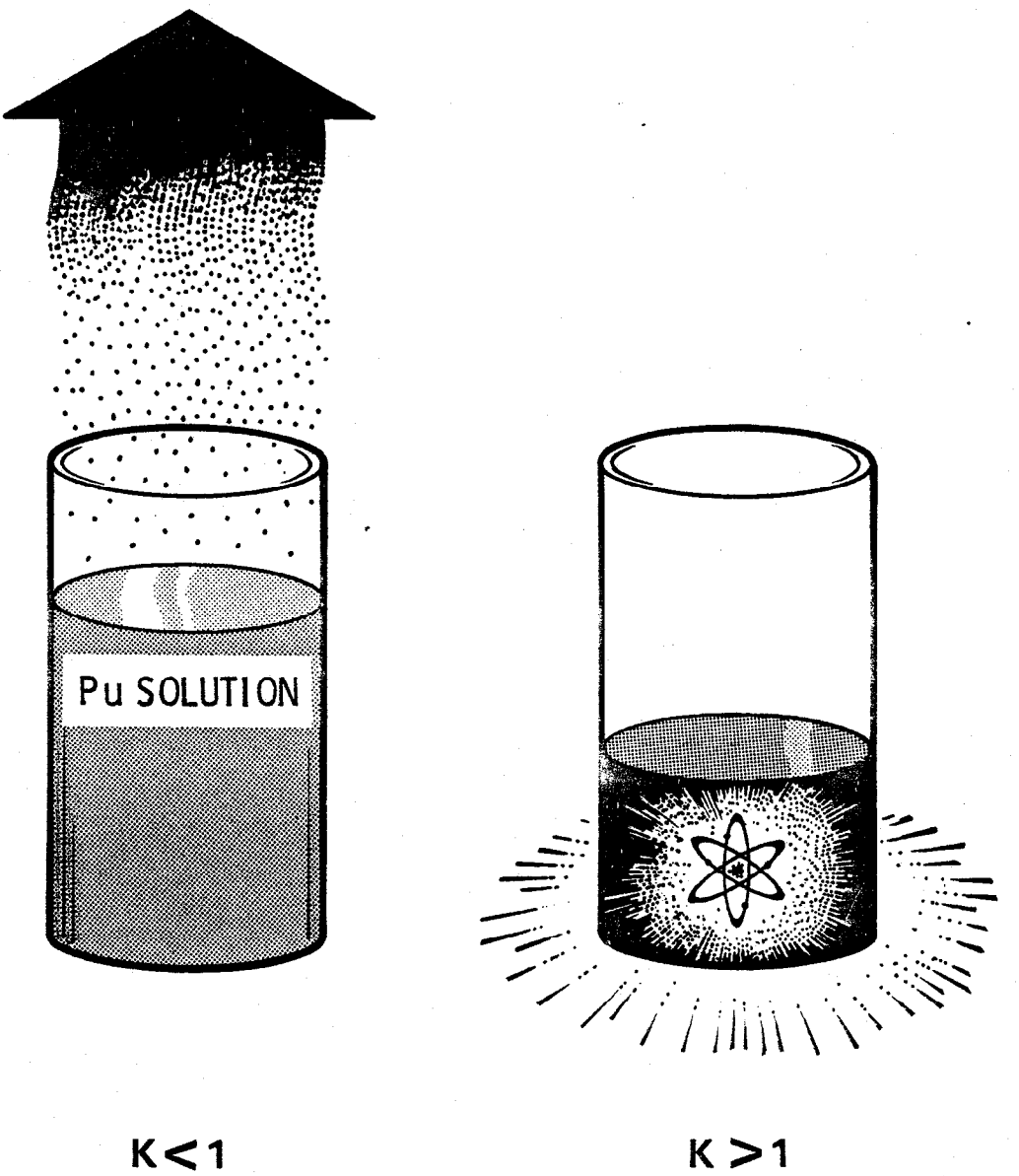


Figure 18. Evaporation - Concentration and Criticality

I. CRITICALITY AND THE NAGATIVE BUCKLING CORE

The example under "External Moderation" served to illustrate that, under certain circumstances, the critical mass could be reduced by a reduction in core density, but for the cases described, k_∞ (the reproduction factor for an infinite size core) would always have exceeded unity. We shall now cite an example that is contrary to the usual expectation that k_∞ for the fuel mixture has to be greater than unity if criticality is to be achieved. It follows logically from the simple formula, $k_{eff} = k_\infty P$, where P is the nonleakage probability, that k_{eff} becomes equal to k_∞ for the case of no neutron leakage (an infinitely large system).

It would be reasonable to assume, therefore, that, if the system could be made infinitely large and remain subcritical, a reduction in size could hardly be cause for concern. Yet, an example can be given in which k_∞ of the core is less than unity (the core buckling is negative), but criticality can be achieved nonetheless.

In 1968, a study was made of the possibility of inducing criticality in unmoderated, negative buckling cores of slightly enriched uranium by using moderating reflectors.[32] It was demonstrated (by means of calculations) that, given certain reflector conditions, a finite, reflected system with negative core buckling ($k_\infty \leq 1.0$) could have a $k_{eff} \geq 1.0$. Some of M. L. Blumeyer's results are included in the following table, which illustrates the point in question and shows k_{eff} to be greater than k_∞!

TABLE VI

Computed k_{eff} for Spheres of 1000 cm Core Radius With 500 cm Thick D_2O Reflector

Material	Enrichment	H/U in Core	k_∞	k_{eff}
UO_2	3.10 wt%	0.59	0.999	1.109
Uranyl Nitrate, $UO_2(NO_3)_2$	2.26 wt%	5.90	0.999	1.012
Metal - Full Density	2.96 wt%	zero	1.006	1.170

The results for light water reflectors were inconclusive, but negative for the few cases examined. It, therefore, remains problematical as to whether a system with a negative buckling ($k_\infty < 1$) could be found that could be made supercritical in finite size with light water reflection.

J. THE COMPLEX REFLECTOR

The critical mass, or dimension, is reduced as a result of neutron reflection from materials external to the fissile bearing core. There are wide differences in the effectiveness of reflectors, but in a relative sense, the best reflector is that which results in the smallest critical size. Reflectors frequently consist of more than one layer of reflecting material such as steel and water or steel and concrete, etc. Although it might appear logical to assume that a combination of reflecting layers would not be better than the best reflector separately, there are noted exceptions.

Experiments with a $^{235}UH_3C$ sphere, reflected with layers of nickel and natural uranium, show a composite reflector consisting of 1/2 inch thick nickel next to the core, surrounded by natural uranium, gives a significantly smaller critical mass than either reflector alone.(33)

There is no verified explanation for the effect, but is suspected that it may be associated with a strong scattering resonance that nickel has at about 16 KeV.(34)

There also is data on reflector combinations from critical experiments performed in 1978 by S. R. Bierman, Staff Scientist at Battelle - Pacific Northwest Laboratory, on interacting arrays of 2.35 and 4.29 wt% enriched UO_2 rods in water that shows the following: A reflector composed of a layer of water about 2 cm, thick backed by a 7.6 cm wall of depleted uranium is more efficient than that of either a thick water reflector by itself or of the uranium when backed by water. For the case of a composite reflector composed of lead and water a similar (although much smaller effect) was observed only with the 4.29 wt% ^{235}U fuel rods. The observed effects are shown in Figure 19.

CRITICAL SEPARATION BETWEEN FUEL CLUSTERS OF 2.35 wt% AND 4.29 wt% ^{235}U ENRICHED UO_2 RODS IN WATER WITH DEPLETED URANIUM OR LEAD REFLECTING WALLS

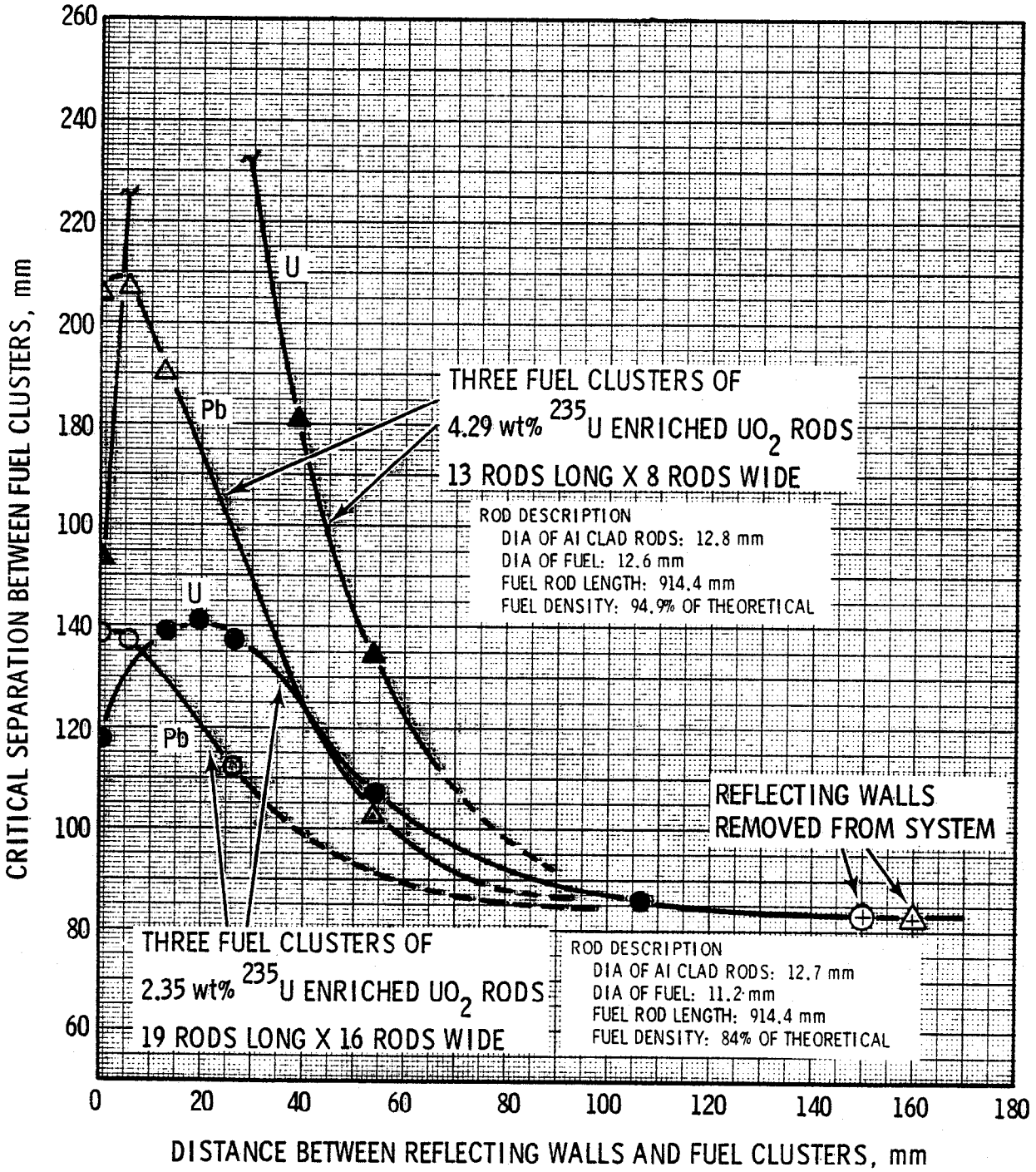


FIGURE 19

K. THE DISSOLVER PARADOX

The dissolution process can involve fissile material in the form of metal or oxide initially; whereas, during the intermediate stage, the material will be surrounded by a solution containing the partially dissolved material. In the final step, all of the material is dissolved, with the concentration being determined by the quantity of material starting the process.

One might conclude that if the material were subcritical initially, and when fully dissolved, the process could safely proceed, but this does not necessarily follow, as during the intermediate state of the coupled fast-thermal system, criticality may occur.

In the case of an idealized plutonium dissolver, it has been shown that, at least for the conditions assumed, it is possible to begin dissolution in a system that is subcritical at both the starting and ending configurations and yet achieve supercriticality somewhere in between although the total mass of Pu in the form of solution and/or in metal or oxide has remained constant (see illustration, Figure 20.[35]

The computed curves in Figure 21 show the critical masses (total of Pu-239 in metal and sphere solution) and the corresponding critical volumes of the dissolver (regions I and II). The critical envelope is drawn tangentially to the various curves and the subcritical region is the region below this envelope. The possibility of a system being subcritical at the beginning and end of dissolution, yet being supercritical in between is further illustrated in Figure 22 for the case of a 3-kg mass dissolving into a 5-liter volume. The critical mass becomes less than 3 kg total mass at or near 40 g/liter in solution and reaches a minimum of 2.8 kg at about 100 g/liter. In the example given the system would then have become supercritical at a solution concentration > 40 g/liter and would have remained so until the concentration reached about 350 g/liter.

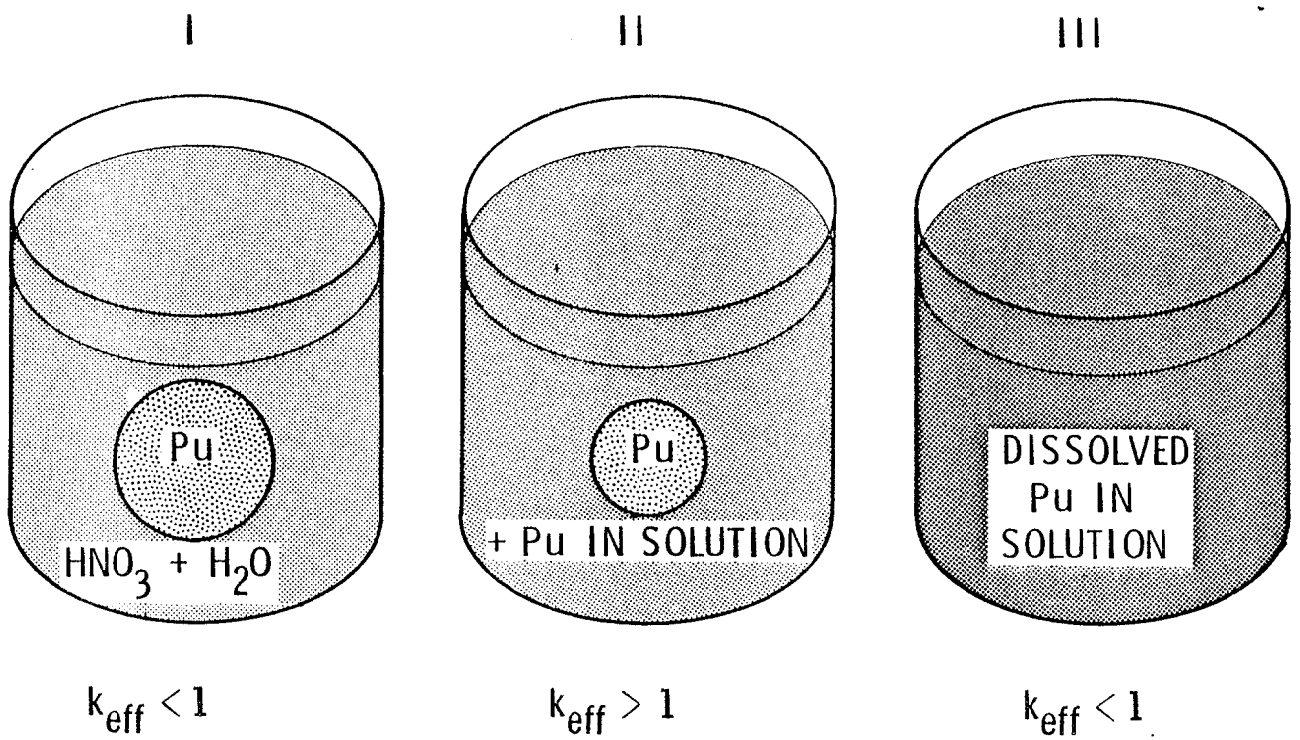


Figure 20. Pu Metal Dissolution

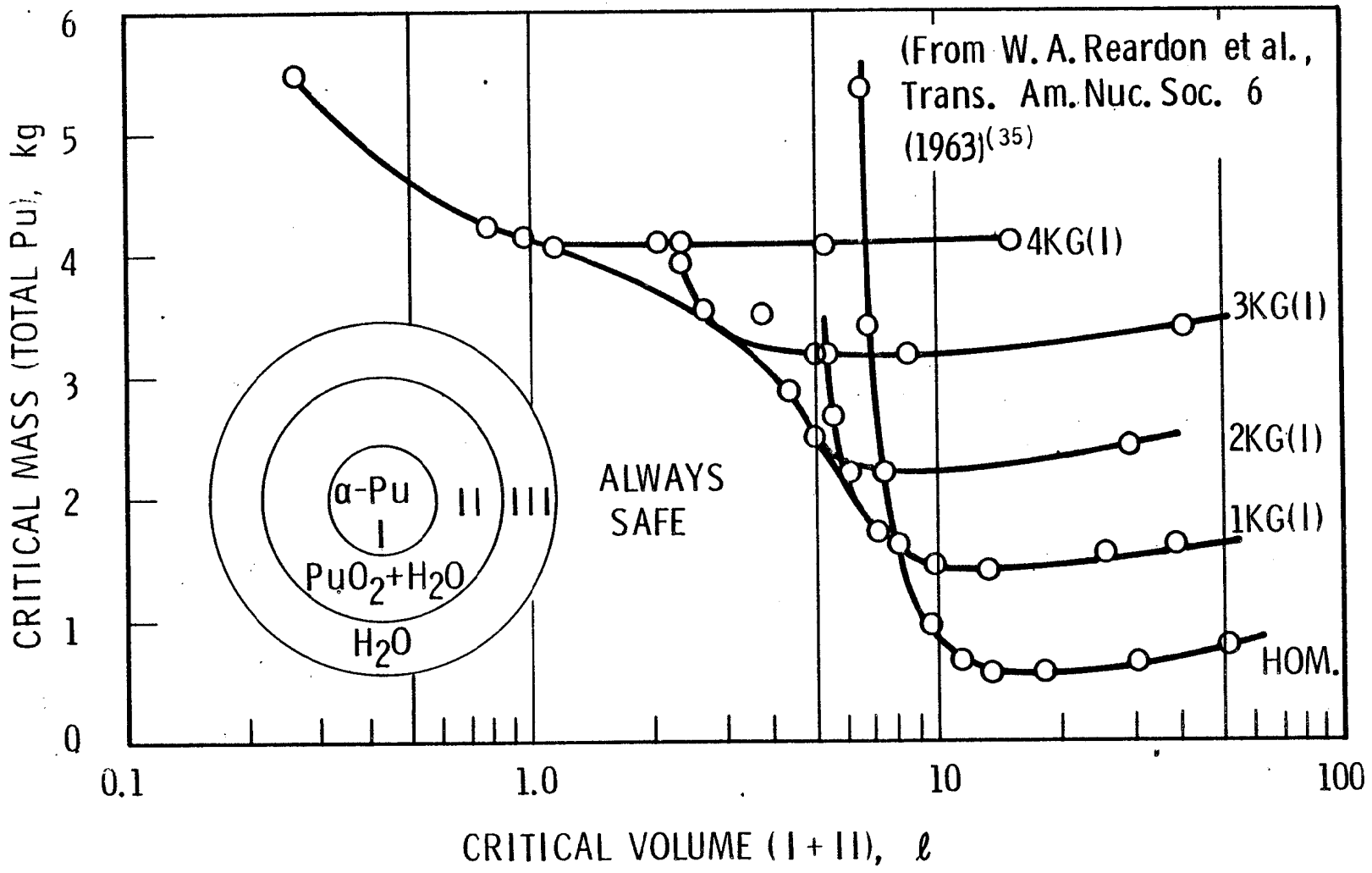


Figure 21. Computed Critical Mass (Total ^{239}Pu) Versus Volume (^{239}Pu in $PuO_2 + H_2O$ Solution)

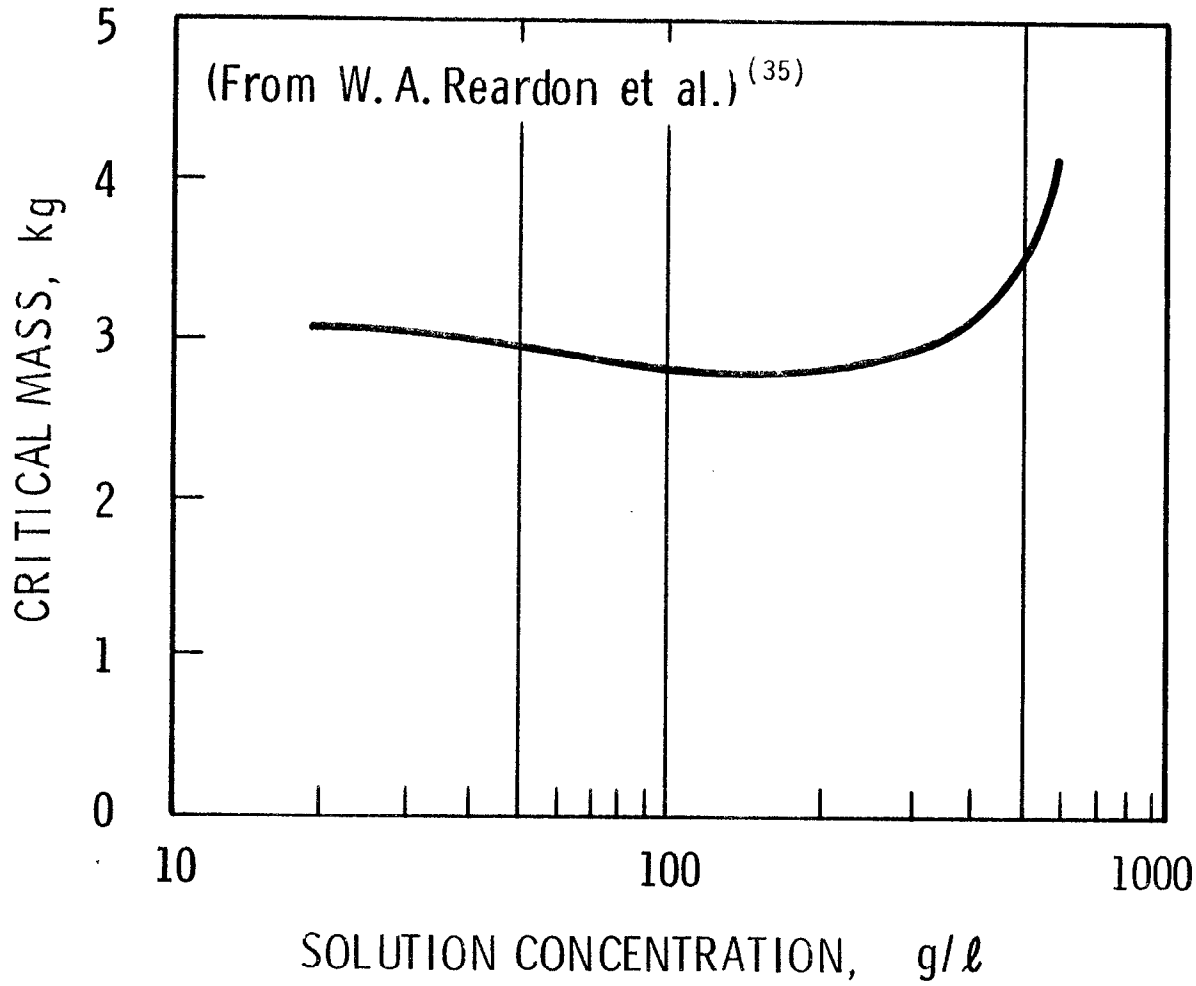


Figure 22. Computed Critical Mass Versus Solution Concentrations; 3 kg Dissolving into 5 Liter Volume

L. ^{235}U, ^{239}Pu-^{238}U MIXTURES (CONCENTRATION OR MODERATION - EFFECT ON CRITICAL MASS

At first hand, it may seem surprising that, in the case of a homogeneous aqueous mixture of low enriched uranium, the ^{235}U mass required for criticality can be significantly less than for fully enriched U (93.5%) within a narrow H/U range at the same total (^{235}U + ^{238}U) concentration.[36] This is evident from Figure 23, which gives computed ^{235}U critical masses for 5.0 wt% enriched U and 93.5 wt% U plotted against H/U (^{235}U + ^{238}U) atom ratios. However, in cases such as these, it is always the H/^{235}U ratio, rather than concentration per se, that is the controlling factor. If both curves were plotted against H/^{235}U ratio (instead of total U), the lower enriched uranium case would be seen to have the larger ^{235}U critical mass for the same H/^{235}U ratio.

