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E. D. Clayton

ANOMALIES OF NUCLEAR CRITICALITY

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ANOMALIES OF NUCLEAR CRITICALITY, REVISION 6

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ANOMALIES OF NUCLEAR CRITICALITY

A. INTRODUCTION

An anomaly is defined as something not in keeping with expected notions of fitness and order, a departure from the regular arrangement or general rule. In reviewing the literature and experimental data on nuclear criticality and the conditions under which a fission chain reaction can be achieved, a number of anomalies have come to light. The actinide group is composed of the fourteen elements beginning with thorium, Atomic No. 90, and ending with lawrencium, Atomic No. 103.[1] There are about 220 known actinide isotopes, most of which are believed capable of supporting neutron chain reactions. Although every element in the actinide group has at least one isotope believed capable of supporting a chain reaction, from a practical point of view regarding the time for chemical processing, only those nuclides with half-lives more than several weeks are of obvious concern in criticality safety. Forty-six of the nuclides are known to have half-lives greater than six weeks. (Note: The selection of six weeks is somewhat arbitrary. There may well be shorter-lived nuclides of concern in criticality safety). These 46 nuclides are identified in Figure 44. Of the 46 nuclides, 4 are known and 37 are believed to be capable of supporting chain reactions, and 5 are known or believed to *not* support a chain reaction. Figure 44 was constructed in the format used for the *Chart of the Nuclides*.[1] Figure 44 also shows the distinction between fissile and fissible nuclides (more on that later).

The actinides derive their name from actinium (Atomic No. 89) because they tend to maintain an actinium-like electron structure as the fourteen inner electrons are added to the 5f electron shell. Actually, electrons do not appear in the 5f shell until protactinium (Atomic No. 91). It is, however, the inner 14 electrons in the 5f electron shell that distinguish this series from other elements of the periodic table. The first three elements occur naturally and the next eleven are man-made. Trace amounts of Np and Pu have been found, however, in naturally-occurring uranium ore samples as a result of the absorption of neutrons from both cosmic rays and spontaneous fission.

Since the cross sections for the various neutron interactions differ widely between the isotopes, it is to be expected that anomalies will occur. The safety record pertaining to criticality control and prevention of accidental chain reactions outside reactors has been outstanding to date considering the many challenges of a new industry.

The anomalies are presented to add to the awareness of those persons involved with criticality control and prevention and so contribute to a high level of safety within the nuclear industry, which is expected to play a more significant role in the future in view of the currently projected energy shortages.

A large body of knowledge has been accumulated on criticality, and the factors affecting criticality, since inception of the first chain reaction in 1942.[2-6] Criticality, however, is fraught with complexities, and it may be well 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, the discussion begins with a listing of physical characteristics followed by a qualitative description of the concepts and complexities of nuclear criticality.

B. PHYSICAL CHARACTERISTICS[a]

Some physical and nuclear properties of the 46 nuclides with half-lives greater than six weeks are presented in Table I. A property is missing in the table if it has not yet been measured. The density is an important factor affecting the criticality of the metal. Heat generation from radioactive decay is also an important consideration because, for large values, the heat generated from radioactive decay and concomitant excessive temperature would almost certainly preclude the assembly of a critical mass. The material would rapidly melt. Therefore, in these cases, criticality becomes more of an academic consideration than a physical reality. Naturally, this does not preclude criticality in an interacting array made up of small subcritical units (for example, thermoelectric generators in space applications).

[a] The calculations of heat emission are by C. T. Rombough, of CTR Technical Services, May, 1996.

Table I. Some Physical and Nuclear Properties of 46 Actinides

Nuclide	Half-Life	Type of Decay	Heat[(a)] Emission after 1 yr (watts/g)	Spontaneous Fission Half-Life (yrs)	Elemental Density (g/cm^3)	Melting Point (˚C)
^{228}Th	1.913 y	α – 100%	115.5	-	11.72	1750
^{229}Th	7880 y	α – 100%	.0387	-		
^{230}Th	75380 y	α – 100%	.0006	1.51×10^{15}		
^{232}Th	1.41×10^{10} y	α – 100%	< .0001	1.41×10^{19}		
^{231}Pa	32760 y	α – 100%	.0017	1.09×10^{16}	15.37	1600
^{232}U	68.9 y	α – 100%	2.04	-	18.95	1132
^{233}U	159200 y	α – 100%	.0003	2.27×10^{17}		
^{234}U	245500 y	α – 100%	.0002	1.44×10^{16}		
^{235}U	7.04×10^{8} y	α – 100%	< .0001	1.01×10^{19}		
^{236}U	2.34×10^{7} y	α – 100%	< .0001	2.44×10^{14}		
^{238}U	4.47×10^{9} y	α – 100%	< .0001	8.94×10^{13}		
^{235}Np	396.1 d	ε – 100%	.502	-	20.25	640
^{236}NP	154000 y	ε – 87.3% β – 12.5% α – 0.15%	< .0001	-		
^{237}Np	2.14×10^{6} y	α – 100%	< .0001	1.07×10^{18}		
^{236}Pu	2.858 y	α – 100%	14.96	2.04×10^{9}	α phase – 19.6 σ phase – 15.8	641
^{237}Pu	45.2 d	ε – 100%	.0314	-		
^{238}Pu	87.7 y	α – 100%	.564	4.06×10^{10}		
^{239}Pu	24110 y	α – 100%	.0019	8.04×10^{15}		
^{240}Pu	6564 y	α – 100%	.0071	1.15×10^{11}		
^{241}Pu	14.35 y	β – 100%	.0089	7.18×10^{16}		
^{242}Pu	373300 y	α – 100%	.0001	6.79×10^{10}		
^{244}Pu	8.08×10^{7} y	α – 100% sf – 0.12%	< .0001	6.73×10^{10}		
^{241}Am	432.2 y	α – 100%	.115	1.08×10^{14}	13.67	994
^{242}Am[(b)]	141 y 16 h	it – 99.5% α – 0.45% β – 82.7% ε – 17.3%	.268	2.82×10^{14}		
^{243}Am	7370 y	α – 100%	.0067	1.99×10^{14}		

Table I (cont'd.) Some Physical and Nuclear Properties of 46 Actinides

Nuclide	Half-Life	Type of Decay	Heat[(a)] Emission after 1 yr (watts/g)	Spontaneous Fission Half-Life (yrs)	Elemental Density (g/cm^3)	Melting Point (˚C)
^{242}Cm	162.79 d	α – 100%	26.3	7.19x10^6	13.51	1340
^{243}Cm	29.1 y	α – 99.7% ε – 0.27%	1.80	5.49x10^{11}		
^{244}Cm	18.1 y	α – 100%	2.73	1.39x10^7		
^{245}Cm	8500 y	α – 100%	.0057	1.39x10^{12}		
^{246}Cm	4730 y	α – 99.97% sf – 0.03%	.0101	1.58x10^7		
^{247}Cm	1.56x10^7 y	α – 100%	< .0001	-		
^{248}Cm	340000 y	α – 100% sf – 8.26%	.0005	4.12x10^6		
^{250}Cm	9700 y	sf – 80% α – 11% β – 9%	.141	1.21x10^4		
^{247}Bk	1380 y	α – 100%	.0366	-	≈ 14	-
^{248}Bk	9 y	α – 100%	6.56	-		
^{249}Bk	320 d	β – 100%	.243	1.87x10^9		
^{248}Cf	333.5 d	α – 100%	29.4	3.15x10^4	-	-
^{249}Cf	351 y	α – 100%	.152	6.75x10^{10}		
^{250}Cf	13.08 y	α – 99.9% sf – 0.08%	3.86	1.64x10^4		
^{251}Cf	898 y	α – 100%	.0581	-		
^{252}Cf	2.645 y	α – 96.9% sf – 3.09%	29.9	85.6		
^{254}Cf	60.5 d	sf – 99.69% α – 0.36%	153.5	0.166		
^{252}Es	471.7 d	α – 76%	35.4		-	-
		ε – 24%				
^{254}Es	275.7 d	α – 100%	33.3	2.52x10^7		
^{257}Fm	100.5 d	α – 99.8%	35.2	131.12	-	-
^{258}Md	51.5 d	α – 100%	42.6	4703.2	-	-

Key: it – Isomeric transition (through or conversion-electron decay)
α – Alpha decay ε – Electron capture
β – Beta (β^-) decay sf – Spontaneous fission

(a) This value also includes the heat emission from daughter products. For nuclides having a half-life much less than one year, most or all of the heat emission will be due to the decay of the daughter products.

(b) The half-life of the isomeric state, ^{242m}Am, is 141 years, while the ground state half-life is only 16 hours. The total heat generation from both states, when originally formed, is low at ~0.017 watts/g. The daughter product of the β-decay of ^{242}Am is ^{242}Cm, which has a half-life for α decay of 163 days. Consequently, the heat emission in a sample of ^{242}Am builds up to 0.268 watts/g after 1 year and 0.333 watts/g after 3 years.

C. 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 that may be used; on the nature of any solvents or diluents; 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 Economies and Criticality in Uranium

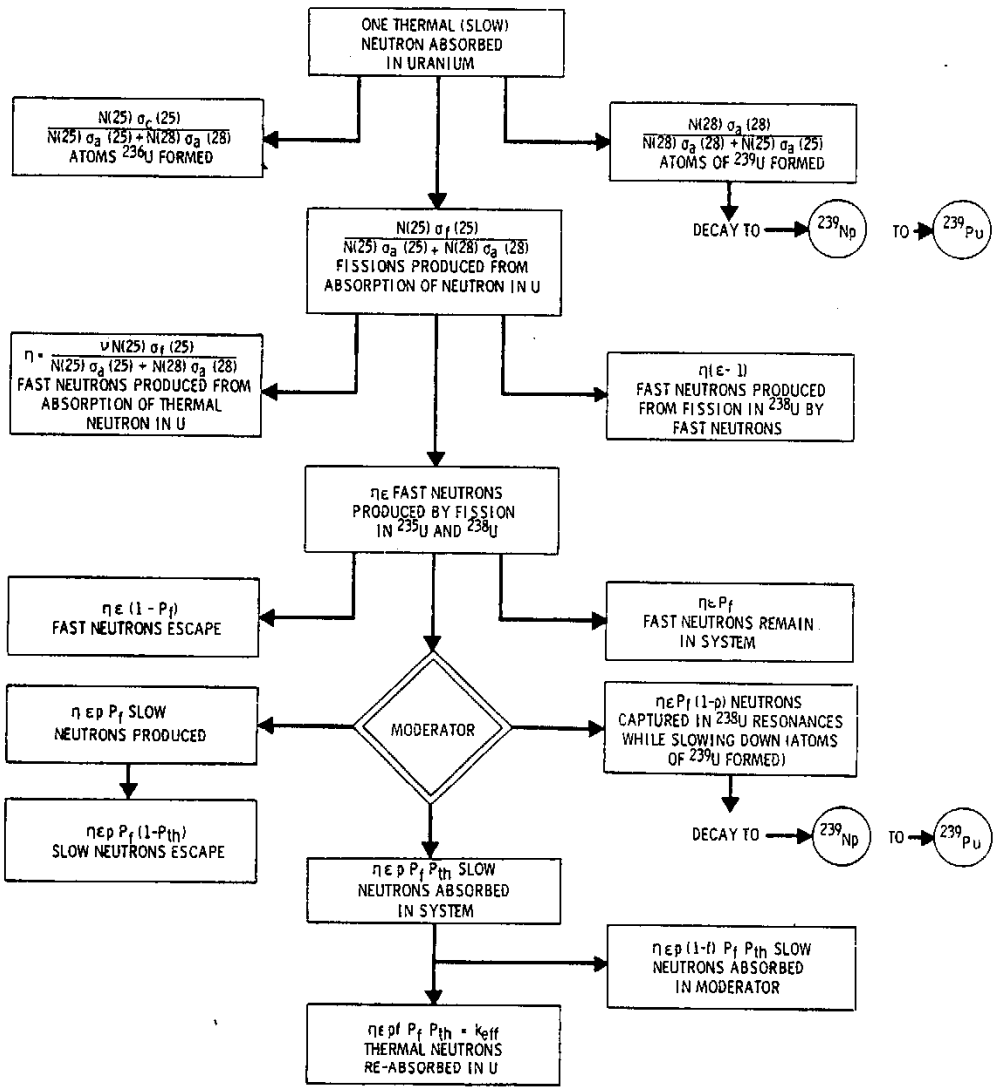
To illustrate, the neutron economy for a homogenous mixture of U and diluent is presented in abbreviated form in Figure 1.

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 = εpfη, 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.

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

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 collision of a neutron 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 hydrogenous materials) that can moderate or slow down the neutrons. Fast neutrons lose energy through collision 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 (~ 0.025 e.v.)



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

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, ~ 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} ~ 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, safety is commonly evaluated in terms of a given value of k_{eff}; i.e., the system is safe provided 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 that have the same fraction of critical mass may have different values of k_{eff}, e.g., 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.

The weird complex variation in k_{eff} of fraction of critical mass vs. critical mass for spheres, is shown in Figure 2, where k_{eff} has been computed by R. D. Carter, et al,[2] for two cases: 50% of critical mass and 75% of critical mass, 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.

Figure 3 gives k_{eff} of the fractional critical cylinder diameter vs. the critical cylinder diameter for infinite cylinders.[2] 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 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.

Figures 4 and 5 illustrate these effects for uranium-235 – water mixtures. Figure 4 shows the variation in k_{eff} for spheres containing 50% and 75% of the critical volume. Figure 5 gives the variation in k_{eff} for infinite slabs that are 85% of the critical slab thickness. For one of the latter cases (Figure 4, bare sphere), k_{eff} ranges from a low value of about 0.75 to a high one of about 0.96 at one-half the critical volume.

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 be 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:

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

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

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

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. Table II is presented for purposes of illustration.[7]

Table II. 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

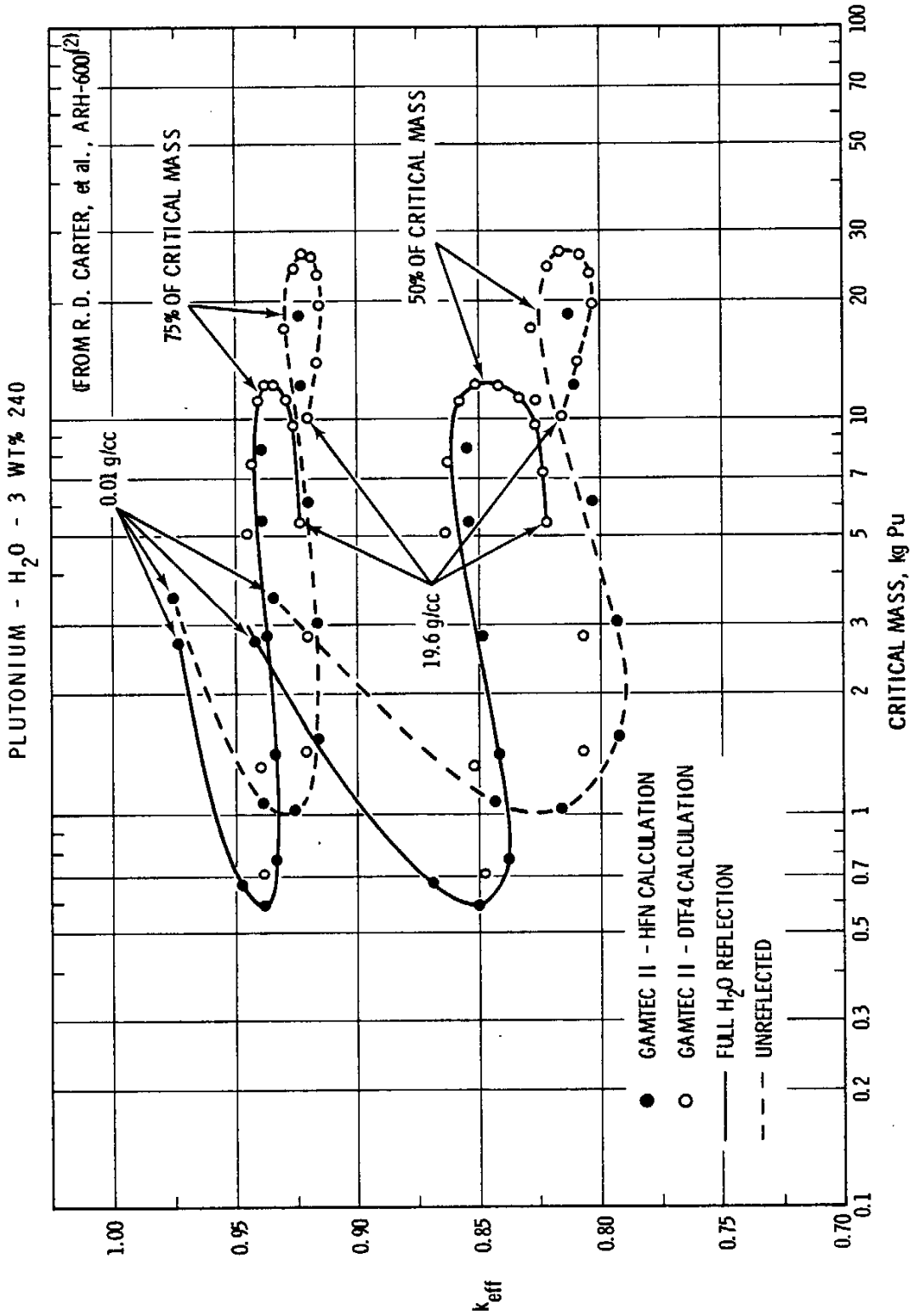


Figure 2. k_{eff} of Fractional Critical Mass vs. Critical Mass

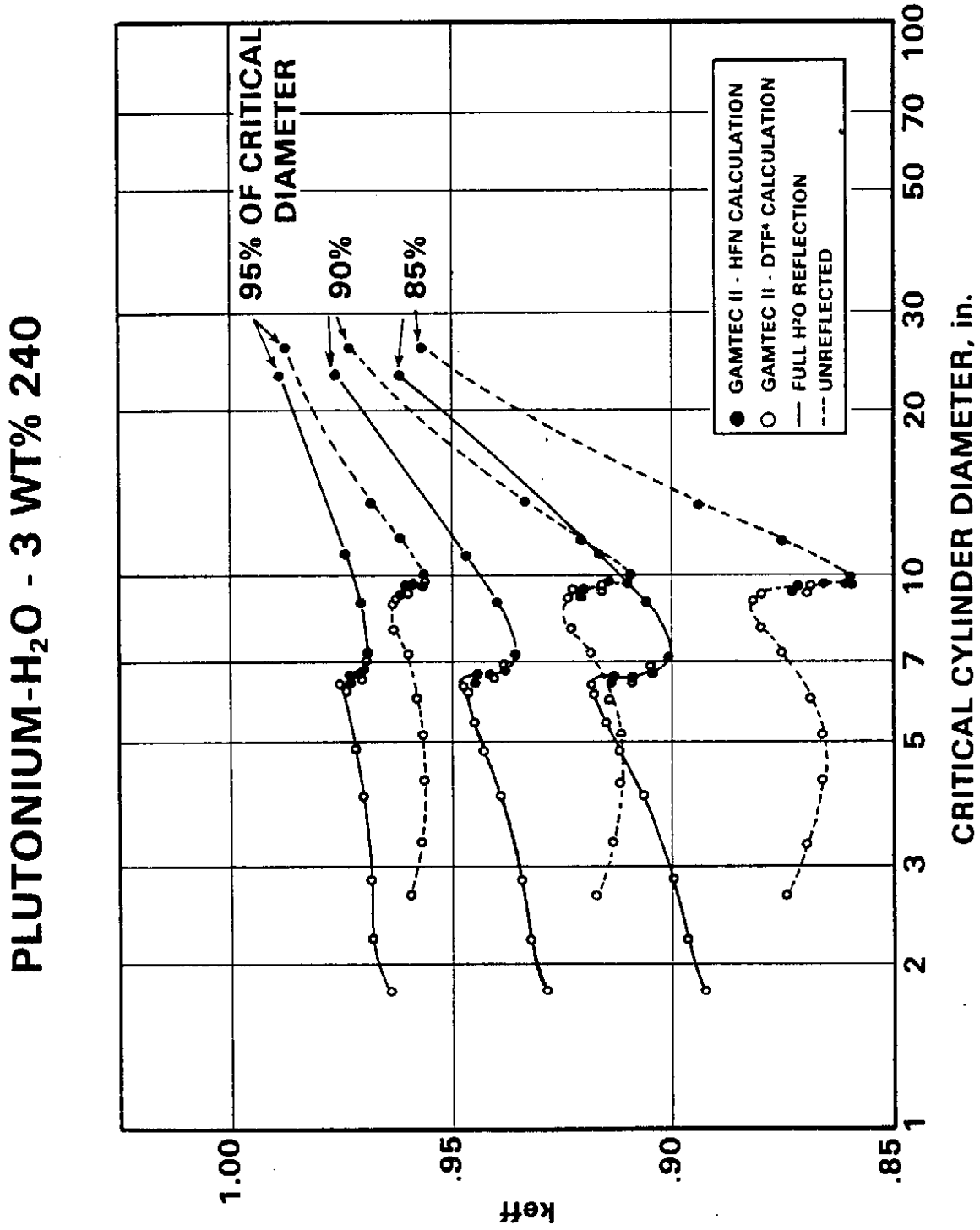


Figure 3. k_{eff} of Fractional Critical Cylinder Diameter vs. Critical Cylinder Diameter

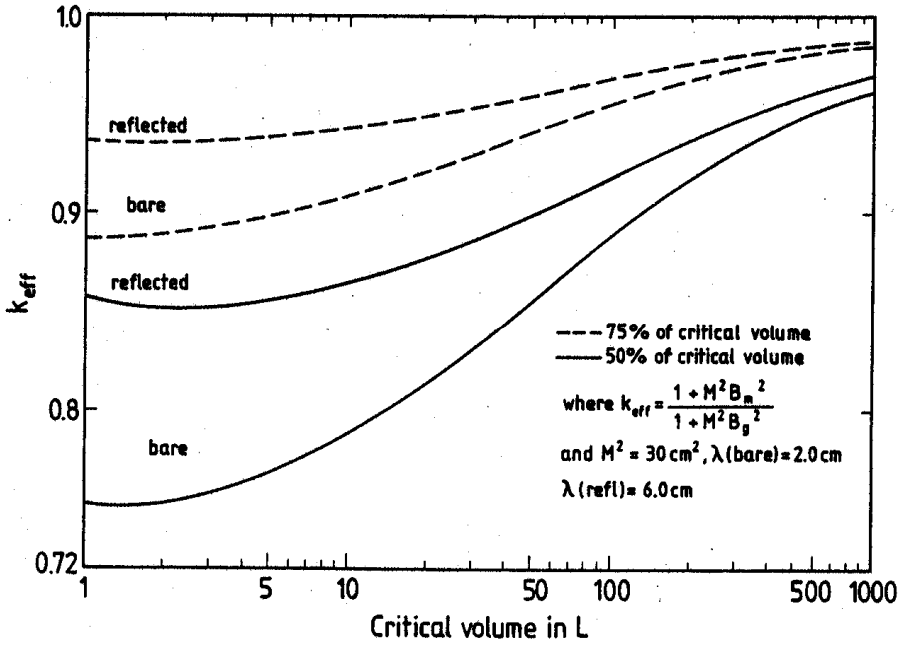


Figure 4. Relationship between Spherical Critical Volume and k_{eff} for Spheres that Contain 50 and 75% of Critical Volume

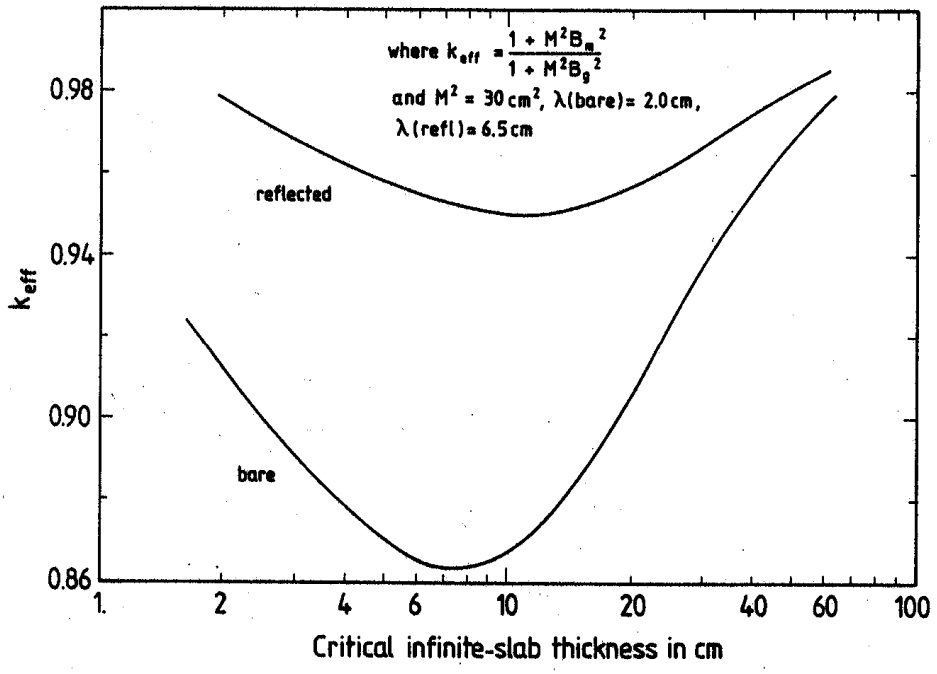


Figure 5. Relationship between k_{eff} and Infinite-Slab Thickness for Slabs that are 85% of Critical Thickness

The calculations of Table II were made for homogenous 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 position 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 principal, the value of k_{eff} may be determined from the ratio of the number of neutrons in successive generations, but this is also difficult to accomplish.