It should be borne in mind, however, that in nongeometrically safe situations (wherein vessels are not safe by geometry) it would be possible to achieve criticality (albeit over limited concentration ranges) with a smaller quantity of ^{235}U in the form of low enriched U than if the ^{235}U were in the highly enriched form!

The curves presented in Figure 24 are similar to those in Figure 23, except that comparisons are made for 100% $^{239}PuO_2$ solutions and for 8 wt% PuO_2 in PuO + U(NAT)O_2. In this case, critical masses are given in terms of kg PuO_2 and are plotted as a function of the toal concentration of Pu + U. The same conclusions are evident.

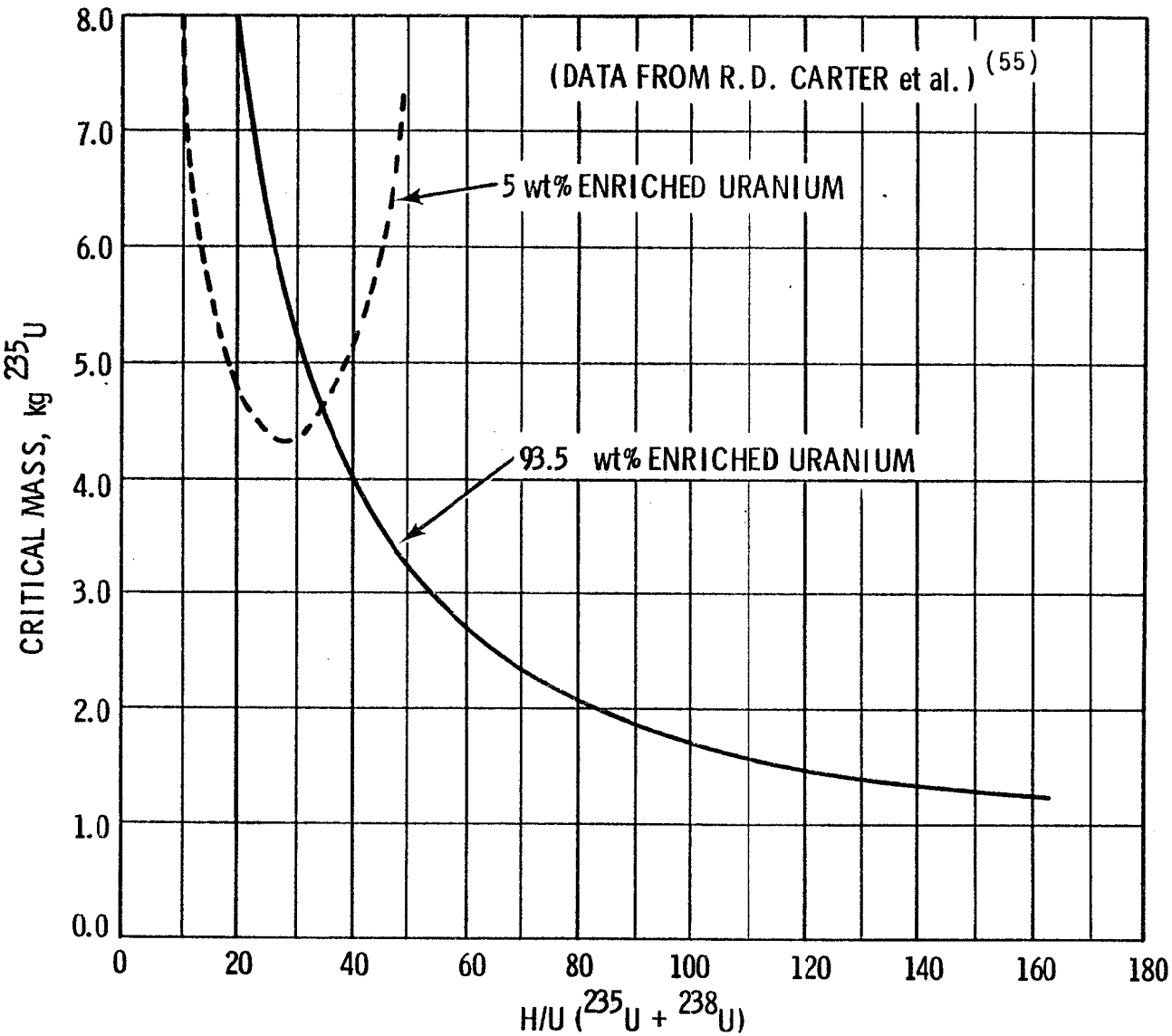


Figure 23. Computed Critical Masses of Water Reflected Spheres of Uranyl Nitrate Solutions (NO Excess HNO_3)

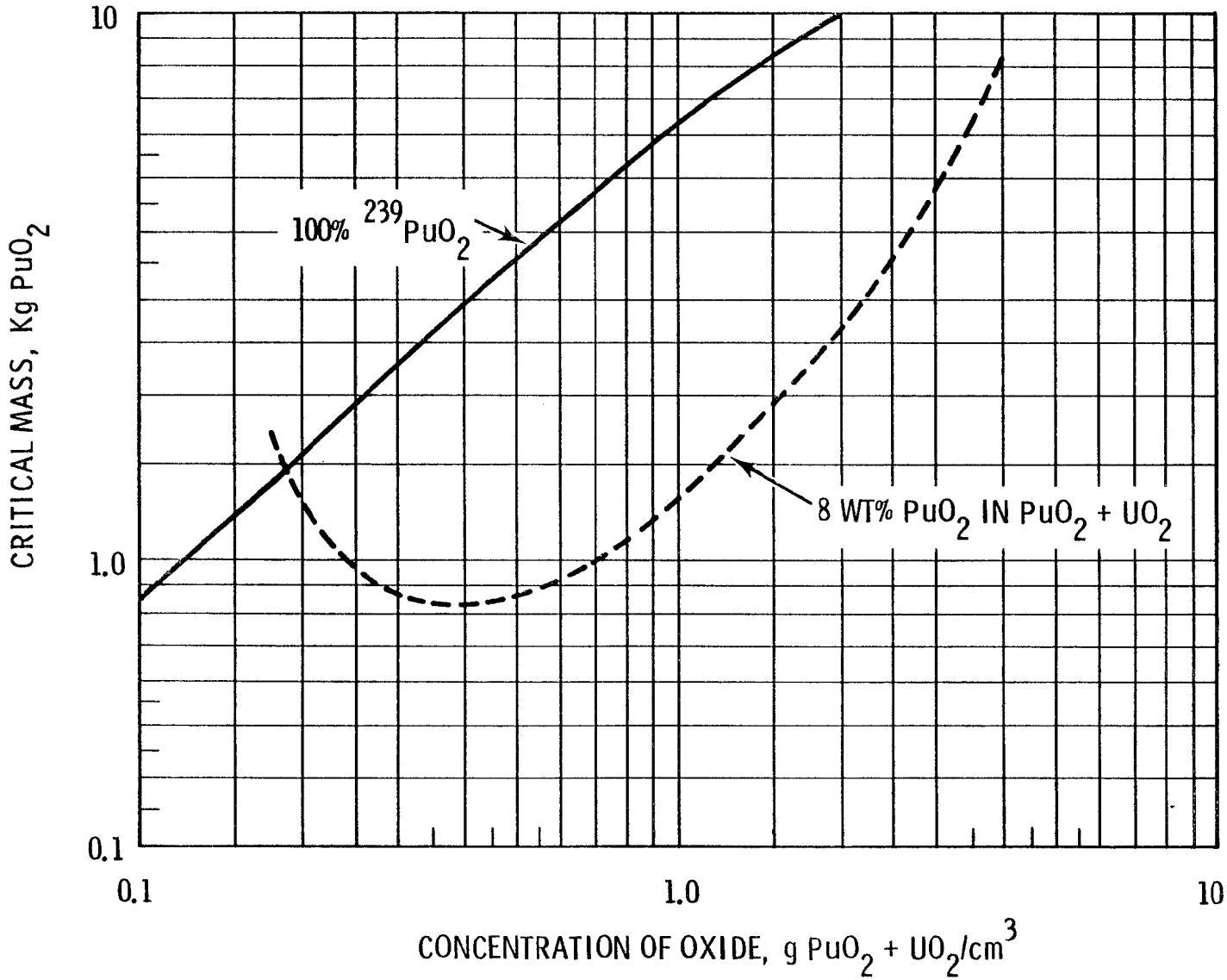


Figure 24. Computed Critical Masses of Water-Reflected Spheres of PuO_2 and $U_{NAT}O_2$ - H_2O Mixtures

M. THE CRITICALITY OF ^{239}Pu-^{240}Pu METAL MIXTURES

Critical mass values have been recalculated for the even-even nuclide, $^{240}_{94}Pu$, which lie in the range 33 to 19 kg, depending on the type of reflection (see Table VIII). (Note: This represents a substantial reduction in the previously estimated critical values; the critical mass for a bare sphere of ^{240}Pu metal ($\sim$ 33 kg) is now significantly less than that of ^{235}U metal!) The critical mass for ^{239}Pu metal is given as 5.4 and 10 kg for water-reflected and bare spheres, respectively. Pertaining to mixtures of the above isotopes, Figure 25 gives the percent change in critical mass per percent change in ^{240}Pu content as a function of the total Pu concentration in homgeneous water mixtures, spanning the range from dilute solutions to that of full metal with no water wherein the H/Pu ratio is zero.[37] The Figure shows the ^{240}Pu to have its maximum effect as a neutron absorber at a Pu concentration of about one g/cm^3 (H/Pu ratio $\sim$ 25). Up to the point of the metal mixture, spectrum changes will occur because of the variation in hydrogen content. In examining the case for ^{240}Pu metal, it should be remembered that the quantity of ^{239}Pu contained in the mixture must vanish as the ^{240}Pu concentration approaches 100%, at which point the ^{240}Pu metal would be critical by itself. As seen from the Figure, the percent change in critical mass approaches a value of about 2% change in ^{240}Pu content for concentrations up to 20%. To illustrate, in the case of 20 wt% ^{240}Pu, the total critical mass would be some 40% greater than that for ^{239}Pu metal by itself (^{239}Pu content $\sim$ 6.0 kg). Calculations also indicate the critical assembly to contain more than a critical mass of ^{239}Pu until the ^{240}Pu content is near 30%. This anomalous behavior, or peak in the ^{239}Pu content in the unmoderated metal mixture, at the point of criticality, can possibly be explained on the basis of a change in neutron spectrum on addition of ^{240}Pu to the ^{239}Pu metal system. Since odd nuclei are expected to give more inelastic scattering, the spectrum in the ^{239}Pu system can be expected, on the whole, to be slower than that in a ^{240}Pu metal system; the latter point has not been examined in detail. Calculations show, however, the critical mass of ^{240}Pu to be extremely sensitive to changes in neutron spectrum and that moderation equivalent to an H/Pu atom ratio of only about one would prevent criticality.

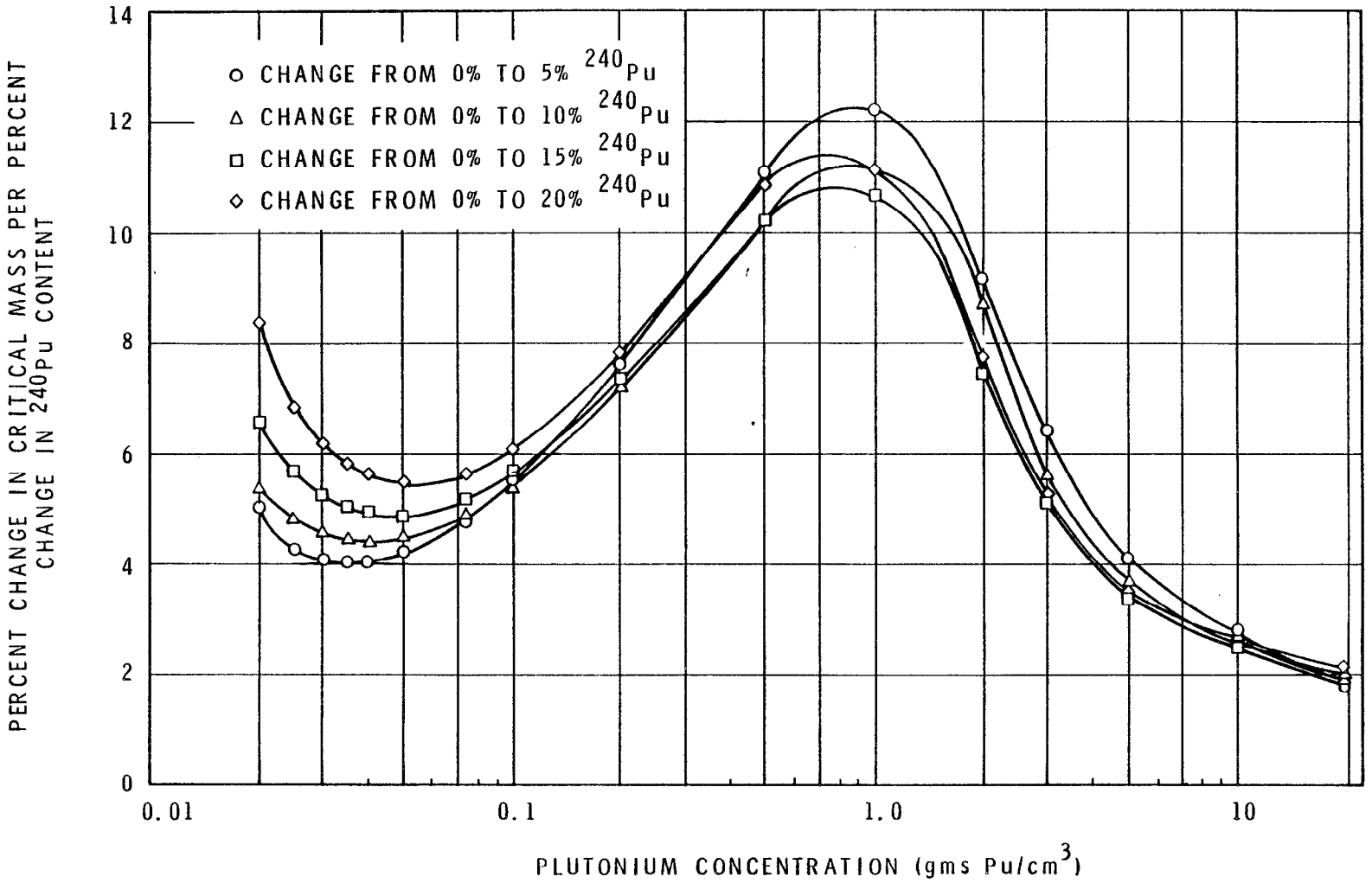


Figure 25. ^{240}Pu Effects on Water Reflected Spherical Critical Masses

N. ARRAY ANOMALIES

1. Mixed Units in Storage

The criticality of an array of units involves the effect of neutron interaction between like or dissimilar quantities of other nuclear materials that may be in the vicinity. Suffice it to say, the problem of computing criticality becomes more complex when interacting arrays of units are to be considered, such as may occur in storage areas and in shipments of containers of nuclear materials. In any operation, not only must subcriticality be established for a single unit, but the degree of subcriticality of the system as a whole; for example, the effects of interconnected and adjacent pipes must be evaluated in processing plants.

An interesting problem concerns the mixing of units within an array. If it has been determined that an array can safely handle A units of metal by itself, and B units containing dissolved fissile material in solution by itself, then is it logical to assume that these units could be mixed together in the array at the same lattice spacing, providing the combined number were less than either A or B? Surprisingly, the answer can be "no", as borne out by the following simple example (see Figure 26 and Table VII.)[38]

Note that the total number can be significantly less, depending on the pattern of positioning used.

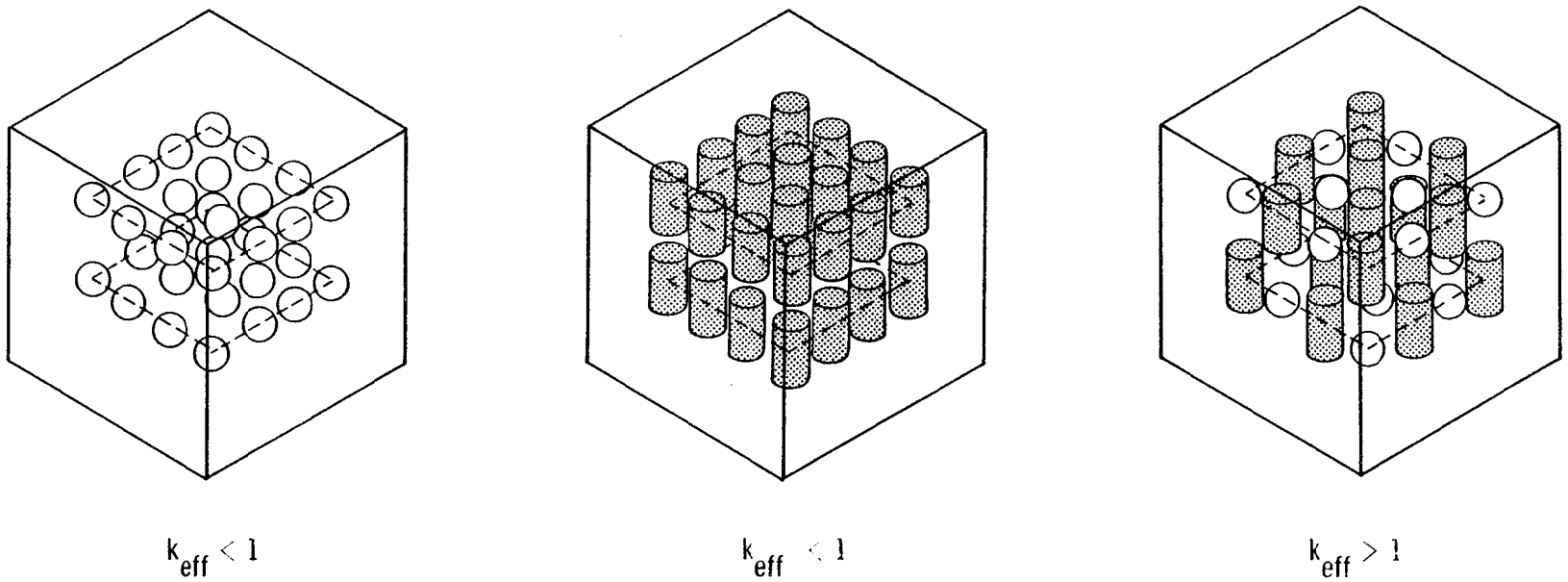


Figure 26. Criticality of Mixed Arrays

TABLE VII

Mixed Units of 3.5 kg Pu Metal and
125 g Pu in Solution (H/Pu Ratio of 500)

(Cubic Array)

Spacing Between Units Center-to-Center	Critical Numbers		Total Number
	Metal Units	Solution Units	
26 cm	93	None	93
26 cm	None	80	80
26 cm	31	29	60
26 cm	32	34	66

2. Bare Metal Arrays - A Case Wherein Criticality Can be Achieved by Diluting ^{235}U With Non-Fissile ^{238}U

Some interesting results were reported by C. E. Newlon on the criticality of unreflected cubic arrays of enriched uranium metal.(39) The calculations indicate that an interacting array of 30.0% ^{235}U enriched metal spheres could have a lower critical lattice density of contained ^{235}U than an array of 93.2% ^{235}U enriched spheres, and thus a smaller critical ^{235}U mass in the lower enrichment array. (See Figure 27). In the case of these interacting arrays, the calculations imply that a situation might be obtained whereby a smaller critical ^{235}U mass could be achieved in a given array volume by mixing the ^{235}U with ^{238}U! The array would contain fewer, but larger, units distributed over its volume with a net reduction in total ^{235}U content.

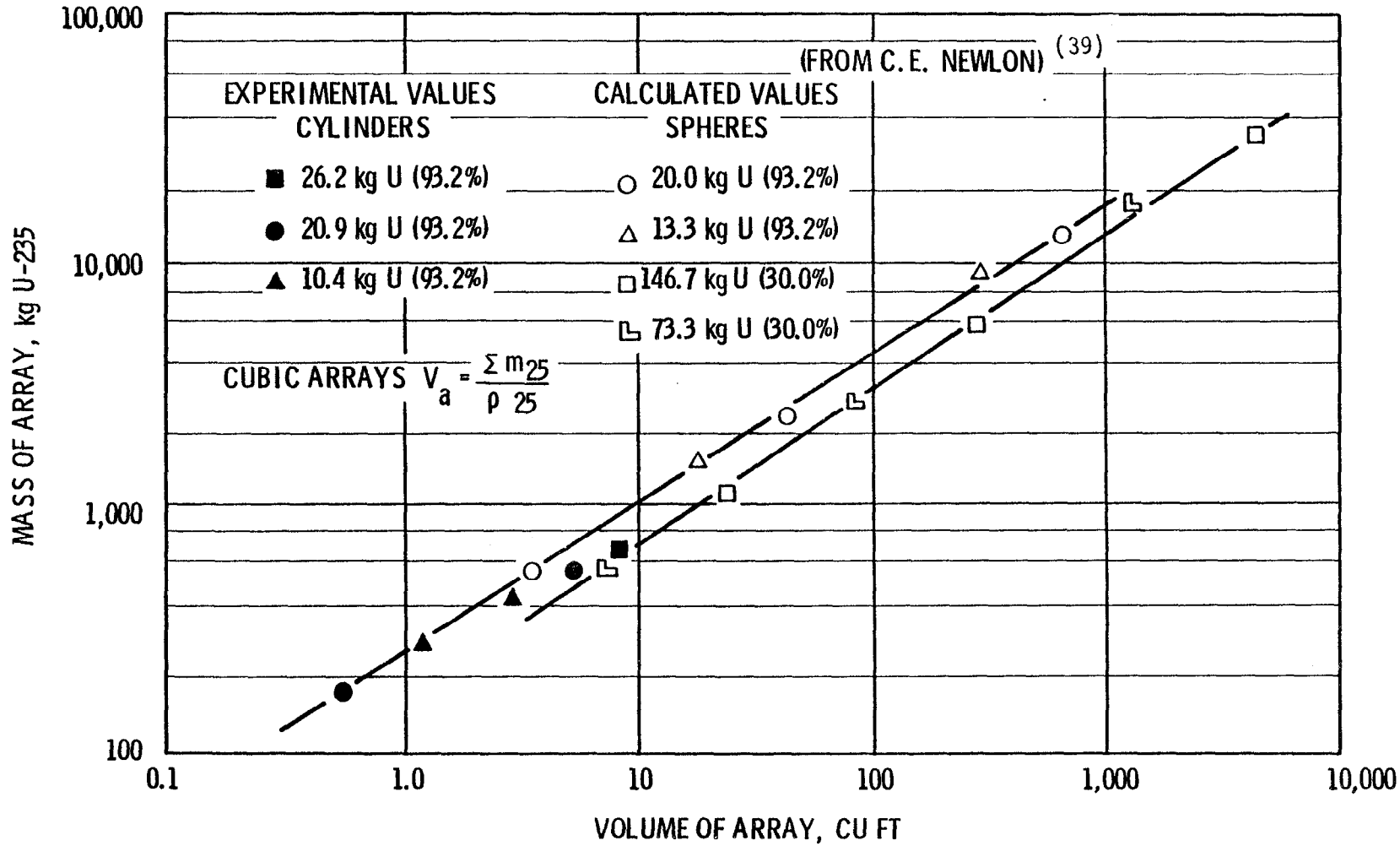


Figure 27. Critical Mass and Volume of Unreflected Metal Arrays

3. Reactivity Enhancement Due to Density Reduction in Units of Arrays (When a Reduction in the Unit k_{eff} Can Enhance Array Criticality)

In 1977, B. L. Koponen reported on a series of Monte Carlo calculations that show some storage and transportation arrays will become more reactive if the fissile material density is reduced. In particular, his calculations show that a subcritical array of shipping containers, with solid metal units, can become supercritical under certain conditions if the density of fissile material in the container is reduced.[40] Similar results were obtained with arrays made up of ^{235}U metal spheres. (See Figure 28). In the case of the arrays with spherical units, the sphere radius was varied in four steps, with a corresponding variation in density in each case so as to preserve the original uranium mass; the arrays consisted of equally spaced spheres. Calculations were done for both unreflected and water reflected arrays including variable interstitual water moderation.

For those cases studied, it was found that the most reactive unreflected arrays were those containing solid metal units, regardless of the amount of interstitual water moderation. Under the condition of a full density external water reflector, and with optimum moderation, however, the lowest density unit array was the most reactive.

The primary reason for the increase in the reflector worth of low-density fissile units is the increased utilization of thermal neutrons in the outer regions of the low density spheres. The mass of uranium in the region accessible to thermal neutrons is effectively increased, as there is an increase in the penetration of thermal neutrons into the low density core. With solid uranium spheres at full density Koponen gives the median fission energy as $\sim$ 0.5 MeV., and in the sphere with a radius four times

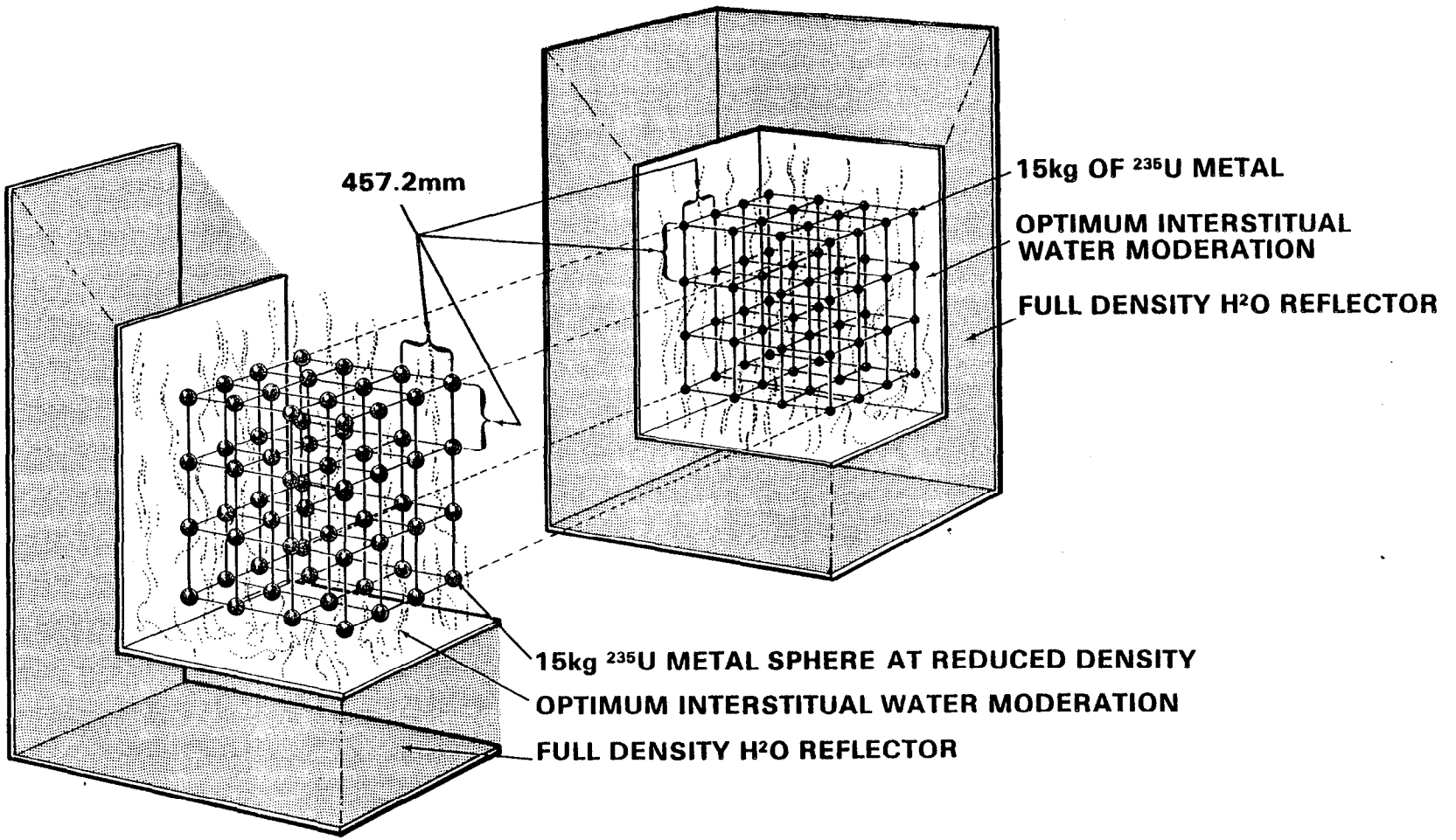


Figure 28. Reactivity Enhancement Due to Density Reduction in Units

larger, but with a density reduction of 64 in order to preserve the unit mass, the median fission energy becomes, $\sim$ 0.1 MeV. It should also be evident that as the size of the individual fissile units increase, the chances of a neutron encountering another unit before escaping from the system by leakage is increased (larger solid angle between units). Also, neutrons returning from the reflector have a better chance of interacting with a large low-density unit than a small high density unit.

It is evident from Kopenen's paper, that the enhancement of reactivity of arrays of low-density units is dependent upon the presence of an external reflector and on internal, or interstitual, neutron moderation. As pointed out by Kopenen, it may be worth considering that some shipping containers that have been approved for shipping compact fissile units may not be in compliance with criticality safety requirements if the fissile units are very low in density.

Previous calculations in two earlier papers, one by W. R. Stratton[17], and also by C. B. Mills[27] show that by surrounding a single reactor unit of fissile material with a thick weakly absorbing reflector such as graphite, heavy water or beryllium, it is possible to affect a reduction in the critical mass by a decrease in core density, but no such effect has been clearly demonstrated to date in the case of a single fissile unit with a thick light water reflector.

There also may be a condition whereby a reflected array of fissile units, that is subcritical initially, could become supercritical from either an increase or decrease in density of the individual units of the array - even though the mass of each unit were conserved and the separation between units remained unchanged. (In a theoretical sense, at least, this would be quite possible).

4. Reciprocalness (When a Reduction in the Unit k_{eff} Can Diminish Array Reactivity)

The following example shows that the reverse of the previous effect can be achieved if the nuclear material is contained within steel (neutron absorbing) drums. R. A. Carter and W. A. Blyckert of Rockwell Hanford Operations have made a study on the change in k_∞ with fissionable material volume and moderation for plutonium contained in arrays of steel waste drums. Without the steel walls in the drum, the plutonium density or spacial mass would be immaterial for a k_∞ determination, as only the moderation would change k_∞. With the drum walls added, however, decreasing the density (enlarging the volume) of the plutonium increases the neutron leakage from the mass within the drum, increasing the probability of neutron absorption in the drum walls. This is shown by the following calculations of k_∞ with a fixed quantity of 200 g Pu per drum.

	Computed k_∞	
Plutonium Density	With Drum Walls	Without Drum Walls
50 g/ℓ, H/Pu = 529	1.3748 ± .0045	1.7068 ± .0033
0.92 g/ℓ, H/Pu = 529, Full Drum	0.9691 ± .0045	1.7148 ± .0038

In this example, distributing the material throughout the drum, reduces its density, and the k_{eff} of the unit and the probability of criticality is indeed less (k_∞ is less); whereas, in the previous example of reflected finite interstitially moderated arrays, the probability of criticality was enhanced.

5. Unit Shape and Array Criticality (Units of Same Nuclear Material, k_{eff}, and Average Lattice Density in Array - But Critical Number Can Differ)

Some interesting calculations were reported by J. T. Thomas in which the effect of unit shape on the criticality of unreflected arrays of enriched uranium (93 wt% ^{235}U) was examined.[41] The three basic geometries, or shapes, of the subcritical units composing the arrays were, the cube, sphere, and the cylinder. Now it should be clear that units of identical material that have the same value of k_{eff} will have the same neutron leakage fraction irrespective of shape. It was noted, however, that if a sphere were replaced with a cube of the same k_{eff} that the k_{eff} of the array would be increased; the cube consistantly resulted in a larger k_{eff} for the array. This is understandable since an array of cubes results in less free space in the lattice than do spheres. At the same value of k_{eff}, a cube or cylinder will have a larger volume, and contained mass therein. At the same spacing in the array, the density of fissile material in the lattice will then be larger than in the case of the reference sphere. If a larger spacing is utilized for the cubes (cubes with the same k_{eff} as that of the spheres) so as to preserve the average uranium density in the lattice, k_{eff} for the array of cubes continues to be larger than that of the spheres. Arranging a given number of units, of equal k_{eff}, but different shape, to have the same average lattice density was, therefore, not sufficient to provide the same value of k_{eff} in the overall array.

It would appear, however, that if an array were to be made up of subcritical spheres, and if units of different shape, but equal mass (equal mass ensures k_{eff} of the replacement units to be less than that of the spheres) were then substituted at the same lattice spacing then k_{eff} of the replacement array would not exceed that of the reference spheres.

In Figure 29, data from J. T. Thomas of ORNL are presented wherein reflected arrays of cylinders are compared to that of spheres. These curves show that cylinders for some H/D values may result in a lower total solid fractional angle than do spheres of the same unit k_{eff}. This indicates that the cylinders with the H/D of unity would require a lattice density less than that of the spheres. In this case, a smaller fractional angle would be required to give the same k_{eff} in the array.

It might be concluded that if shapes other than spheres are to be stored, and if larger mass values are required than that permitted for the spheres, that the array criticality (or its subcriticality) should be carefully examined for the specific shape and spacing of the subcritical components involved. The k_{eff} of the replacement unit must be made smaller than that of the sphere.

6. Fire (Fog, Mist or Flooding: A Potential for Triple Criticality)

It was previously shown that, on mixing metal and solution units together, the total critical number could be less than that for either of the units when stored separately in the same configuration.

An interesting problem concerns the effect on criticality for an array of interacting units if the water content of the intervening air spaces within the array were increased. This could be brought about by the use of water for fire control or possibly through the use of automatic sprinkler systems in buildings so equipped.

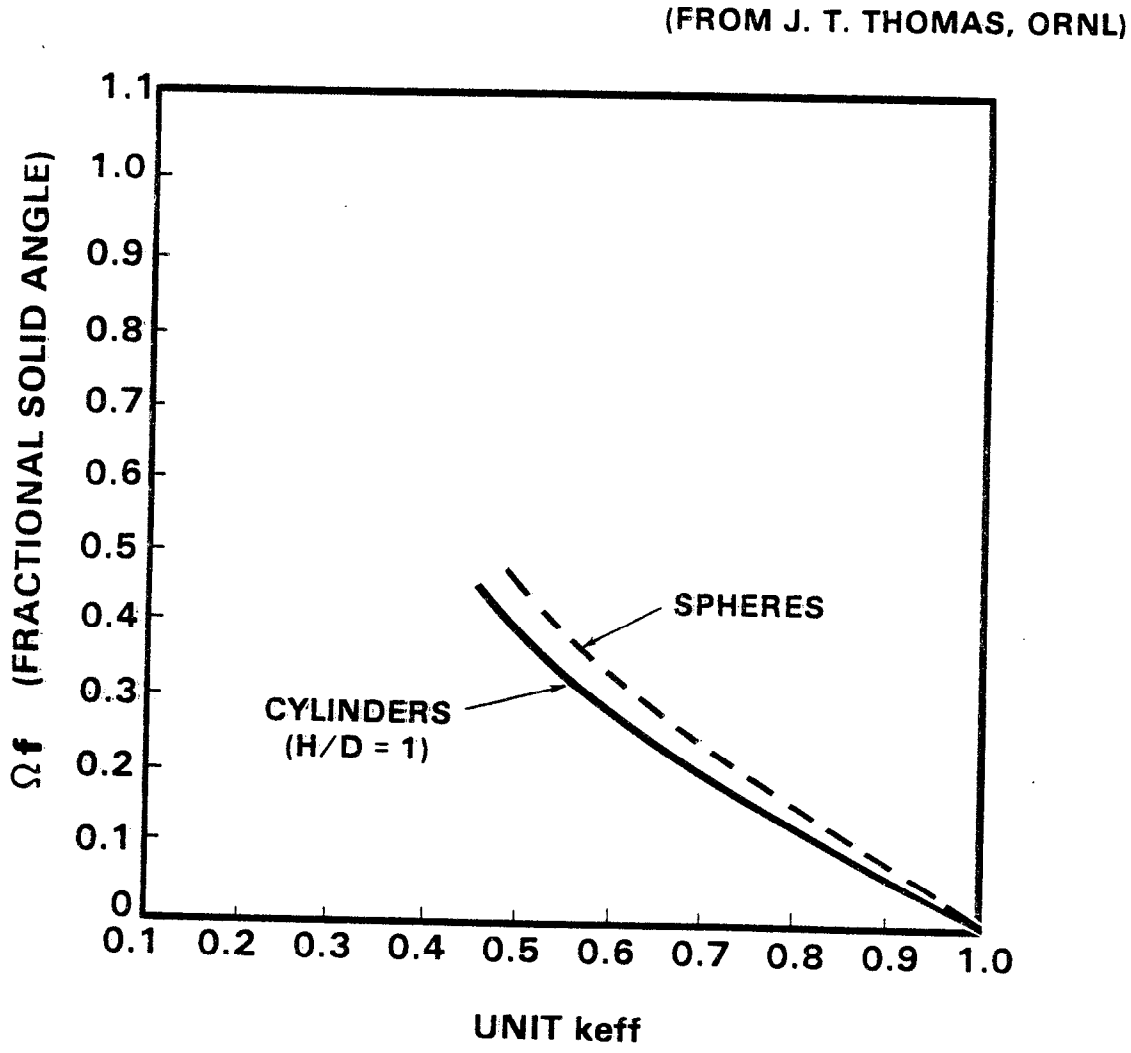


Figure 29. Comparison of Critical Reflected Arrays of U (93.1) Metal Spheres and Cylinders

In the case of storing mixed oxide fuels of PuO_2 and $U_{NAT}O_2$, or slightly enriched uranium, three effects (shown schematically in Figure 30) will be paramount. For purposes of illustration, let us assume the Pu content, or ^{235}U content in the U, to be less than 5% such that criticality would not be possible without the addition of moderator, taken in this case to be water. The array is well subcritical initially. Depending on the fuel composition making up the fuel bundles, and the storage arrangement used - it is possible by means of Monte Carlo calculations to generate the type of curve shown. The three effects involve 1) internal moderation of the fuel elements within each fuel bundle, 2) reflection about the array as a whole and also about each individual unit and 3) interaction between units. Initially, the value of k_{eff} increases rapidly with increased water density due to internal moderation, external reflection and enhanced interaction. Interaction is enhanced because a small amount of water (typically a few percent of full water density) in the space between units will slow down some of the neutrons in the interaction process. The number of neutrons actually arriving at a second unit will be less, but there will be a higher probability for fission if the neutron energy is reduced, however, with too much moderation or intervening water, too many neutrons will be absorbed between the units and the effect of interaction will be reduced. The value of k_{eff} is rapidly increased at first, then falls, due to the decrease in neutron interaction. If the surface-to-surface distance between fuel bundles is some 8 - 12 inches or more, then, on full flooding, the reactivity of the array would become merely that for a single bundle of fuel immersed in water. With full flooding, the neutron interaction would be reduced to zero.

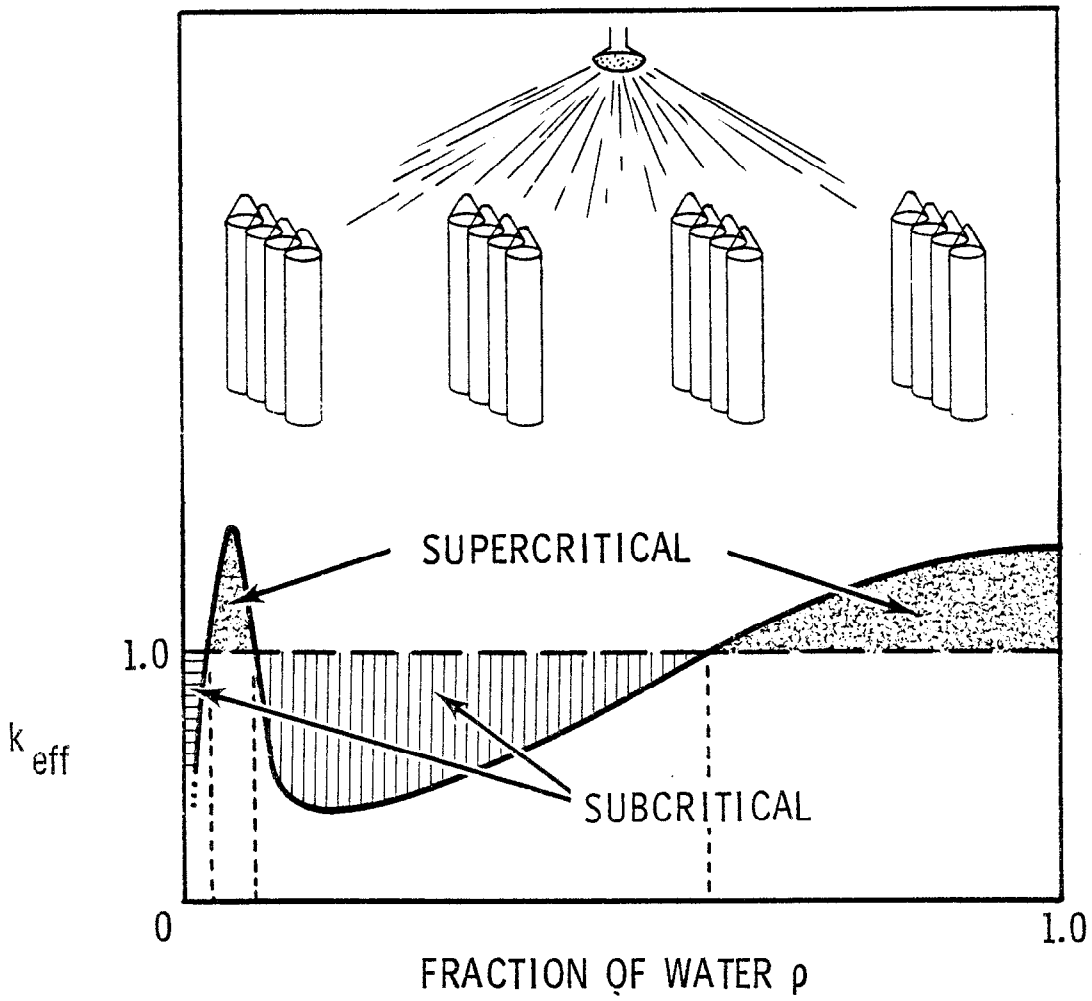


Figure 30. Fire (Fog, Mist, or Flooding: The Potential of Triple Criticality in a Storage Array)

Note that in going from the completely dry case to the fully flooded condition, criticality could occur at three different water densities being separated by two subcritical regions of water density. It is important, therefore, for determining the safety of a given storage array that the effect of sprinkler systems, and the use of water for fire control, be fully examined over the full range of water densities that may be encountered.

O. A PRECAUTION ON SOLUBLE NEUTRON ABSORBERS

If comparisons are made between an aqueous homogeneous plutonium nitrate solution ($Pu(NO_3)_4 + H_2O$) and a homogeneous mixture of Pu atoms or PuO_2 in water at like Pu concentrations, the $Pu(NO_3)_4$ solution will have the larger critical dimension and mass, due to the presence of the nitrogen and neutron capture therein. At first hand it might be presumed, irroneously, that if a sufficient quantity of soluble neutron absorber were added to render a mixture of Pu atoms in water subcritical, that a Pu nitrate solution with the same concentration of Pu in g/ℓ also would be subcritical. Calculations by S. W. Heaberlin of Battelle - Pacific Northwest Laboratories, are presented in Figure 31 that show the boron concentration required to reduce k_∞ to unity for aqueous Pu solutions. At lower Pu concentrations the boron content required for the mixture of Pu atoms in water also is sufficient for the nitrate solution. This is what would normally be expected since the nitrate is an additional neutron absorber. At higher plutonium concentrations, however, more boron is required for the nitrate system. This seemingly anomalous result is caused by the larger volume of the nitrate molecule. At the same plutonium concentration the nitrate solution contains less water than the mixture of Pu atoms in water. The reduction in hydrogen content, displacement of water by nitrate, reduces the effectiveness of the boron poison and more is required.