4. Variation of Critical Mass with Sphere Radius for Homogenous ^{239}Pu – Water Mixtures

All factors that influence the interaction of neutrons with matter affect criticality. The following curves (Figure 6), show the complex variation of critical mass with sphere radius for homogenous ^{239}Pu-water mixtures, and serve to illustrate several effects.[8] 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, homogenous plutonium-water spheres and the lower one for plutonium-water spheres immersed in water. Striking changes occur in the critical mass as the plutonium is diluted with water. Beginning with alpha-plutonium metal (ρ = 19.6 g/cm^3), the

critical mass and radius both increase upon dilution with water. The mass passes through a maximum value at an H/Pu ratio of about 4 (Pu density ~ 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 so 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 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.[9] Finally, both the critical radius and mass increase 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).[10] At this point, about half the neutrons released in fission are absorbed in the diluent.

Figure 6 also shows the effect of neutron reflection. For the sphere immersed in water, some of the neutrons that 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 6 illustrates that the same critical mass could be achieved with three different Pu concentrations, but that the critical volume of size would differ with each concentration.

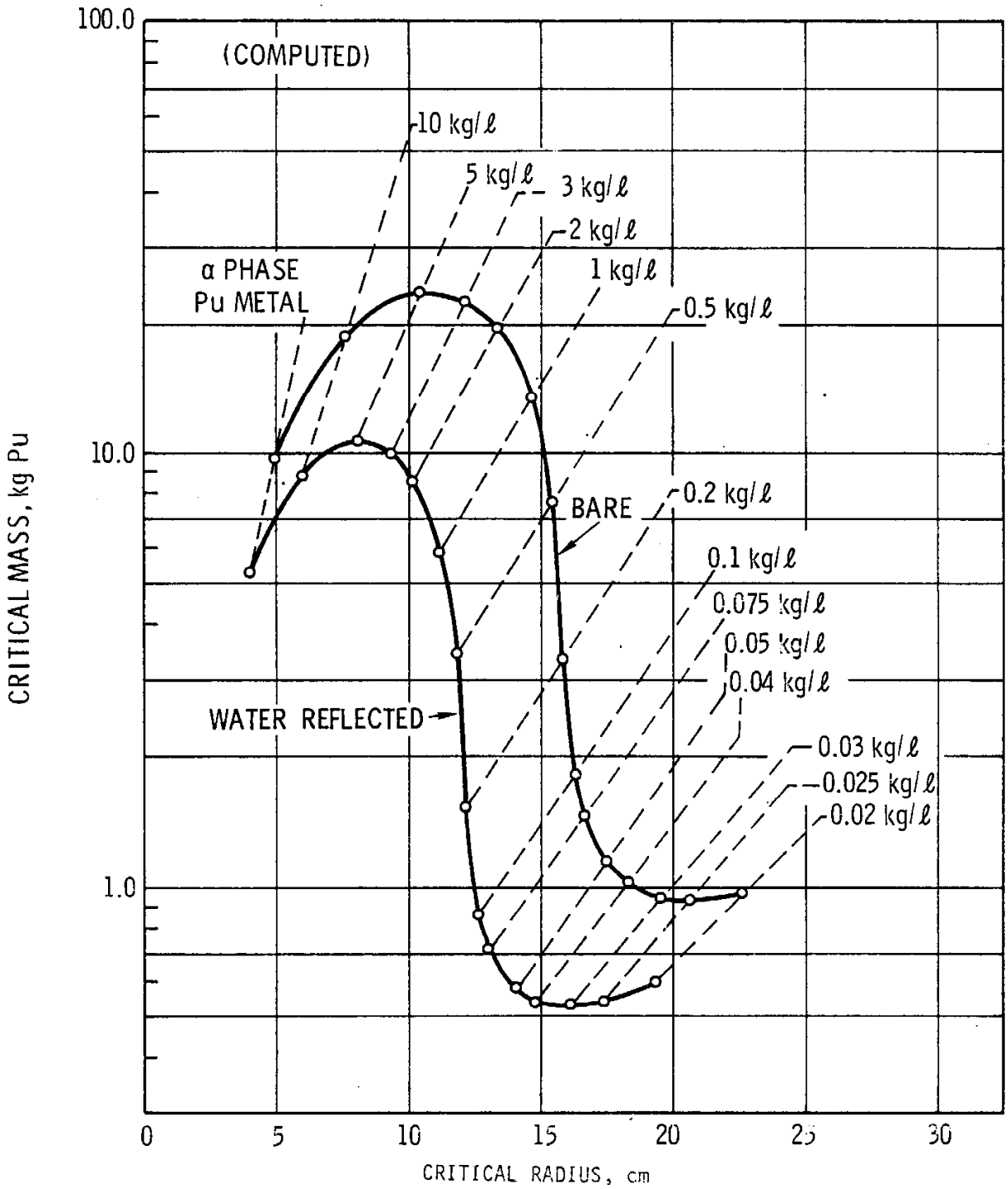


Figure 6. Estimated Mass and Radius of Critical Plutonium-Water Spheres

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

Figure 6 illustrates some of the factors such as moderation and reflection that affect the criticality of systems containing a single actinide isotope (^{239}Pu for purposes of illustration).

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 7.[11] 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. Therefore, the effect of ^{240}Pu on the criticality of ^{239}Pu will be largely dependent on the neutron spectrum, which 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 8.

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 addition or removal of fuel that changes the hydrogen to fuel ratio and consequently, the neutron spectrum.

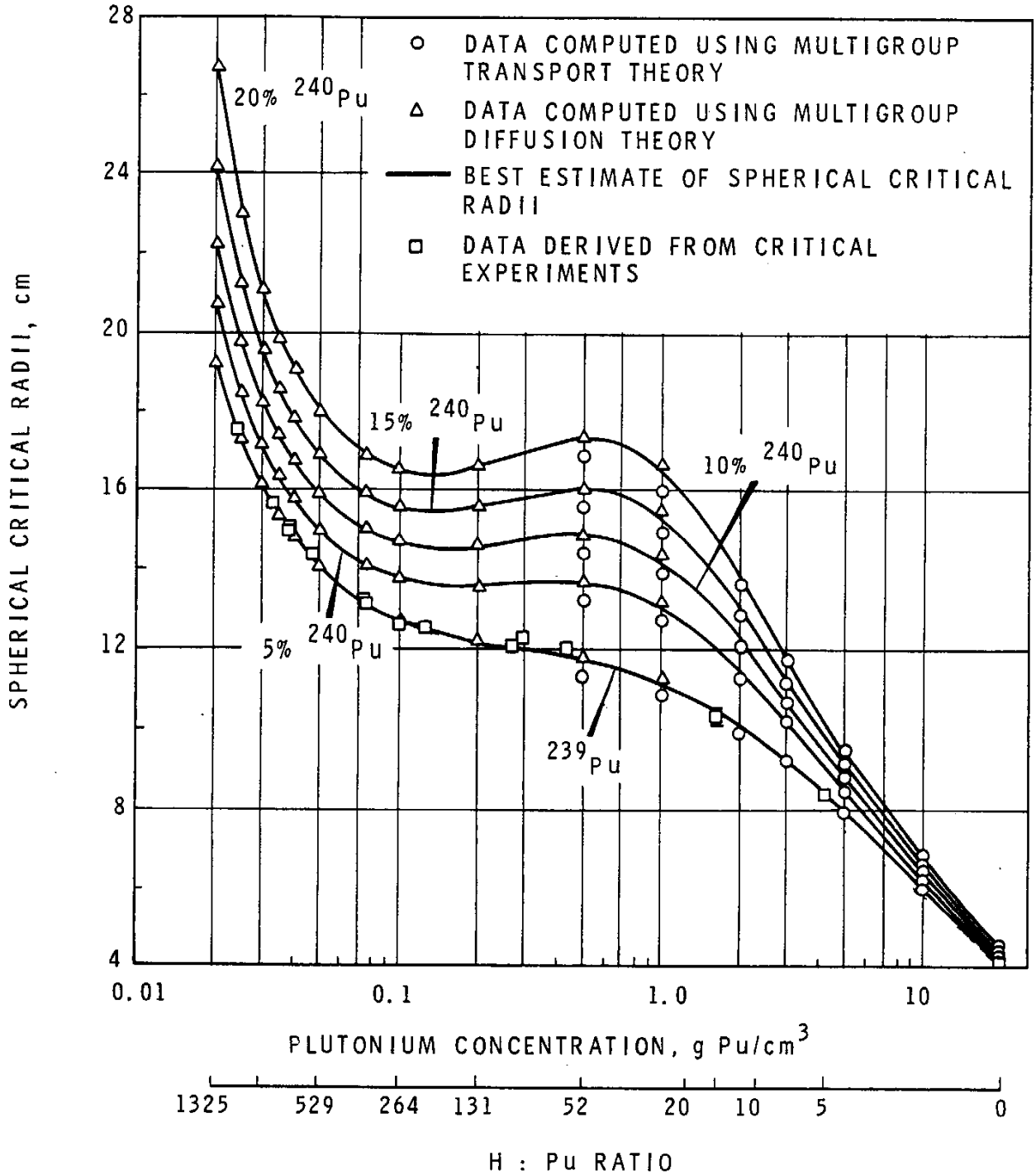


Figure 7. Water Reflected Spherical Critical Radii of Pu(Metal)-Water Mixtures

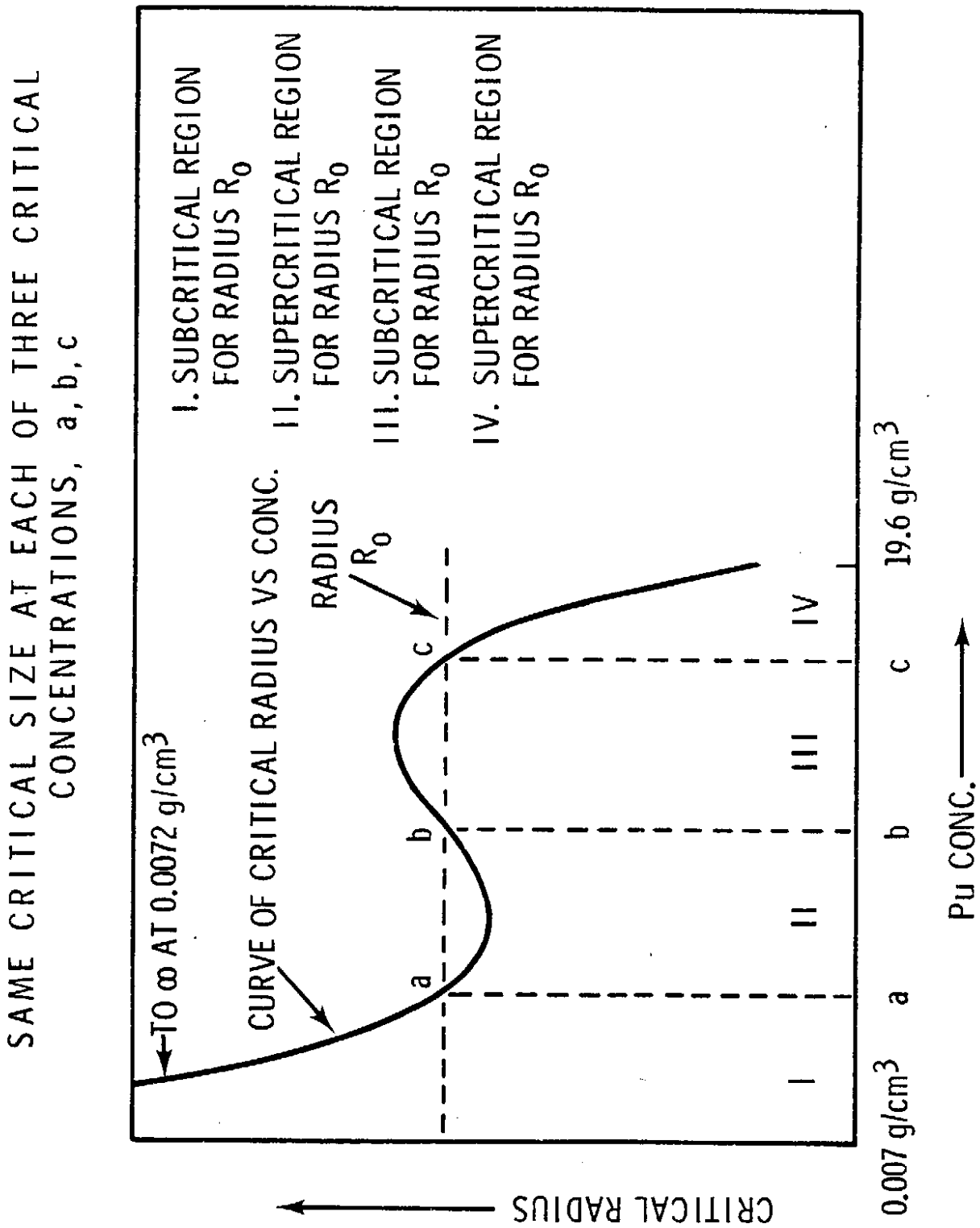


Figure 8. Illustration of Triple Point of Criticality

6. Identical Critical Mass at Four Concentrations

Critical mass calculations are presented in Figure 9 for mixed oxides of U and Pu in which the Pu contains 25 wt.% ^{240}Pu.[12] If the peculiar curve shape at the higher concentration range is correct, (note curves for 15 and 30 wt.% in Pu and 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 8), there was one critical volume at three different fuel concentrations.

7. Limiting Critical Enrichment of Uranium for Aqueous Homogenous Solutions

A series of calculations by B. M. Durst of Battelle – Pacific Northwest Laboratories, are presented in Figure 10 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 homogenous UO_3 water mixtures.[13]

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

For homogenous 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.[14] At this enrichment, the largest value that can be obtained for the reproduction factor, k_∞, for an infinite system under optimum conditions of moderation, is unity.

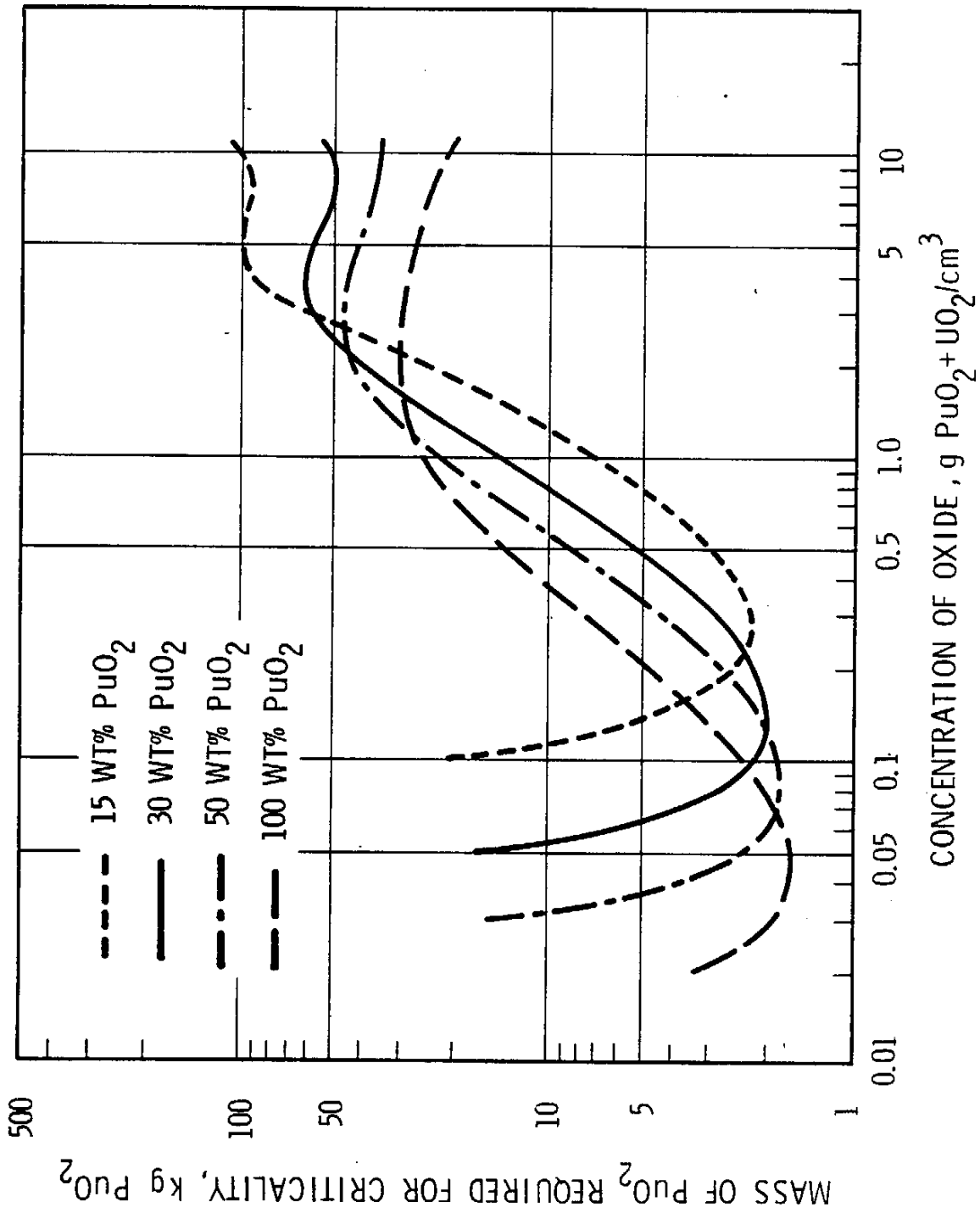


Figure 9. Computed Reflected Spherical Critical Mass of PuO_2-UO_2-Water Mixtures with 25 Isotopic Percent ^{240}Pu

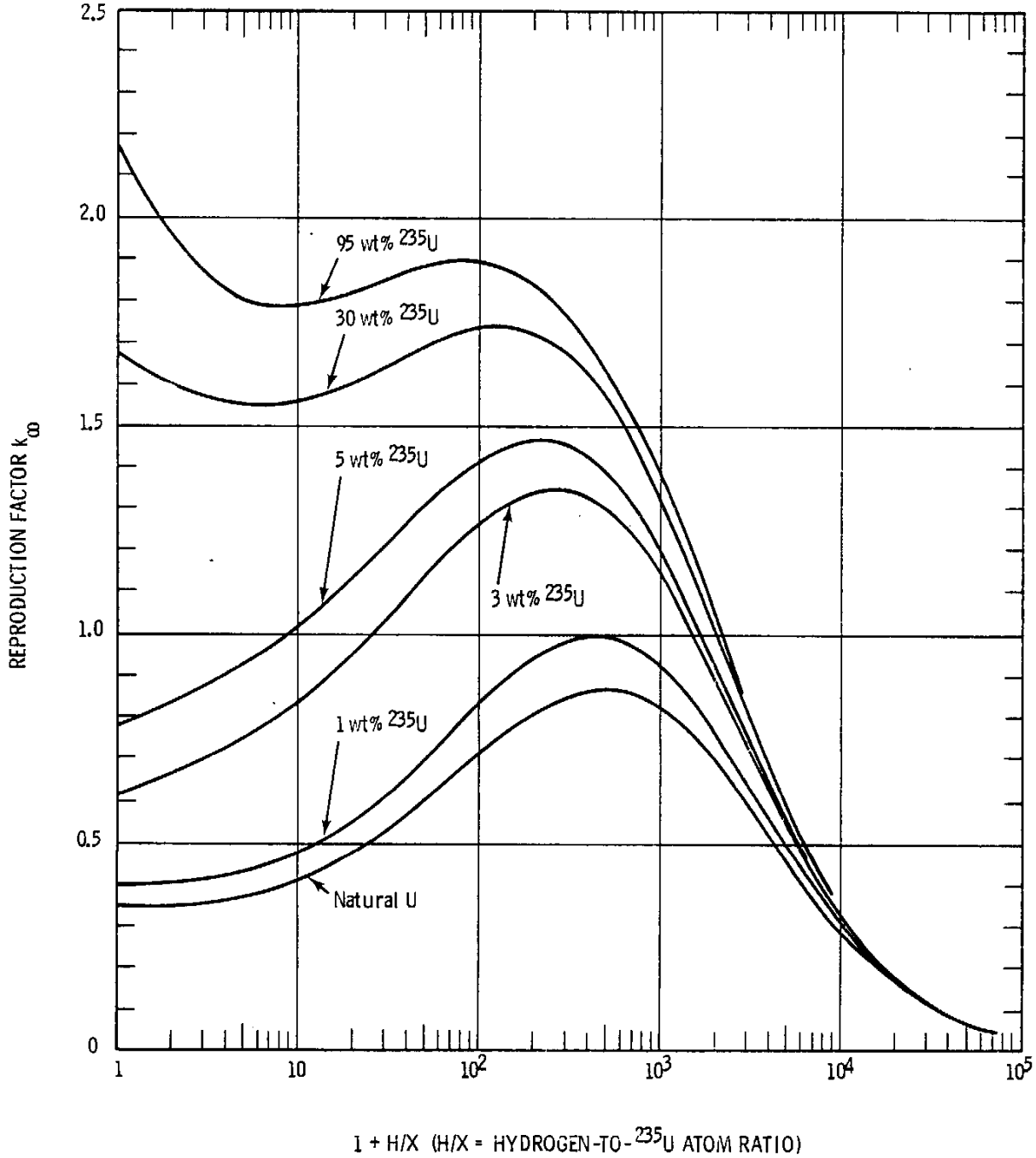


Figure 10. Computed Values of Reproduction Factors for Homogeneous UO_3 Water Mixtures at Various ^{235}U Enrichments

8. Sixty-Six Years and the Criticality of Intermediate Uranium Enrichments – What is Known

Data on the criticality of uranium in the intermediate enrichment range (~ 6 wt.% to < ~ 93 wt.% ^{235}U) from which to deduce subcritical limits for criticality control are limited. [Subcritical limit is defined basically as the limited value assigned to controlled parameter that results in a system known to be subcritical (see Reference 15)]. It is surprising that in the years since the first man-made nuclear chain reaction (December 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," ANSI/ANS-8.1-1998.[15] The data on uranium are for uranium enriched to no more than 5% ^{235}U, and then for 10% enriched and highly 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 homogenous system. The critical volume is also 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 achievable 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. Although the minimum critical mass may be less for the homogenous case, for enrichments above 6 wt.%, the minimum critical volume will not be. It is possible then 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 III.

Table III. 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.%	Heterogeneous[(a)]	Heterogeneous[(a)]
~ 6 wt.% to ~ 34 wt.%	Heterogeneous[(a)]	Homogenous[(b)]
> ~ 34 wt.% to 100 wt.%	Single Metal Unit[(c)]	Homogenous[(b)]

(a) Heterogeneous 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 spacing.

(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.

Due to 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 (~ 6% to < ~ 93%) as shown in Figure 11.[(16)] 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. This 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 as indicated on the figure in the lower dashed curve.

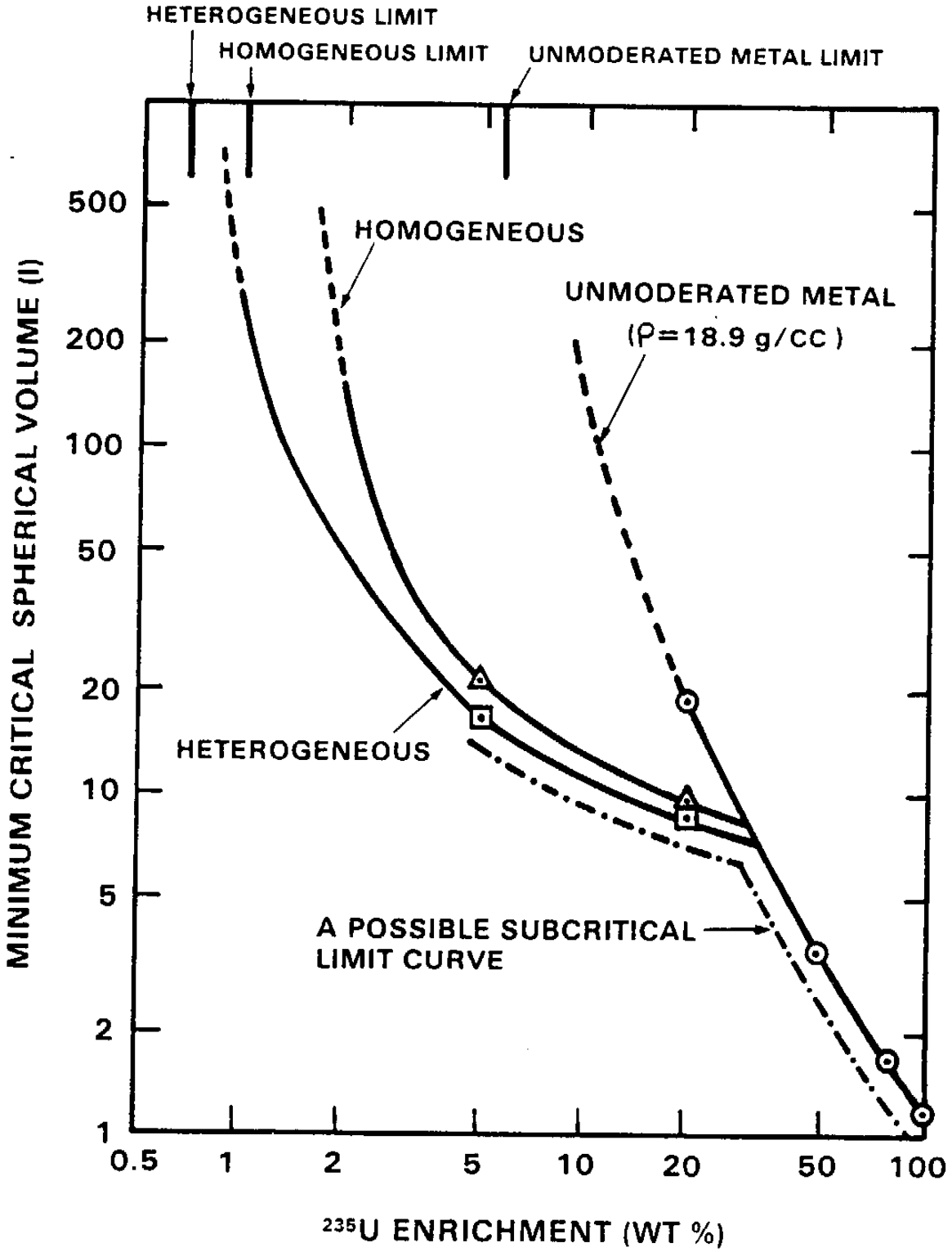


Figure 11. Minimum Critical Volume vs. Uranium Enrichment

9. Criticality Fundamentals and Fissible Nuclides (The Dilemma of k_∞ and P)

A basic concept of reactor theory is that the effective multiplication constant, k_{eff}, can be expressed as $k_{eff} = k_\infty P$ where:

k_∞ = the multiplication factor in the infinite medium for no neutron leakage and

P = the non-leakage probability

The fraction of neutrons that escape via neutron leakage is then = 1 – P. For an "infinite" system P=1 and $k_{eff} = k_\infty$. In the case of an actinide element in the form of metal with no diluents, or for a fast unmoderated system,

$$k_\infty \equiv \text{eta } (\eta) = \upsilon\sigma_f/(\sigma_f + \sigma_c).$$

The fraction of neutrons that leak out of a critical assembly when k_{eff} = unity is then,

$$L = 1\text{-}P = 1\text{-}1/k_\infty$$

Let us consider the special case of a typical fissible nuclide, or so-called threshold fissioner.[17] The neutron leakage will in general not be equal to

$$L = 1\text{-}1/k_\infty$$

but in some cases will be substantially different. This gives rise to an interesting dilemma. Why does this simplest of equations not apply to the fissible actinide nuclides, or the threshold fissioners, but seems to be quite adequate for the well-known fissile nuclides, such as ^{233}U, ^{235}U and ^{239}Pu?