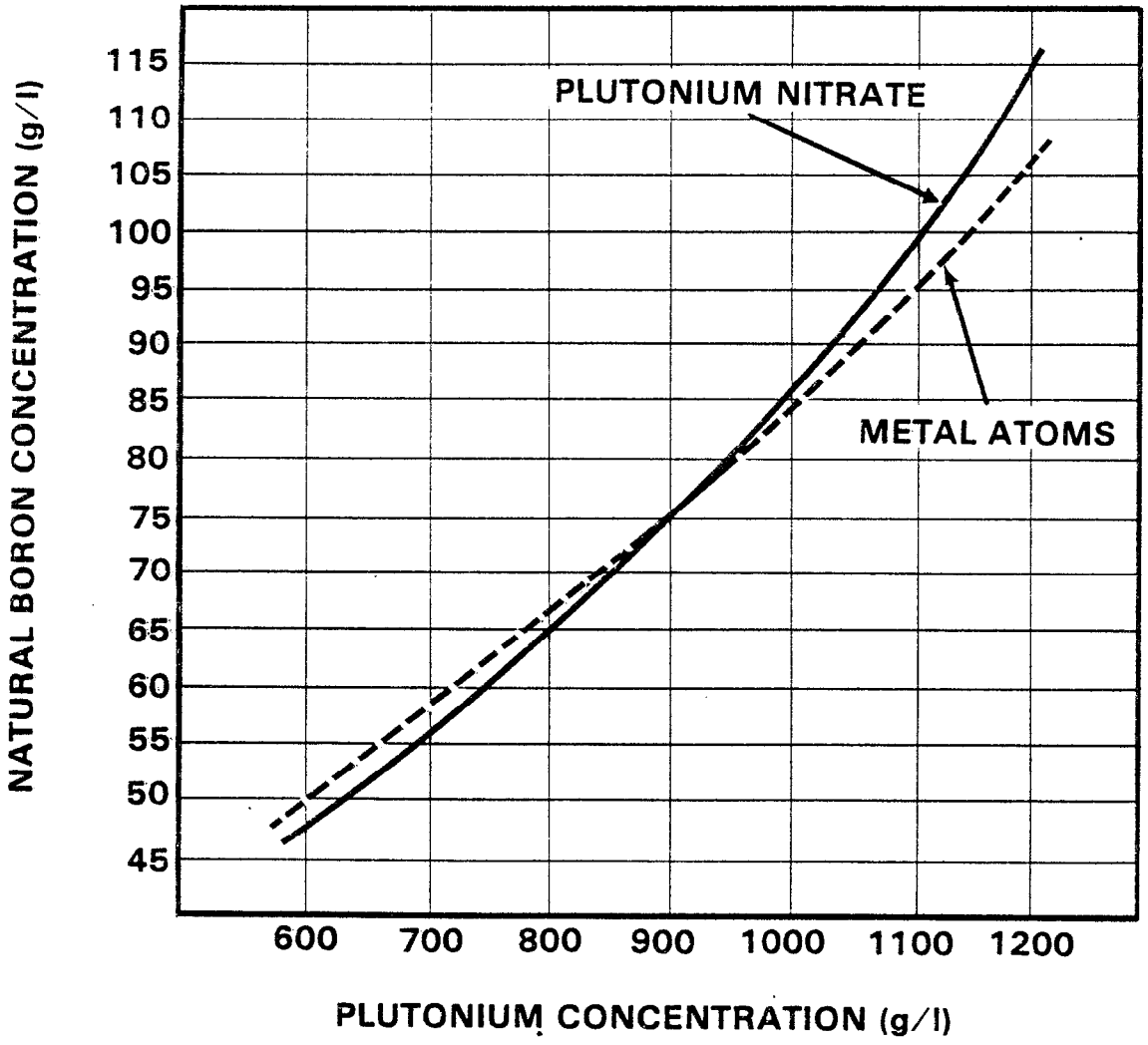


Figure 31. Quantity of Boron Required to Reduce k_∞ of Homogeneous Aqueous Pu Solutions to Unity

If the comparison be made at the same H/Pu atom ratios, not equal Pu concentration, then the quantity of boron will be sufficient, in every case, to cover the nitrate system as well.

An example also is given in a paper by R. D.Carter pertaining to criticality considerations in reprocessing wastes and contaminated soils wherein a smaller critical mass could be found for a mixture which had more cadmium and less plutonium than another because of differences in the H/Pu ratios of the mixtures.(42)

For example, at 6 grams of plutonium per liter in soil, a mixture containing 0.2 gram of cadmium per liter and 20 percent water had a calculated critical mass of 7.6 kilograms while a mixture of 15 grams of plutonium per liter with no cadmium had a critical mass of 10.6 kilograms at 10 percent water.

In any event, it is not the concentration of Pu per se, but the hydrogen content that is the controlling factor in determining the effectiveness of soluble absorbers in aqueous solutions.

P. AN ODDITY OF POISON (THE CONTROL ROD AND THE SOLUTION SPHERE)

In early criticality experiments with Pu solutions it was noted with some surprise that, as a hollow cylindrical control rod was moved into the solution of a sphere along its axis, the reactivity actually increased during the initial phases of rod insertion and then reversed itself, contrary to the usual expectation that k_{eff} should be continuously reduced in such cases.(43) A copy of the chart recording (heretofore unpublished) showing the strange variation in neutron flux with control rod movement is shown in Figure 32. As noted, when the control rod (tube) entered the solution, for spheres that were not quite full, the flux was first observed to rise and then fall. This peculiar behavior might be

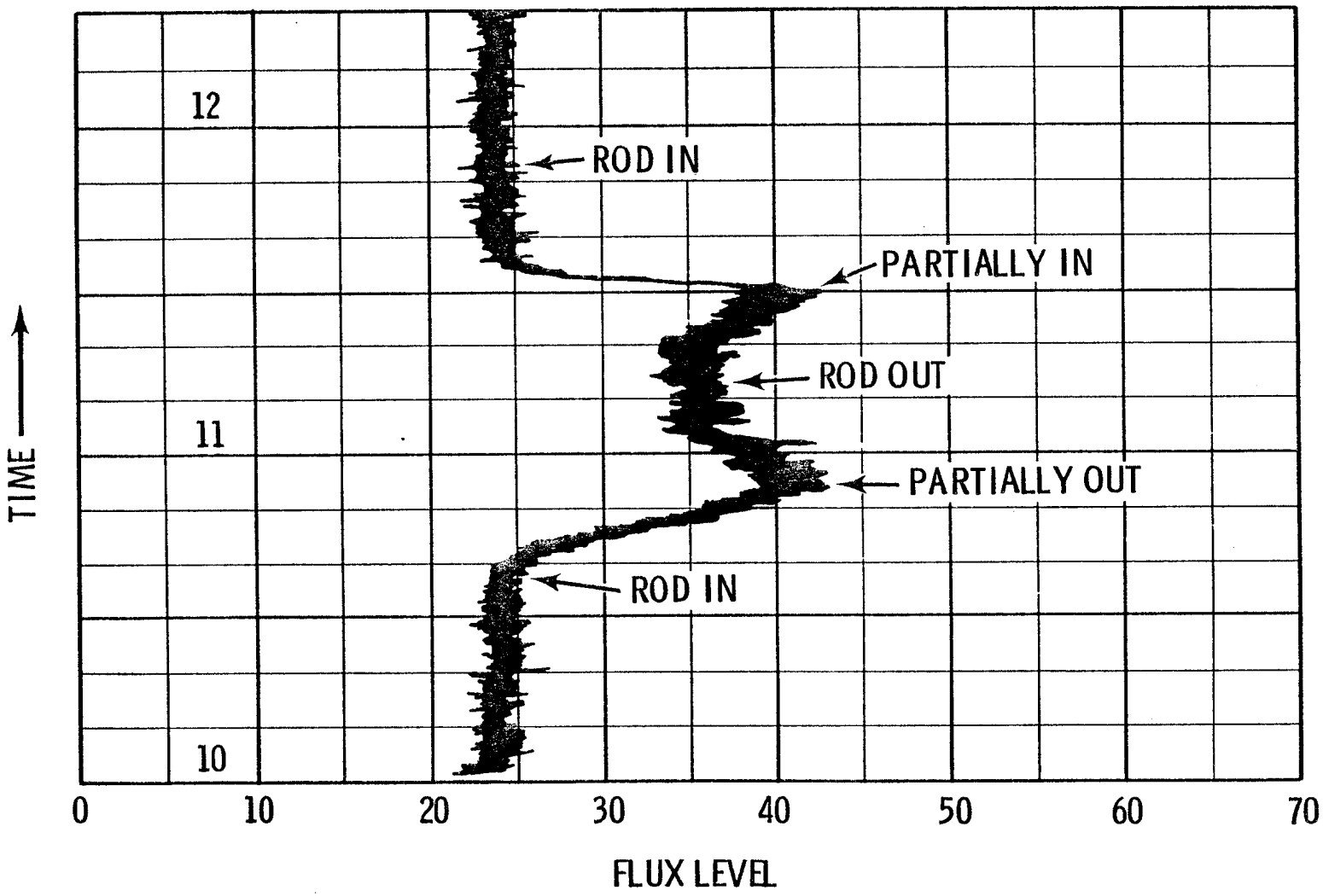


Figure 32. Control Rod Effect on Flux Level

expected if the rod's poison worth were small, for in that case the first portion of the rod is worth more in terms of a volume displacement of solution (the sphere is becoming effectively more full) than as a neutron absorber. The effect was estimated to be worth approximately ten cents from the multiplication curves plotted with the control rod in the full out position and then partially inserted. A perturbation calculation subsequently provided an estimate of 8.4 cents.

Q. NATURE OF FISSION AND THE CRITICALITY PROCESS (From Protactinium to Californium and Beyond)

The actinide group is composed of 14 elements with atomic numbers 90 to 103; the first three are naturally occurring and the other eleven are man-made. Although the principal interest in criticality to date has centered on isotopes of the elements uranium and plutonium, a number of isotopes of other elements within the actinide group also are capable of supporting a chain reaction. Calculations or measurements of criticality show extreme variations in critical masses, ranging from gram to kilogram quantities under like environmental conditions. These variations are to be expected on the basis of simple qualitative considerations and observations concerning nuclear structure and stability.

Neutron fission, a prerequisite for a self-sustaining chain reaction in these elements, depends on the interaction of neutrons with the nucleus. Even for a single given nuclide, extreme variations are likely to occur in the critical mass, subject to a multiplicity of factors affecting the interaction.

Nuclear physics is said to be made up from "big ideas" about exceedingly small things. An artists rendition of this concept of the atom is protrayed in Figure 33 by H. E. Krueger of Battelle - Pacific Northwest Laboratories. (Should the reader wish to indulge, he may actually locate the different numbers of electrons that appear at various

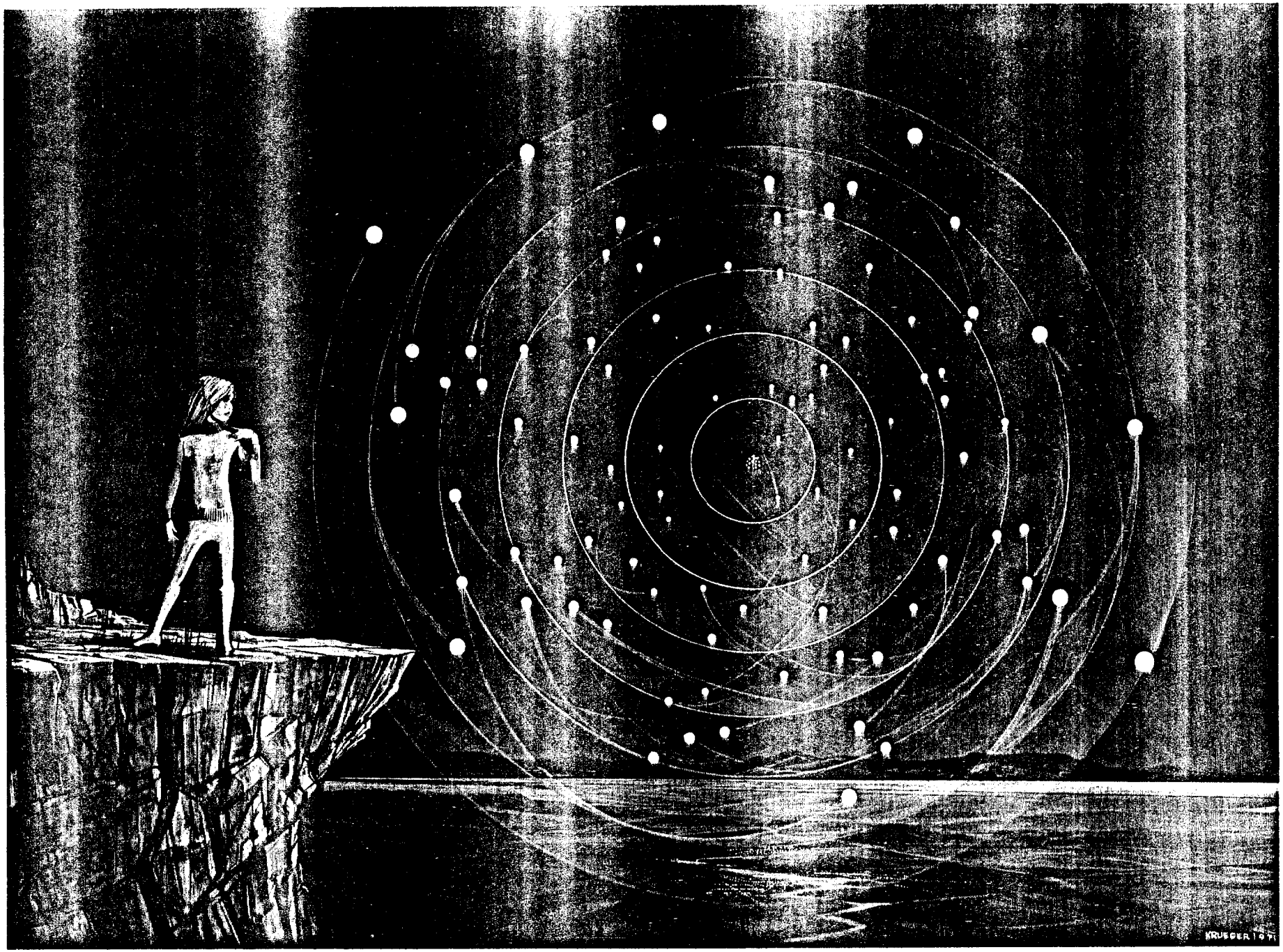


Figure 33. The Atom (A "Big Idea" About An Exceedingly Small Thing)

distances from the nucleus in each of the seven shells for uranium and will note they add up to 92, beginning with two in the innermost shell and ending with two in the outermost shell; (see Figure 34).

Classically, the atom has then been likened to a miniature solar system in which electrons orbit about a heavy, dense nucleus composed of neutrons and protons. Quantum theory continues the subdivision, and, according to some theories, the neutrons and protons (See Figure 34) could possibly in turn be composed of three quarks each, the neutron consisting of two negative quarks and one positive quark, with the proton consisting of two positive quarks and a negative quark. To obtain correct charges for the neutron and proton, the negative quark would have to carry a charge equal to one-third that of the negative electron, whereas the charge of the positive quark would have to be plus two-thirds.

External to the nucleus, the electrons build up in shells, two in the first, eight in the second, eighteen in the third (the equation for the total number of electrons permitted in each shell is No. = $2n^2$), etc. The attraction of one atom for another is due to the interaction of the outer electrons and is known as the valence force, wherein lies the basis for the field of chemistry. The source of chemical energy is the rearrangement of the electrons about the nucleus, whereas nuclear energy comes from the rearrangement of nucleons within the nucleus. The closure, or completion of sets of electron shells or subshells within the principal quantum number, results in atoms that are particularly stable and chemically inactive; under these circumstances stable compounds with other elements simply do not exist. The "cross section" for chemical reaction could then be considered zero in this case. The inert, or noble, gases, He, Ne, Ar, Kr, Xe, Rn, are examples of these types of atoms. These atoms have high ionization potentials; i. e., the energy required to detach an electron has been sharply raised, a state indicating that the electrons are more tightly bound in these cases.

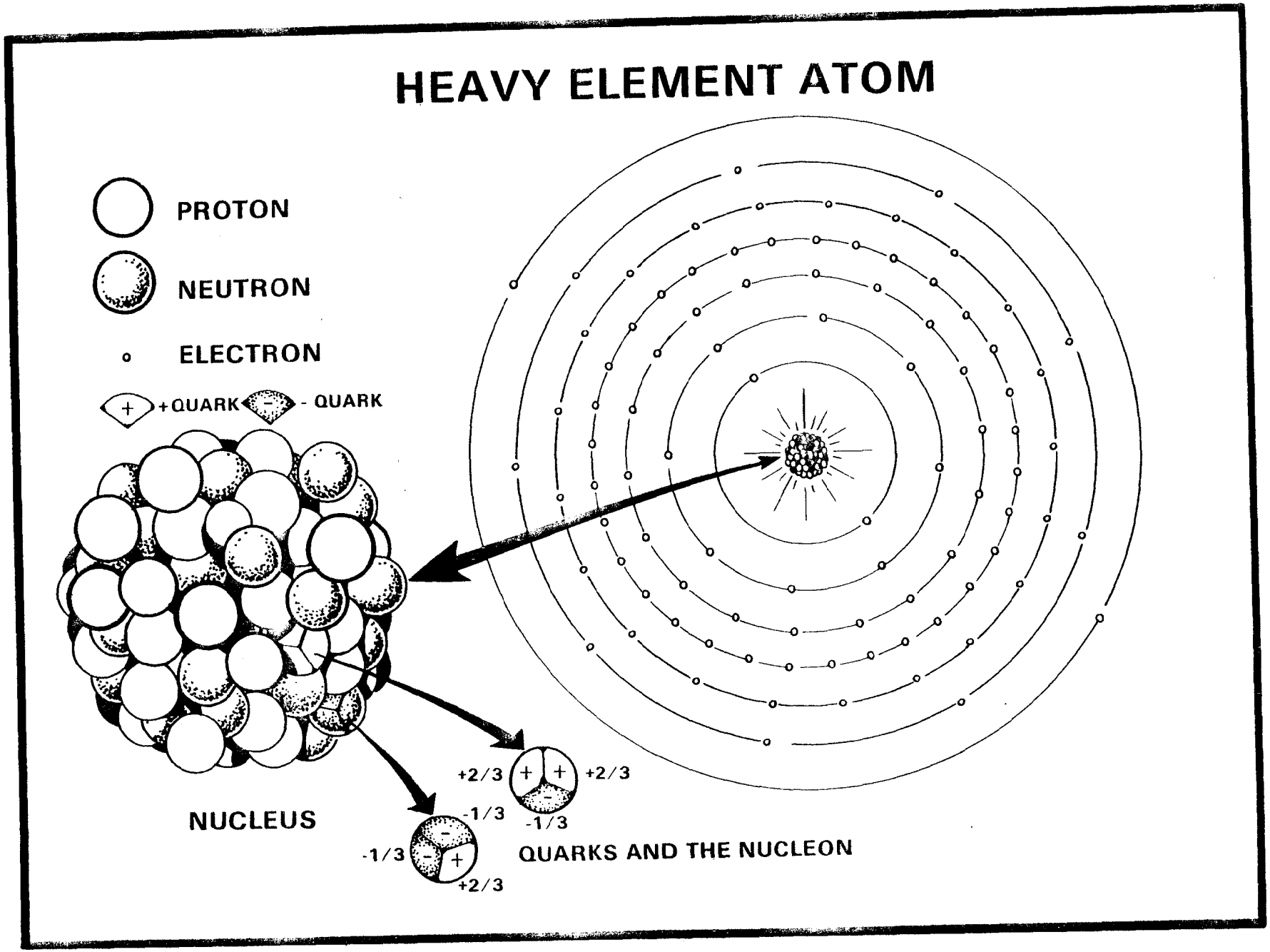

HEAVY ELEMENT ATOM
PROTON
NEUTRON
ELECTRON
+QUARK
- QUARK
NUCLEUS
+2/3
+2/3
-1/3
-1/3
-1/3
+2/3
QUARKS AND THE NUCLEON

FIGURE 34

It is not surprising that the most reactive chemical element is fluorine, an atom short one electron in completing the second shell. Fluorine was discovered in 1886 by Henry Moissan (Recipient of the Nobel Prize in Chemistry in 1906 - the sixth one given). He died in 1907 at the age of 55 and often said, "Fluorine has shortened my life by ten years." Further, owing to its high chemical reactivity, fluorine is extremely difficult to separate from its compounds. Fluorine will even form a compound with Xe, albeit unstable.

The highly successful shell model of the atom serves to explain the sharply increased ionization potentials on the closure of the shells. A somewhat similar situation prevails in the structure of the nucleus. For certain values of N and Z, discontinuities appear in the binding energy of the nucleus.[44] These discontinuities occur[45] at the so-called "magic" numbers, 2, 8, 14, 20, 28, 50, 82, and 126. Nuclei having these numbers of N, Z, or both, are called magic nuclei. The magic numbers appear to be associated with the completion of sets of shells within the nucleus, analogous to the closure, or completion, of sets of electron shells about the nucleus. The closure of a single major nucleon shell results in about 5 MeV of additional binding energy, whereas the closure of two shells, as in the case of $^{208}_{82}Pb$ (doubly magic nucleus, Z = 82, N = 126), gives rise to about 10 MeV of additional binding energy.[46] Neutron capture cross sections drop sharply on completion of the nuclear shells. The magic nuclei are relatively "inert" for interaction with neutrons, in the same sense that the noble gases (with their completed electron shells) are inert chemically. It also is not surprising, then, to find that the Xenon-135 isotope, which is short one neutron in completing a closed shell of neutrons, i.e., $^{135}_{54}Xe$, has the highest absorption cross section (2.6×10^6 barns) for interaction with the neutron. This is analogous to the case of the electron counterpart, fluorine (short one electron in completing a shell). Consistent with their greater stability, nuclei with magic numbers are anomalously abundant.

Also, changing from an odd number to an even number of neutrons releases 1 or 2 MeV (analogous to the completion of a subshell of electrons), wherein lies the explanation as to why some of the actinides are easily fissionable with slow (low-energy) neutrons, but, for others, criticality is possible only with unmoderated, or fast, neutrons.

1. Even-N vs. Odd-N Nuclides - A Reversal on the Effect of Moderation

In Figure 4, striking changes were observed to occur in the critical mass as the fissile isotope was diluted with water. The effect of moderation was to enhance criticality and to reduce the critical mass by a factor of ten. This same type of curve prevails for other odd-n nuclides, ^{233}U and ^{235}U, etc. We shall now cite examples in which the smallest mass occurs for the unmoderated system, where the effect of water is to prevent, rather than enhance, criticality.

Nuclides such as $^{231}_{91}Pa$, $^{237}_{93}Np$, $^{238}_{94}Pu$, $^{240}_{94}Pu$, $^{241}_{95}Am$, $^{244}_{96}Cm$, and $^{252}_{98}Cf$ contain even numbers of neutrons and all are fissionable, though not fissile. Fissionable nuclei are considered those for which a chain reaction can be set up, with the majority of fissions caused by high energy neutrons. Fissile nuclei are those for which it is possible to set up a chain reaction in which the majority of fissions are caused by thermal or slow neutrons. It follows that the known nuclei which are fissile are fissionable, but not the converse. Calculations show that criticality can be obtained with each of the above listed fissionable even-n nuclides but only under essentially unmoderated conditions. Results of some of the calculations that have been made are summarized in Table VIII, which also includes critical mass values for the odd-n nuclides.

TABLE VIII

Neutron Fissionability and Criticality

Nuclide	Type[a]	Z^2/A	E_a[b] (MeV)	B_n[c] (MeV)	$B_n - E_a$ (MeV)	Criticality Aspects: Slow Neutron Chain Reaction	Criticality Aspects: Fast Neutron Chain Reaction	Computed or Measured Critical Masses of Solution Spheres[d]; Thermal Systems at Optimum Moderation (Ref. 47, 48, 50-54): Bare (kg)	Computed or Measured Critical Masses of Solution Spheres[d]; Thermal Systems at Optimum Moderation (Ref. 47, 48, 50-54): Water Reflected (kg)	Computed or Measured Critical Masses of Metal Spheres; Fast, Unmoderated Systems (Ref. 47, 48, 50-54): Bare (kg)	Computed or Measured Critical Masses of Metal Spheres; Fast, Unmoderated Systems (Ref. 47, 48, 50-54): Water Reflected (kg)	Computed or Measured Critical Masses of Metal Spheres; Fast, Unmoderated Systems (Ref. 47, 48, 50-54): Steel Reflected (kg)
$^{227}_{89}Ac$	Odd-Even	34.89	6.29	5.04	-1.25	No	No	-----	-----	-----	-----	-----
$^{228}_{90}Th$	Even-Even	35.53	5.77	5.24	-0.53	No	Questionable	-----	-----	-----	-----	-----
$^{229}_{90}Th$	Even-Odd	35.37	5.42	6.8	1.37	No	No	-----	-----	-----	-----	-----
$^{230}_{90}Th$	Even-Even	35.22	5.88	5.1	-0.75	No	No	-----	-----	-----	-----	-----
$^{232}_{90}Th$	Even-Even	34.91	6.0	4.8	-1.2	No	No	-----	-----	-----	-----	-----
$^{231}_{91}Pa$	Odd-Even	35.85	5.94	5.6	-0.34	No	Indicated	-----	-----	750[50]	-----	-----
$^{232}_{91}Pa$	Odd-Odd	35.49	5.7	6.5	0.8	Questionable	Indicated	-----	-----	-----	-----	-----
$^{233}_{91}Pa$	Odd-Even	35.54	6.06	5.2	-0.86	No	No	-----	-----	-----	-----	
$^{232}_{92}U$	Even-Even	36.48	5.42	5.74	0.32	Yes	Indicated[e]	-----	-----	-----	-----	-----
$^{233}_{92}U$	Even-Odd	36.33	5.1	6.8	1.7	Yes	Yes	1.2	0.59	16.5	7.3	-----
$^{234}_{92}U$	Even-Even	36.17	5.53	5.31	-0.23	No	Indicated[e]	-----	-----	-----	-----	-----
$^{235}_{92}U$	Even-Odd	36.02	5.2	6.5	1.3	Yes	Yes	1.5	0.82	49.0	22.8	-----
$^{236}_{92}U$	Even-Even	35.86	5.64	5.13	-0.52	No	Questionable (estimated k_∞ is ~ 1.0)[54]					
$^{238}_{92}U$	Even-Even	35.56	5.7	4.8	-0.9	No	No	-----	-----	-----	-----	-----
$^{237}_{93}Np$	Odd-Even	36.49	5.7	5.5	-0.2	No	Yes	-----	-----	88[51]	83 [f]	55 [f]
$^{239}_{93}Np$	Odd-Even	36.19	5.83	5.17	-0.66	No	No	-----	-----	-----	-----	-----
$^{238}_{94}Pu$	Even-Even	37.13	5.2	5.6	0.4	No	Yes	-----	-----	12[51]	10.5[f]	6.9[f]
$^{239}_{94}Pu$	Even-Odd	36.97	4.8	6.4	1.6	Yes	Yes	0.905	0.53	10.0	5.42	-----
$^{240}_{94}Pu$	Even-Even	36.82	5.3	5.24	-0.06	No	Yes	-----	-----	33[52]	29[52]	19[52]
$^{241}_{94}Pu$	Even-Odd	36.66	5.0	6.3	1.3	Yes	Yes	-----	0.244[53]	-----	6.0[47]	-----
$^{242}_{94}Pu$	Even-Even	36.51	5.4	5.0	-0.37	No	Yes	-----	-----	90[51]	84[f]	56[f]
$^{241}_{95}Am$	Odd-Even	37.54	5.4	5.5	0.1	No	Yes	-----	-----	58[51]	51[f]	34[f]
$^{242}_{95}Am$	Odd-Odd	37.29	5.1	6.4	1.3	Yes	Indicated[e]	-----	0.019[53]	-----	-----	-----
$^{243}_{95}Am$	Odd-Even	37.14	5.5	5.4	-0.1	No	Yes	-----	-----	54[54]	50[54]	36[54]
$^{243}_{96}Cm$	Even-Odd	37.93	4.5	6.8	2.3	Yes	Indicated[e]	-----	0.122[53]	-----	-----	-----
$^{244}_{96}Cm$	Even-Even	37.77	5.0	5.5	0.5	No	Yes	-----	-----	13.5[52]	11.5[52]	7.6[52]
$^{245}_{96}Cm$	Even-Odd	37.62	4.6	6.5	1.9	Yes	Indicated[e]	-----	0.041[53]	-----	-----	-----
$^{246}_{96}Cm$	Even-Even	37.46	5.1	5.2	0.09	No	Indicated	-----	-----	-----	-----	-----
$^{247}_{96}Cm$	Even-Odd	37.31	4.7	6.2	1.5	Yes	Indicated[e]	-----	2.05[53]	-----	-----	-----
$^{249}_{98}Cf$	Even-Odd	38.57	4.3	6.6	2.3	Yes	Indicated[e]	-----	0.070[53]	-----	-----	-----
$^{250}_{98}Cf$	Even-Even	38.42	4.7	5.1	0.4	No	Indicated[e]	-----	-----	-----	-----	-----
$^{251}_{98}Cf$	Even-Odd	38.26	4.4	6.2	1.8	Yes	Indicated[e]	-----	0.022[53]	-----	-----	-----
$^{252}_{98}Cf$	Even-Even	38.11	4.8	4.8	0.0	No	Indicated[e]	-----	-----	-----	-----	-----
$^{254}_{99}Es$	Odd-Odd	38.59	4.7	6.0	1.3	Indicated[e]	Indicated[e]	-----	-----	-----	-----	-----

a) Proton number; neutron number.

b) E_a = activation energy for fission (fission time 10^{-14} sec); E_a is 0.9 MeV less than fission barrier (Ref. 49).

c) B_n = neutron binding energy for nuclide of mass A + 1 (Ref. 49).

d) Minimum mass for homogenous aqueous solutions.

e) No question concerning the possibility of criticality, but no calculations are known to have been made.

f) Estimated from bare value (Ref. 51).

In considering the criticality of the even-n nuclides, it is noted that the optimum condition for criticality in terms of minimum mass is reversed. It is reversed in the sense that mixing moderator with an odd-n nuclide (such as ^{239}Pu or ^{241}Pu) produces the condition resulting in the smallest critical mass. For the even-n nuclides, moderation is found to prevent - rather than enhance - criticality. The guaranteed presence of a small amount of moderating diluent would then ensure against accidental criticality and serve as a method of criticality control.

The effect is illustrated in Figure 35, which gives the value of k_∞ and critical radius for $^{237}_{93}Np$ as a function of metal concentration. As moderating material is added to a metal-atoms-in-water system, k_∞ decreases steadily, as shown in the Figure, from that for a pure metal at 20.45 kg/liter through a value of unity at a ^{237}Np concentration of about 12.7 kg/liter. Below 12.7 kg/liter, the reproduction factor for an infinite system (k_∞) becomes less than unity and criticality is no longer possible.

The effect of energy degradation also becomes evident in the reflector savings of such a system. (Reference Table VIII). A good moderating material, such as water, returns neutrons of reduced energy to the core. In the case of $^{237}_{93}Np$, many of the reflected neutrons are degraded in energy below the effective fission threshold and, hence, these returning neutrons contribute little to the fission process.

2. ^{238}Pu - No Oddity After All

A prima facie condition for criticality in any finite system is that k_∞ (the reproduction factor for an infinite system) be greater than unity. An empirical correlation of k_∞ versus energy difference B_n-E_a (difference between neutron binding energy and activation energy for fission) is presented in Figure 36 for the even-neutron nuclides

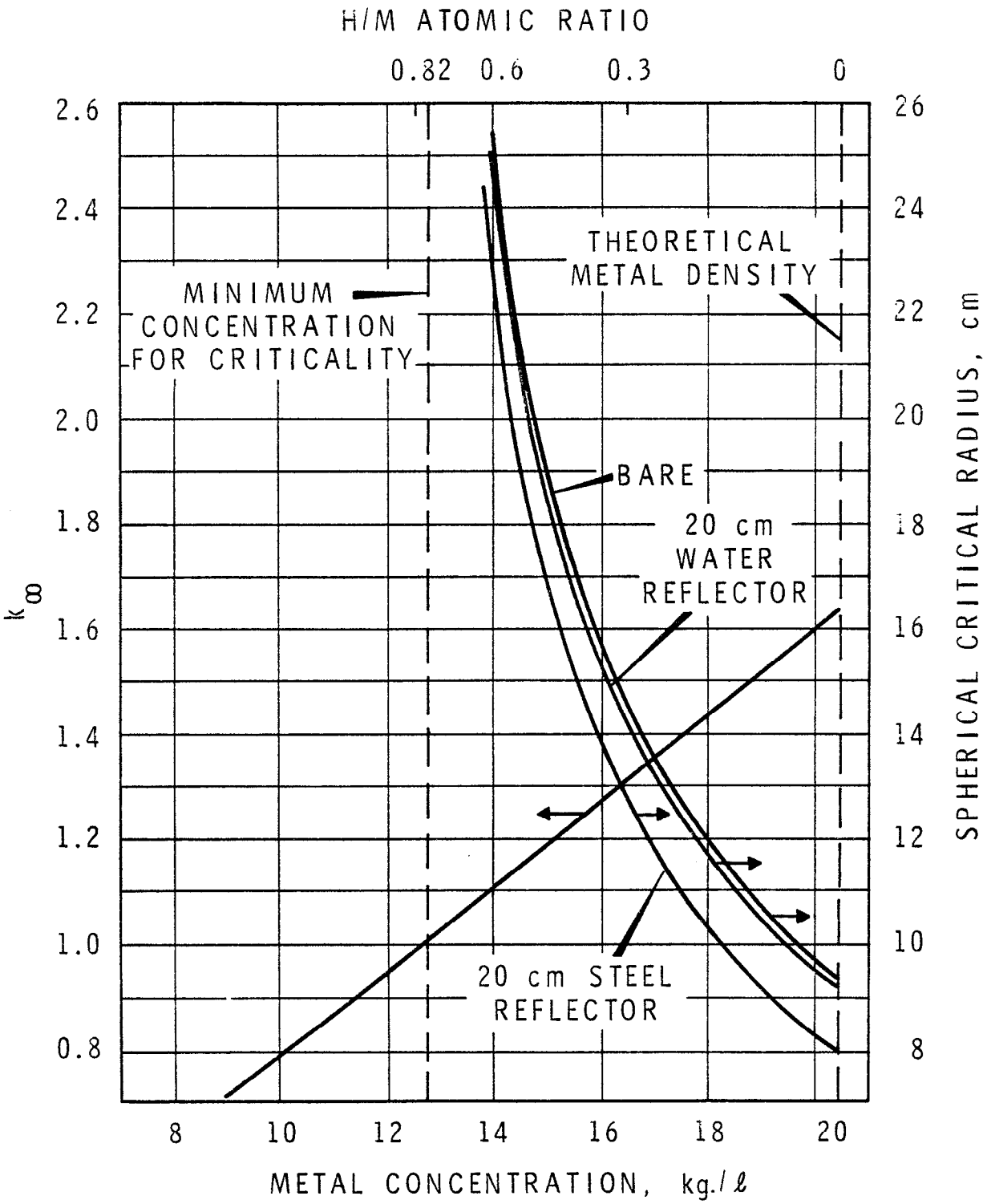


Figure 35. Criticality of Homogeneous ^{237}Np-Water System

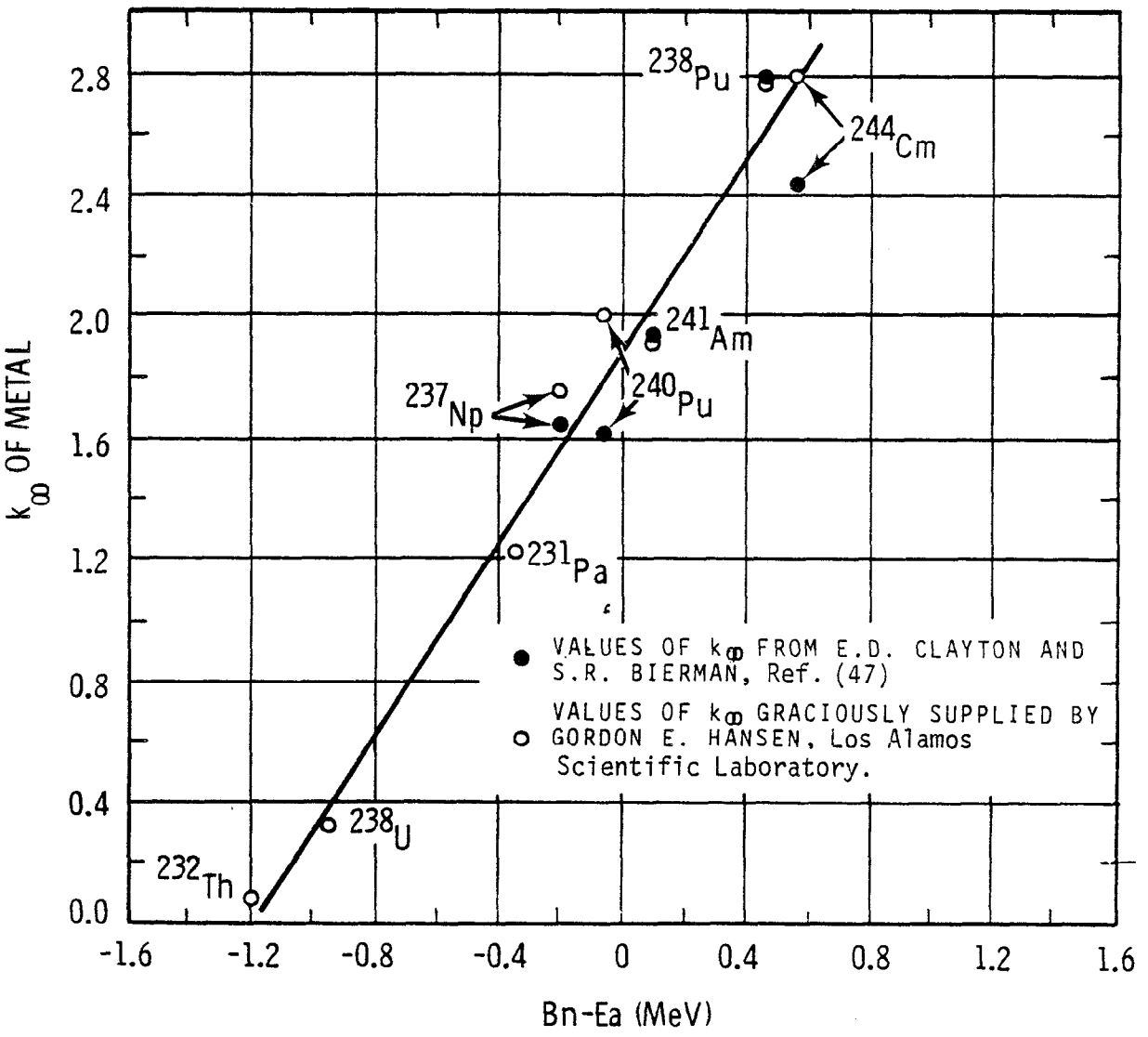


Figure 36. Empirical Correlation of k_∞ vs Energy Difference Bn-Ea (Even-n Nuclides)

on which computed values of k_∞ are available.[48] This empirical correlation suggests that it is possible to provide an estimate of k_∞ for other heavy even-neutron nuclides without recourse to knowlege other than the B_n-E_a difference. Thus, in a qualitative sense, the even-neutron actinide isotopes having potential for criticality may be more easily sorted.

Although there currently is no theoretical explanation, the points group about a straight line which extends from k_∞ = 0.08 ($^{232}_{90}$Th metal) through k_∞ = 2.8 (^{244}Cm metal).

The probability for fission can be expected to be related to B_n-E_a in a qualitative sense. The value of k_∞ depends on ν (neutrons per fission), σ_f/σ (ratio of the fission cross section to the absorption cross section), and σ_1 (inelastic scattering which can degrade the fission neutrons below the fission threshold). Nonetheless, it is reasonable to expect increased fissioning with higher values of k_∞ as B_n-E_a increases. Note that ^{238}Pu, once considered somewhat of an oddity (its bare critical mass in metal form is comparable to that of ^{239}Pu), now appears in a rather logical position relative to the other actinide isotopes shown.

An upper limit for the uncertainty in the calculated values of k_∞ of the heavier elements for the cases presented is considered, at most, no more than $\pm$ 0.2, or $\sim$ 7%. Some confidence also is manifested in the consistency shown in the k_∞ values independently computed at Battelle - Pacific Northwest Laboratories and obtained from Gordon E. Hansen at the Los Alamos Scientific Laboratory. If the empirical correlation is correct, Figure 36 may be used to predict qualitatively the value of k_∞ for other even-neutron nuclides. This could be done without any knowledge of the nuclear cross sections of the elements involved, through simple application of the B_n-E_a difference. For example, k_∞ for $^{250}_{98}$Cf (B_n-E_a = 0.4 MeV) should be $\sim$ 2.50; for $^{252}_{98}$Cf (B_n-$E_a \sim 0$). $\sim$ 1.9, etc.

On examining the critical masses for the even-neutron nuclides (Table VIII), and on referring to the empirical correlation of k_∞ versus B_n-E_a, the evidence strongly suggests the achievement of criticality to be possible with a second naturally occurring element that is lighter than uranium, i. e., the protactinium isotope, $^{231}_{91}Pa$. This isotope would be fissionable with finite critical mass; its k_∞ could be as high as 1.4. The calculations by George Chung Wu in 1977 that predict a critical mass of 750 kg (range in mass due to uncertainties, 570 kg - 930 kg) for a bare spherical ^{231}Pa metal assembly appear to confirm the possibility of criticality.(50)

R. THE "CASE OF THE VANISHING DOLLAR"

1. Even-N Nuclides

The following anomaly, which we have chosen to call "The Case of the Vanishing Dollar" was kindly brought to our attention by Dr. W. Seifritz of the Swiss Federal Institute for Reactor Research.

Calculations and analysis show that criticality would be possible for a number of even-n actinide isotopes including, $^{231}_{91}Pa$, $^{237}_{93}Np$, $^{238}_{94}Pu$, $^{240}_{94}Pu$, $^{242}_{94}Pu$, $^{241}_{95}Am$, $^{243}_{95}Am$, $^{244}_{96}Cm$, $^{246}_{96}Cm$, and $^{252}_{98}Cf$. The results of calculations were summarized in Table VIII. Although criticality now appears possible for each of the above even-n nuclides, it may only be achieved under essentially unmoderated conditions. It is not possible to achieve criticality in a moderated system, because of the fission cross section thresholds for these isotopes. To illustrate, the threshold in the fission cross section for ^{238}U is at about 1 - 2 MeV. An interesting anomaly now develops, because the reactivity in terms of dollars is defined as,

$$\rho = \frac{k_{eff} - 1}{k_{eff}\,\beta_{eff}} .$$

When k_{eff} exceeds unity, the reactivity in terms of dollars becomes infinite as β_{eff} (the effective delayed neutron fraction) approaches zero, and this would be essentially the case for the above listed actinides.

Let us consider a critical bare sphere of ^{238}Pu metal. Since the delayed neutrons from the precursor nuclei have a mean energy of the order of 300 - 400 KeV, neutrons of this energy will have little effect on the chain reaction, as the fission cross section is practically zero below one MeV. Although the fraction of neutrons which are delayed (β) differs from zero, the effective delayed neutron fraction (β_{eff}) becomes small as the percentage of delayed neutrons with energy above the fission threshold is small. This means "the worth of the dollar" can become vanishingly small. Under such circumstances, no state of "delayed criticality" could prevail, but only "prompt criticality." The difference between delayed and prompt criticality can then be expected to be very small for the all the isotopes listed above.

Due to the near impossibility of controlling a prompt critical assembly, and the fact that the effective delayed neutron fraction may be very small, any future criticality measurements on these types of isotopes will, by necessity, be made with great caution.

2. Kinglet Critical Assembly - The Recirculation of Fuel

An interesting criticality condition occurs in the operation of the Kinglet critical assembly, wherein the effectiveness of the delayed neutrons is reduced.[55] In the Kinglet assembly, an enriched uranium solution is circulated at moderate velocity through a region where criticality is achieved. The solution consists of 93.2 wt% ^{235}U in the form of uranyl sulfate, UO_2SO_4, at a concentration of about 90 g ^{235}U/liter. The solution, which is pumped up a 5.0-in. diameter zirconium tube, becomes critical as it passes through a beryllium reflector. A most interesting point is that the fuel circulation causes reactivity variations differing from those in the static

condition. As the flow rate increases, delayed neutron precursors are more effectively swept out of the core. The maximum fuel velocity is some 22.5 ft/sec (the rate capacity of the solution pump ranges up to 1150 gpm). If the returning precursors are ignored, the effect would be to increase the apparent reactivity at which delayed criticality (constant fission rate) occurs, shifting it toward the unchanged condition for prompt criticality. The effective delayed neutron fraction is decreased because some of the delayed neutrons are produced and lost external to the reactor's core. In this case, "the worth of the dollar" has been decreased artificially, by mechnical means via circulation of the reactor's fuel.

S. THE INFINITE SEA CRITICAL CONCENTRATION

The "infinite sea" critical concentration may be referred to in the sense that if we have a large volume of water, and if fissile atoms are added uniformally to form a homogeneous mixture with the water, then at a certain concentration of these fissile atoms criticality will occur. (This is the "infinite sea," or limiting critical concentration). Since thermal values of eta (neutrons produced per thermal neutron absorbed in the fissile atom) for the three principal nuclides of interest, ^{233}U, ^{235}U, and ^{239}Pu are all about 2.0: 2.29, 2.07, and 2.08, this implies that criticality occurs when about one half the neutrons released in fission are re-absorbed in the fissile nuclide and one half in the water, or diluent. (Somewhat less than one half in the case of ^{233}U because of its higher value of eta).

1. Infinite Sea Concentrations and Minimum Critical Masses (The Smallest Criticality Concentration in an Infinite System, - but not the Smallest Mass in a Finite System, and Vice Versa)

The limiting critical concentration for Pu in water is 7.19 g Pu/ℓ (H/Pu atom ratio of 3680).[9] Certain other nuclides such as the deuterium in heavy water (D_2O), carbon, and Be have extremely small

cross sections for absorption of thermal neutrons. Because of this, very small "infinite sea" critical concentrations are achievable in mixtures with these nuclides. For heavy water the critical D/Pu atom ratio is about two million; the concentration in g/ℓ is only about 0.01. For Pu in graphite, the C/Pu atom ratio is about 300,000 and in the case of beryllium, the Be/Pu ratio is about 100,000. For comparison, recall that for light water the H/Pu ratio is 3680, which is much smaller than any of these values. At limiting critical concentrations, masses, of course, are theoretically infinite. As the concentration of the fissile nuclide is increased, the mass is reduced, and in every case there will be a concentration that results in the smallest, or minimum, critical mass (see Figure 4 for the case of Pu in water).

Critical masses have been calculated for bare spherical reactors containing homogeneous mixtures of the fissile atoms, ^{233}U, ^{235}U, and ^{239}Pu in each of the above diluents or moderators.[56] Referring to these calculations, and to Pu in particular for purposes of illustration, it is noted that the smallest critical mass for a Pu-water mixture, in an unreflected spherical vessel, is about 900 g Pu. For a D_2O - Pu (heavy water) mixture, the comparable value is about 1300 g Pu. For C-Pu, and Be-Pu, the values are about 3700 g Pu, and 1500 g Pu respectively. Although each of the above low neutron absorption moderators have much smaller "infinte" critical concentrations for Pu, wherein the mass would theoretically be infinite (k_∞ = one), the smallest possible critical mass for Pu in mixtures of D_2O, C, and Be is significantly greater in each case than for light water.

The reason is principally due to the fact that neutrons slow down much more rapidly in hydrogeneous mixtures. They travel shorter distances in becoming slow or thermal and the fraction of neutrons that escape is less for equal size assemblies. This more than compensates for the larger absorption cross section of hydrogen in the

water mixture at the concentration for minimum critical mass. Then in a finite world and finite situations, water, which is the most frequently encountered diluent, is the "worst" afterall from the viewpoint of inadvertant criticality with these fissile nuclides - the water - Pu mixture, afterall, has the smallest critical mass.