Weinberg and Wigner shed some light on this subject in their text "The Physical Theory of Neutron Chain Reactors," pp 169-174.[18] They define the criticality factor, C, as = k*P where k* is said to be a "finite" multiplication factor. k* is then defined as the value of k_∞ in an infinite medium – if the neutron spectrum were the same as in the finite system, which it isn't. The quantities k* and P are awkward to calculate since they must then be averaged over the persisting neutron distribution in the finite assembly and change with the assembly size. It is only when

$P \equiv$ one (an effectively infinite reactor) that $k_\infty = k^*$ and $k_{eff} = k_\infty P$.

It is further stated that although the difference between k_∞ and k^* is not great, the distinction between them is important conceptually, but the difference between k_∞ and k^* in reactors is almost academic and is usually disregarded.

Though it may be convenient to have criticality theory based on k_∞ in lieu of k^* (the value of k_∞ if the neutron spectrum were the same as in the finite system), it is only those cases where $k_\infty = k^*$ that P^* equals the non-leakage probability P. It is concluded that in all practical cases, P^* and P are also very nearly equal.

Although these statements apply to fissile nuclides, the same cannot in general be said of the so-called fissible nuclides, or threshold fissioners, that have been identified in recent years.[19-22] This is because the value of eta averaged over the finite critical system (and the neutron spectrum that persists therein) can differ very significantly from the value of eta (k_∞) based on the spectrum in the infinite medium, for example, up to some 29% different in the case of ^{242}Pu metal.

This was illustrated for a number of fissible nuclides in a paper by Srinivasan, et al., in 1989.[23] The pronounced variation in eta with radius for the fissible nuclide, ^{242}Pu, in the form of metal, is beautifully illustrated in recent calculations graciously supplied in May of 1999 by Calvin Hopper of ORNL.[24] Note Figure 12. Extrapolating to zero radius in Figure 12 gives the watt spectrum average value of eta. The value of eta at the critical radius of the bare sphere (10.2 cm) is 2.60, which decreases to 1.86 as the radius becomes larger. If the usual definition of k_∞ is used in the expression, $k_{eff} = k_\infty P$, and the non-leakage probability calculated for ^{242}Pu, from

$$P = 1/ k_\infty$$

then the error at $k_{eff} = 1$ would be about 40%, and the error in leakage

$$L = 1 - 1/ k_\infty,$$

some 25.3%.

The curve in Figure 12 clearly shows the pronounced variations of eta with size of assembly. It is apparent that the distinction between k_∞ and k* can be considerably more than academic in the case of the fissible nuclides or threshold fissioners. The criticality properties of the "fissible" nuclides were essentially unknown at the time "The Physical Theory of Neutron Chain Reactions" was published (1958).[18]

In reference to nuclides with significant half-lives, there are currently known to be more fissible nuclides (nuclides which have their smallest critical masses in the form of metal-fast neutron chain reactions, than fissile nuclides that have their smallest critical masses under conditions of optimum moderation, i.e., thermal neutron chain reactions). Considering those nuclides with half-lives ≥ six weeks, 23 fissible nuclides have been identified, whereas there are only 18 fissile nuclides.[22] In terms of numbers, the fissible isotopes are the ones that predominate over the Actinide Group.

As is well known, fissible nuclides characteristically exhibit rather sharp fission thresholds in their fission cross sections in the 0.5-2-Mev neutron energy range. There is little or no probability for fission at thermal neutron energies.

Inelastic scattering degrades the neutron energy spectrum and shifts neutrons below the fission threshold.

Then, in the case of fissible nuclides, the number of neutrons produced per neutron absorbed (eta) becomes significantly smaller as the assembly is increased beyond the critical state.

For fissile nuclides that do not have fission thresholds the variation of eta with size from the critical point is not very significant, for example, less than 3% in the case of ^{233}U, ^{235}U or ^{239}Pu metal. The decrease is due principally to the decrease in υ as a consequence of inelastic scattering that degrades the spectrum. For a solution (moderated) system of ^{235}U, the difference between k_∞ and k* is negligible, as well as for ^{239}Pu.[23] It is to be noted that well-moderated solution systems of fissible nuclides will not even support chain reactions!

As already noted, the variation in eta for the ^{242}Pu fissible nuclide is much larger than 3%, nearly ten times, at 29%.[24] The calculated critical mass for a bare metal sphere of ^{242}Pu at

density 19.84 g/cc is 88 kg. This result was obtained with SCALE 4.4, KENO-V.a with 238 energy groups.[24] The critical mass for a bare ^{242}Pu metal sphere, as deduced from critical experiment data at LANL, was recently reported as 80 kg at density of 20.05 g/cc.[25] Though there are some 23 fissible nuclides with half-lives ≥ six weeks in the Actinide Group, ^{242}Pu is the only fissible nuclide for which a critical mass has been reported to date based on critical experiment data.

The data indicate the expression, $k_{eff} = k_{\infty}P$ is inappropriate for the fissible nuclides that by definition have fission thresholds and support only fast neutron chain reactions. k_{∞} in principle cannot by definition be defined as the value of the reproduction factor in the infinite medium if, in order to use it in calculations of k_{eff} for a finite system, it must be based on a neutron spectrum that does not persist in the infinite medium. The fissible nuclides, which constitute a majority (23 out of 41) of isotopes of interest in the Actinide Group, are not in keeping with the accepted notions of fitness and order which apply to the common fissile nuclides, etc. It may, therefore, be said that the fissible nuclides constitute an anomaly, and the simple expression, $k_{eff} = k_{\infty}P$, as defined in the “early days”, does not apply to these nuclides, but rather to the “awkward” expression of k*P!

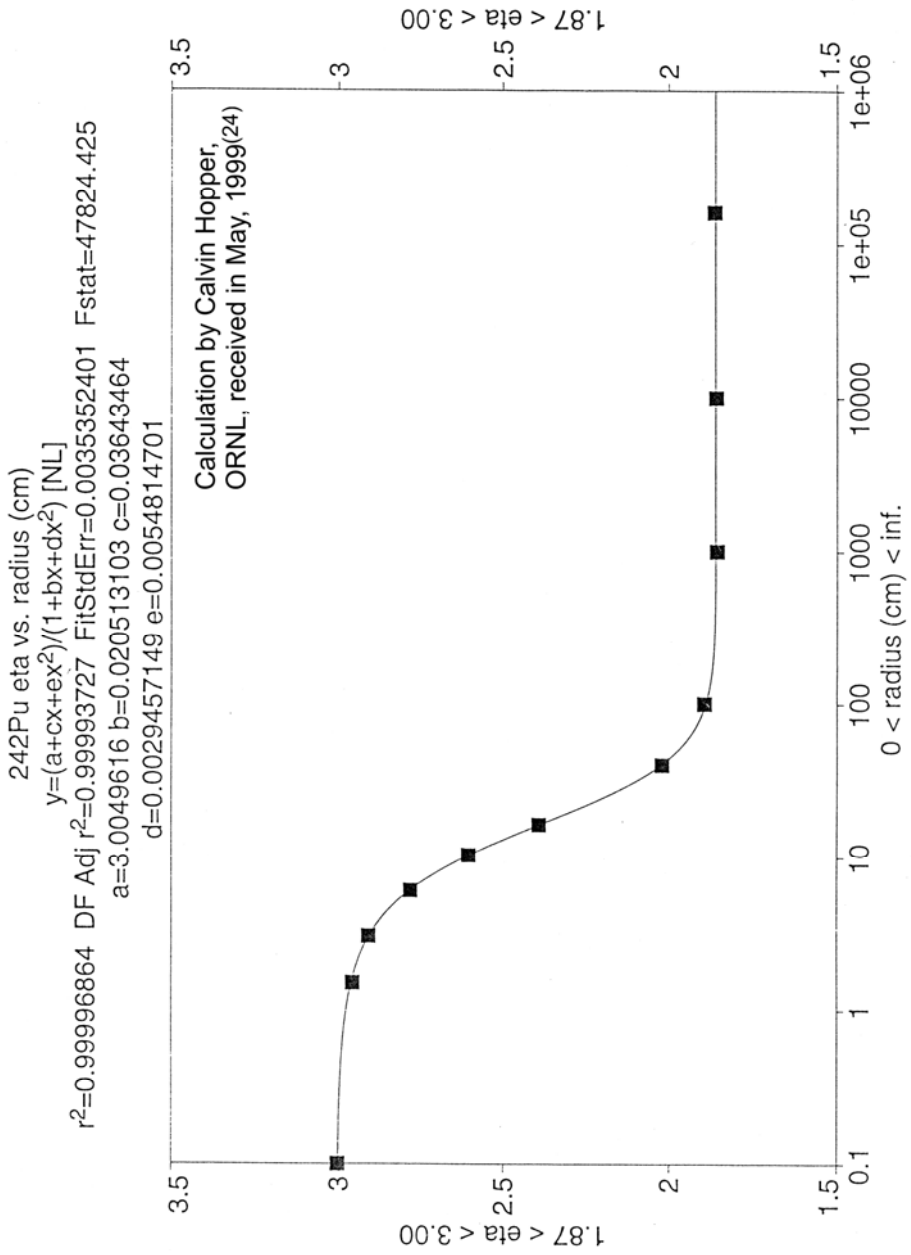


Figure 12a. ^{242}Pu eta vs. Radius (cm)

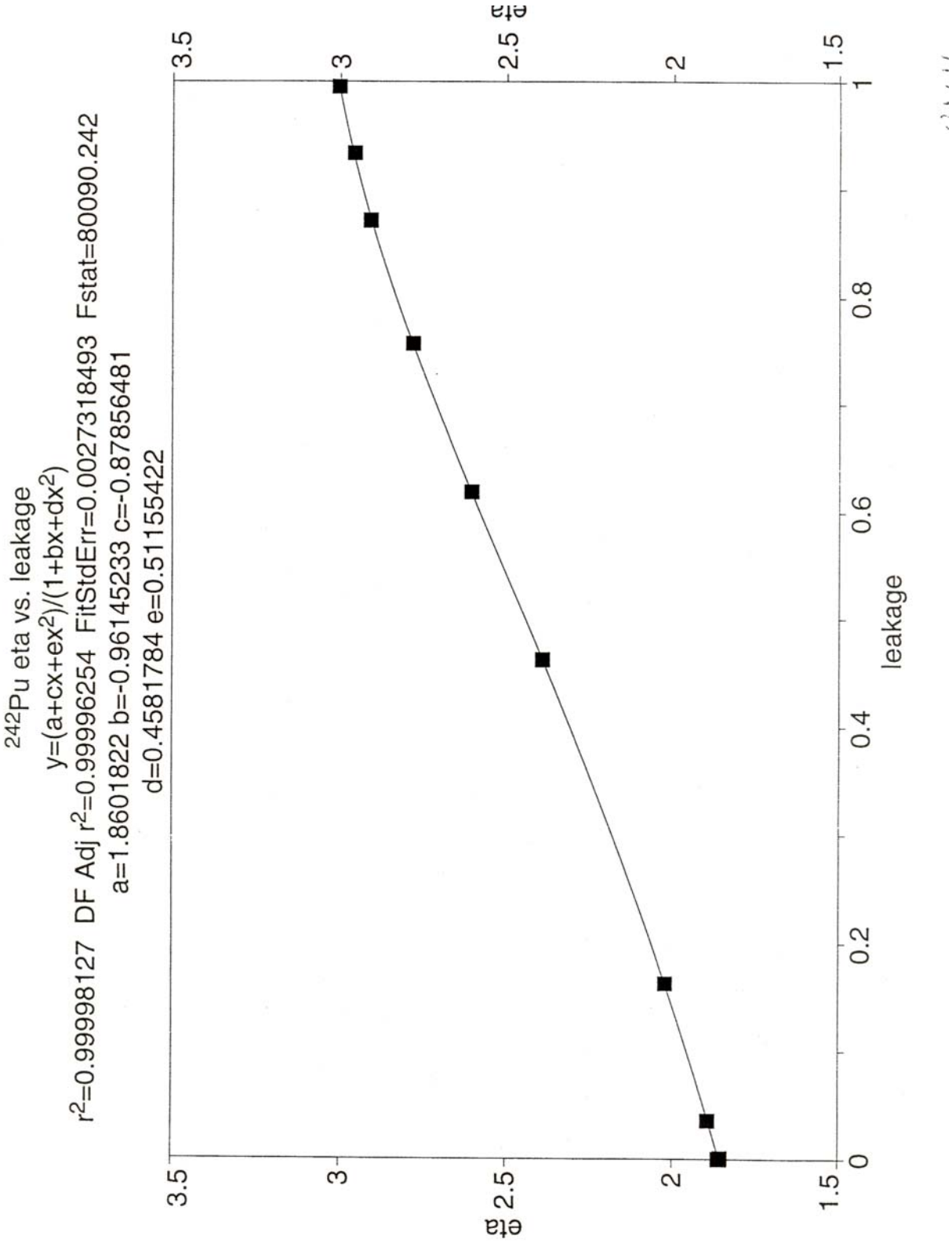


Figure 12b. ^{242}Pu eta vs. Leakage

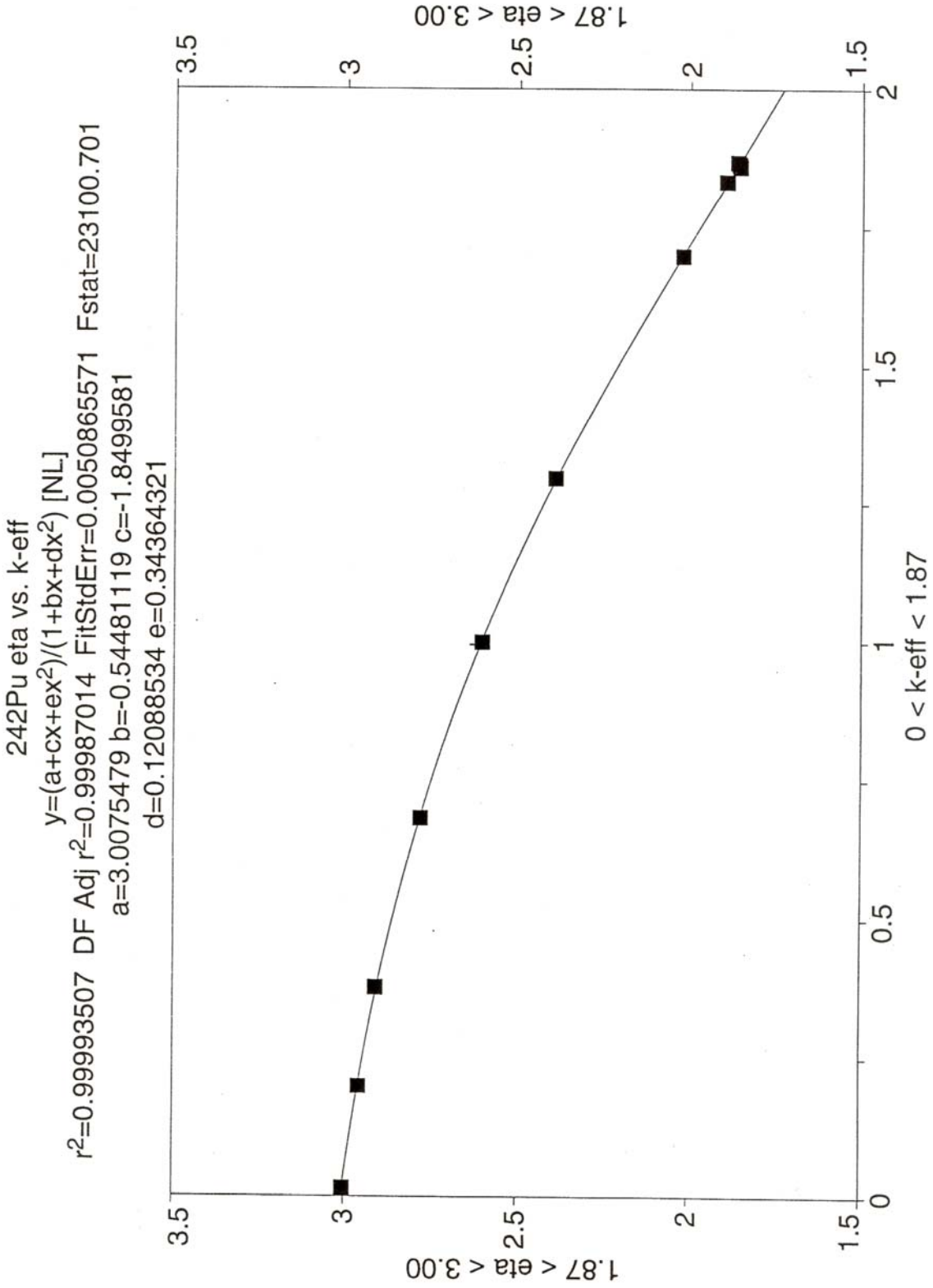


Figure 12c. ^{242}Pu eta vs. k-eff

D. 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 "infinite 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 ~ 3680).[9] Fuel processing operations involve cylindrical vessels 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 IV 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 answer is no. Note that Pu has the smallest limited 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 (see Table X). The critical ^{235}U concentrations are smaller, however, than those of ^{239}Pu, by up to a factor of ~3 for cylinders in the 5½ to 6 inch diameter range. Below about 5½ inches the critical concentrations exceed those for ^{239}Pu. For cylinders about 7 inches in 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 IV. Estimated Critical Concentrations of Fissile Isotopes in Infinite Length, Water-Reflected Cylinders[2, 5]

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	25	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 that scaling factors might be developed from the more extensive data that was available on ^{235}U solutions, which 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. As the data of the table show no consistent scaling factor to exist, it is probably good that this procedure was not attempted. The differences in variation of critical concentrations are due to the variations in eta, and in the cross sections, with changing spectrum that also depends on the concentration or H/X ratio of the fissile isotopes in the aqueous solutions.

E. 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 certain circumstances have a smaller critical volume and mass than if the fissile material were in the form of a sphere. This result stems from experiments performed with PuO_2-plastic compacts arranged in cubic geometry and reflected with Plexiglas.[26, 27]

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[28] 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 uncertainty in the Monte Carlo calculations rules out firm conclusions. In examining data from a number of other experiments involving cubes of Pu-bearing fuels, it should 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 13.

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. The Monte Carlo calculations are in support of these results.

It has also been concluded that for some unmoderated 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.[5, 29] These conclusions lend additional support to our conclusions regarding the cube and the sphere.

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.

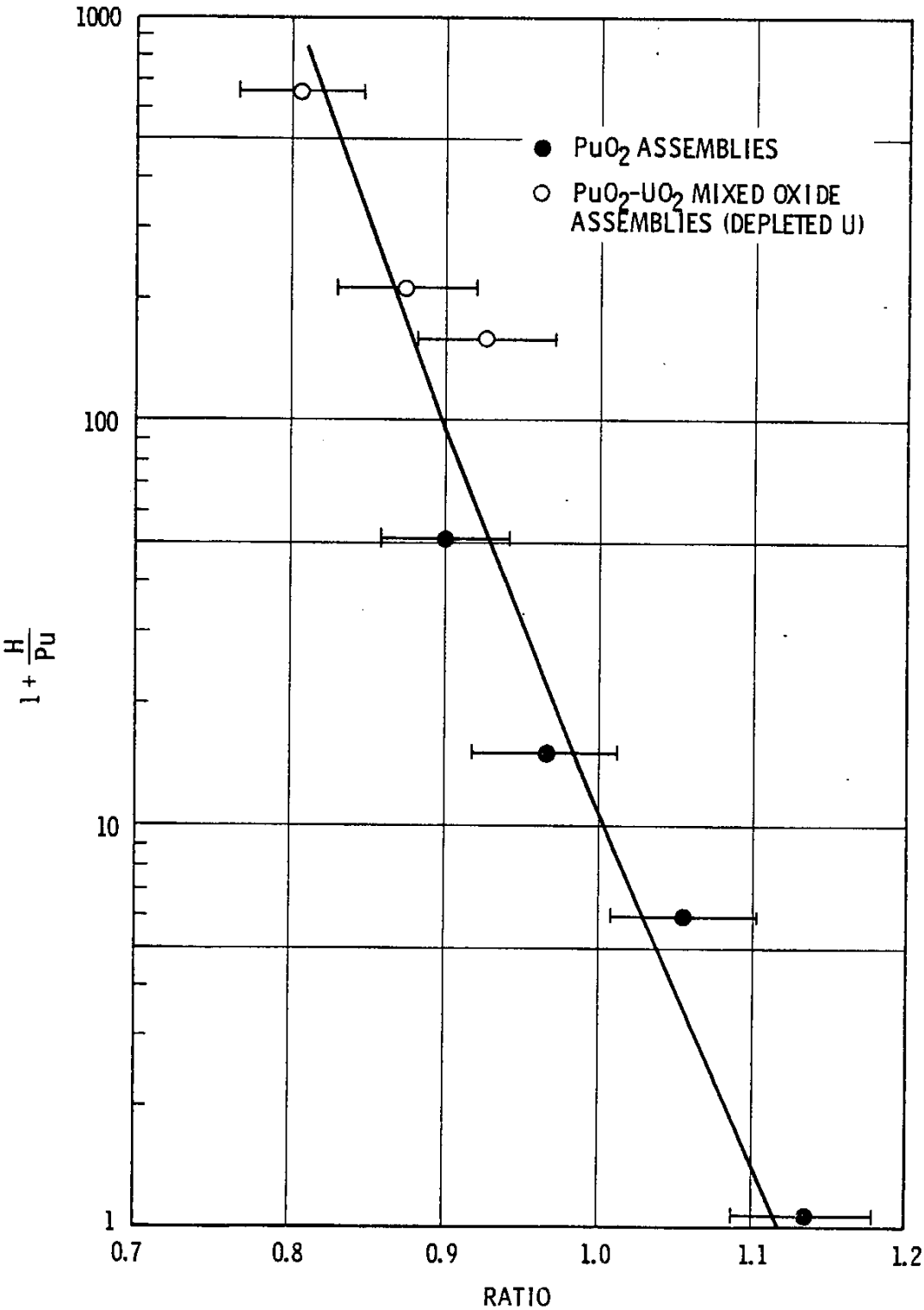


Figure 13. Ratio of Critical Sphere Volume to Cube Volume – Plexiglas Reflected Assemblies

F. THE CRITICALITY OF LARGE BILLETS vs. SMALL RODS – CONDITIONS FOR MINIMUM MASS (Triangular 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[30, 31] (see Figure 14). 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 those 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. For the same outside diameter (7.2 in.), however, 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.