2. Interpretation and Application of Limiting Critical Concentrations of Fissile Nuclides in Water

The American National Standard ANSI N16.1-1975, "Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors," provides in its Table I limits for aqueous solutions.[13] The "infinite sea" concentrations for ^{235}U and ^{239}Pu are given as 11.5 and 7.0 g/ℓ, respectively. It is interesting to note that the limit given for ^{235}U is perfectly valid for any uranium enrichment less than fully enriched, i. e., down to the subcritical uranium enrichment for a homogeneous aqueous solution, which is given as 1.00 wt% ^{235}U. At this enrichment, the maximum value of k_∞ will be near unity, occurring at a uranium concentration of $\sim$ 30 g $^{235}U/\ell$. It is obvious that as the uranium enrichment decreases, the ^{235}U content required for criticality steadily increases. That is, starting with highly enriched uranium in an aqueous homogeneous solution, the value of k_∞ will be less than unity at 11.5 g/ℓ ^{235}U. This limit is also valid for all lesser enrichments, as the presence of ^{238}U will require ^{235}U concentrations $>$ 11.5 g/ℓ if criticality is to be achieved. It should be recalled that an aqueous homogeneous solution of uranium having an enrichment below the limiting critical value of 1 wt% ^{235}U will have a k_∞ less than unity for any concentration whatsoever.

The limit given for plutonium (7.0 g Pu/ℓ), however, will not necessarily be subcritical if the plutonium is mixed with natural uranium. The computed value of k_∞ is given in Figure 37 as a function of the weight fraction of plutonium in Pu + U for homogeneous aqueous

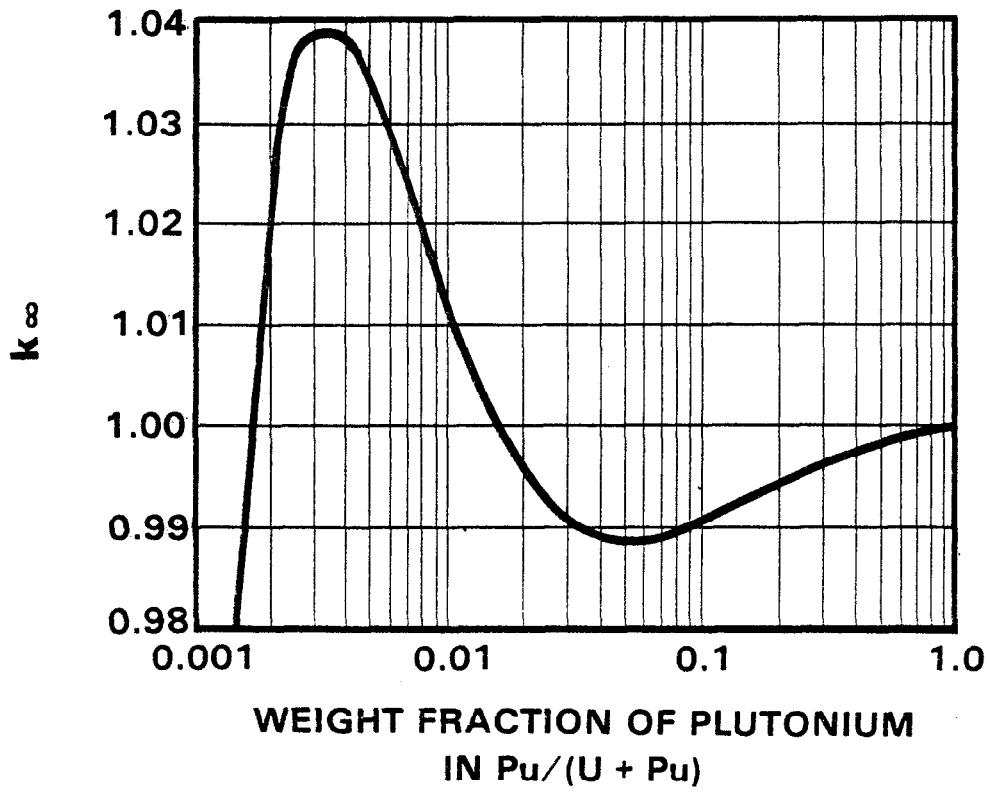


Figure 37. Computed k_∞ vs. Weight Fraction of Plutoniun in Pu + U Homogeneous Aqueous Solutions of ^{239}Pu + U (nat) UO_2 (Plutonium Concentration Held Fixed at 7 g Pu/ℓ)

solutions.[57] Moving from the right (weight fraction of one) to the left, the value of k_∞ is intially seen to decrease from unity until the weight fraction, Pu/(Pu + U), is decreased to $\sim$ 0.05. This is so, even though the total fissile content is being increased [i. e., keeping the plutonium at 7 g Pu/ℓ, the addition of natural uranium (0.71 wt% ^{235}U) adds ^{235}U to the system]. The fall in k_∞ means that, to achieve criticality in this region of the weight fraction curve, the plutonium concentration would have to exceed 7 g Pu/ℓ in the solution. At weight fractions of plutonium < 0.05, the trend is reversed, and the value of k_∞ begins to increase steadily, and at a weight fraction of only 0.0035, it is estimated that the value of k_∞ might be as high as $\sim$ 1.04 for a plutonium concentration of 7.0 g Pu/ℓ. To ensure subcriticality in the solution, the Pu/(Pu + U) weight fraction must be reduced; the limiting subcritical weight fraction for mixed oxides in water is 0.0013. At this weight fraction the ^{239}Pu concentration will be $\sim$ 5 g ^{239}Pu/ℓ; the H/fissile atom ratio giving the highest value of k_∞ is $\sim$ 500.

There are several reasons for the anomalous behavior of the k_∞ curve. First, it should be borne in mind that it is the H/Pu ratio that is the controlling factor in these cases, not the concentration per se. In the aqueous PuO_2, at 7 g Pu/ℓ, this ratio is 3789. If uranium is added, water will be displaced and the H/Pu ratio decreased. The uranium contains 0.71 wt ^{235}U; therefore, for a weight fraction [Pu/(Pu + U)] of only 0.005, but with the Pu + U concentrations such that 7 g Pu/ℓ are contained therein, the ^{235}U concentration will be some 10 g ^{235}U/ℓ. The total fissile content (Pu + ^{235}U) is then some 17 g/ℓ, which will have a k_∞ greater than unity, but the H/fissile atom ratio is now only 1330. Absorption of neutrons in the ^{238}U that has been added will cause k_∞ to decrease, but there also are two factors that will cause an increase:

1. The ^{235}U contained in the natural uranium, although only 0.71 wt%, is important and cannot be neglected.

2. The H/X ratio is simultaneously decreased through displacement of water by uranium.

This increases the value of k_∞ above that for ^{239}Pu alone in water at 7 g Pu/ℓ. Hence, the subcritical limit, as expressed in terms of ^{239}Pu, does not apply to Pu + U (natural) mixtures, contrary to the usual expectation that, if the limit is safe for ^{239}Pu or ^{235}U by itself, the addition of natural uranium should not increase the potential for criticality.

What we are saying is that the limit for plutonium is applicable only if no natural uranium is present - and it is not entirely certain that this point has been made sufficiently clear in the past. If the H/fissile atom ratio is preserved, there will be no problem, but to preserve the H/fissile atom ratio in the aqueous U + Pu mixture, it will be necessary to reduce the concentration of plutonium below the value prescribed in the Standard, i. e., to values < 7 g Pu/ℓ. The Standard does not give the H/X ratios corresponding to the subcritical (safe) limits for either ^{235}U or ^{239}Pu.

3. Criticality in Earth

As noted previously, the limiting critical concentration in water was defined as that uniform concentration of the fissile isotope that is required to obtain k_∞ of unity. In the case of plutonium the "infinite sea concentration is 7.19 ± 0.1 g/liter (H/Pu ratio ≃ 3680).[9] At this point, about half of the neutrons released in fission are absorbed in the diluent (H_2O), since

$k_\infty = 2.08 \cdot f = 1$, where 2.08 is the η for Pu.

It should be remembered that it is not the concentration per se that is important, but rather the ratio of absorbing atoms to fissile atoms which determines this limit, and is the controlling factor.

Let us consider the discharge of dilute plutonium solutions to earth (such as in the case of an underground waste trench or sludge-filled vessel). Initially, there would be no problem of criticality, providing the Pu concentration in the aqueous solution were uniform and less than 7.2 g/liter, for below this concentration k would be less than unity even for an infinitely large system.

Let us assume the Pu to build up uniformly and be held as in a matrix within the sand or soil. Subsequently, the soil begins to dry out and our earth system could become supercritical, even though the concentration of Pu in the soil were significantly less than 7.2 g/liter!

This seemingly anomalous happening may be explained as follows: It is well known that a dilute aqueous solution containing less than 7.2 g Pu/liter could be contained in a large vessel and be well subcritical initially, and subsequently achieve criticality through the simple process of evaporation. In this case, however, the fissile atom density would automatically increase beyond 7.2 g/liter as the water evaporated and the solution concentrated.

The problem of criticality is unique as given herein, because the density of fissile atoms could, theoretically, remain unchanged as the soil dried and yet criticality could occur at concentrations significantly below 7.2 g/liter. The reason is that the soil displaces water and the absorption cross section for "pure" sand is relatively small. Thus, as Pu builds up in the soil (perhaps from solutions containing only milli-gram/liter quantities initially) a concentration could be achieved that is well below 7.2 g/liter, yet on simple evaporation of water from the soil (at a later date) the

system might well become critical. Thus, an abandoned crib could (under the proper circumstances) become critical months or even years later. Note that such a system would likely be autocatalytic in the event of criticality - for the reproduction factor would be further enhanced as the heat from fission evaporated water from the system. It should be noted, also, that under the conditions given, k_{eff} could be reduced by the readdition of water to the system, or the system would be made further subcritical on flooding.

The bases for these conclusions are presented in Figures 38 and 39 and come from an interesting series of calculations by K. R. Ridgway and R. D. Carter on "Criticality Prevention Parameters of Plutonium in Soils."[58-59]

The calculations were made for plutonium-soil mixtures of two different void fractions (the void fraction is the space available within the soil that might be filled with plutonium-water mixtures). Void fractions of 30 and 40 volume percent were assumed, and both fully saturated, and one-third saturated, soil parameters were calculated. The soil composition used was as follows:

TABLE IX

Hanford Soil Compositions, Weight Percent

Component	Dry Soil
SiO_2	81.0
Al_2O_3	6.0
Fe_2O_3	2.0
FeO	2.0
CaO	4.0
MgO	2.0
K_2O	1.0
Na_2O	1.0
H_2O	1.0

Full soil density, no voids: 2.43 g/cm_3

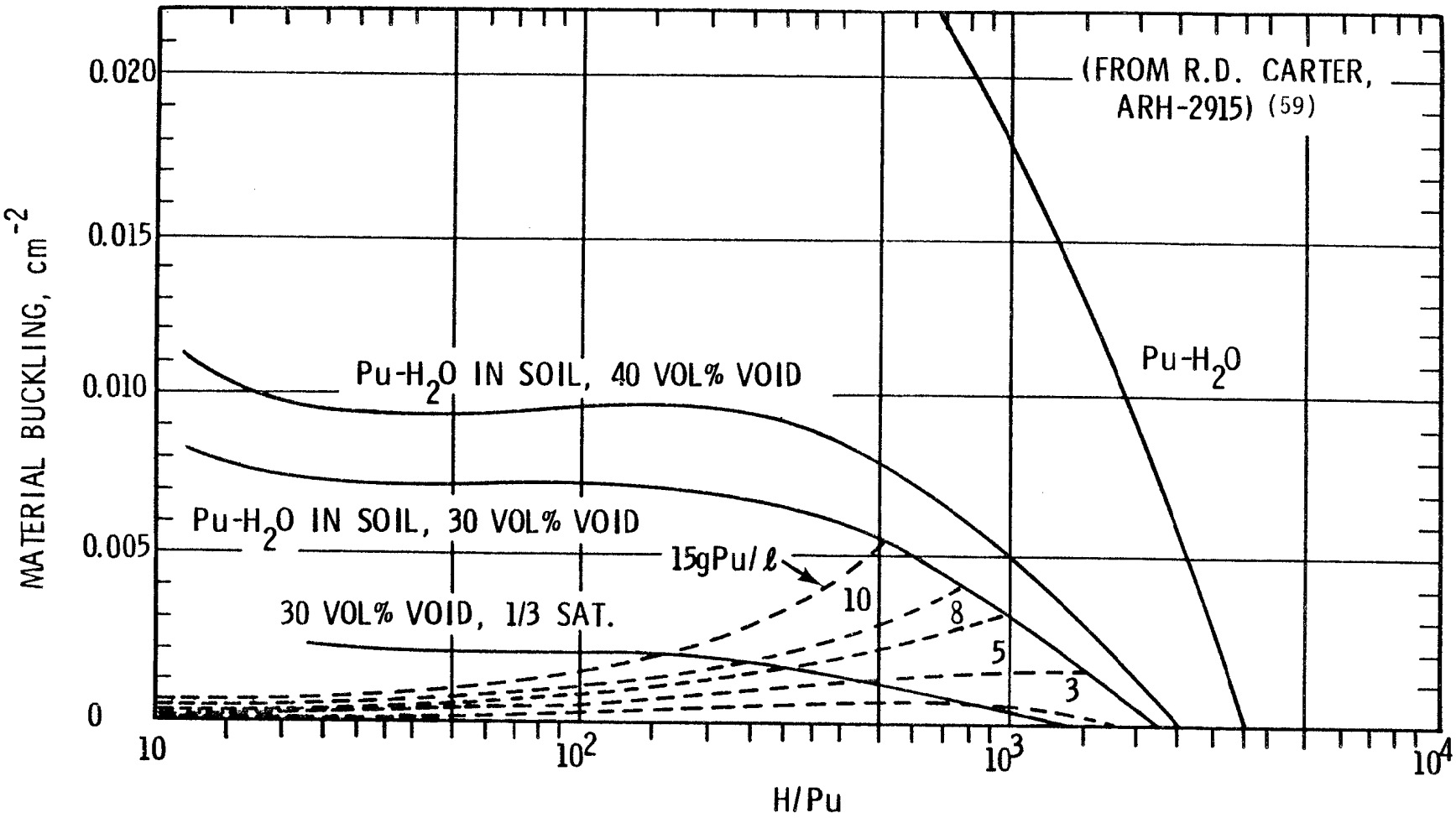


Figure 38. Material Buckling vs. H/Pu (Pu-H_2O) in Soils; 3 wt% ^{240}Pu in Pu, GAMTEC-II Calculation)

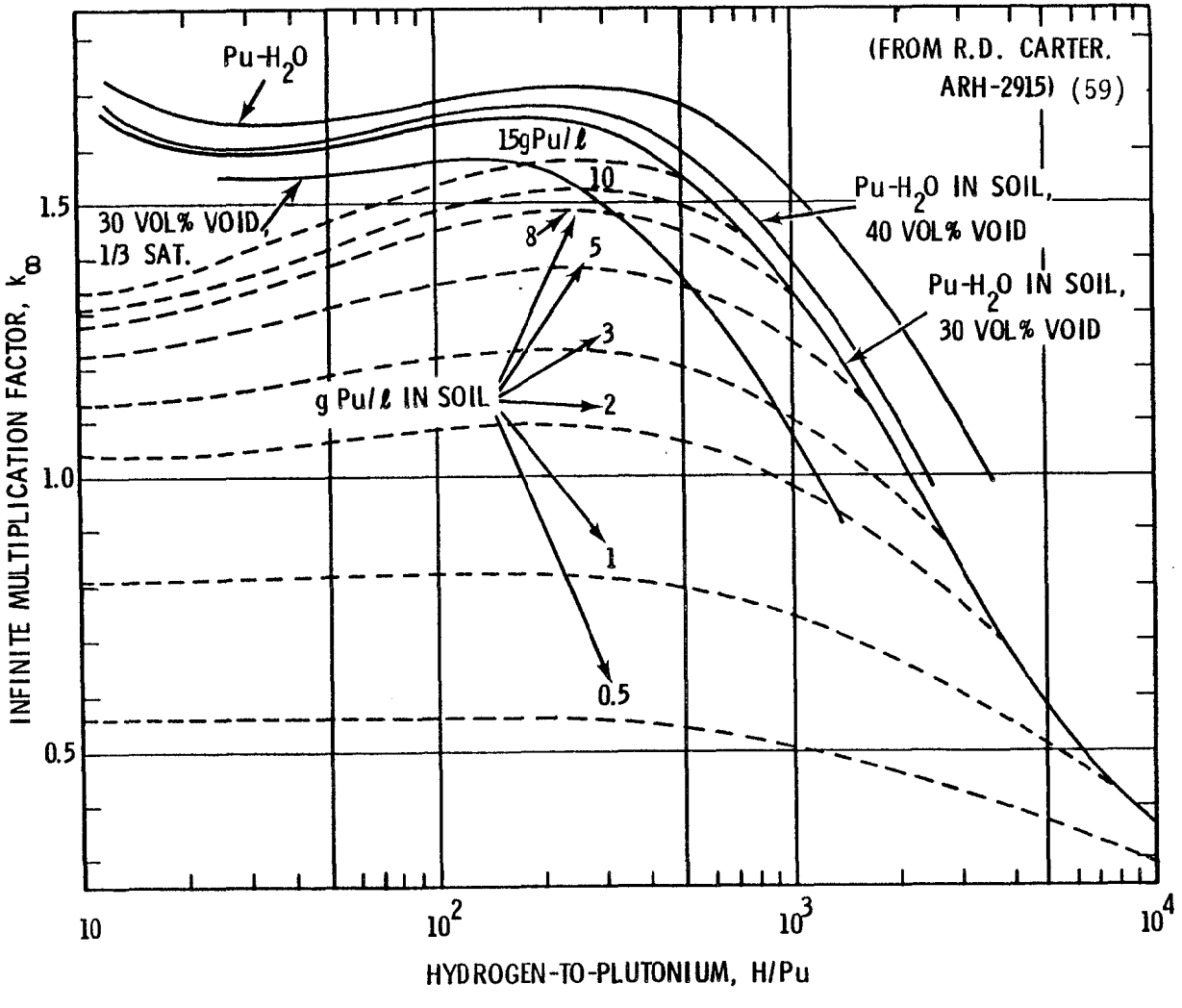


Figure 39. k_∞ vs. H/Pu ($Pu-H_2O$ in Soils; 3 wt% ^{240}Pu in Pu; GAMTEC-II Calculation)

The calculations for soil (assuming 30 vol% void) fully saturated show the value of k_∞ to be about 1.3 at a concentration of 7.2 g Pu/liter (critical mass about 4.2 kg Pu). If water were to be removed however, such as through the process of evaporation, k_∞ would subsequently increase and go through a maximum value ≥ 1.4 during the process. This means that criticality would be possible at concentrations below 7.2 g/liter - perhaps as low as 1.75 g/liter in the soil with the proper dryness (H/Pu $\sim$ 200).

4. The Universe - The Beginning

" . . . the universe is not only queerer than we suppose, but queerer than we can suppose."(a)

The 235 U atomic percentage in U, as found on earth, is remarkably constant, 0.7200 ± 0.006%, with the exception of that found in the Oklo mine (see Section T). The well known half-life for ^{235}U is 7.13×10^8 years; whereas that for ^{238}U is 4.507×10^9 years. The ^{235}U atomic percentage has, therefore, changed continuously throughout the age of the universe. If the concept that the universe evolved from a dense concentration of primeval material some ten billion years ago is correct, then a simple calculation gives the ^{235}U atomic percentage as $\sim$ 96% at the beginning of time; the uranium would have been highly enriched at the time of its formation if it occurred ten billion years ago!

Pertaining to the "infinite sea" concentration and criticality in earth, it was noted that it was not the concentration per se that was the controlling factor, but rather the ratio of fissile atoms to absorbing atoms that was paramount. We may now specualte as to whether there is any lower limit on the critical concentration in the absence of

(a) J. B. S. Haldane, late British Scientist.

any non fissile absorbing atoms; for example, in infinite space - or the "ether." Although of academic interest only, there is technically such a limit.

In this situation, the infinite multiplication constant is the ratio of neutron generation rate by fission to the rate of neutron loss by both β-decay and absorption within a critical system whose lower limit of nuclei density is determined by the radioactive decay constant of the neutron. Under these conditions, we have deduced the equation for estimating the minimum critical concentration for criticality in "infinite ether" (assuming the neutrons are at the average energy of fission, $\sim$ 2 MeV).

Equation for Criticality in Infinite Ether

$$n \, v \, N \, \sigma_a \, \eta = n \, v \, N \, \sigma_a + \lambda_\beta \, n$$

The losses (on the right) come from absorption as well as β - decay of the neutron, since the neutron is radioactive (half-life about 12 minutes) with a mean life of abut 17 minutes. Because of the latter, a neutron that might be released through fission into infinite space would subsequently appear, or be detectable, only as a high energy proton, or a proton of like energy, elsewhere within the universe. In the above, λ_β is the decay constant for β emission by the neutron, taken as 9.6×10^{-4} sec^{-1}. The realativistic velocity for the neutron with 2 MeV of energy is about 2×10^9 cm/sec, or some 12,200 mi/sec. Assuming 1.8 barns for the absorption cross section averaged over the fission spectrum of Pu, and taking η as 2.94, the mean free path, $\lambda = \frac{1}{N\sigma}$, is estimated to be 4.0×10^{12} cm. The latter corresponds to a distance some 100 times that between the earth and the moon, as symbolically portrayed in Figure 40.

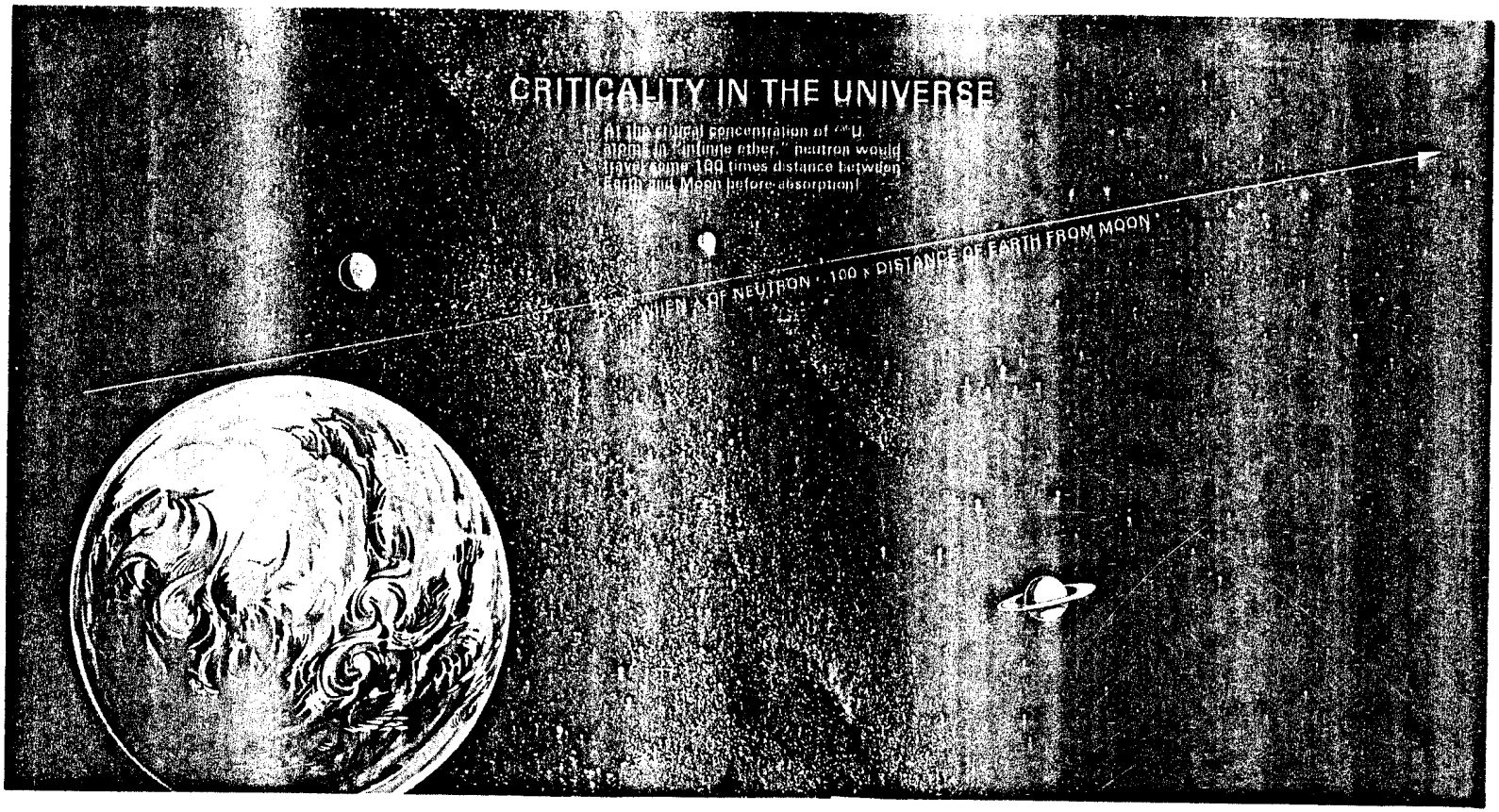

CRITICALITY IN THE UNIVERSE
AVERAGE LIFE OF NEUTRON = 100 x DISTANCE OF EARTH FROM MOON

FIGURE 40

The density of the fissile atoms (or "infinite ether" concentration) would, under these circumstances, be incomprehensibly small - a fraction of a billionth of a gram/cc. Under these conditions (if Pu were considered as fuel) there would only be about 1 α-decay/cc every 8 seconds. In passing, the quantity of material contained within a sphere whose radius is equal to the above mean free path, even at the nuclei density of 0.054×10^{-9} g/cc, would be some 200 times the lunar mass, or about 2 1/2 times the mass of Earth.