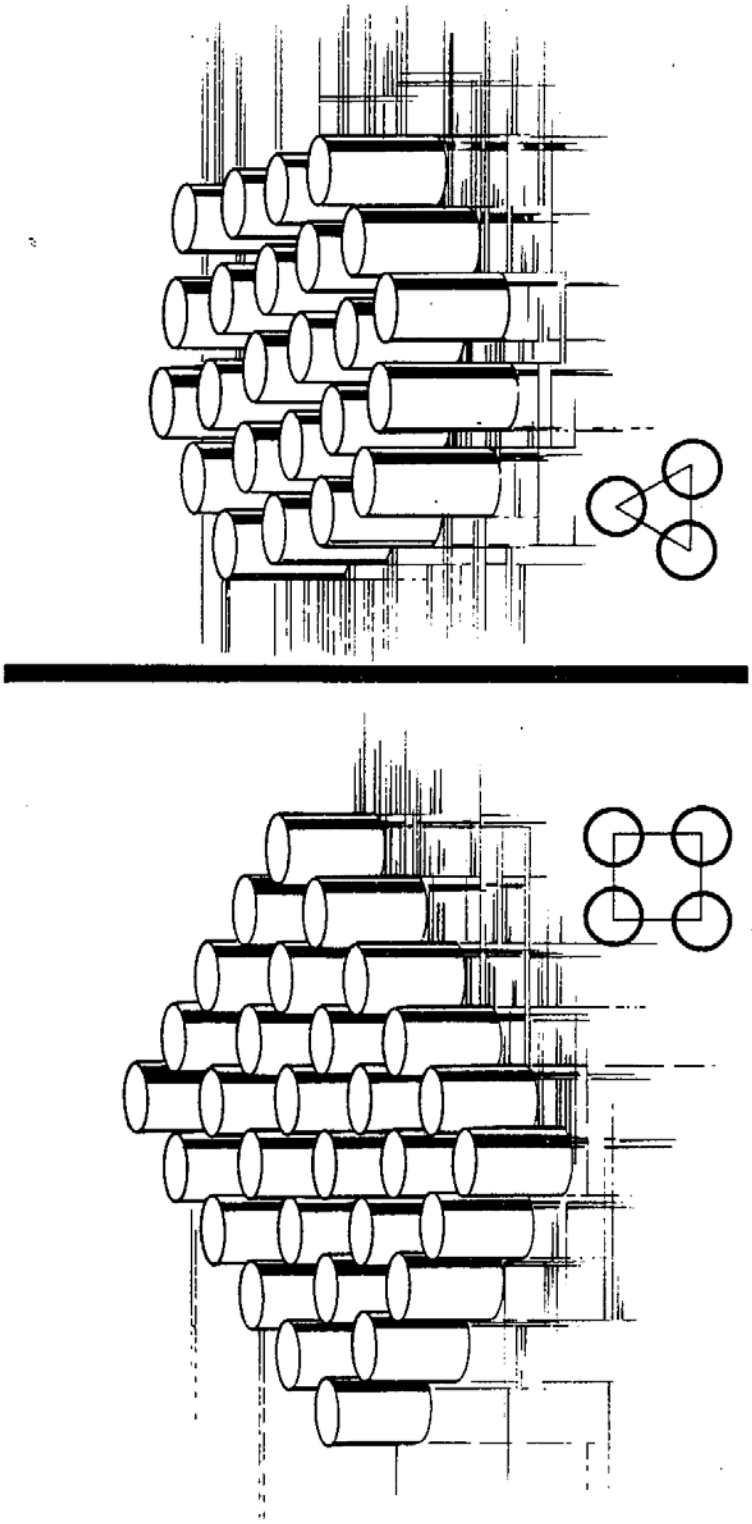


Figure 14. Square Lattice vs. Triangular Lattice Pattern

Data are presented below (Table V) 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 V. Comparison of Minima for Lattices of U(3.85)
Arranged in Square and Triangular Patterns

Change between Patterns		
Outside Diameter (in.)	%	
	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, 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.

G. 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 affects 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 homogenous 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. As 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, because 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 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 thoroughly 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 homogenous 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[32] and critical experiments performed at ORNL with 1.95 and 3.85 wt.% ^{235}U large annual cylinders and solid rods.[30, 31] The outside diameter of the annual cylinders was 7.2, 6.2 and 5.2 in., and the inside diameter was 2.6 in. In the case of the 3.85 wt.% ^{235}U, experiments were also completed with solid rods of 2.6, 6.2, 6.2 and 7.2 in. diameter. The results of the calculation of these large rods and billets proved to be 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 x 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 x 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 that can be made critical in a water lattice as a function of enrichment. The results are shown in Figure 15.[33] 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 15 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 16, 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 each fuel bundle, or if water were excluded from the bundle, and if each fuel bundle were of sufficient diameter, then an infinite number of fuel bundles containing an infinite number of individual rods, could conceivably be subcritical in any arrangement whatsoever in water, for example a storage pool.

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

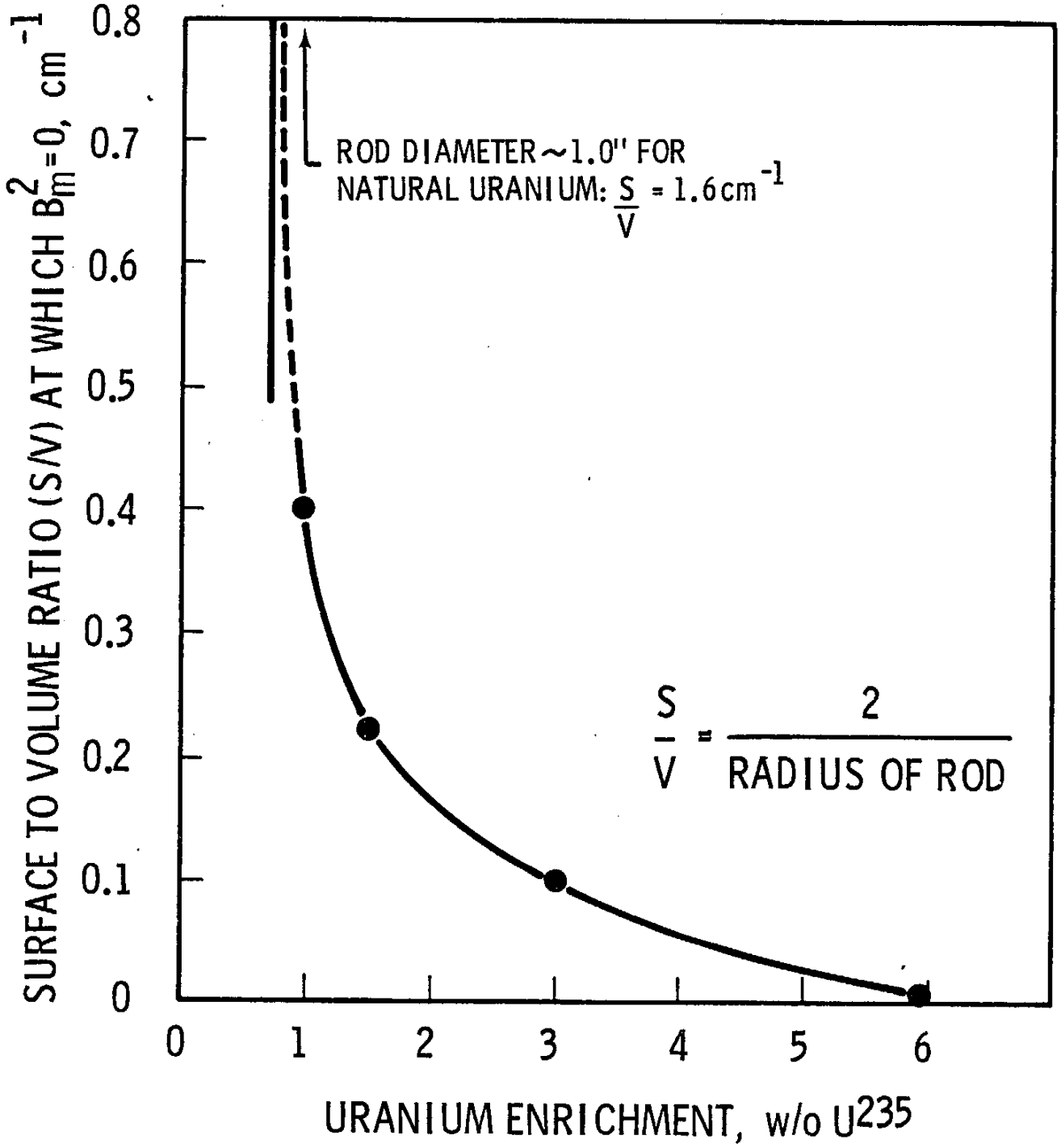


Figure 15. Estimated Surface-to-Volume Ratios of Large Rods which Result in Zero Bucklings (Infinite Critical Masses)

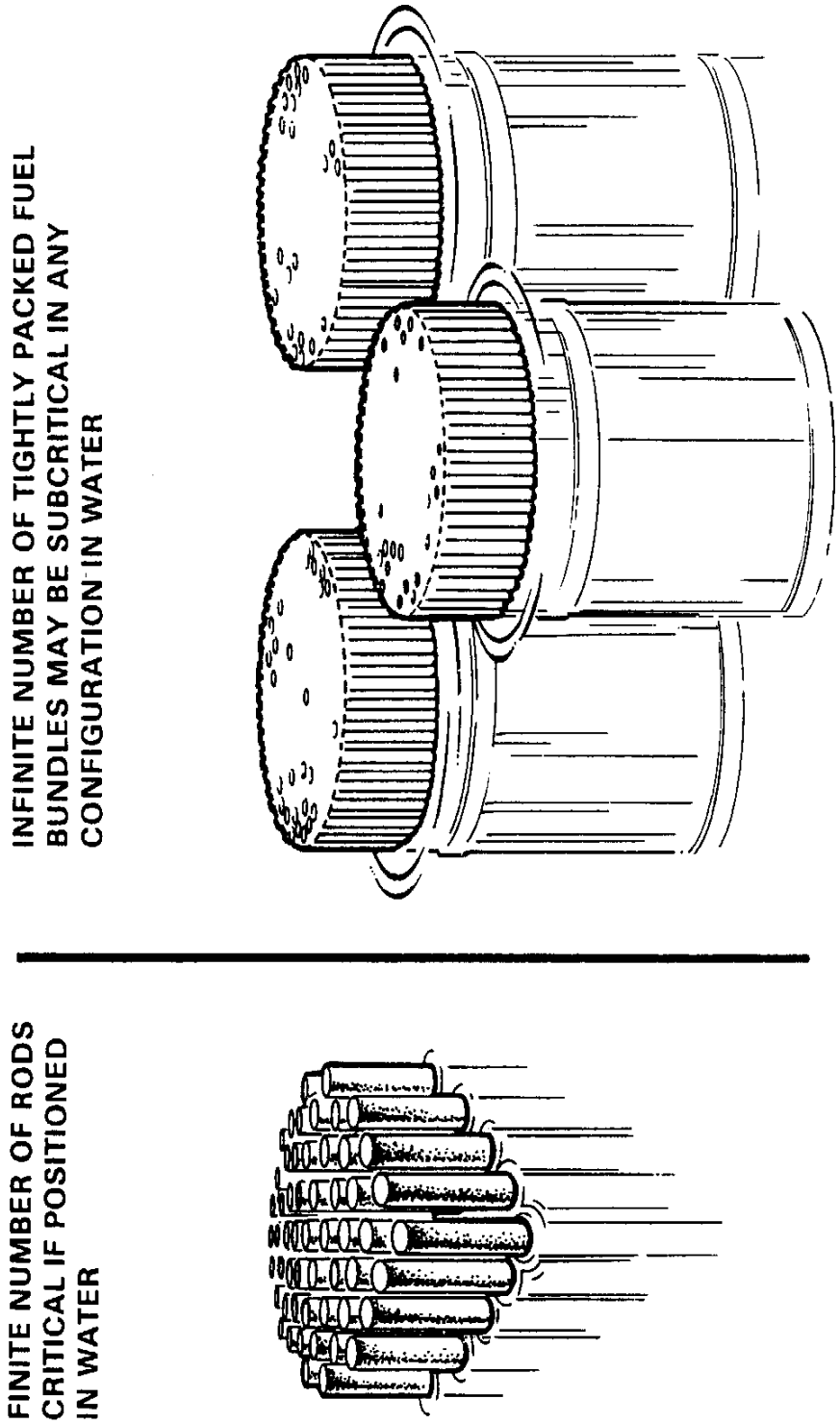


Figure 16. The Criticality of Tightly Packed Low Enriched ^{235}U Fuel Bundles in Water

H. ADDED SCATTERERS AND MODERATION

In Figures 6 and 7, 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 17 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.[34] 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 18 (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.

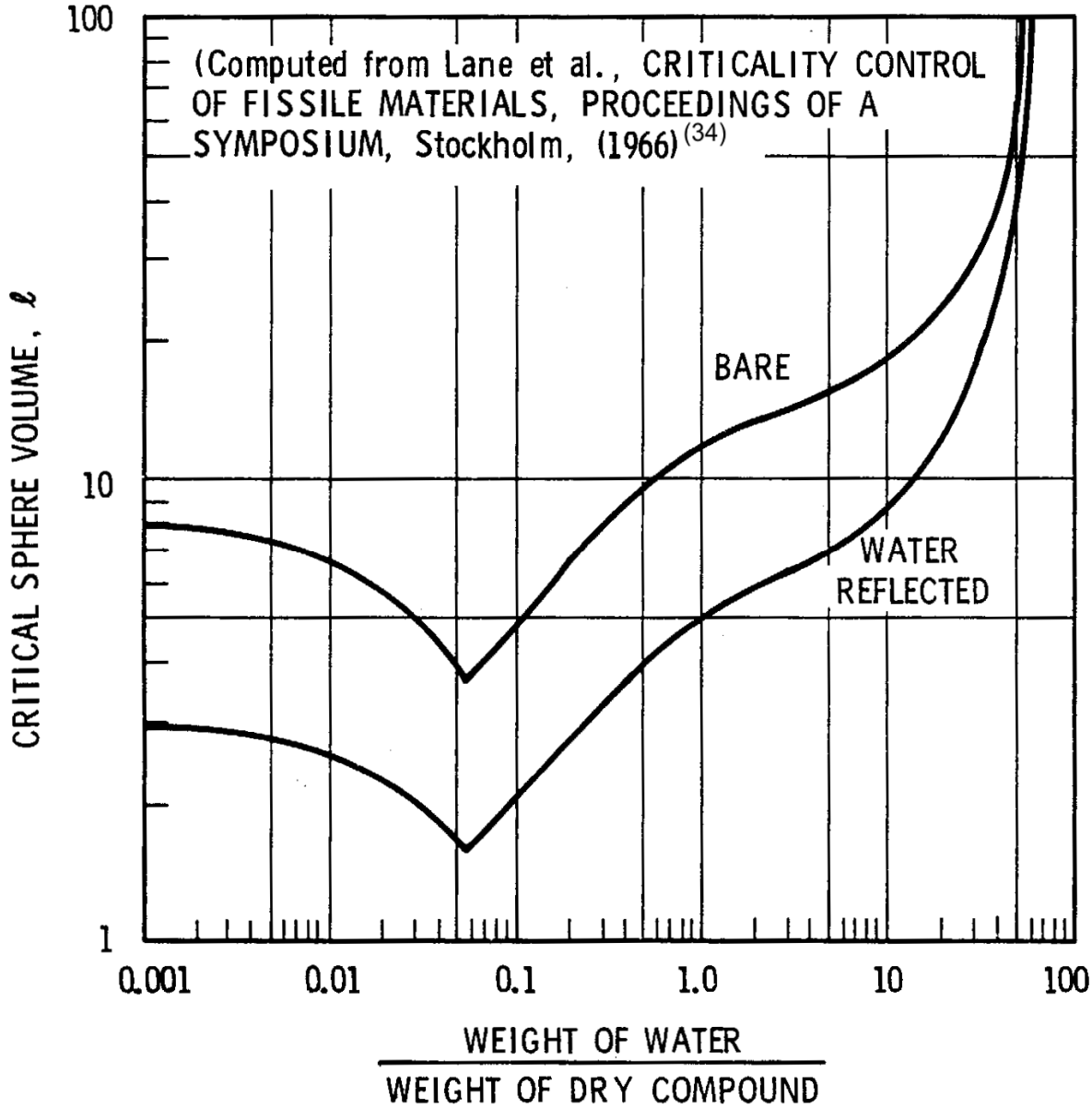


Figure 17. Computed Critical Volume as Water is Added to 30/70 $^{239}PuO_2/^{235}UO_2$ at 7 g/cm^3

2. The Reduction in Mass of the Sphere

The next example (Figure 18) 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.[34] 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[35] and later studied in detail by Makoto Iwai.[36]

E. R. Woodcock 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.

Makoto 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.[37] His study pertained to plutonium compounds likely to be encountered throughout the nuclear industry in fuel processing and fabrication processes. His results do indeed confirm that, in some cases of unmoderated thin slabs with hydrogenous 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.

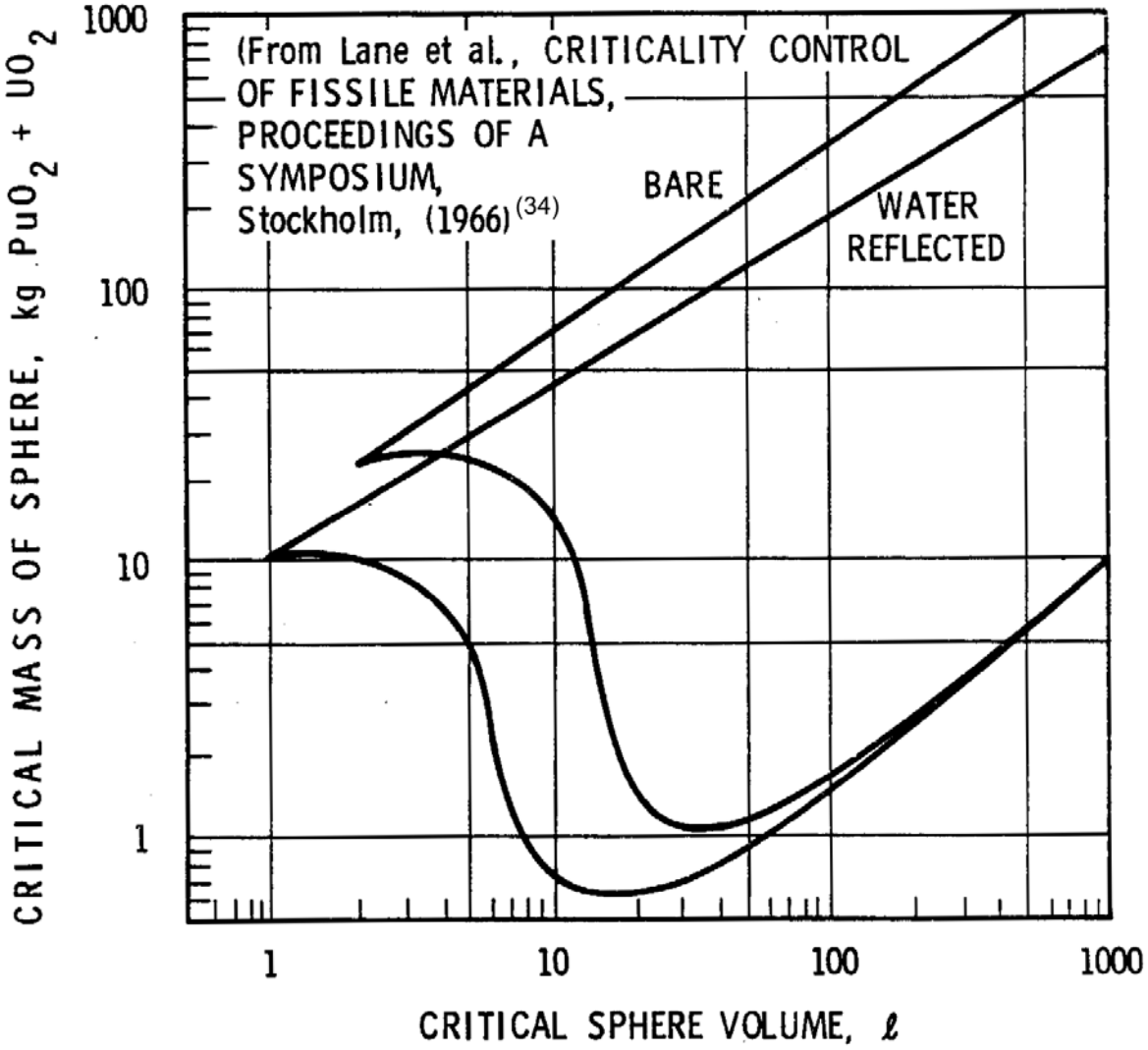


Figure 18. Computed Mass/Volume Curves for 30/70 $^{239}PuO_2/^{235}UO_2$

I. 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 to the density. To maintain the same non-leakage 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 unity length will vary inversely with density, $M_c \sim \rho^{-1}$. In the case of infinite slabs, the mass per unity area, $M_c \sim \rho^{-0}$ = constant, and remains unchanged. An unreflected infinite slab that 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

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

with the provision that m + n = 2.[38]

The following is 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.[29, 39] The effect is illustrated in Figure 19, 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. In this instance, 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.[40, 41] 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 extremely low-density interiors.

In his report, G. Safonov calculates the critical mass of cavity type reactors fueled with ^{235}U, ^{233}U, and ^{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 thickness of moderating exteriors.[41]

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 asymptotically with radius cubed once a limiting moderated-to-fuel ratio is obtained."

2. <u>Internal Moderation – Unbounded Regions and Multiple Infinity</u>

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 20. 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.

In 1967, L. B. Engle and W. R. Stratton[42] made a parametric study of bare homogenous spheres containing ^{235}U, ^{238}U and carbon in various mixtures. Figure 20 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 ~ 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%.[43] 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.

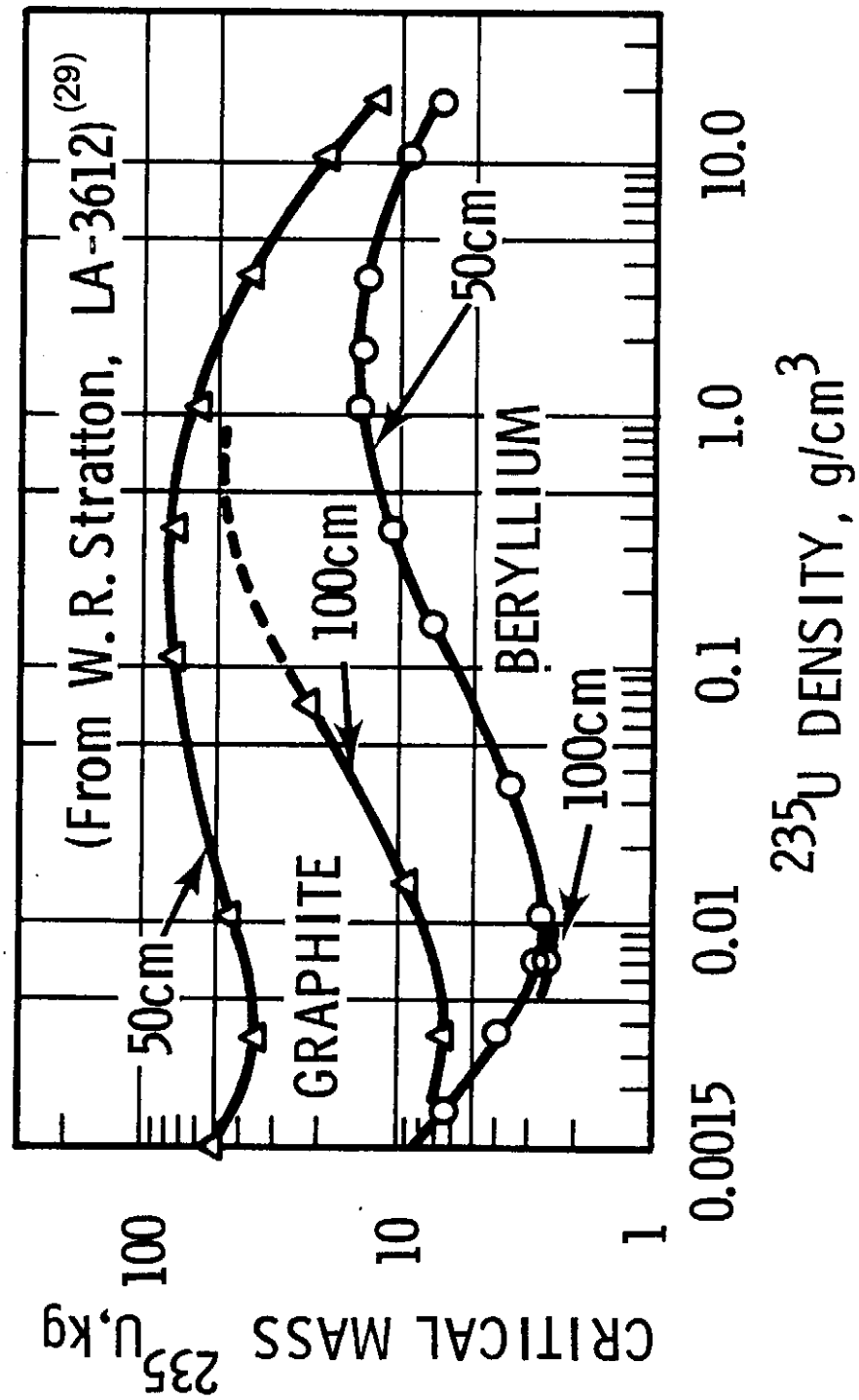


Figure 19. Computed Critical Masses of U(93.5) Metal Reflected by Thick Graphite or Beryllium for a Wide Range of ^{235}U Densities

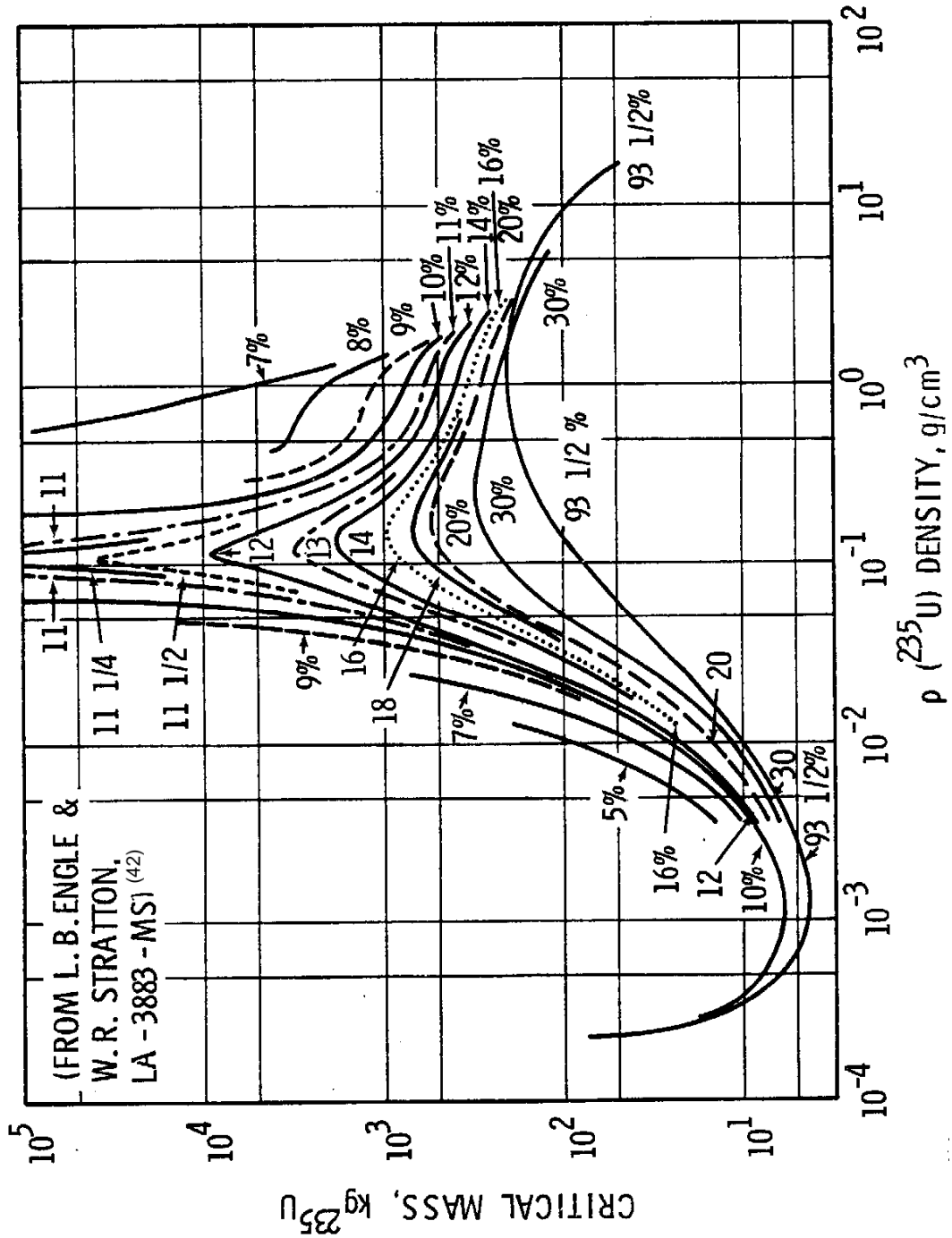


Figure 20. Critical Mass (kg ^{235}U) of Homogeneous Spheres Containing ^{235}U, ^{238}U and Carbon vs. ^{235}U Density for Various ^{235}U Enrichments

3. Moderation and Density Effects in Dry and Damp Powders

The curves of Figure 6, previously discussed, show that a significant increase occurs in the critical mass of Pu as the metal is initially 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 made in 1975, on dry and damp mixed oxide powders, by J. H. Chalmers, Health and Safety Executive, Nuclear Installations Inspectorate, England, bear this out.[44] Data taken from calculations made on mixed oxides containing 15 wt.% PuO_2 are presented below (Table VI):

Table VI. 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 6, 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 hydrogenous mixture is 1.35 = 0.013, which occurs at an H/U ratio of about 7 (H/^{235}U ratio about 24.)[14]

Some interesting results were reported on Monte Carlo calculations of k_∞ for unmoderated 3.04% enriched uranium metal and for UO_3.[14] 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 approximately 135 mk. With the proper amount of hydrogen, however, k_∞ for the 3% oxide is raised from 0.58 to 1.35.

4. <u>The Dilute Fissile Bearing Solution (Criticality and Evaporation)</u>

Apart from reactors, with a few exceptions, all of the nuclear criticality incidents have involved uranium or plutonium in the form of solutions.[43] Solutions can concentrate, leak, siphon, or be inadvertently 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 “infinite 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 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 ~ 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 concentrations as the vessel height is increased indefinitely. 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 columns 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 19 for artists rendition). Further, in the event that criticality was to occur by this process, the reaction might well be autocatalytic depending upon 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 vessel's 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 infinitely 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.

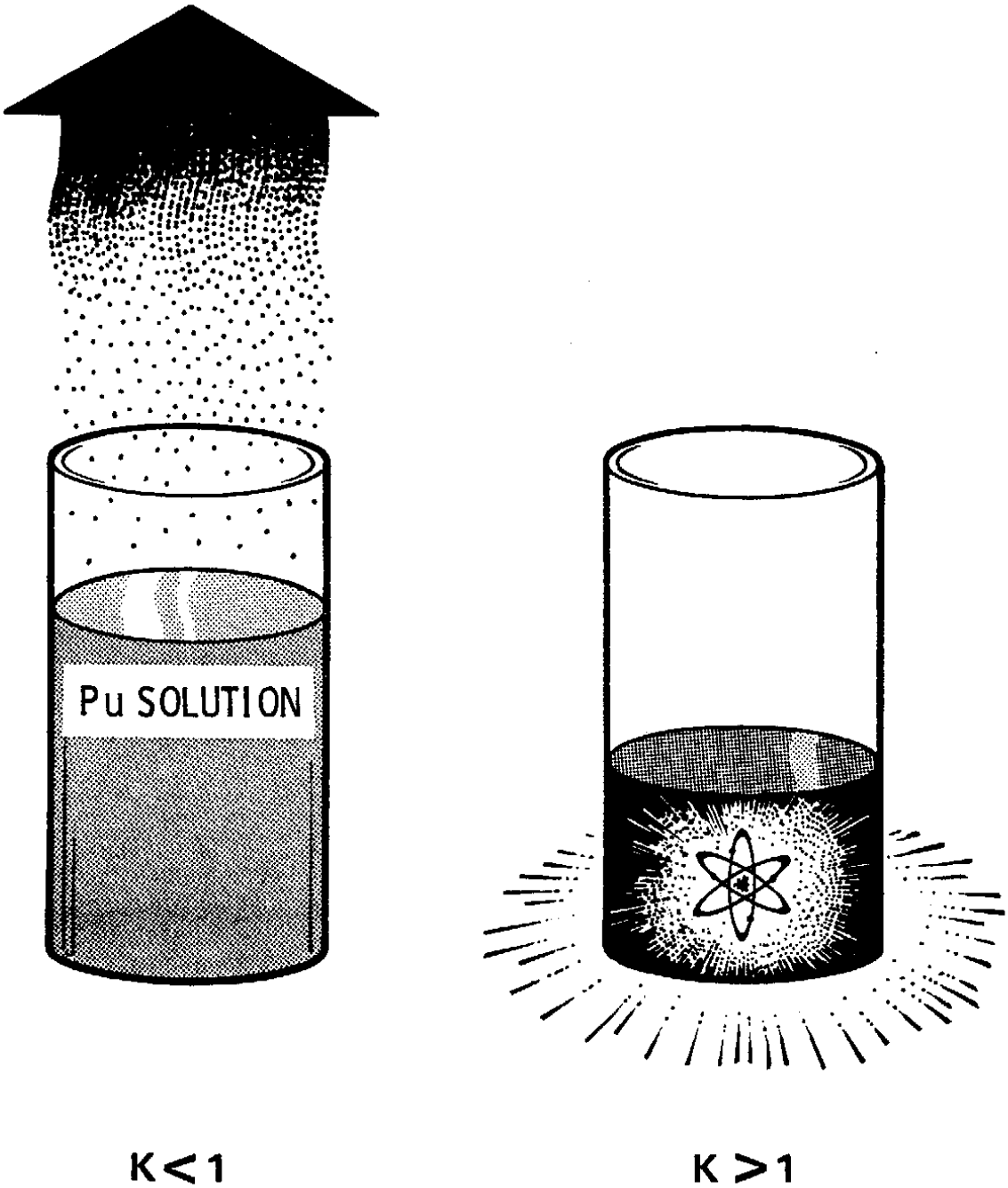


Figure 21. Evaporation – Concentration and Criticality

J. CRITICALITY AND THE NEGATIVE 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. Following is 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 non-leakage probability and 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 not be cause for concern. Yes, 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.[45] It was demonstrated (using 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[45] results are included in the following table, which illustrates the point in question and shows k_{eff} to be greater than k_∞.

Table VII. 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 ($k_\infty \leq 1$) could be found that could be made supercritical in finite size with light water reflection.

K. 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 ½ inch thick nickel next to the core, surrounded by natural uranium, and give a significantly smaller critical mass than either reflector alone.[46]

There is no verified explanation for the effect, but it is suspected that it may be associated with a strong scattering resonance that nickel has at about 16 KeV.[47]

There are also 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 show 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 a thick water reflector by itself, or 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 22.

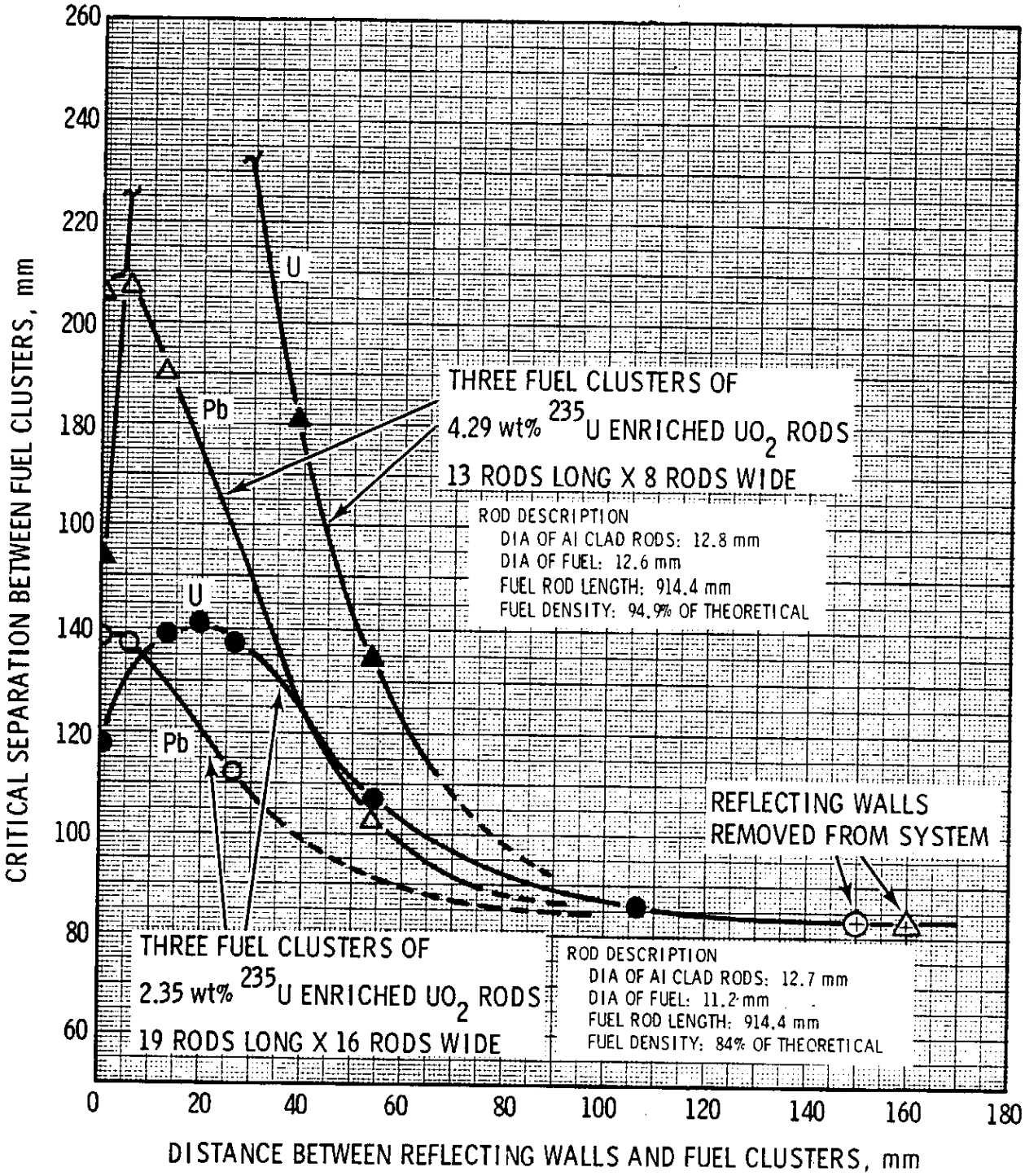


Figure 22. Critical Separation between Fuel Clusters of 2.35 wt.% and 4.29 wt.% ^{235}U Enriched UO_2 Rods in Water with Depleted Uranium or Lead Reflecting Walls

L. THE DISSOLVER PARADOX

The dissolution process can involve fissile material in the form of metal or oxide initially, whereas, during the intermediate state, 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 was 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 23).[48]

The computed curves in Figure 24 show the critical masses (total of ^{239}Pu 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 25 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.

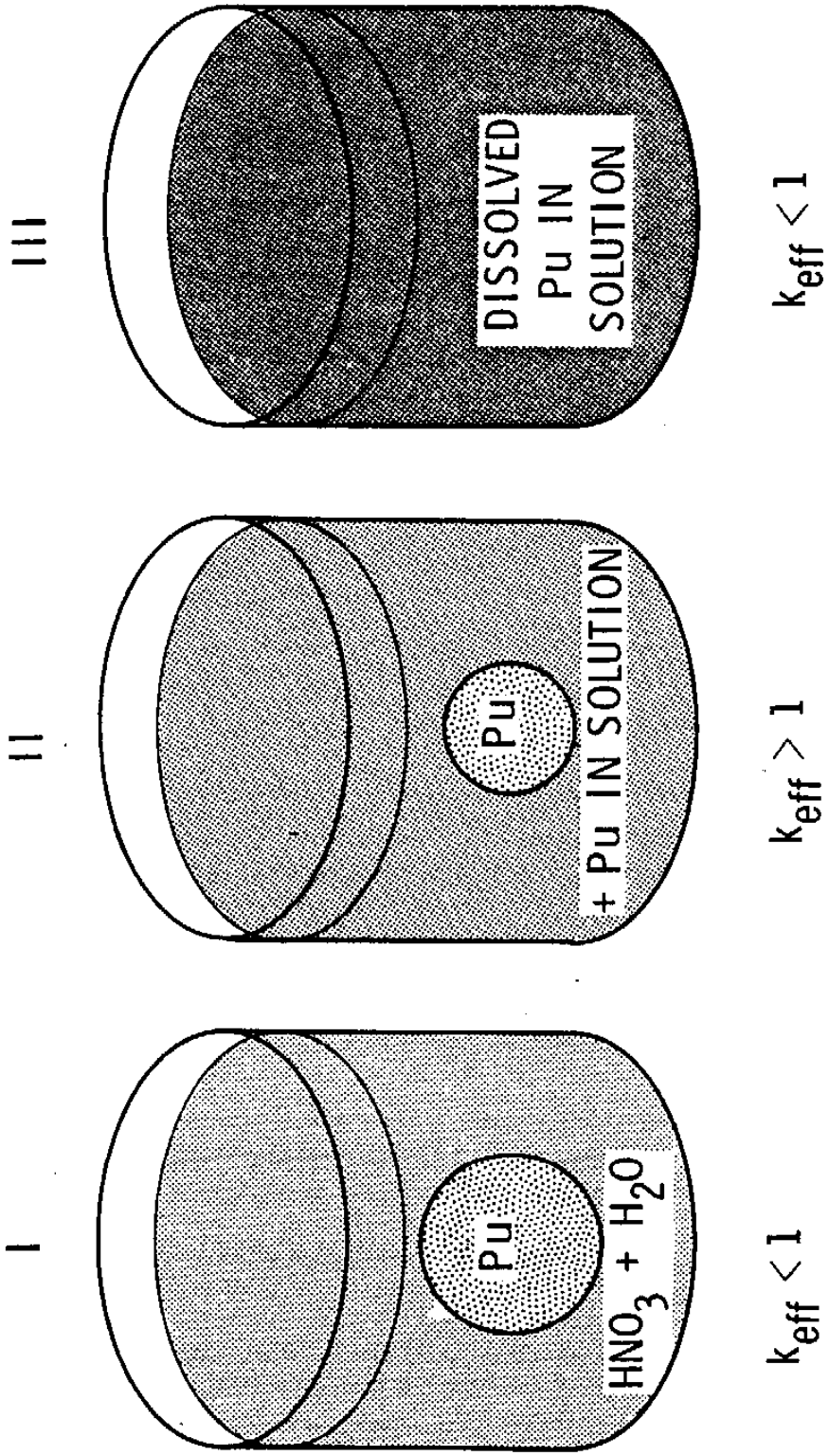


Figure 23. Pu Metal Dissolution

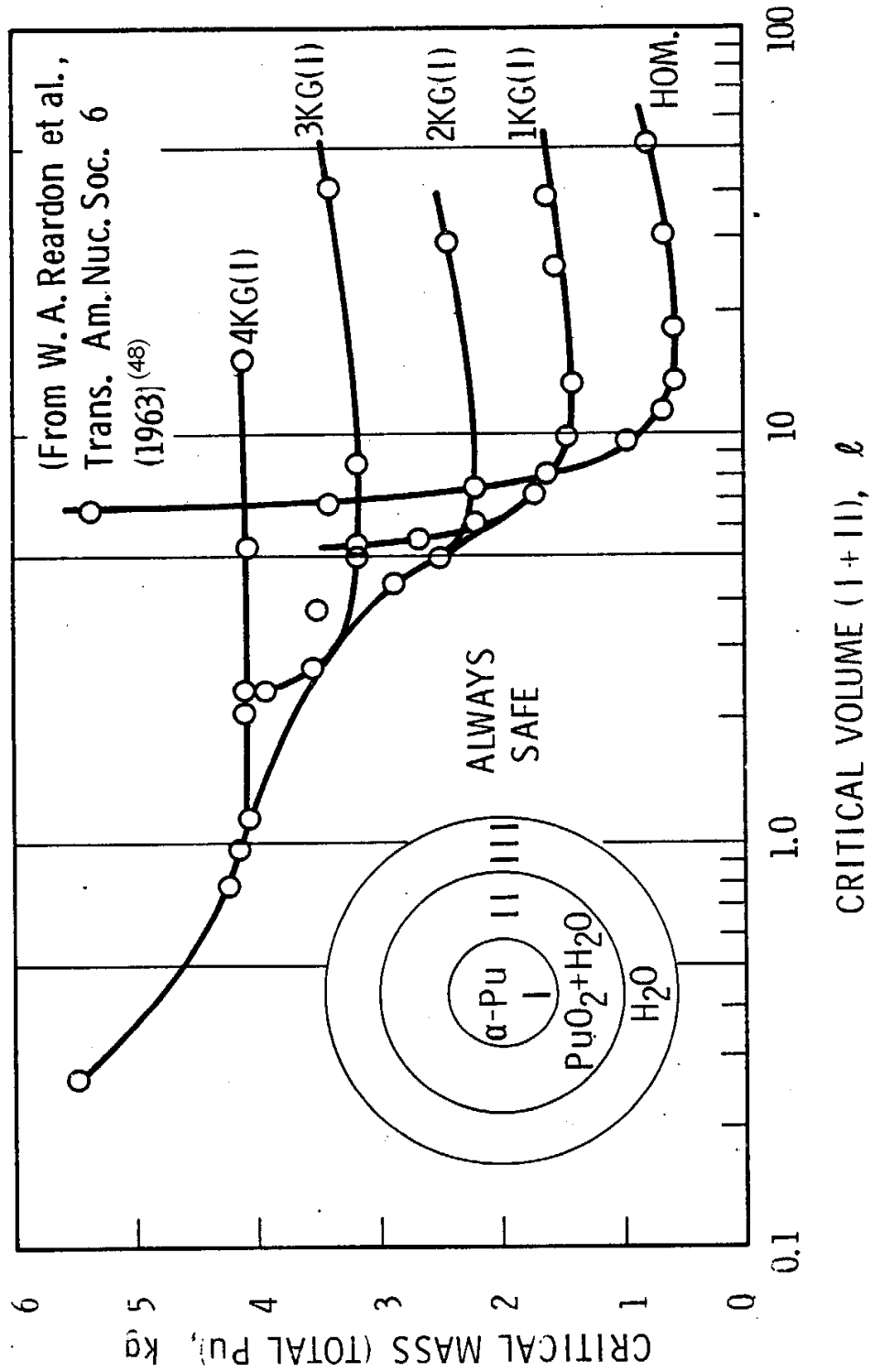


Figure 24. Computed Critical Mass (Total ^{239}Pu) vs. Volume (^{239}Pu in PuO_2 + H_2O Solution)

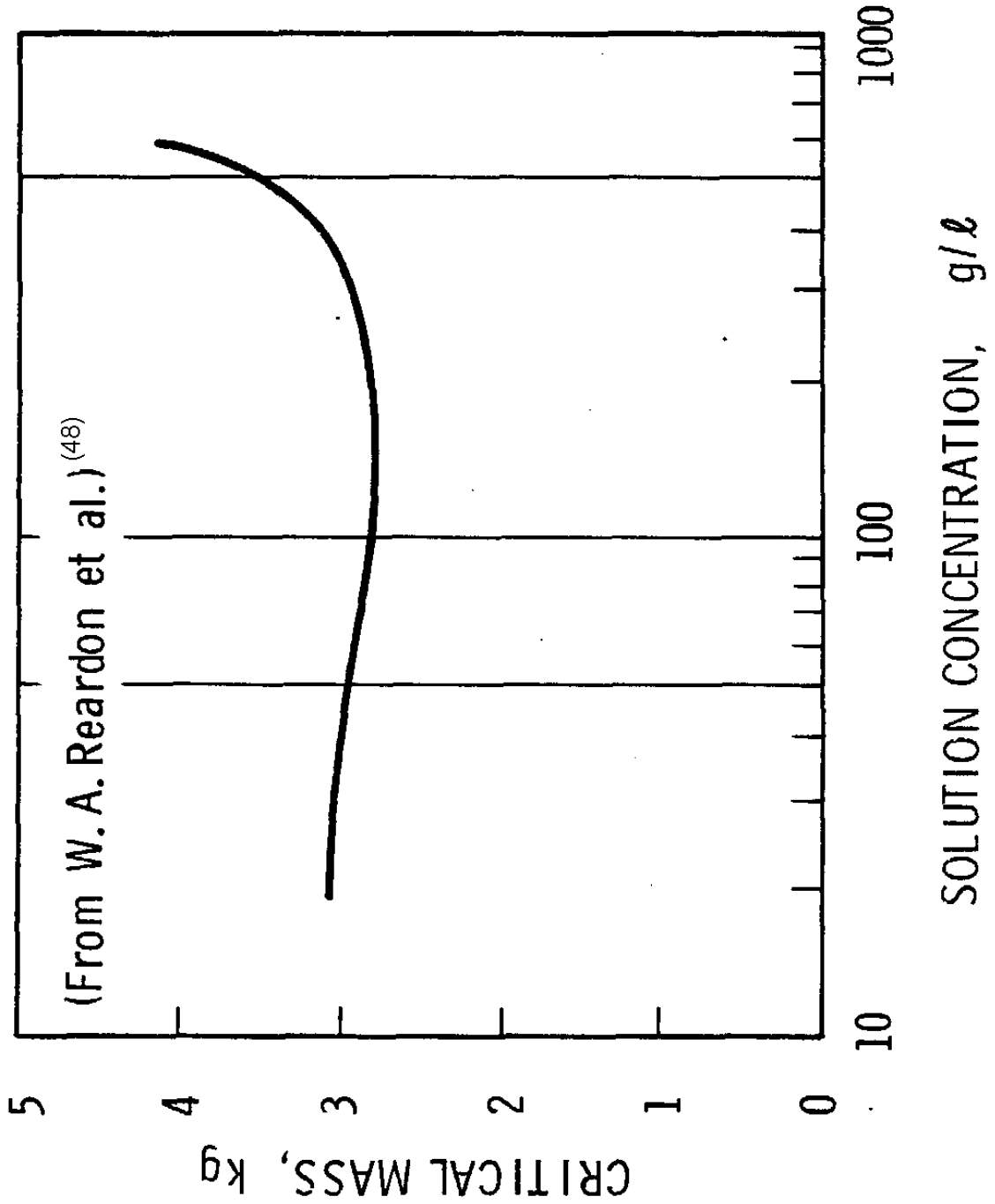


Figure 25. Computed Critical Mass vs. Solution Concentrations; 3 kg Dissolving into 5 Liter Volume

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

Initially, it may seem surprising that in the case of a homogenous 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.[49] This is evident from Figure 26, 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 remembered, 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 27 are similar to those in Figure 26, 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 total concentration of Pu + U. The same conclusions are evident.