To be more precise, it should be considered that some of the fast neutrons will scatter inelastically per collision event. These neutrons will move with less velocity and, hence, will have more time to decay before encountering another Pu atom. In other words, the effect of inelastic scattering is to increase the number of neutrons lost by β-decay. To compensate for this effect, the nuclei density would have to be slightly increased. The criticality equation(s) become considerably more complex under the latter consideration.

5. Criticality Possible in Universe With Fissile Nuclides at Concentrations in Ether Near Permissible Airborne Limits on Earth

The permissible limit for occupational exposure (covering a 50-year time span!) for airborne contamination in the case of the naturally occurring ^{235}U isotope ($T_{1/2} = 7.13 \times 10^{8}$ years) is set by Federal Regulation (10 CFR 20, Appendix B) in activity concentration units ($\mu C/cm^3$). The value expressed in terms of g/cm^3 for ^{235}U in insoluble form is 4.67×10^{-11}. This amounts to 3.7×10^{-6} d/sec per cc; at this level one could expect only about one α-decay from a single cc sample of air to occur every 3 days! (The only practical means of measurement is by sampling large volumes of air). At the limiting concentration for criticality in the universe (near 2.6×10^{11} atoms/cc in the case of ^{235}U) the number of α-decays from

^{235}U would only be about 7.9×10^{-6} d/sec per cc, which only differs by a factor of two from the above "50 year" earth-bound limit for airborne contamination!

T. NATURE'S ANOMALY IN WEST AFRICA

One of the strangest happenings to have been uncovered since the first man-made criticality (December 2, 1942, by E. Fermi and co-workers) was the discovery of nature's criticality in the Republique of Gabonaise.[60] This event is believed to have taken place in primeval times, and the reaction is thought to have remained critical for perhaps one million years. An artist's rendition of the event is portrayed in Figure 41 by M. S. Ferguson of Battelle - Pacific Northwest Laboratories.

This strange phenomenon was brought to light when it was noted that the ^{235}U content from the Oklo mine was much less than normal, ranging in values down to as low as 0.440% in some places where the concentration of uranium in the mine exceeded 20% by weight; also, in some cases, a few samples of very slightly enriched uranium were actually found, making the situation even more puzzling. Now, the isotopic composition of natural uranium is known to be remarkably constant throughout the world. The ^{235}U atomic percentage is $0.7200 \pm 0.0006\%$, with the possible variation being less than the experimental accuracy.

After a detailed and careful analysis, it was concluded that the modifications in the isotopic compositions of the uranium could only be the consequence of nuclear fission reactions. It was found that, in all samples in which the uranium was depleted, the isotopic composition of the rare earth elements differed completely from that of naturally occurring elements and was strikingly representative of fission product yields.

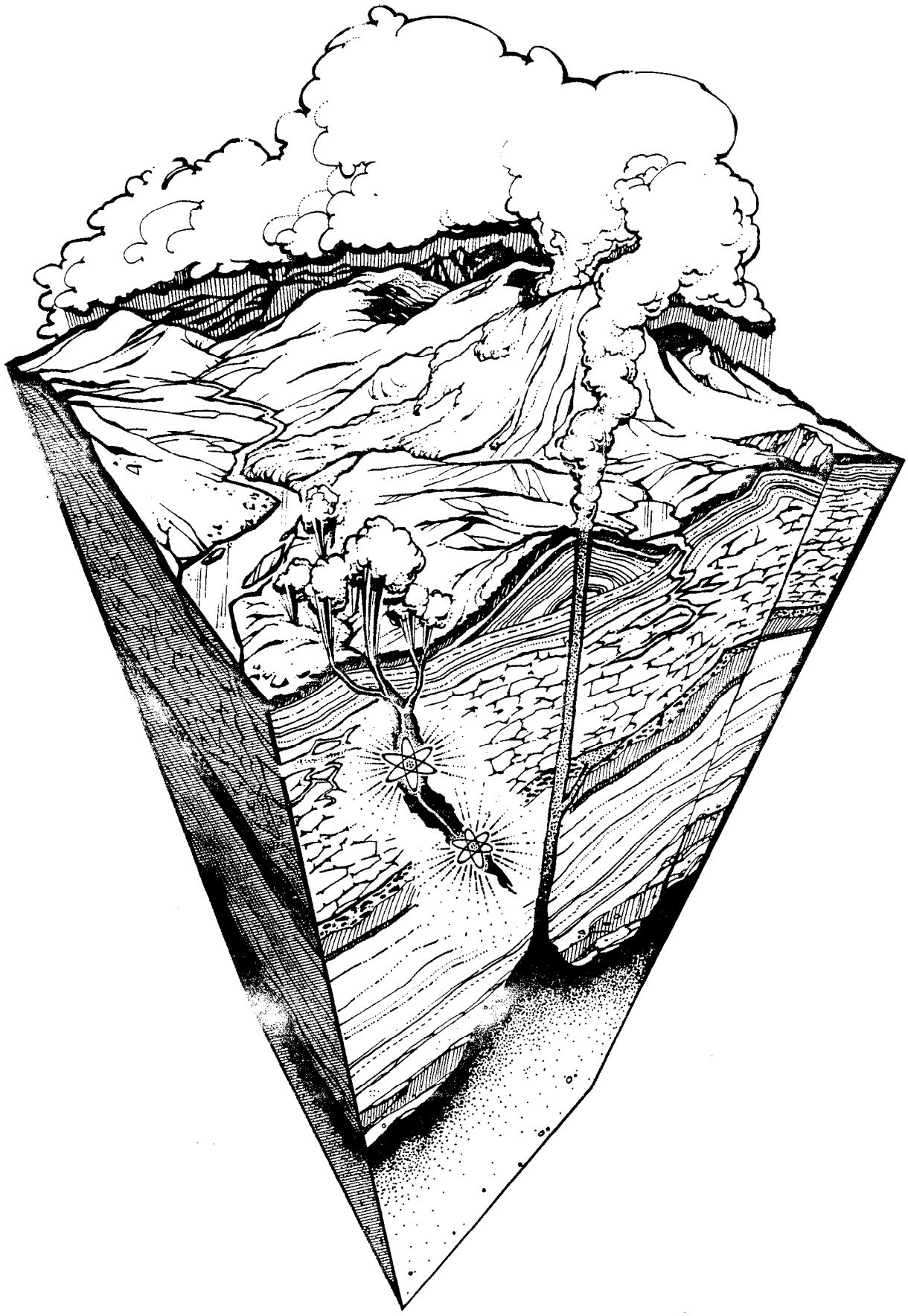


Figure 41. Criticality in Earth - Site of OKLO Mine Circa Two Billion BC

The analysis suggests that the uranium deposited and concentrated at Oklo some 1.74×10^9 years ago was actually close to 3% enriched. The amount of fissionable material consumed during criticality was computed to be of the order 1 to 1.5 tons of ^{235}U. The corresponding energy produced amounted to 2 to 3000 MW-year. This prehistoric reactor would have been somewhat akin to today's light water reactor with a burnup of the order 20,000 MWd/T. The evidence for the event is most convincing, as a result of the elegant analysis made, and there can be little doubt the reaction actually took place - a billion years or so prior to man's achievement of criticality.

U. "SMALL MASS" CONCEPTS

1. Thin Foils and Nonabsorbing Low Temperature Moderating Reflectors

It is now known how, in principle, to obtain a critical configuration with less than an ounce of ^{239}Pu, or with only about one ounce of ^{235}U.[61] These quantities are only about 1/20 of the minimum critical masses prescribed for these nuclides in criticality safety handbooks or safety guides. The study by R. S. Olson and M. A. Robkin was predicated on the surprising observation that an infinite slab of material with $\eta > 1$ immersed in an infinite nonabsorbing moderator would have an essentially zero critical thickness.

As reported by Olson and Robkin, a series of calculations were made with single sheets of ^{235}U and ^{239}Pu metal foils reflected by thick regions of D_2O. The temperature of the core and D_2O were lowered to 4^oK to rethermalize neutrons striking the core (thin fuel sheets) and to take advantage of the absorption characteristics of the fuel. Under these circumstances, a minimum critical mass of only 35 g was obtained for the ^{235}U foil, and only 22 g for the ^{239}Pu foil. The results demonstrate the theoretical possibility of obtaining remarkably small critical masses with the fissile material in the form of

a single foil $\tilde{-}$ 0.2 mil thick. (See R. S. Olson's Thesis for Master's Degree in Nuclear Engineering, University of Washington, 1970, for details).

This concept was further examined in a paper presented in 1977 by K. R. Yates, entitled, "Criticality of Thin Flat Foils Versus Spherical Shells of ^{239}Pu."(62) These concepts are illustrated in Figure 42.

Yates performed a series of KENO calculations to determine the minimum critical mass of ^{239}Pu as a thin square slab and a thin spherical shell immersed in a large volume of D_2O. The calculations assumed that the fissile core and D_2O reflector were at room temperature as provision was not available to adjust the neutron cross sections to 4°K as in the calculations of Olson and Robkin. Hansen-Roach cross sections were used in the calculations.

In Yates's calculations the greatest value of k_{eff} was found to occur with a 390 g spherical shell of 50-cm o.d. (0.0025 cm thick) filled with and centered in a 240-cm cube of D_2O. From these calculations, it appeared that a thin spherical shell required only about 70% of the mass of a thin square slab to achieve criticality under essentially identical conditions of D_2O reflection at room temperature. If, at 4°K, the thin spherical shell also required only 70% of the mass of a thin slab to achieve criticality, perhaps only 22 g x 390/550 = 15.6 g of ^{239}Pu would be necessary for criticality.

2. The Laser-Induced Micro-Explosion

As noted previously, the critical mass of an unreflected sphere will vary inversely as the square of the density, $M_c \sim \rho^{-2}$. For example, if two systems, differing only in density, were critical

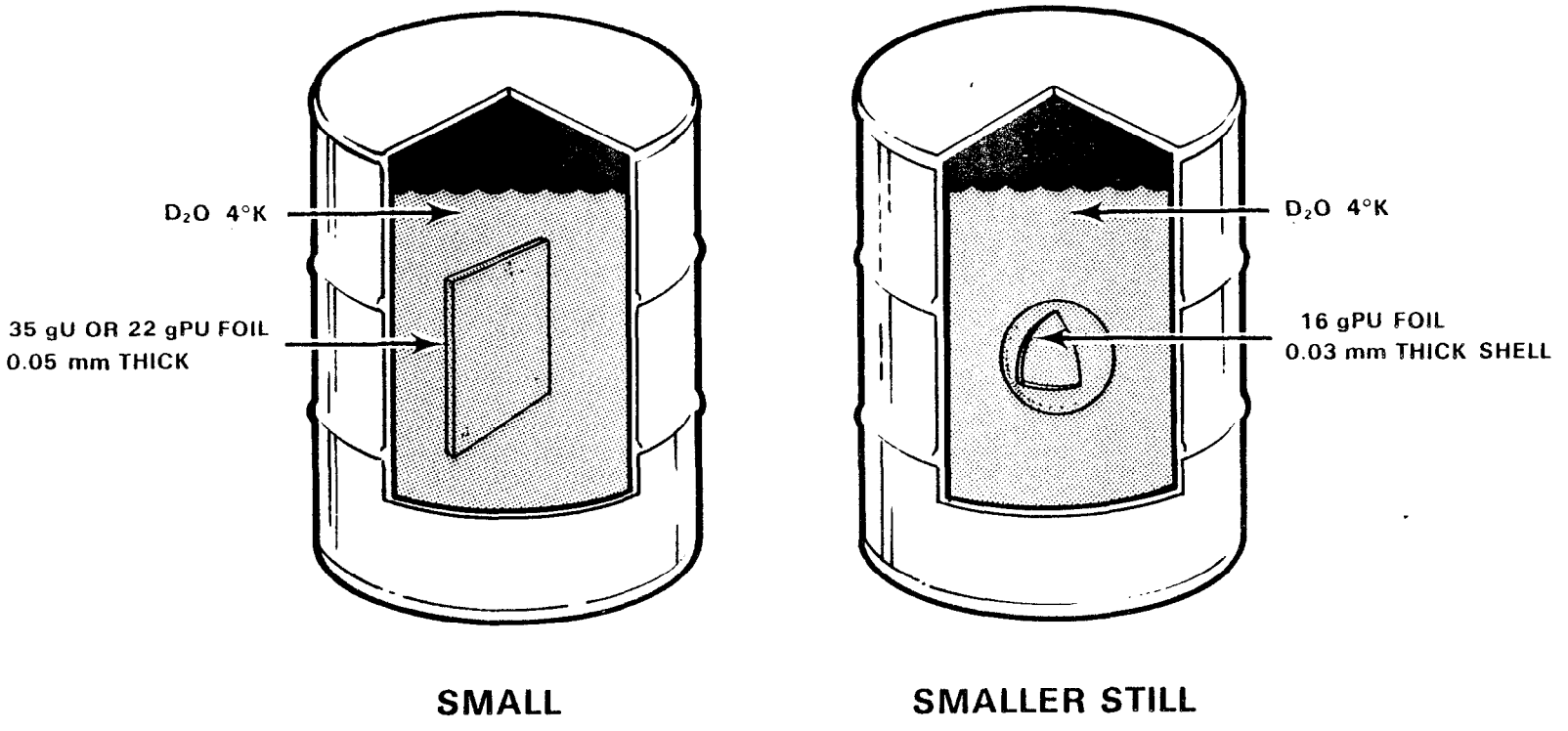


Figure 42. Small Mass Concepts (Thin Foils and Nonabsorbing Low Temperature Moderating Reflectors)

(k_{eff} = unity) but the density of the first was 100 times that of the second, its critical mass would be only 1/10,000 that of the second.

It has recently been suggested that it may be possible, by means of powerful laser beams or intense relativistic electron beams, to compress a small fissionable sphere, of the order of a mm, into a highly supercritical assembly.[63-64] Pressures up to 10^{12} atm, comparable with the pressure in the center of the sun, are believed achievable with advanced giant lasers or electron beams irradiating the small pellet simultaneously from all sides. Under these circumstances the fissile material is said to be compressed to about 250 times normal density. It is further reported by W. Seifritz and J. Ligou that a pellet containing 0.2 g Pu (95% ^{239}Pu and 5% ^{240}Pu of radius 1.35 mm) reflected by a 1.77 mm thick Li^6D shell, could be compressed to a supercritical state having $k_{eff} \tilde{=} 1.25$ if a laser pulse-energy of some 4.7 MJ were absorbed in the outer oblative layer.[64] The number of fissions that would occur during the 0.8 nsec burst is given as 2.33×10^{20}, which is equivalent to an energy release of 1.61 tons TNT (a burnup of nearly 50% would be achieved). Further, in the case of T-D reflector the initial fissionable pellet diameter could be even smaller (containing only 10^{-2} g Pu). (See Figure 43 for an illustration of the laser induced micro-explosion).

The preceding illustration constitutes an example for the effect of ultra-extreme density change on criticality. It would be of great interest, should it prove feasible during the next few years, to perform a successful irradiation confirming the above. Under the conditions stated it would be possible to reduce the critical mass of Pu (or other fissionable material) by tremendous factors; in the case of Pu, some one-half million below that required for Pu metal at normal density.

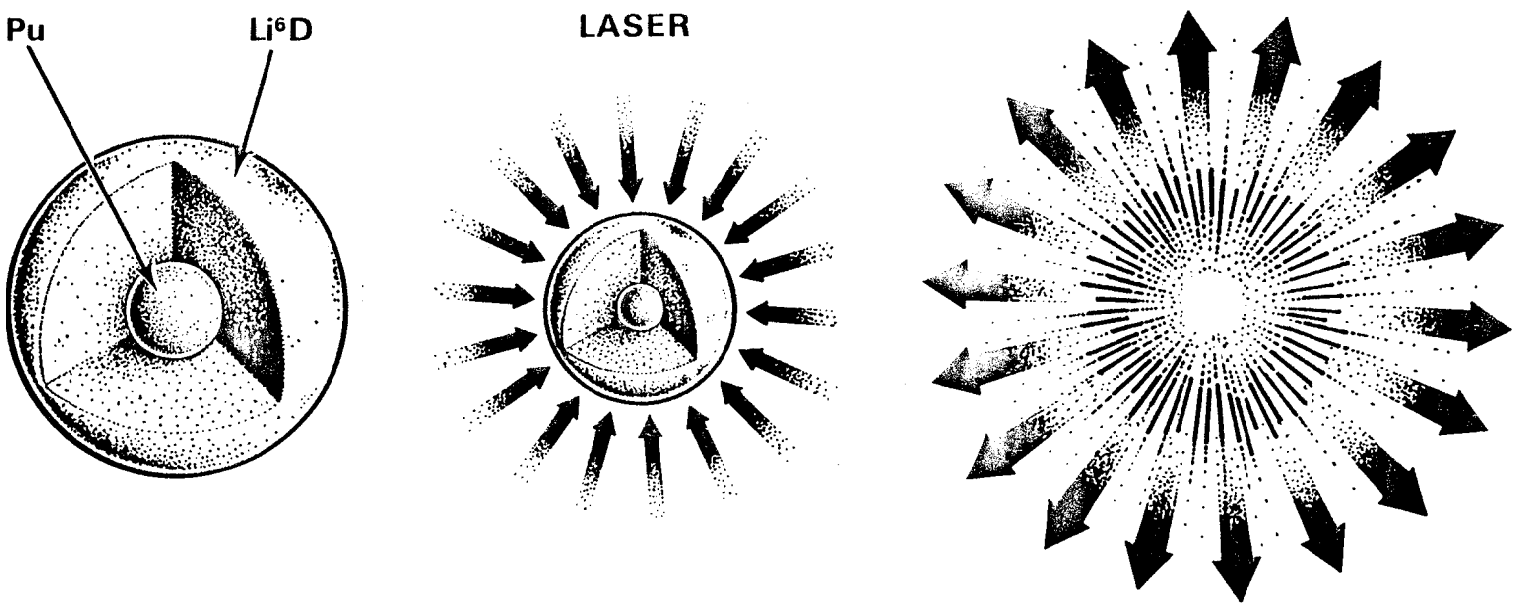


Figure 43. Laser Implosion of Fissionable Pellet

3. The Beryllium Reflector: Polyethylene Moderation and Spacial Redistribution

Low mass concepts have been of special interest since the earliest days of the nuclear reactor, and perhaps more recently in space or satellite applications. Calculations and experiments have been carried out that indicate significant reductions in critical masses can be achieved through use of polyethylene moderation and beryllium reflection. Experiments performed at Los Alamos show the minimum critical mass of ^{235}U in a hydrogeneous core with a thick beryllium reflector ($\sim$ 32 cm. thick) can be reduced to 250 - 300 g.[65] (This is a factor of 3.3 to 2.7 below the commonly quoted minimum critical mass of 820 g ^{235}U for a homogeneous aqueous solution in a water reflected sphere). The Los Alamos experimental assemblies consisted of ^{235}U foils (93% enrichment), polyethylene sheets, and beryllium blocks (as reflector) in a cubic array. Three sizes of fuel cells were studied, being approximately 8, 6.5 and 6 in. square. It was noted that a further reduction in critical mass could be made by spatial redistribution of the ^{235}U fuel. Changing from a constant density throughout the core, i. e., with 20% of the fuel moved to the outer 0.4 cm., a 3% decrease in critical mass could be affected.

Pertaining to alpha-phase plutonium metal, experiments performed at the Lawrence Radiation Laboratory have been reported in which critical thicknesses of beryllium reflectors were determined for five different alpha-phase plutonium metal spheres with masses in the range 2.47 to 5.43 kg.[66] The average density of the Pu used in these experiments was 19.25 g/cc. Extrapolation of the experimental results of the Pu critical mass vs. beryllium reflector thickness indicates that for a beryllium reflector 48 cm thick the minimum critical mass (although poorly determined) would possibly be in the range of only one to two kg Pu. (This may be compared with the water reflected critical mass for alpha-phase Pu of 5.4 kg).

Some calculations were recently reported on criticality parameters for ^{239}Pu in organic media.[67] These calculations show the minimum critical mass for a homogeneous mixture of ^{239}Pu in polyethylene - but with a water reflector, to be 370 g. This may be compared to 530 g for the minimum critical mass of an aqueous Pu solution contained in a water reflected sphere. Had a beryllium reflector been utilized for the polyethylene moderated core, the 370 g value would undoubtedly have been substantially smaller.

There is nothing exotic in the methods (or examples given) by which the reductions in critical masses can be made below the commonly quoted minimum values for water moderated and reflected assemblies. Therefore, in the handling of nuclear material, it is well to consider the possibility of the low mass configurations being encountered.

V. BEYOND CALIFORNIUM - AN ISLAND OF STABILITY - THE SUPERHEAVY ELEMENT "X"

Increased binding energy at closed, or in the region near-closed, shells of nucleons results in increased stability of the nucleus.[44] [46] Shell closures beyond lead are predicted at Z = 114, and N = 184 and possibly at Z = 164 and N = 318.[68] Due to the additional binding, doubly-closed shell, superheavy "magic" nuclei, if somehow formed, might be relatively stable; whereas, nuclei lying in the region beyond the end of the periodic table other than at "islands of stability" would not exist with any significant half-life. This leads to the prediction of the relatively stable superheavy element $^{298}_{114}X$, and to others with closed or nearly closed, neutron and proton shells as graphically illustrated in Figure 44. This particular portrayal was adopted from a paper by G. T. Seaborg.[68]

J. R. Nix has predicted some of the properties associated with the fission of the hypothetical superheavy nuclei.[46] A few of these properties are included in Table X.