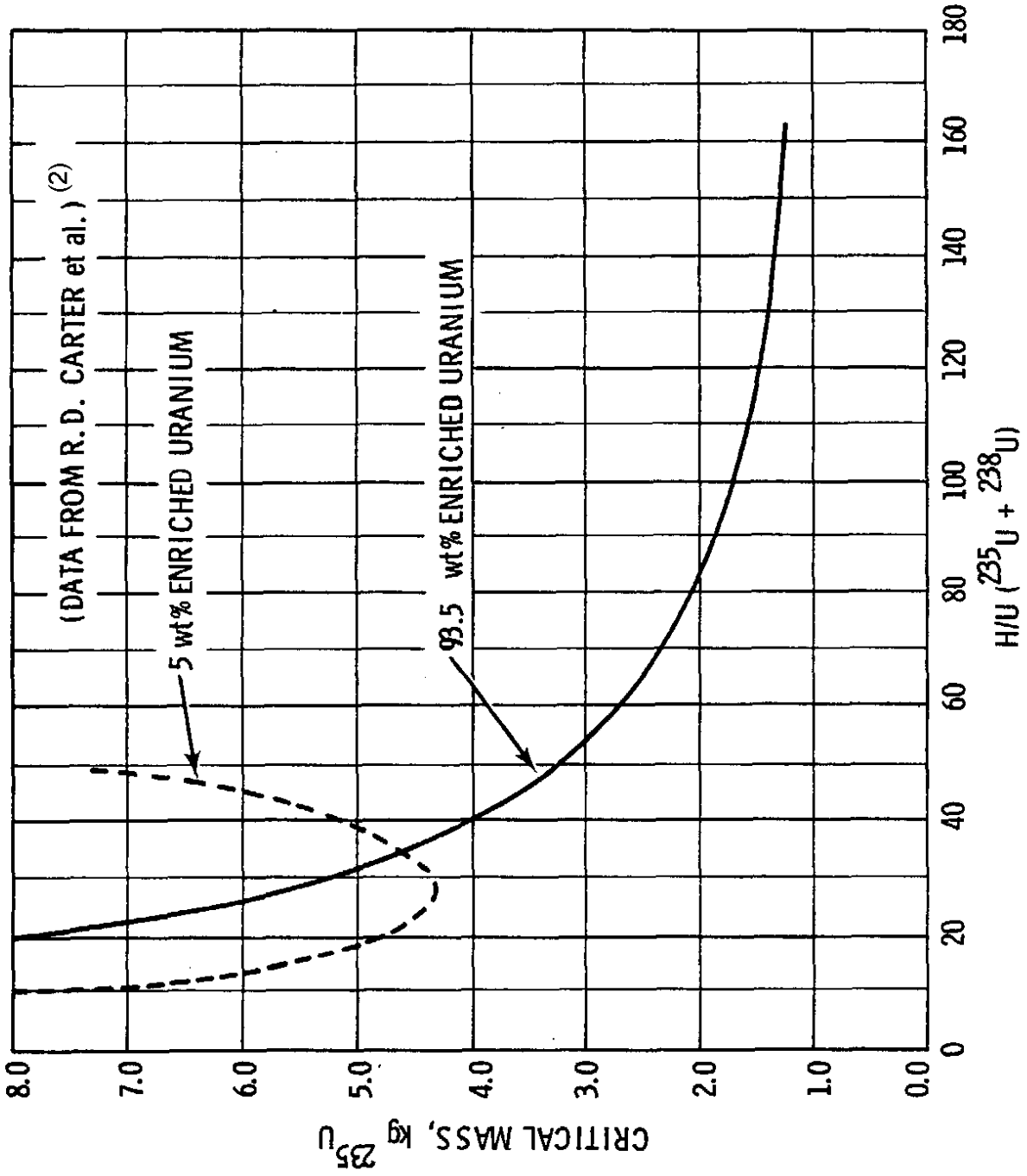


Figure 26. Computed Critical Masses of Water Reflected Spheres of Uranyl Nitrate Solutions (No Excess HNO_3)

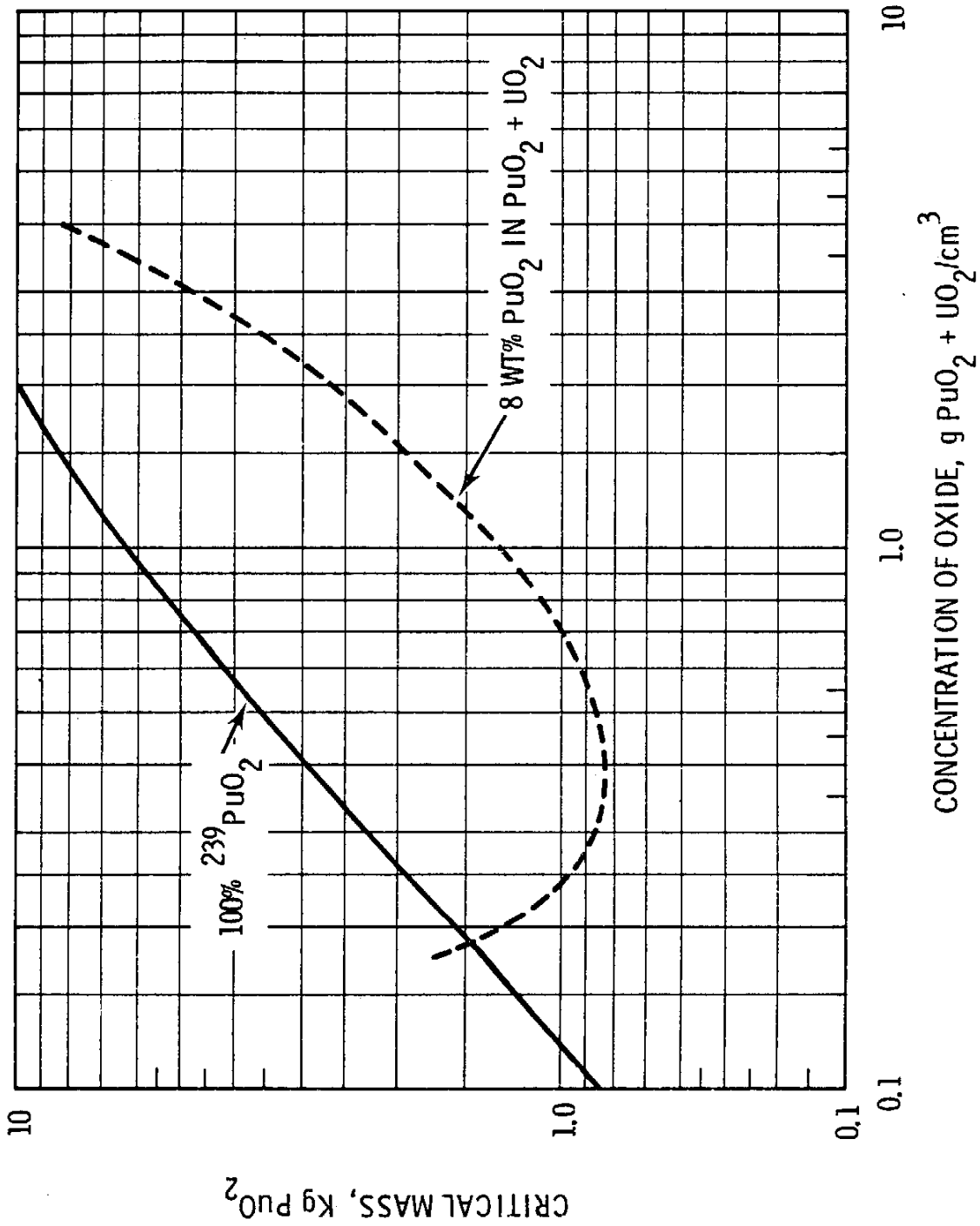


Figure 27. Computed Critical Masses of Water-Reflected Spheres of PuO_2 and $U_{NAT}O_2$-H_2O Mixtures

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

Critical mass values have been recalculated for the even-even nuclide, ^{240}Pu, which lies in the range 33 to 19 kg, depending on the type of reflection (see Table X). *Note: This represents a substantial reduction in the previously estimated critical values; the critical mass for a bare sphere of ^{240}Pu metal (~ 33 kg) is now significantly less than that of ^{235}U metal.* The critical mass for ^{239}Pu metal is given as 5.2 and 10 kg for water-reflected and bare spheres, respectively. Pertaining to mixtures of the above isotopes, Figure 28 gives the percent change in critical mass per percent change in ^{240}Pu content as a function of the total Pu concentration in homogenous water mixtures, spanning the range from dilute solutions to that of full metal with no water, wherein the H/Pu ratio is zero.[50] 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 ~ 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 ~ 5.8 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. As 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.

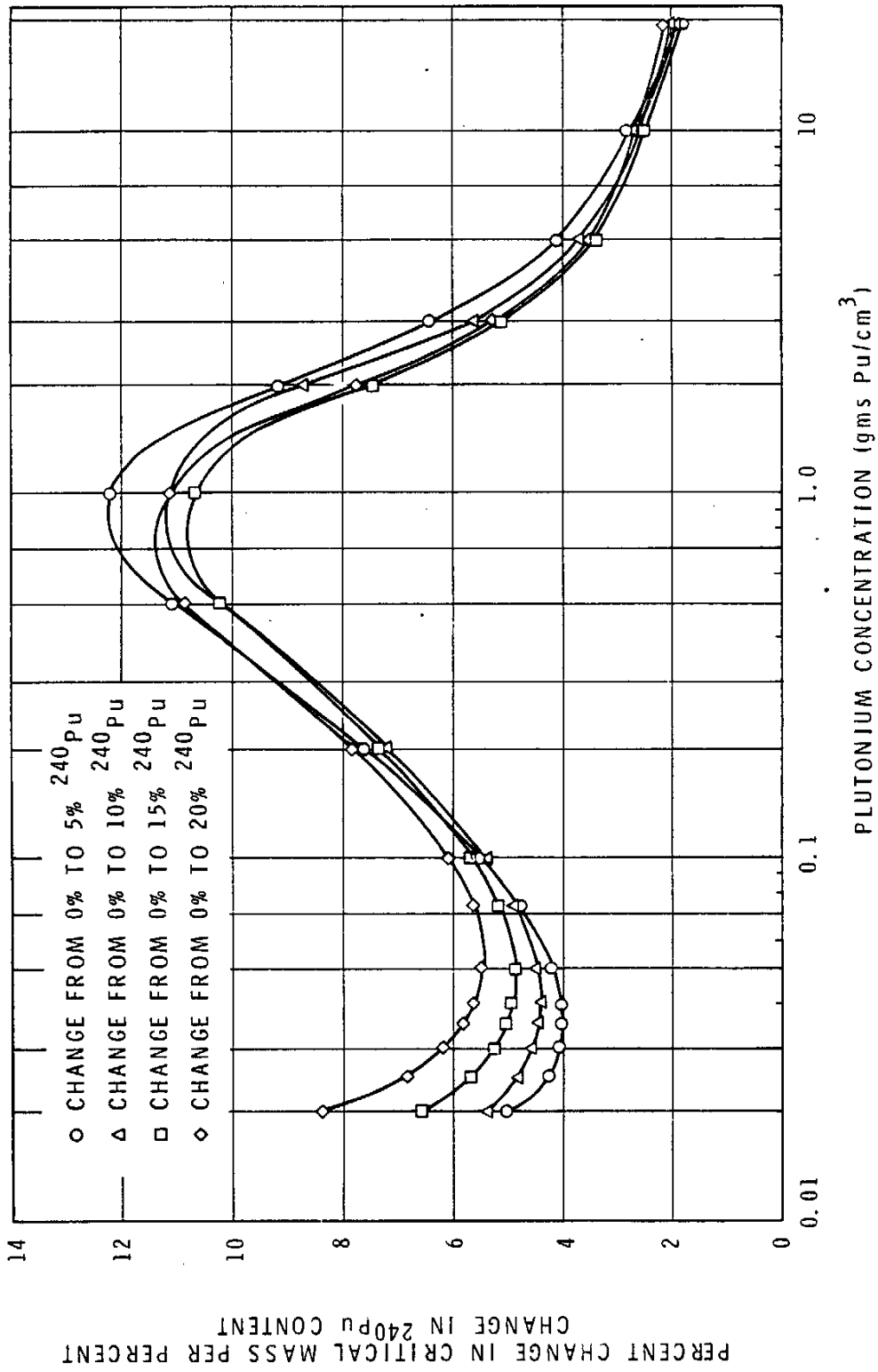


Figure 28. ^{240}Pu Effects on Water Reflected Spherical Critical Masses

O. 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. 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 can it be logically assumed 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 29 and Table VIII).[51]

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

Table VIII. 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

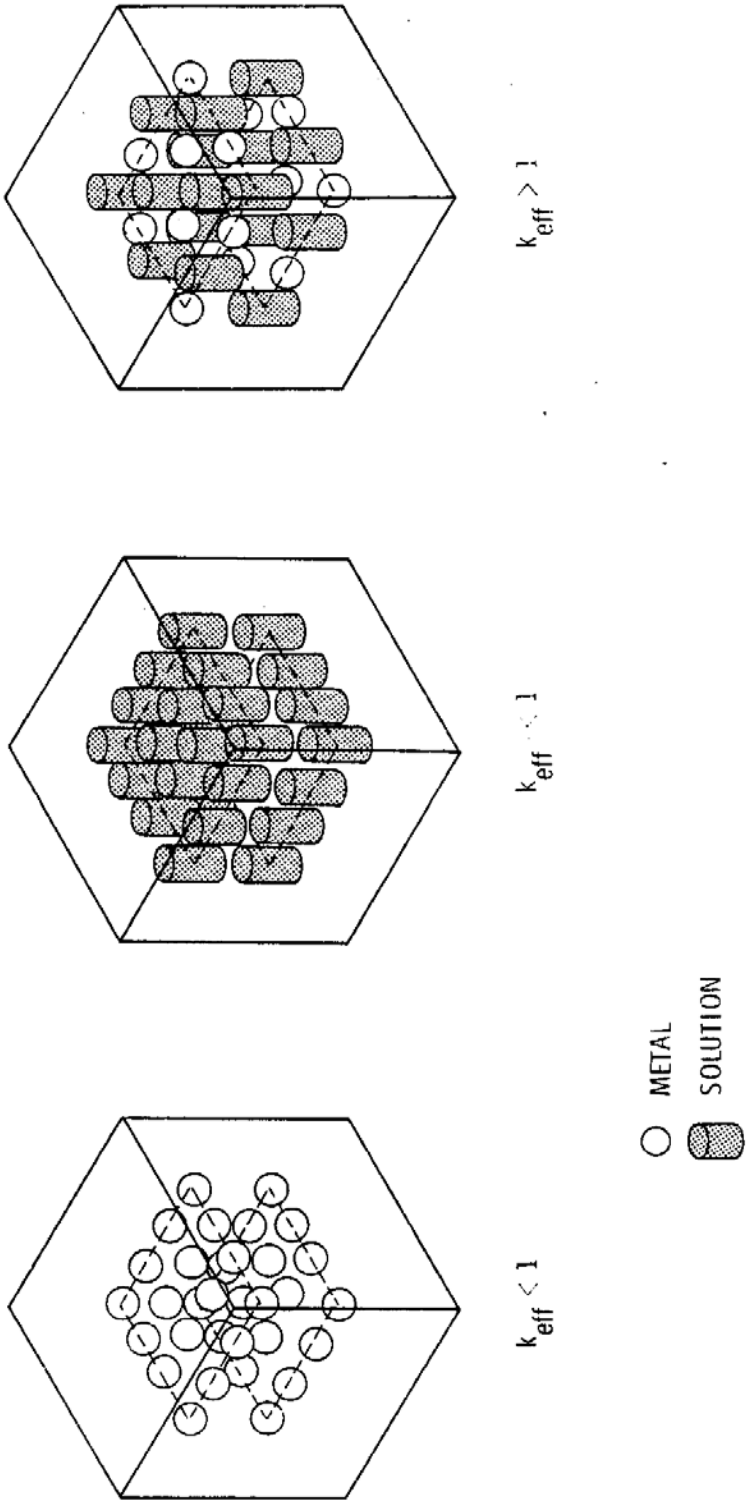


Figure 29. Criticality of Mixed Arrays

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.[52] 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 30). 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.

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

The anomalous effects of moderation in transportation and storage arrays of nuclear materials present challenging calculational problems.[53] The nuclear criticality safety of fuel storage arrays requires that the potential of low density moderation within the array be considered. Over the years, several anomalies have been described that pertain to, 1) the effect of internal low-density interstitial moderation on the criticality of storage arrays,[54] and 2) the reactivity enhancement that can be caused by a density reduction in the units composing an array with interstitial moderation.[55, 56]

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.[55] Similar results were obtained with arrays made up of ^{235}U metal spheres (see Figure 31). 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 interstitial water moderation.

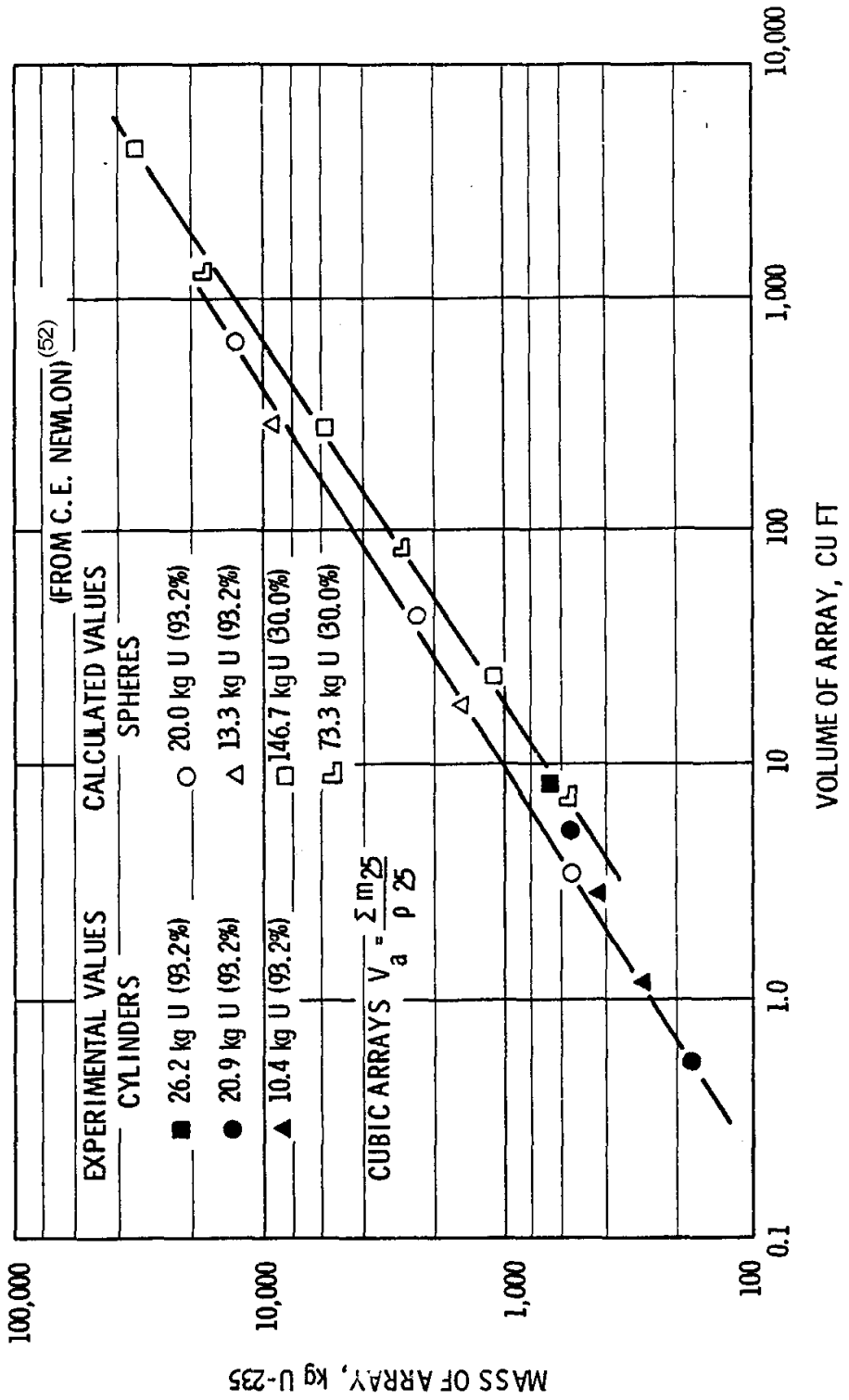


Figure 30. Criticality Mass and Volume of Unreflected Metal Arrays

For those cases studied, it was found that the most reactive unreflected arrays were those containing solid metal units, regardless of the amount of interstitial 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 of about 0.5 Mev., and in the sphere with a radius four times larger, but with a density reduction of 64 in order to preserve the unit mass, the median fission energy becomes about 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 Koponen'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 interstitial, neutron moderation. As pointed out by Koponen, 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[29] and another by C. B. Mills[39] 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).

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[57] 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 special 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.

There have been a number of papers written to assess the criticality safety of proposed and existing storage arrays, and to examine the effects of low-density moderation.[55, 58-64]

The availability of appropriate benchmark experiments for low-density moderation is quite limited. The French, however, have performed experiments at Valduc in which four PWR-type assemblies were made critical in water with various hydrogenous compounds interposed between the assemblies.[65] The interposed materials were water, polystyrene balls, polystyrene powder, expanded polystyrene and air. Expanded polystyrene $(C_8H_8)n$ was reported to have a hydrogen concentration equivalent to about 2% full-density water whereas polyethylene powder $(CH_2)n$ was equivalent to about 38% full-density water. Attempts to validate calculations against the one set of suitable experiments at low density moderation were reported as disappointing.[62]

It has been reported that the maximum k_{eff} for a typical PWR fuel storage array will occur for interstitial moderation equivalent to 5% of full density water or 0.05 to ~0.1 g H_2O/cm^3

depending on the array.[59, 62] These densities, although relatively small, are still quite large compared with the density of water provided by an overhead sprinkler system.

Experiments to measure the water density from sprinklers and fire hoses have recently been reported in detail.[63] Since the maximum water density was only 0.004 g/cm^3, achieving a density in the range of 0.05 to 0.1 g/cm^3 was considered unachievable or incredible. Most of the papers pertaining to the effect of density reduction and/or low density interstitial moderation on storage arrays show the proposed or existing arrays to be "OK," but this is principally due to the fact that the maximum achievable water density from the overhead sprinkler system is not high enough to increase the k_{eff} of the proposed finite array above unity. If the array were large enough, however, and the enrichment of the uranium near 5% or greater, this would not necessarily be the case. Thus, interacting arrays of storage materials require detailed examination for the effect of possible interstitial moderation and density reduction on the criticality of the units composing the array. It is often required to show that the fuel array is subcritical for the aqueous atmosphere of all water densities from 0.0 to 1.0 g/cc.

Although the effect of most sprinkler systems may be unimportant due to the very low density of the moderator – it has been observed[66] that a quantity of mist moderation judged to be safe might still be unacceptable due to water film formation on the fuel material. The film thickness is due to the viscosity of water and possibly an updraft during a fire. The effective film thickness should increase also if the fuel rods are stored horizontally. KENO V.a displayed this effect for fuel assemblies containing 256 rods, composed of UO_2 at 4.1 wt.% enrichment, in a 16 x 16 array. The assemblies were in 19 x 34 storage array. The KENO results are plotted in Figure 32.

Most arrays show a maximum k_{eff} with low-density water moderation. The study by Koponen in 1977,[55] however, did not show this maximum for unreflected 5^3 and 10^3 arrays of 15-kg ^{235}U spheres. Repeat calculations were made in 1993 for some of the unreflected arrays reported by Koponen in 1977[55] with some interesting results. Figure 33 shows the results of calculations for the 10^3 arrays with the MCNP neutron Monte Carlo code with the pointwise X6XS.0 cross-section library.[67] Low density water moderation is now seen to produce maximum reactivity at water densities near 0.1 g/cc. Calculations on unreflected 10^3 arrays with KENO V.a – CSAS4 in SCALE 4.1, with the 27-group cross-section library, matched the MCNP results.[53]

Another interesting effect becomes apparent by comparing results plotted at the left end (no interstitial moderation) of Figures 33 (a) and (b). A considerably higher k_{eff} is obtained for the low-density units in unmoderated reflected arrays than for unmoderated bare arrays, but for arrays with interstitial moderation, the difference is quite small. This can be explained by the action of the interstitial moderation in keeping neutrons from leaking from the array by acting as a *internal reflector* as well as providing some degree of reflection at the array edges of "unreflected arrays," due to the unit cell setup which includes water in the region external to the edge units of the array.

The reactivity enhancement due to fissile material density reductions exists for both unreflected and water-reflected arrays.

As pointed out by Koponen,[55] 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[68] and also by C. B. Mills[69] 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.)

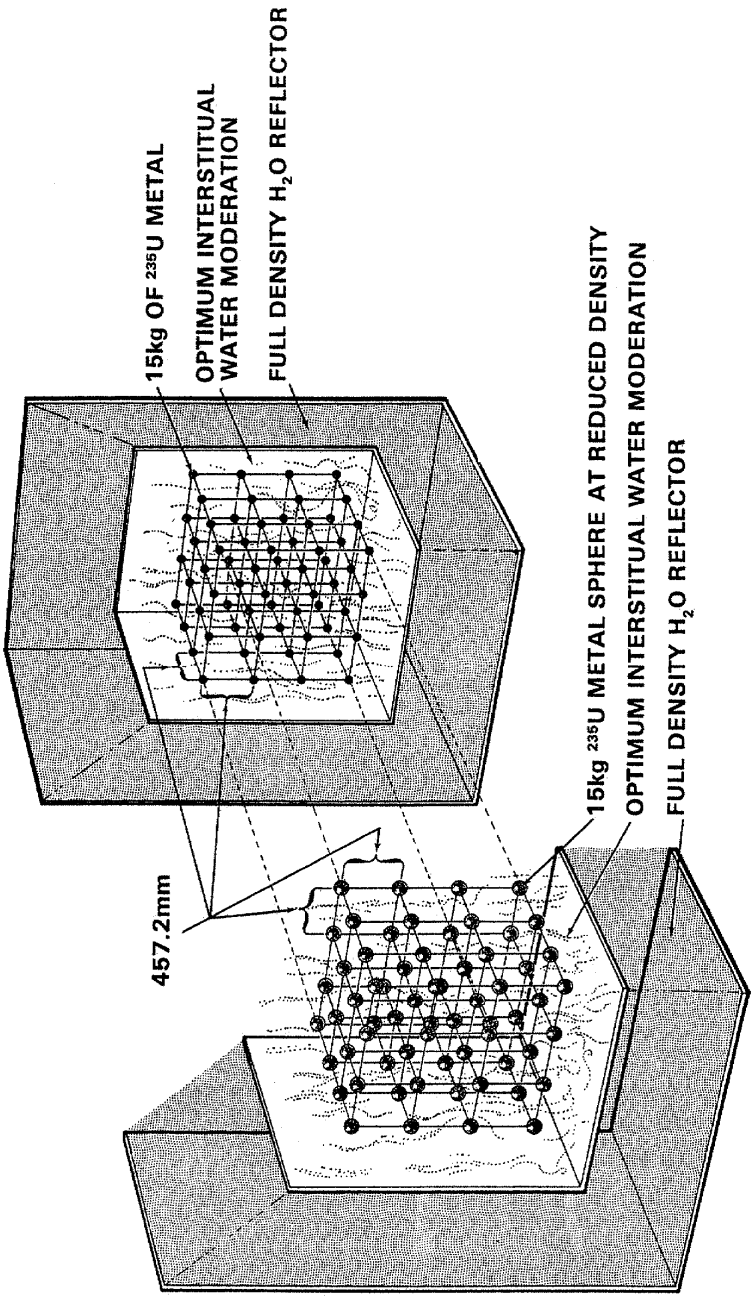


Figure 31. Reactivity Enhancement due to Density Reduction in Units

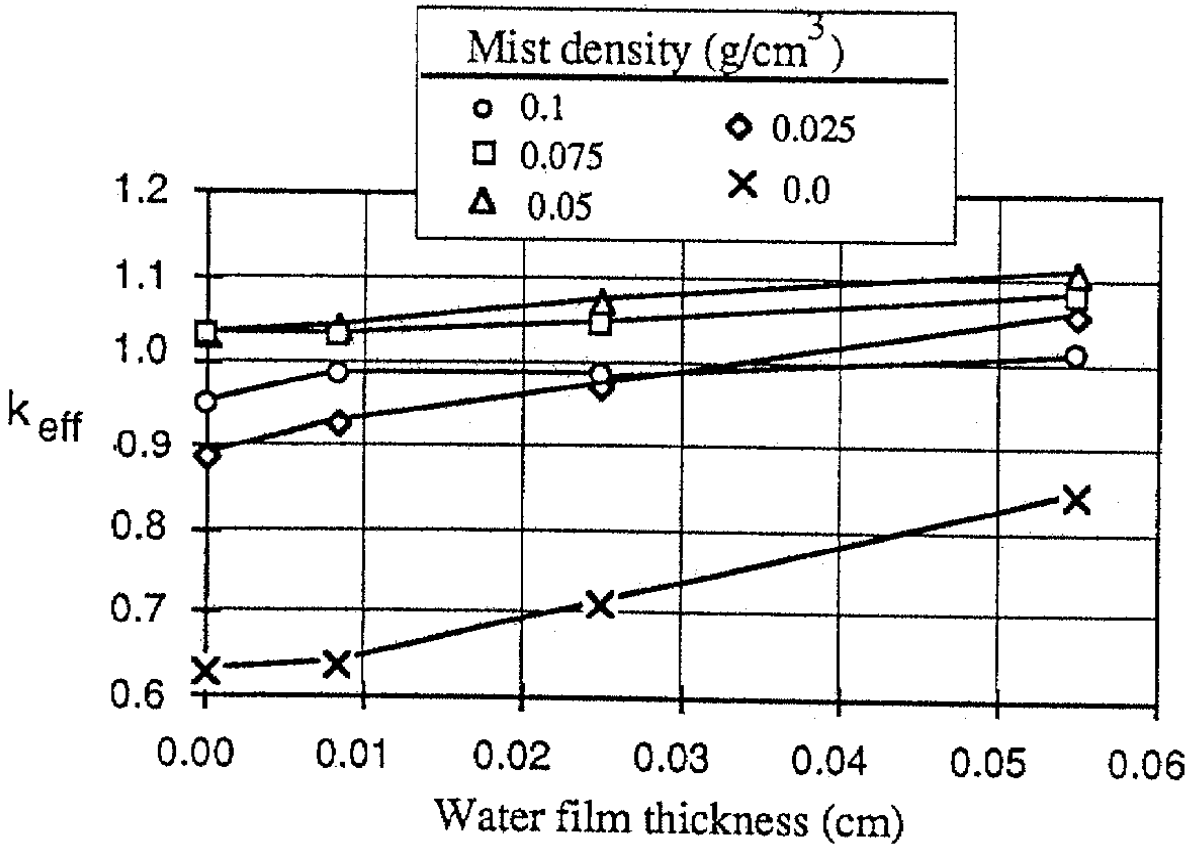
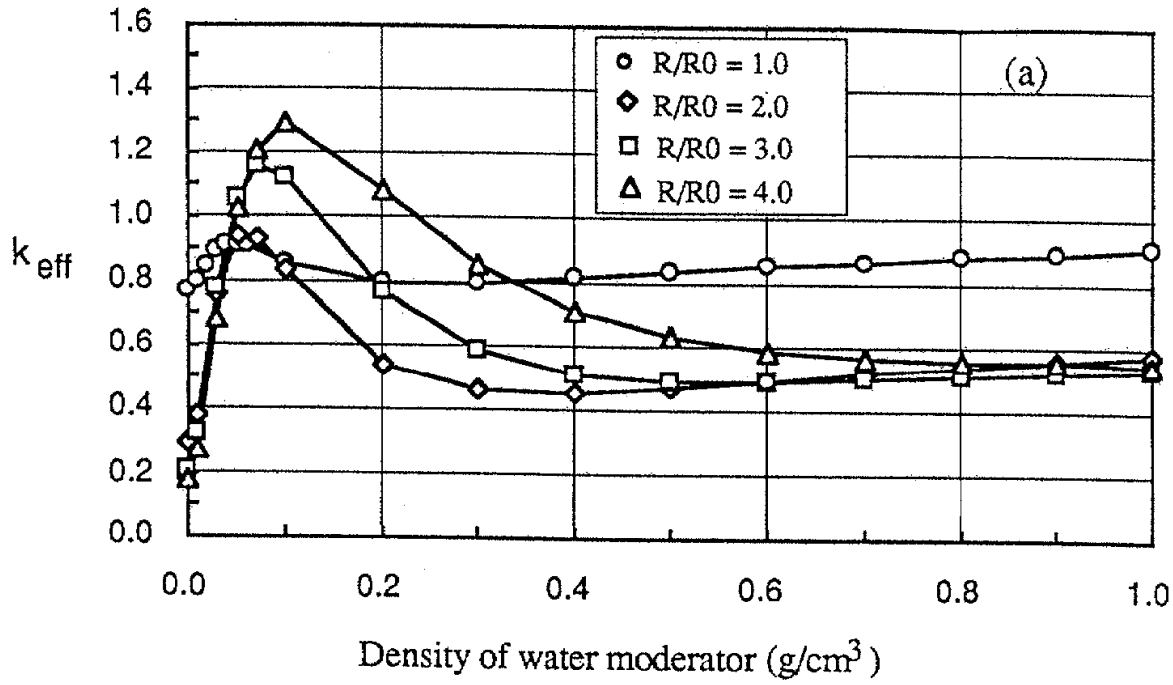


Figure 32. Film Effects of Water Sprinklers on Storage Array of 4.1%-enriched UO_2 Rods

(Assemblies consist of 256-rod-assemblies in 19 x 34 storage array.)
(Calculations are with KENO V.a. with 27-group SCALE cross-section library[66])



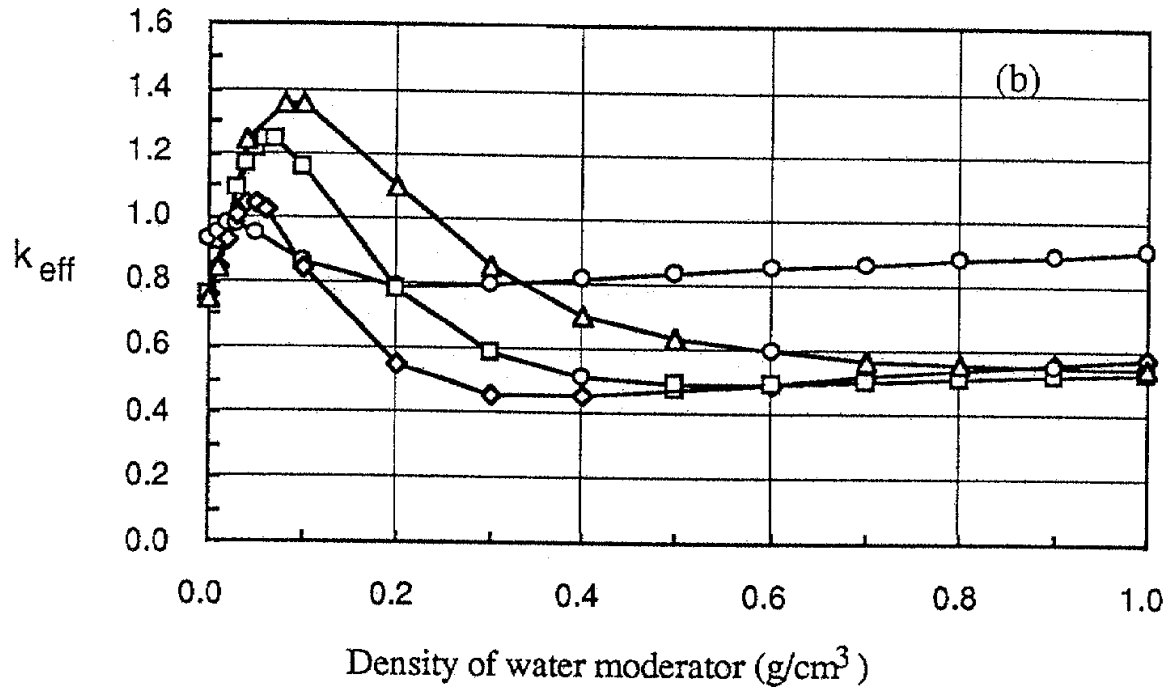


Figure 33. Effect of Unit Density Variations and Interstitial Water-moderator Density Variations in 10^3 Arrays of Dry 15-kg ^{235}U Units at 60.96-cm CTC Separations Calculated by the MCNP Neutron Monte-Carlo Code with the Pointwise X6XS.0 Cross Section Library: (a) Calculations for an Unreflected Array, and (b) Calculations for an Array surrounded by a Full-density Water Reflector[67]

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

An interesting problem concerns the effect on criticality for an array of interacting units if the water content of the intervening airspaces within the array was 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.

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 34) 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 a 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 involved 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, and 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.

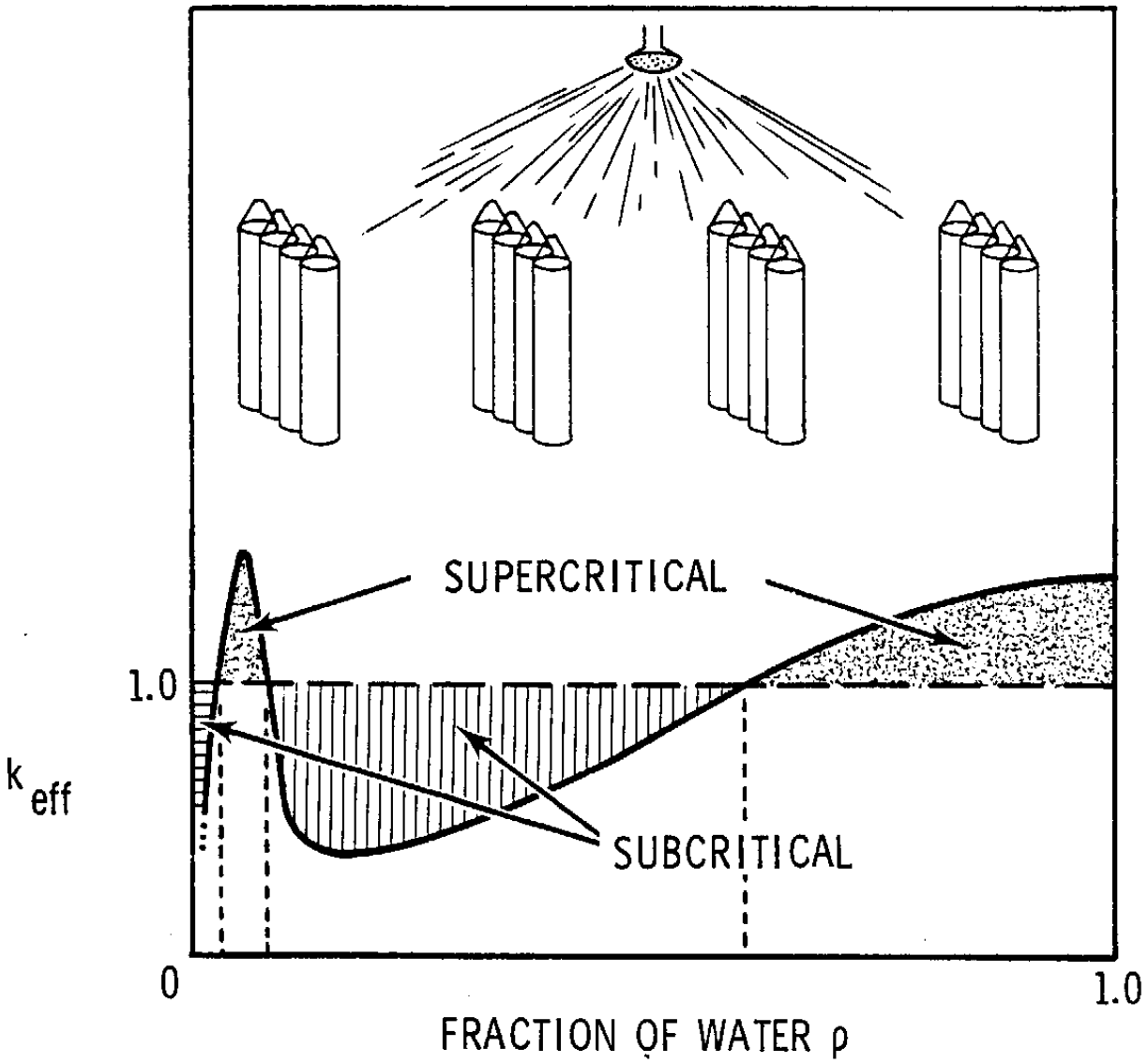


Figure 34. Fire (Fog, Mist, or Flooding: The Potential of Triple Criticality in a Storage Array)

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.[57]

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 (23 wt.% ^{235}U) was examined.[70] The three basic geometries or shapes of the subcritical units composing the arrays were the cube, sphere and cylinder. To clarify, 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 was replaced with a cube of the same k_{eff} that the k_{eff} of the array would be increased. The cube consistently 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 an 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 that if an array were to be made up of subcritical spheres, and if units of different shapes 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 35, 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.

(FROM J. T. THOMAS, ORNL)

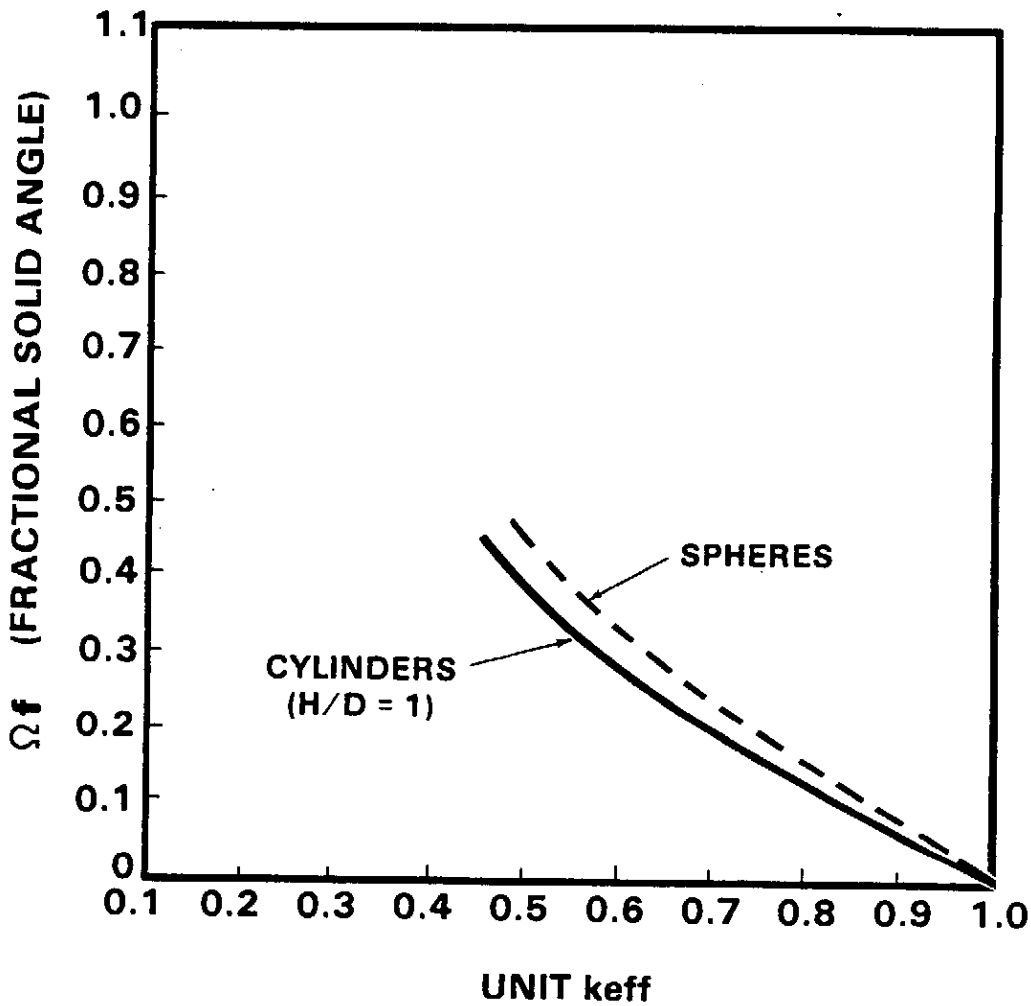


Figure 35. Comparison of Critical Reflected Arrays of U (93.1) Metal Spheres and Cylinders

P. SAFETY IMPLICATIONS OF ANOMALOUS EFFECTS OF NEUTRON ABSORBERS ON CRITICALITY

A number of apparent "anomalies" have been disclosed in recent years,[54, 56] and as new data have become available, additional anomalies have come to light. An anomaly, once disclosed, is amenable to explanation since there is a valid reason for the occurrence of any happening and a scientific way to understand any phenomena. Application of existing data, without knowledge of the "anomalies" could lead to undesirable events, or diminished criticality control. Neutron absorbers are frequently used for criticality control in nuclear fuel cycle operations. In the following, several anomalies have been selected that have principle application in nuclear fuel processing.

Common neutron absorbers include: boron, cadmium, and gadolinium. Other materials are frequently present in the constituents that may act in the capacity of neutron absorbers such as NO_3, ^{238}U and ^{240}Pu. ^{240}Pu is a "fissible" nuclide that may serve either as a strong neutron absorber to inhibit a chain reaction or contribute neutrons to a chain reaction through fast fission, depending on the condition encountered.

1. Use of Soluble Absorbers for Criticality Control of Power Reactor Fuels in Water

The presence of large quantities of neutron absorbing nuclei can alter the neutronics of a system. High concentrations of thermal neutron absorbers such as boron, cadmium or gadolinium cause a shift in the neutron energy. For example, calculations by C. R. Marotta on the re-criticality potential of the TMI-2 core show that the peak value of k_∞ is shifted toward lower values of water-to-fuel volume ratios as the boron concentration is increased in the water moderator.[71] (see Figure 36).

It is apparent that if the concentration of boron in the water moderator of two different lattices be the same, the lattice with the larger spacing (and water-to-uranium ratio) will have a higher ratio of boron to uranium. In addition, as the spectrum will be faster in the lattice of least water, the boron also can be expected to have a smaller effect on the criticality of that lattice.

It also is apparent that compacting a lattice of fuel rods from optimum spacing in water (reducing the separation between fuel rods in the assembly) from that, which gives the maximum buckling, can result in a larger critical size and number of fuel rods for criticality and

lead to a safer condition. In the case of heavily borated lattices, however, it is possible for the reverse to occur, i.e., with the absorber present, compacting or consolidating, the lattice spacing can result in a smaller critical size or volume and number of fuel rods for criticality.

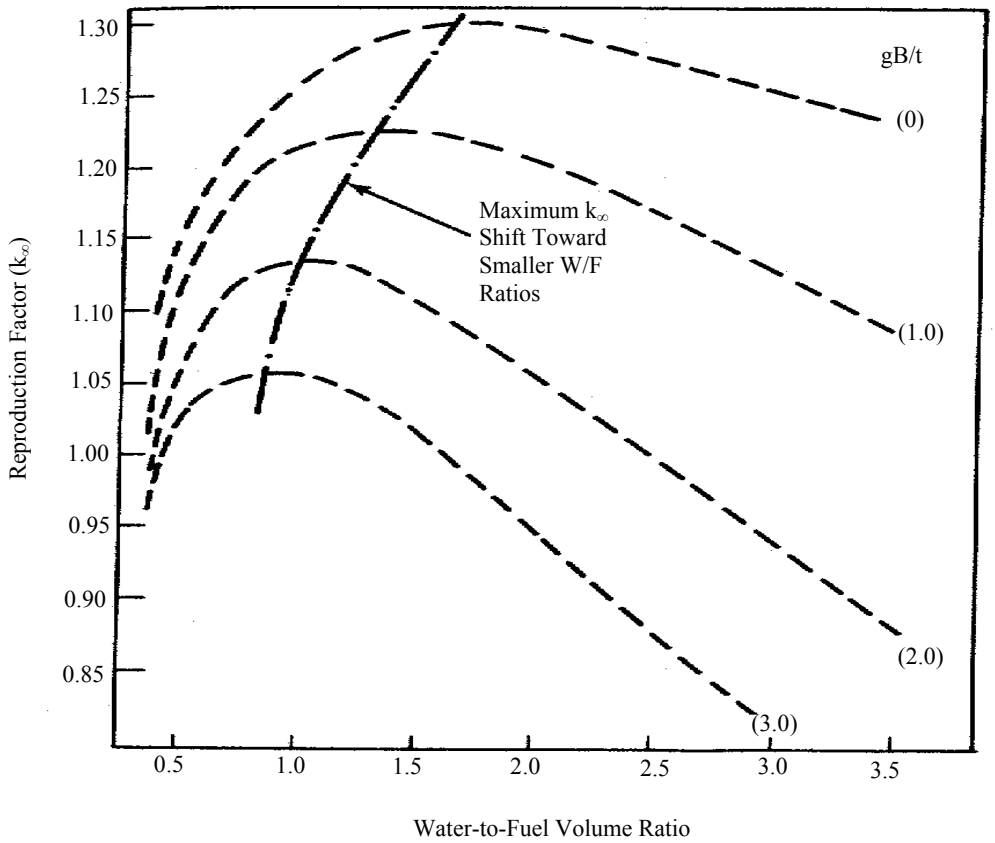


Figure 36. Variation in Reproduction Factor (k_∞) of Water Moderated Lattices as Function of Water-to-Fuel Volume Ratio and Boron Content [Bottom Three Curves from Marotta[71]]

This should not be confused with the fact that adding a "neutron absorber" to any given lattice (providing this absorber does not of itself substantially moderate neutrons or displace the moderator that does) will always render that lattice assembly further subcritical.

2. <u>Use of Borated Glass Raschig Rings for Criticality Control in Vessels containing Fissile Solutions</u>

An American National Standard (ANSI/ANS-8.5)[72] provides guidance for the use of borosilicate glass Raschig rings as a neutron absorber for criticality control. In connection, with the preparation of this standard values of k_∞, they were calculated for various glass volume

fractions versus the volume fraction outside the glass (glass tube OD = 38.10 mm). The volume fraction outside of the glass is the fraction of the cylindrical cell that is outside of the glass tube, and is a measure of the open space between the rings. One of the curves from these calculations is reproduced in Figure 37.[73] It is to be especially noted that although the volume of solution occupied by the glass is the same in each case, the rings are less effective when either in contact or spread out.

Although the volume of the solution occupied by borated glass Raschig rings of different thickness can be the same, the rings may be less effective when either in contact or spread out. Thus, in using Raschig rings for criticality control, not only must the glass volume fraction be specified, but also the outside diameter of the rings. It should be understood that for a Raschig ring of given dimensions, an increase in glass volume fraction (or stacking density) will always cause k_∞ to decrease.