KNOWN & PREDICTED REGIONS OF NUCLEAR STABILITY, SURROUNDED BY A SEA OF INSTABILITY

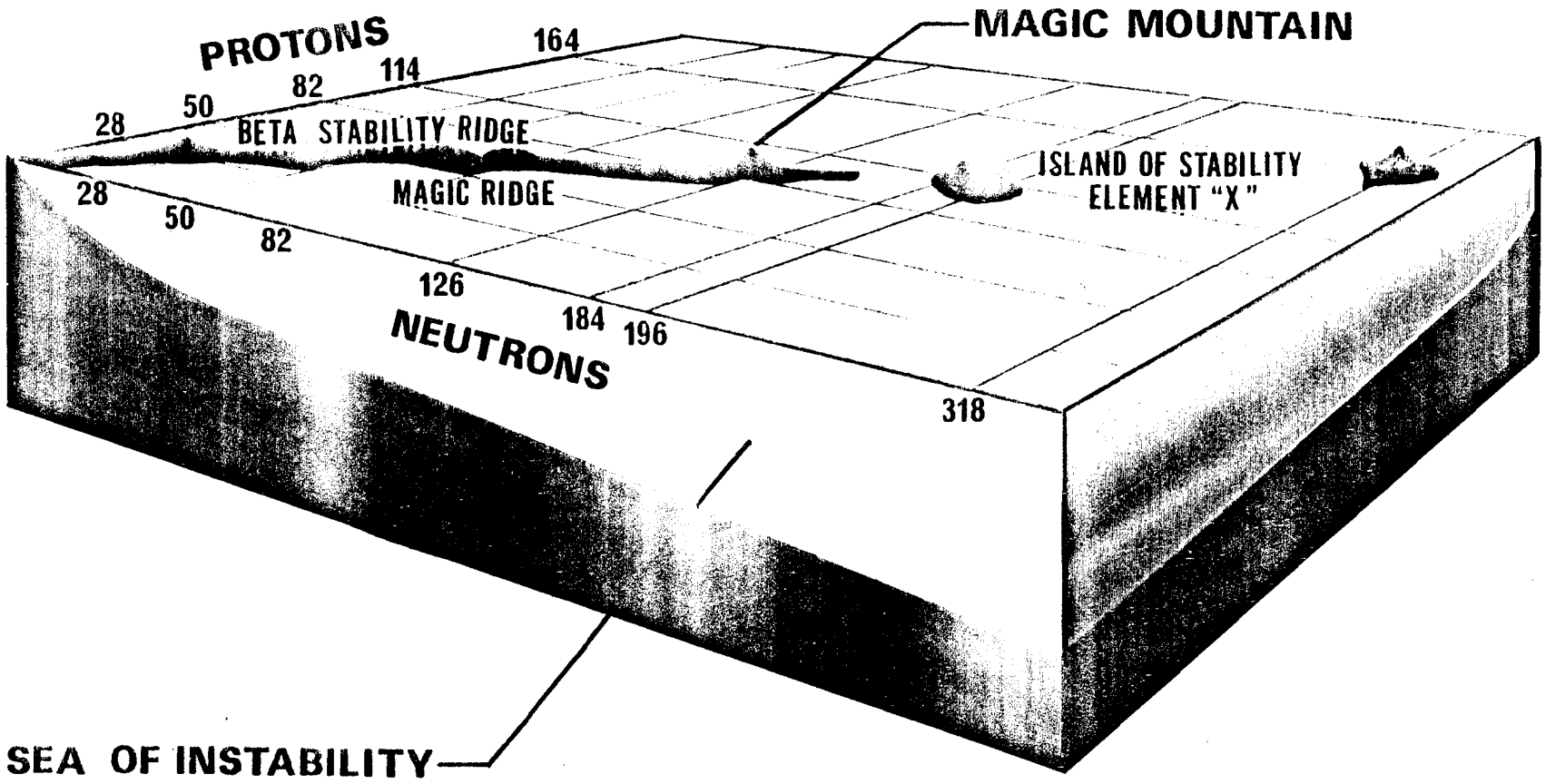


FIGURE 44

TABLE X

Properties of Superheavy Nuclei

Fissioning Nucleus	Energy Release Per Fission	Number of Neutrons Per Fission	Lifetime*
$^{298}_{114}X$	317 MeV	10.5	$\sim 10^3$ years**
$^{294}_{110}X$	290 MeV	10.6	$\sim 10^9$ years

* Total half-life from decay by spontaneous fission, α-decay, β-decay, etc.

** G. T. Seaborg estimate for this nuclide.[68]

Two papers were recently published in SCIENCE, 26 December 1975, Vol. 190, No. 4221, wherein the authors give new evidence for the prior existence of extinct superheavy elements.[69-70] This comes from a study of primitive meteorites in which an anomalous Xe component has been observed. The origin of the Xe component may be the result of spontaneous fission of an extinct superheavy element. Presumably one of the superheavy elements would have an isotope with a half-life in the range 10^7 to 10^8 years, which is too short to survive to the present day, but long enough to leave detectable effects in meteorites. It is suggested that element 115 (or 114, 113) may have been present in a rare chromium when the Allende meteorite was formed. The elegant analyses of the authors in both papers are convincing.

"Micro" Critical Mass

We may now conjecture as to the possible minimum critical mass of superheavy, doubly-closed shell magic element(s) X, as yet undiscovered (although recent analyses on the Allende meteorite provide evidence of previous existence) but theoretically predicted. In making this "qualitative" estimate we have assumed a nucleus one neutron short of the magic number; i. e., a nucleus with an odd number of neutrons but in the region of a closed shell.

$_{114}X$: critical radius $\sim$ 3.44 cm

critical volume $\sim$ 0.17 ℓ

critical mass $\sim$ 0.5 g

Assuming minimum concentration of $\sim$ 3 g/ℓ in aqueous solution; water-reflected sphere. [If a beryllium reflector is used in lieu of water, lets make the critical mass $\sim$ 0.2 g (0.007 oz)].

W. THE POWER REACTOR - FOUR-BILLION WATTS AND SUBCRITICAL

At the entrance of the N-Reactor, on the Hanford Reservation, there is a sign which informs the visitor he is about to enter the largest nuclear steam generating plant in the world. As of August, 1974, the dual-purpose N-Reactor held the Free World's record of steam production for electrical generation at 26.5 billion kW-hr. Yet, this reactor, even if generating 4 billion watts of thermal power in constant mode, is technically subcritical, as the reproduction factor would be fractionally less than unity. This reactor as is true of all others when at constant power level, will be found to be riding on neutron source multiplication where the origin

of the source neutrons is not the chain reaction itself, but rather from α-n reactions taking place in the reactor's core material and from spontaneous fission in ^{238}U or ^{240}Pu, etc. The multiplication rate is given approximately by

$$M = \frac{S + S\,k_{eff} + S\,k_{eff}^{2} + \cdots}{S} = \frac{1}{1 - k_{eff}}, \quad \text{for } k_{eff} < 1.$$

If, somehow, it were possible to remove the source neutrons, the power level would slowly fall to zero unless control rod adjustments were made to compensate. The multiplication rate at full "subcritical" power level could be in the range 10^{11} to 10^{13}, or up to some ten trillion.

X. SUNDRY

1. Gold and Uranium

Two of the most controversial political subjects in recent times have involved gold and uranium, but for different reasons - uranium in connection with the energy crisis and the furor (of some) over the construction of reactors for production of energy, and gold in connnection with "inflation" and the monetary crisis that continues unabated throughout the world. Gold, of course, can be used to purchase energy - so there is a connection. Interestingly enough, uranium is a by-product from the mining of gold in South Africa, just as, for the most part, silver is a by-product from the mining of copper, zinc, lead and other base metals.

In going through the files, the author recently came across a series of criticality calculations, since forgotten, that had been made on gold-uranium mixtures long before either of the current energy or monetary crises. The only "anomaly" here is perhaps the reason as to why such calculations would have ever been made in the first place. The reader may use his own imagination as to the latter. In order that these results be preserved, for whatever value they may now have, some data from these calculations are presented below:

TABLE XI

Multigroup Calculations of k_∞ for Au-U Mixtures

Gold:	^{235}U:	^{238}U	H/U	k_∞	
89:	5.5	5.5	0	0.696	(0.614 ± 0.009 from Monte Carlo Code)
55:	5.5	5.5	0	0.949	
89:	5.5	5.5	5	0.399	
89:	5.5	5.5	10	0.369	

The results indicate that a gold, ^{235}U, ^{238}U atomic ratio of about 50: 5.5: 5.5 should have a k_∞ of unity. Note from the above that as hydrogen is added to the system, k_∞ drops quite rapidly due to the large gold absorption cross section at low energies. In all of the above cases, the uranium was 50% enriched in the ^{235}U isotope.

2. Criticality Accidents - (The Moon, Light Flashes, and Blue Glow)

One of the U. S. Astronauts told the author that on the way to the moon, "we went to sleep counting the light flashes." In one instance, each of the astronauts on board the space craft simultaneously observed the same light flash, i. e., apparently a shower of charged particles initiated by a very high energy cosmic ray particle from the sun passed through the eye of each astronaut. There have been several investigations designed to determine the physical mechanism behind the phenomenon observed by astronauts exposed to radiation in deep space. The diffuse light flashes are apparently observed only when the nucleus of the charged particle moves through the eye fast enough to generate Čerenkov radiation.[71-72] It was first observed by Čerenkov in 1934 that very high speed electrons, or β particles, in passing through a transparent dielectric medium could give rise to visible light.

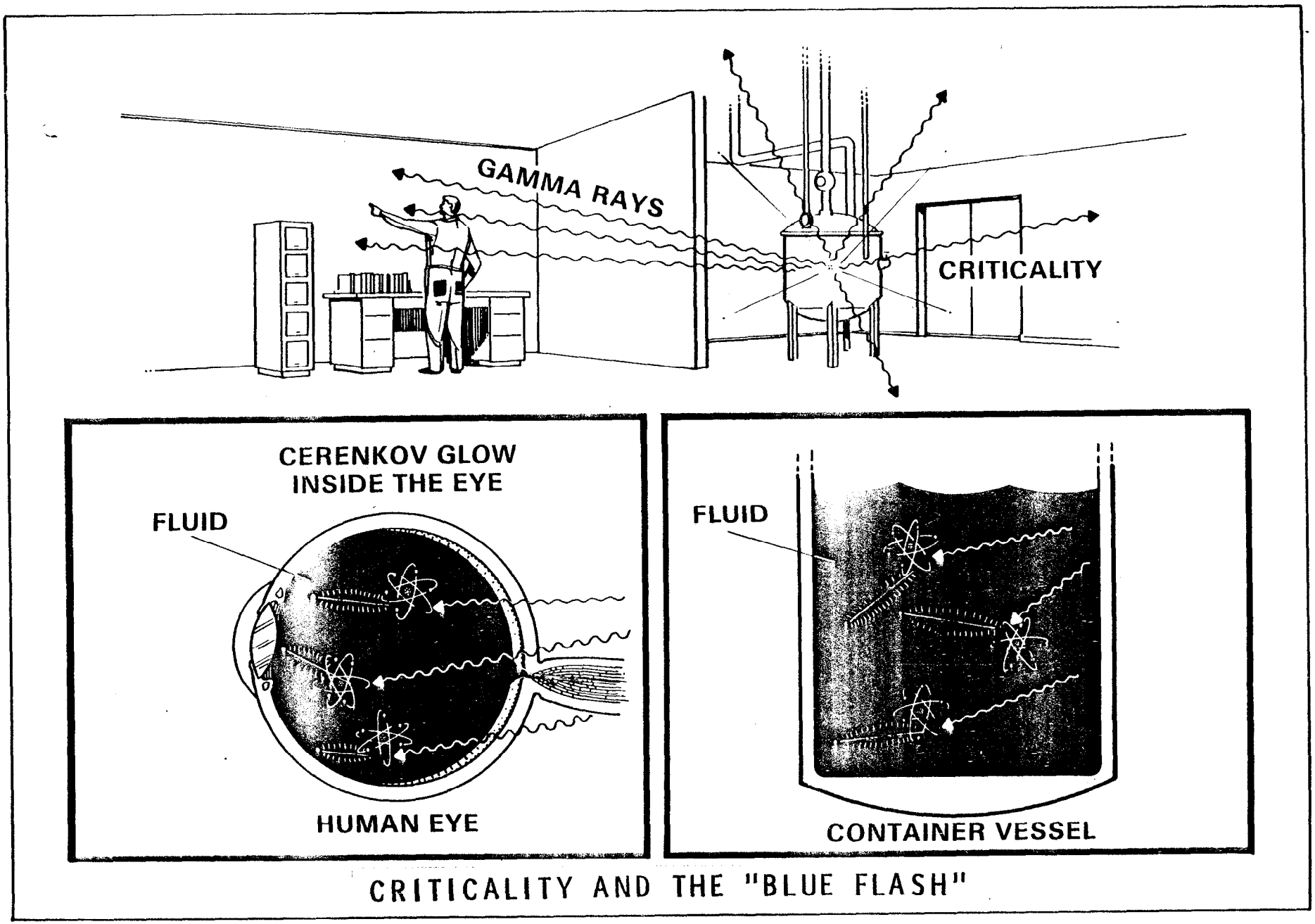
Back on earth, personnel who have been exposed to the radiation from criticality accidents likewise sometimes report seeing a light flash, although from their location it would not at times seem possible for the visible light from the Čerenkov radiation to have directly reached their eye. Yet they claim to have observed a "blue flash." Bear in mind that the visible light of the Čerenkov radiation does not transmit through opague objects such as room walls or the steel walls of a vessel. (In a typical photograph looking down into a reactor core in water, the blue glow is generated under water at the source of the radioactive fuel, and this light is then visible from above). It may be a fruitless endeavor to ask persons who have received a radiation dose, as a consequence of a criticality accident, to identify the source of radiation, or to point out the source of the criticality event following an accident. We suggest that the situation

could be similar to that experienced by astronauts, where in this case the diffuse source of light may again be within the eye itself, but generated through a two stage process; gamma rays from fission interact within the eye tissue to release high energy electrons through such processes as pair production and compton scattering. Neutrons released in fission may also be absorbed in the body tissue, or within surrounding materials, giving rise to capture gamma rays; these in turn may interact to produce high energy electrons within the eye. (For example, the absorption of a thermal neutron in the hydrogen of water gives rise to a maximum gamma ray or 2.23 MeV at the instant of absorption).[73] The electrons in turn give rise to the Čerenkov radiation or "blue flash" observed at the instant of the criticality excursion. The well known condition for the generation of Čerenkov radiation is that the charged particles have a velocity $v > \frac{c}{n}$, where c is the velocity of light in vacuo and n is the index of refraction of light in the medium in which the charged particle is passing. The index of refraction (n) is simply the ratio of the velocity of light in vacuo to that of light in the medium.

Čerenkov radiation is somewhat analogus to the case of sonic booms wherein the aircraft (source of the sound) exceeds the velocity of the sound it creates. A "sonic shock wave" is generated that is heard by an observer on the ground as a sonic boom as the aircraft passes by. Čerenkov radiation is an electromagnetic shock wave phenomenon described as the direct optical analogue of the supersonic boom. For the process to occur it is only necessary for the charged particle to travel through a transparent medium (water, glass, etc.) at a velocity in excess of the electromagnetic waves in the medium. In the case of the Čerenkov radiation the charged particle (source of electromagnetic field) exceeds the velocity of the field associated with the particle. The result

is that a minature "electromagnetic field shock wave" is created and the electrons of the atoms through which the particle moves are accelerated by these fields and so emit radiation. The direction of the light emitted is related to the velocity of the passing particle and makes an angle θ with its direction, wherein $\cos \theta = \frac{c}{nv}$. Bear in mind that it is perfectly possible for the charged particle to move faster than light (electromagnetic radiation) in a medium wherein the index of refraction (n) of the light is greater than unity (according to the laws of physics, it is only the velocity of light in vacuo (c) that may not be exceeded). (For an elegant description of Čerenkov radiation the reader is referred to Reference 72).

Figure 45 qualitatively illustrates the radiation phenomenon as might be observed from a hypothetical criticality incident. Thus, anyone sufficiently close to the source of radiation might be expected to see a "blue flash" or to observe some Čerenkov radiation,, nor would it matter if their eyes were open or closed. To them, (if the light were generated within the eye itself) it might appear to be coming from whatever direction they were looking at the time of the incident.



CRITICALITY AND THE "BLUE FLASH"

FIGURE 45

Y. CONCLUSIONS

An anomaly may be considered a deviation from a common or accepted rule, or something that may be out of keeping with respect to accepted notions of fitness and order. During the course of nuclear energy, a number of apparent anomalies have become evident in nuclear criticality. Some of these have been discussed in the open literature and some have not.

The foregoing examples serve, if for no other purpose, to illustrate the difficulty of attempting to set up a few rigorous, general, rules pertaining to the factors affecting criticality, and illustrate the complexity of criticality itself.

It is shown that there can be as many as three different critical concentrations with the same critical volume, and perhaps four different fuel concentrations having the same critical mass. It is interesting to note the differences and variations that occur in critical concentrations of ^{233}U, ^{235}U, and ^{239}Pu in infinite length cylinders. No single isotope is observed to have the smallest critical concentration over all possible cylinder diameters. On an a priori basis, a plant that was "safe by geometry" for any one of these three isotopes would not necessarily be safe for either of the other two.

Contrary to the usual expectation, the sphere, after all, may not be the configuration of least mass; the reflected cube may be somewhat less under certain circumstances. In some cases, the effect of added scatterers can significantly reduce the critical dimension; whereas, in others, the result can be precisely the opposite. It is noted that reducing the core density can, under some circumstances, actually decrease the critical mass, contrary to the usual expectation that the mass will be increased. Surprising as it may seem, a system with k_∞ < unity

might be made critical by reducing the core size and adding a finite reflector of D_2O, etc. (in the latter case $k_{eff} > k_\infty$!)

In some cases, the effect of moderation results in the smallest critical mass; whereas, in others (depending on the evenness or oddness of the nuclide) the effect is again precisely the opposite. Also, because of the fission cross section thresholds of the even-n actinide isotopes, the "worth of the dollar" can approach zero; due to the lower energy of the delayed neutrons the effective delayed neutron fraction can approach zero. Under these circumstances, no state of "delayed criticality" could prevail, but only prompt criticality as k_{eff} approached unity.

We have seen where a homogeneous aqueous mixture of ^{235}U and ^{238}U could have a smaller ^{235}U critical mass (over a limited concentration range) with low enriched uranium than if the uranium were fully enriched (93.5 wt% ^{235}U).

A number of peculiarities are manifest in the criticality of interacting arrays of subcritical units, that relate to the unit shape, its density, isotopic fuel composition, the lattice density within the array, and the degree of internal and external moderation and reflection involved. Calculations indicate an interacting array of 30.0% ^{235}U enriched metal spheres could have a lower critical lattice density of contained ^{235}U than an array of 93.2% ^{235}U enriched spheres, and thus a smaller critical ^{235}U mass in the lower enrichment array. There also is a case wherein units composed of the same fissile nuclide, unit k_{eff}, and average lattice density in the array can have a different critical number. In addition, an example is given wherein a reduction in k_{eff} of the subcritical units composing the array can actually enhance the overall array criticality.

There is an example wherein the effect of inserting a neutron absorbing rod into a Pu-solution-bearing sphere is to cause the reactivity to initially increase rather than decrease.

Pressures up to 10^{12} atm, comparable with the pressure in the center of the sun, are now believed achievable with advanced giant lasers or electron beams which could change the density of a small pellet of fissionable material under irradiation by a factor of some 250, thus making it possible to achieve a supercritical event in a small pellet of Pu containing as little as 10^{-2} g Pu.

One of the more interesting events in the annals of criticality was the discovery of a possible prehistoric chain reaction (Nature's Anomaly) that took place in the Republique of Gabonaise in primeval times with 3 wt% ^{235}U enriched uranium.

Finally, it is interesting to recall that, in the presence of inherent neutron sources, even the power reactor will be technically subcritical ($k_{eff} < 1$) when operating in a constant power mode at any power level. The list continues, and there are doubtless many other seemingly apparent anomalies that can be cited in the field of criticality.

Z. AND THEN THERE WERE NONE

"The moving finger writes and, having written, moves on;
Nor all your piety or wit shall lure it back to cancel
half a line, Nor all your tears wash out a word of it."(a)

In the early days of aircraft and flying it was recognized that most accidents occurred during take off and landing (something true as well today). In those times the statement was made (and rightly so) that, take offs and landings were all the same, "highly hazardous."

Pertaining to the measurement of nuclear criticality in earlier times, or the assembly of a critical mass, before the factors affecting the chain reaction were so well understood, the "take off" or final stage at which the assembly became chain reacting was always a point of more than casual interest. Many years ago, the author and co-workers performed experiments on graphite-uranium subcritical piles showing for the first time that a natural uranium, grahite-moderated, water-cooled, reactor could be safely designed and operated such that loss of water coolant would not cause an increase in reactivity. This was the so called "cross over point" design. The author also was involved with the initial loading or start-up of a large production reactor based on this design - a reactor that has long since been shut down. To our consternations, it was noted that as fuel was initially being loaded to the graphite core, there was no significant neutron multiplication. If the pattern continued, the reactor might just end up fully loaded with fuel - and be subcritical. A "$200 million" facility might end up as the most expensive repository of uranium in graphite of all time - but it would not be a reactor (bear in mind that in terms of todays depreciated currency the $200 million would be comparable to perhaps a billion). Now the experiments on the small graphite-uranium piles that served as the basis for the lattice design had been carefully

(a) Omar Khayyam - from the Rubiat

performed with high precision and it was believed there was no error. It was then realized that the channels in which the uranium was being loaded were all filled with water. In effect, the reactor was "poisoned down" with a thousand or so "control rods of water." This being understood, there was no further concern, and the neutron multiplication began to appear normal as the fuel loading was further increased; everything finally went according to original prediction. A fuel column was subsequently discharged from the central region of the reactor's core - there was a reactivity gain. A second column was discharged - a further, albeit somewhat less, gain. Now normally one thinks of adding fuel to gain reactivity not by removing it! (Reactors are made critical by adding fuel!) In this case, the behavior is easily explained, but the author will let the reader ponder this "historical anomaly."

At no time in this early work, however, was there ever any hazard to the public at large as in the case of primitive aircraft.

Today, with modern instrumentation, improved knowledge, and well defined operating procedures, the critical experiment may now be performed as safely and routinely, and with as much precision, as the take off or landing of a modern jet aircraft.

The experiments, or calculations, which form the bases of nuclear criticality safety and control were performed by a special breed of persons ("criticality experimenters") many who have vanished, or are now rapidly vanishing from the scene. This is a natural consequence as the critical experiment work is brought to its logical conclusion, and as new critical experiment data requirements have been reduced. The benefits of the contributions of the "criticality experimenters" to nuclear energy will accrue in the course of time largely to their offspring, or the progeny thereof, and will contribute to a higher quality of life for those surviving in the future.

It has been a privilege for the author to have known, and worked with, some of these fine individuals over the course of some years. As has been said, there are those individuals who live out their life in a state of "quiet desparation," but for those of us fortunate enough to have been involved in the field of nuclear energy during its early state of development, there has also been some interesting and highly gratifying moments along the way. But as in the mythical story about the "ten little Indians," one day there was none.

> "I had a dream, which was not all a dream. The bright sun was extinguished, and the stars did wander darkling in the eternal space. Rayless, and pathless, and the icy earth swung blind and blackening in the moonless air."(b)

Perhaps in the end all that any of us can say is that it has been a great privilege for each of us to have lived briefly during a unique cycle of the total history of the cosmos in which nature has been kind enough to have made at least a portion of the cycle knowable to man.

The End

(b) "Darkness" by Lord Byron

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