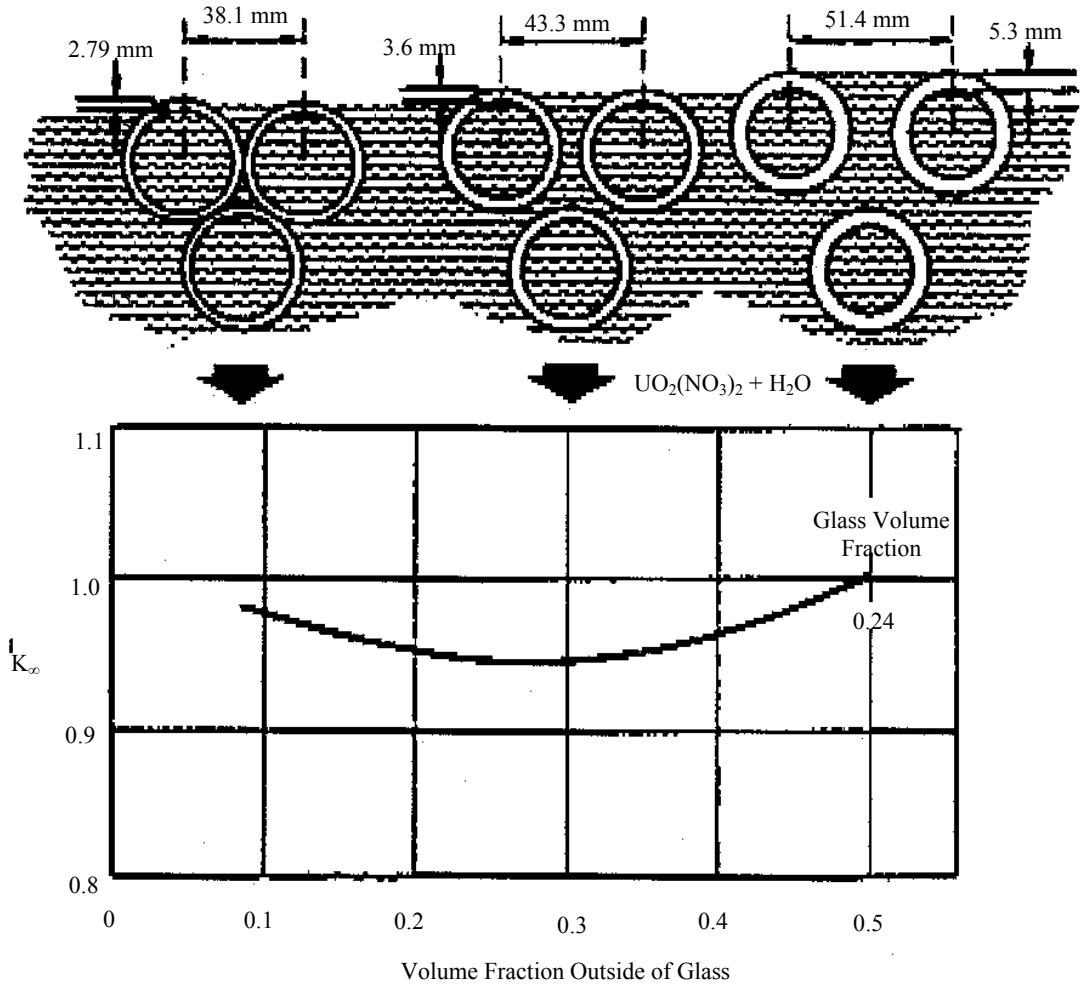


Figure 37. Calculated k_∞ vs. Volume Fraction

3. Effect on Criticality of Mixtures of Soluble Absorbers in Plutonium Solutions

The addition of neutron absorbers in soluble or fixed form can be an effective means of criticality control. Calculations have indicated that mixtures of soluble absorbers may be more effective than single solutes in criticality control.[74, 75] Calculated amounts of boron and gadolinium to reduce k_∞ of Pu + U nitrate solutions (30% Pu in U) to unity are shown in Figure 38. A mixture of two soluble absorbers, of boron and gadolinium, can be more effective than either one separately, i.e., total quantity of B + Gd less, and the mixture ratio of the absorbers can be changed to shift the effectiveness toward either lower or higher concentrations of Pu or U to obtain the most worthwhile effect.

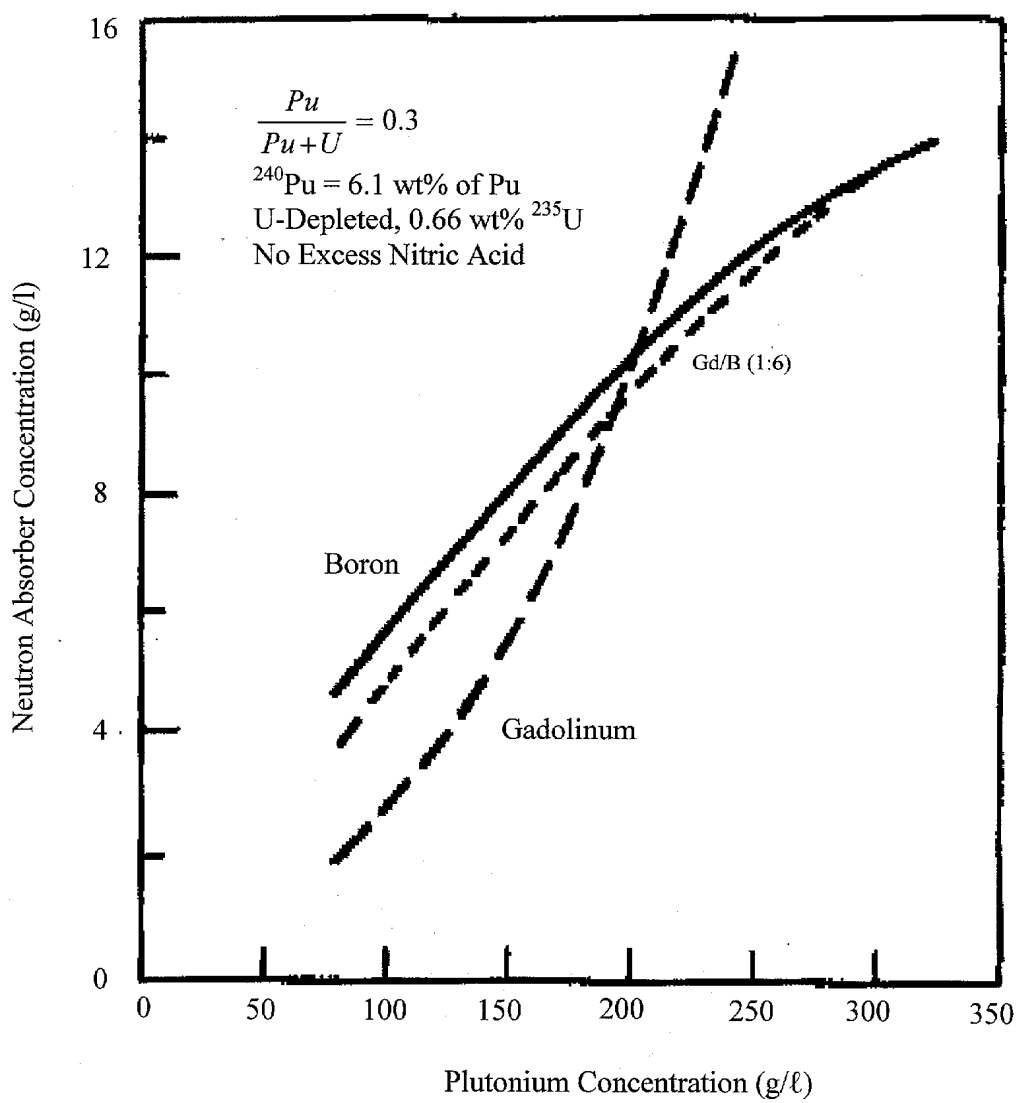


Figure 38. Absorber Concentration needed to Reduce k_∞ of Pu + U Solution to Unity

4. Effect of Boron on Criticality of Plutonium Nitrate Solutions

It has been suggested that a boron concentration required for safety of a homogeneous mixture of Pu atoms in water might not be conservative when applied to plutonium nitrate aqueous solutions.[2] At first glance, this seems to be contrary to normal behavior.

If comparisons are made between an aqueous homogeneous plutonium nitrate solution [$Pu(NO_3)_4 + H_20$] 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 captured therein. It might be presumed, erroneously, that if a sufficient quantity of soluble neutron absorbers 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 would also be subcritical. At higher plutonium concentrations, however, more boron is required for the nitrate system.

The plutonium metal water mixture should always be more reactive than plutonium nitrate. However, the nonconservative behavior does occur. Figure 39 shows the boron concentration required to poison aqueous plutonium solution to $k_\infty = 1.0$. At lower plutonium concentrations the boron content required for the metal systems is sufficient for the nitrate. This is what one would normally expect since the nitrate is an additional neutron poison. At higher plutonium concentrations, more boron is required for the nitrate systems than the metal systems. This seeming anomaly is caused by the larger volume of the nitrate molecule. At the same plutonium concentration, the nitrate solution has a smaller volume of water than the metal solution. The reduction in hydrogen content reduces the effectiveness of the boron poison and more is required.

If the comparison is 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.

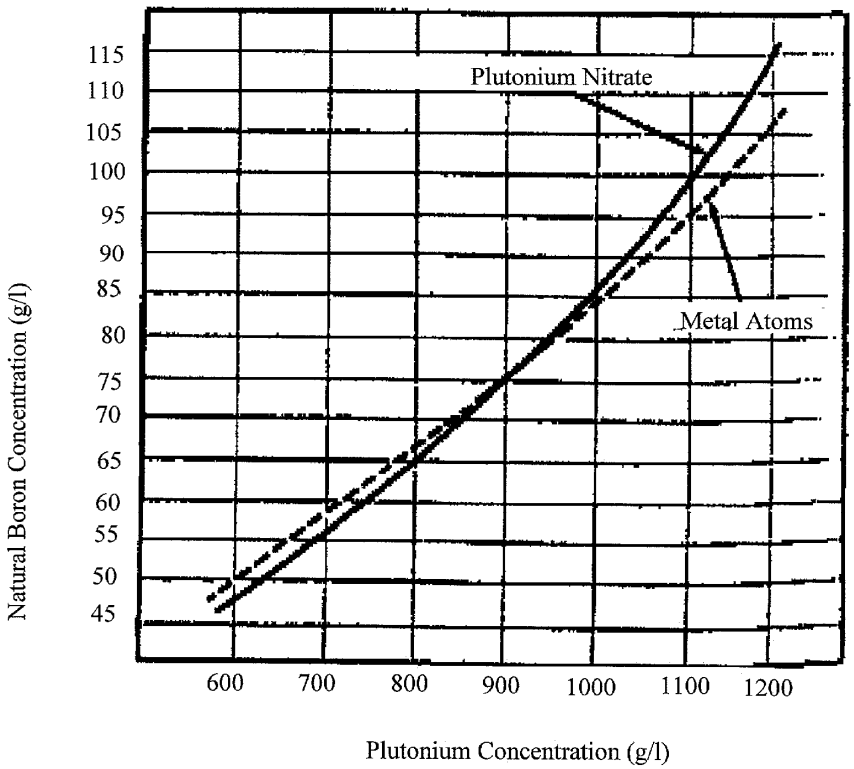


Figure 39. Quantity of Boron Required to Reduce k_∞ of Homogeneous Aqueous Pu Solutions to Unity

5. <u>Enhanced Effect of a Gadolinium Absorber on the Criticality of Plutonium-Uranium Nitrate Solutions with ^{240}Pu Content in the Plutonium</u>

An interesting anomaly (surprising result) was reported wherein the effectiveness of a soluble gadolinium absorber was significantly enhanced by the presence of ^{240}Pu and ^{238}U in a Pu + U (30% Pu) nitrate solution.[76] When the Pu contained 19% ^{240}Pu, the Gd appeared to be up to some three to four times more effective in increasing the minimum critical mass than for the case with no ^{240}Pu. A qualitative explanation for the high efficiency of 2 or 3 coexistent nuclides in the solute is the various resonance peaks that occur in the neutron cross sections of the capturing nuclei over a broad energy range. Data from the French report are shown in Figure 40.

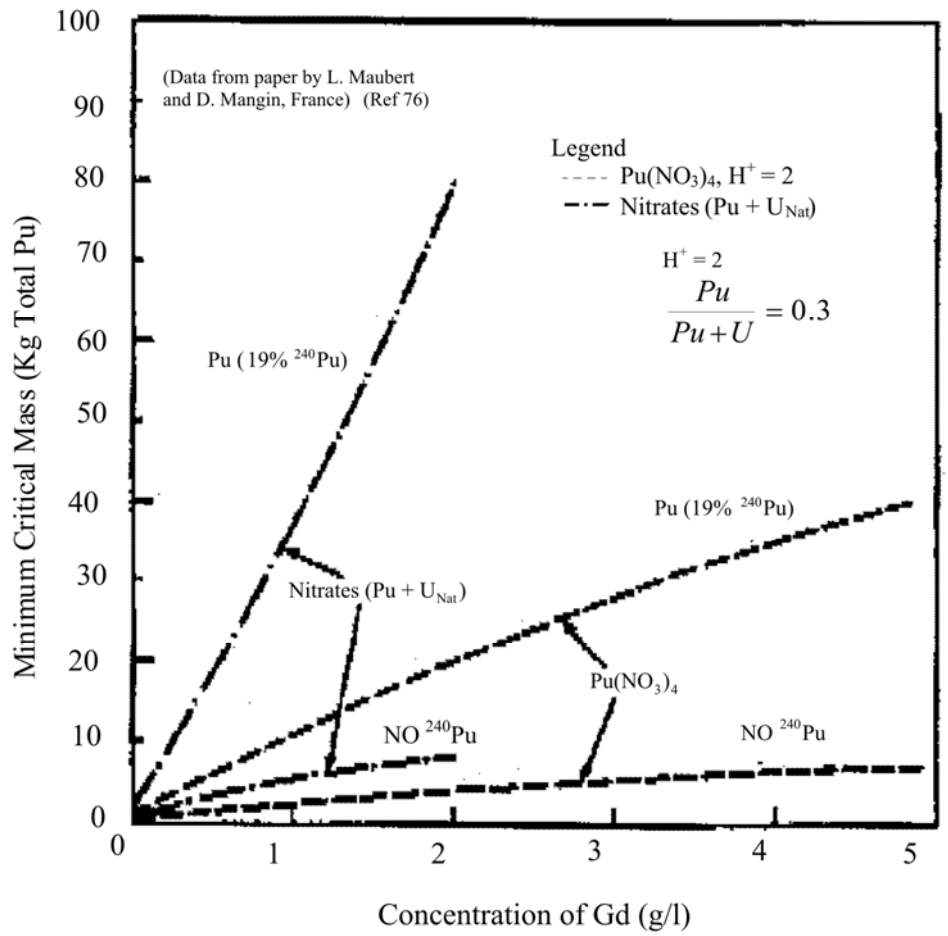


Figure 40. Calculated Minimum Critical Masses for Aqueous Solutions of Pu and U Containing Gd

6. <u>Possible Anomalous Effect of ^{240}Pu on the Minimum Critical Dimension of Mixed Oxide (Pu – Natural U) Fuel Pins in Water</u>

The American National Standard for *Nuclear Criticality and Safety of Homogeneous Plutonium-Uranium Fuel Mixtures Outside Reactors* (ANSI/ANS-8.12) is being revised to include subcritical limits on heterogeneous systems. In connection with this effort, a number of calculations were completed for heterogeneous systems of mixed fuel pins in water.[78]

Some of the calculations, however, have disclosed what may be an anomaly, and if not, then perhaps the failure of existing codes to perform certain types of calculations. The problem concerns the effect of ^{240}Pu on the dimensional limits for heterogeneous systems of mixed

oxides when the PuO_2 concentration in the mixed oxides (PuO_2 + UO_2) is at 30 wt.%. The latter represents the high end of the Pu concentration for Pu in U for which subcritical limits were provided in the revised standard. Figure 41 shows the minimum critical size for 15 wt.% PuO_2 in mixed oxide to increase as higher isotopes of Pu displace ^{239}Pu, as expected. The results in Figure 42 for 30 wt.% PuO_2, however, show the minimum critical dimension initially increases with ^{240}Pu content and then, contrary to expectation, may decrease as higher isotopes of Pu displace the ^{239}Pu.

Although this problem has not been studied in detail, a possible explanation for the phenomena is as follows. In the absence of ^{240}Pu, the minimum critical dimensions occur for the heterogeneous systems under well moderated conditions, a thermal reactor system. If the Pu content in the natural U is substantial, however, for example at 30 wt.% and the ^{240}Pu content of the Pu is as high as ~25 wt.%, the minimum critical dimension is obtained under low or essentially unmoderated conditions. Under the latter circumstances of high Pu content and a relatively fast neutron spectrum, the ^{240}Pu begins to fission in substantial quantity and contributes neutrons to the chain reaction, whereas under moderated conditions, the ^{240}Pu serves principally as a neutron absorber with little or no fission. The latter is understandable when it is considered that ^{240}Pu metal (a fissible nuclide) can be made critical by itself with a finite calculated critical mass of 33 kg for a bare sphere, a value that is less incidentally than that for ^{235}U metal.[17, 18]

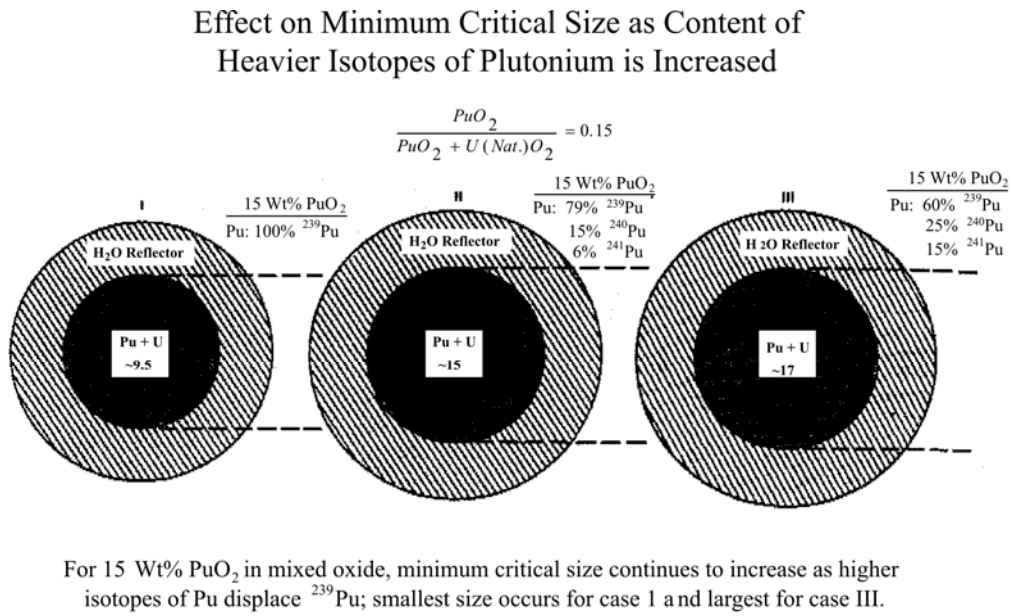


Figure 41. Effect on Minimum Critical Size as Content of Heavier Isotopes of Plutonium is Increased

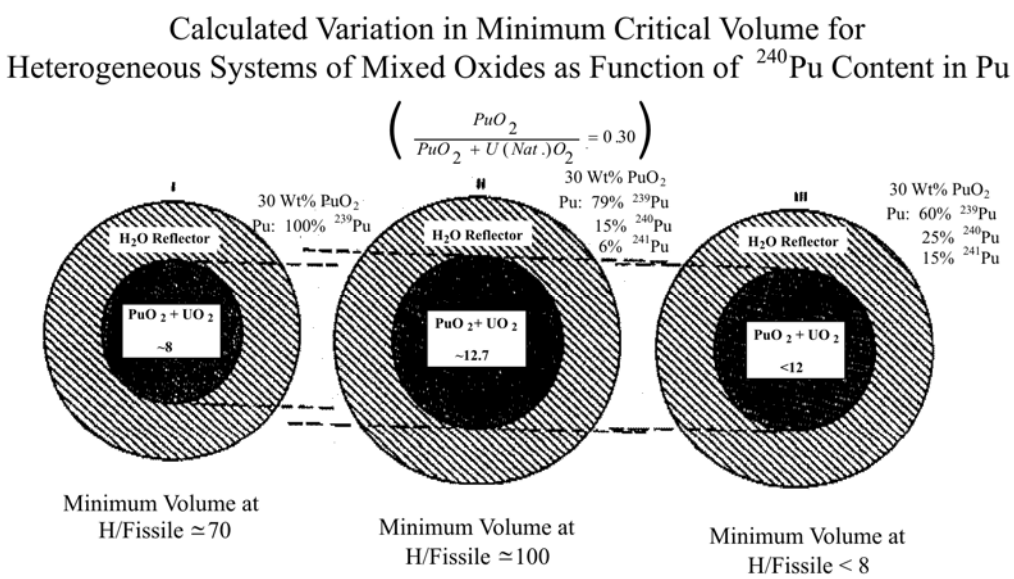


Figure 42. Calculated Variation in Minimum Critical Volume for Heterogeneous Systems of Mixed Oxides as Function of ^{240}Pu Content in Pu

7. A Condition when a Smaller Critical Mass of Pu can be Obtained with more Cadmium Neutron Absorber and Less Pu

An example is also 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.[79]

For example, at 6 grams of plutonium per liter in soil, a mixture containing 0.2 grams 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.

8. General Comments on Soluble Absorbers

This section equally might as well have been entitled, “Some Precautions on the use of Neutron Absorbers.” It has been the intent here, to summarize and briefly discuss several anomalies that pertain to the effect of neutron absorbers on criticality. Some of these deserve further study, which may be the result of inadequate cross-section data and raise questions concerning the validity of computer codes. The presence of large quantities of neutron absorbing nuclei can alter the neutronics of the system causing unexpected results. In particular, mixtures of neutron absorbers in combination with ^{240}Pu and ^{238}U can have surprising results. The nuclide, ^{240}Pu, may serve in the capacity of a resonance absorber or as a fissible nuclide depending on the energy spectrum, or degree of neutron moderation.

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

In early criticality experiments with Pu solutions, it was noted 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.[80] A copy of the chart

recording (heretofore unpublished) showing the strange variation in neutron flux with control rod movement, is shown in Figure 43. 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 expected if the rod's poison worth were small. 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.

R. NATURE OF FISSION AND THE CRITICALITY PROCESS (From Actinium to Californium and Beyond)

1. Background

During recent years, the list of actinide isotopes capable of supporting chain reactions has substantially increased.[21, 22, 25, 54, 56, 81-87]

As noted earlier, however, considering the time for chemical processing, only those nuclides with half-lives more than several weeks are of concern. Forty-six of these actinides are known to have half-lives greater than six weeks. Of these, 41 are known or believed to be capable of supporting chain reactions. These isotopes are identified in Figure 44. Figure 44 was constructed in the format used for the *Chart of the Nuclides.*[1] Figure 44 also shows the distinction between fissile and fissible nuclides that will be discussed later.

Critical configurations have been confirmed experimentally for ^{233}U, ^{235}U, and ^{239}Pu. This body of experimental data confirms that these three nuclides are fissile. Similar critical experimental data do not exist for any of the other actinide nuclides. An experimentally deduced critical mass for $^{242}_{94}$Pu metal, has, however, recently been reported:[25] $^{242}_{94}$Pu is classified as a fissile nuclide. Consequently, classification of the other actinide nuclides is conjectural, based on knowledge of cross-section data and empirical nuclear models.

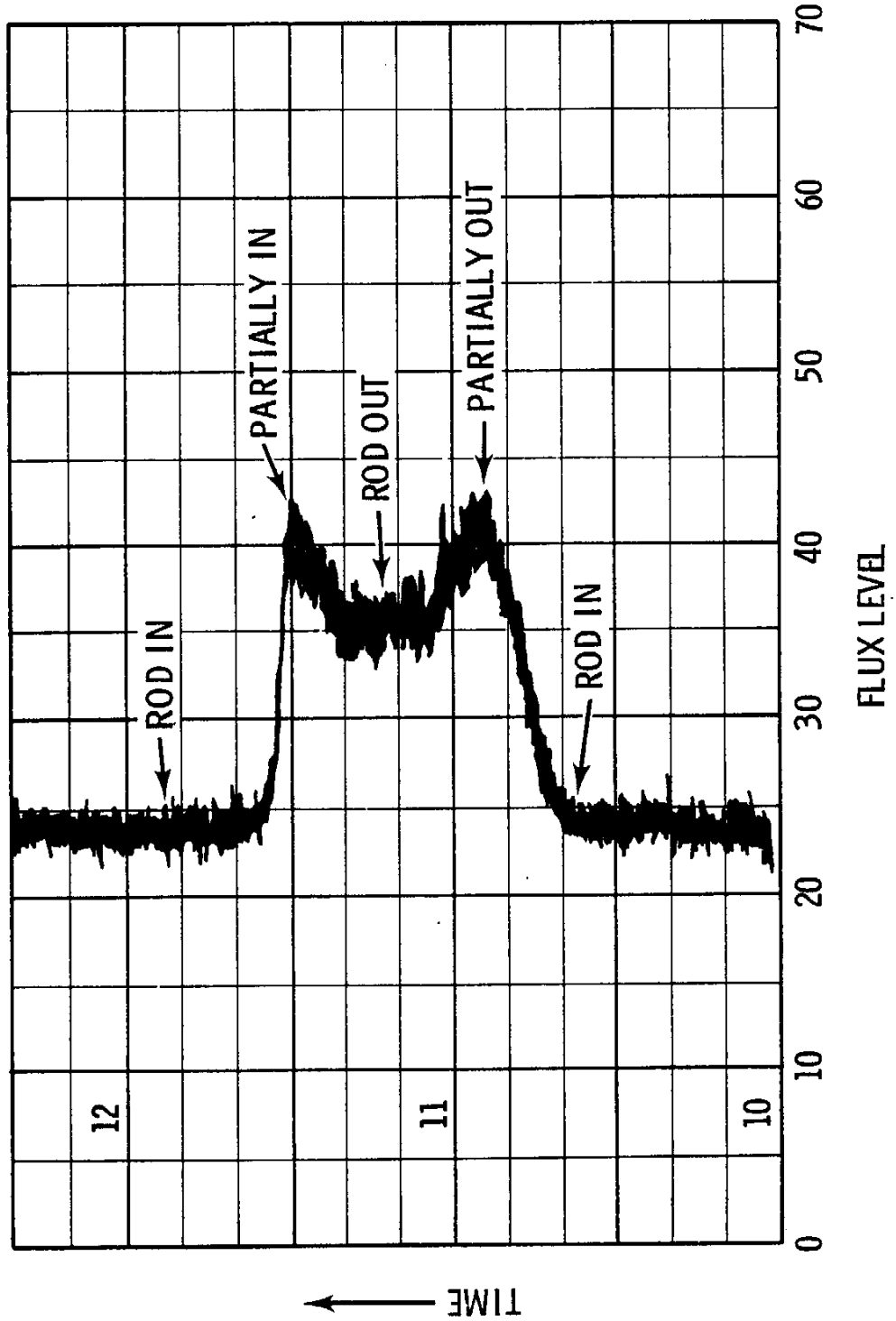


Figure 43. Control Rod Effect on Flux Level

Note 1: ^{236}Pu has an even number of neutrons and protons but the critical mass appears to be smaller for the ^{236}Pu water mixture than for the pure metal, and is therefore, shown as a fissile nuclide. [a]

Note 2: Exception to pattern: ^{229}Th has an odd neutron number but a chain reaction appears possible only with fast neutrons.

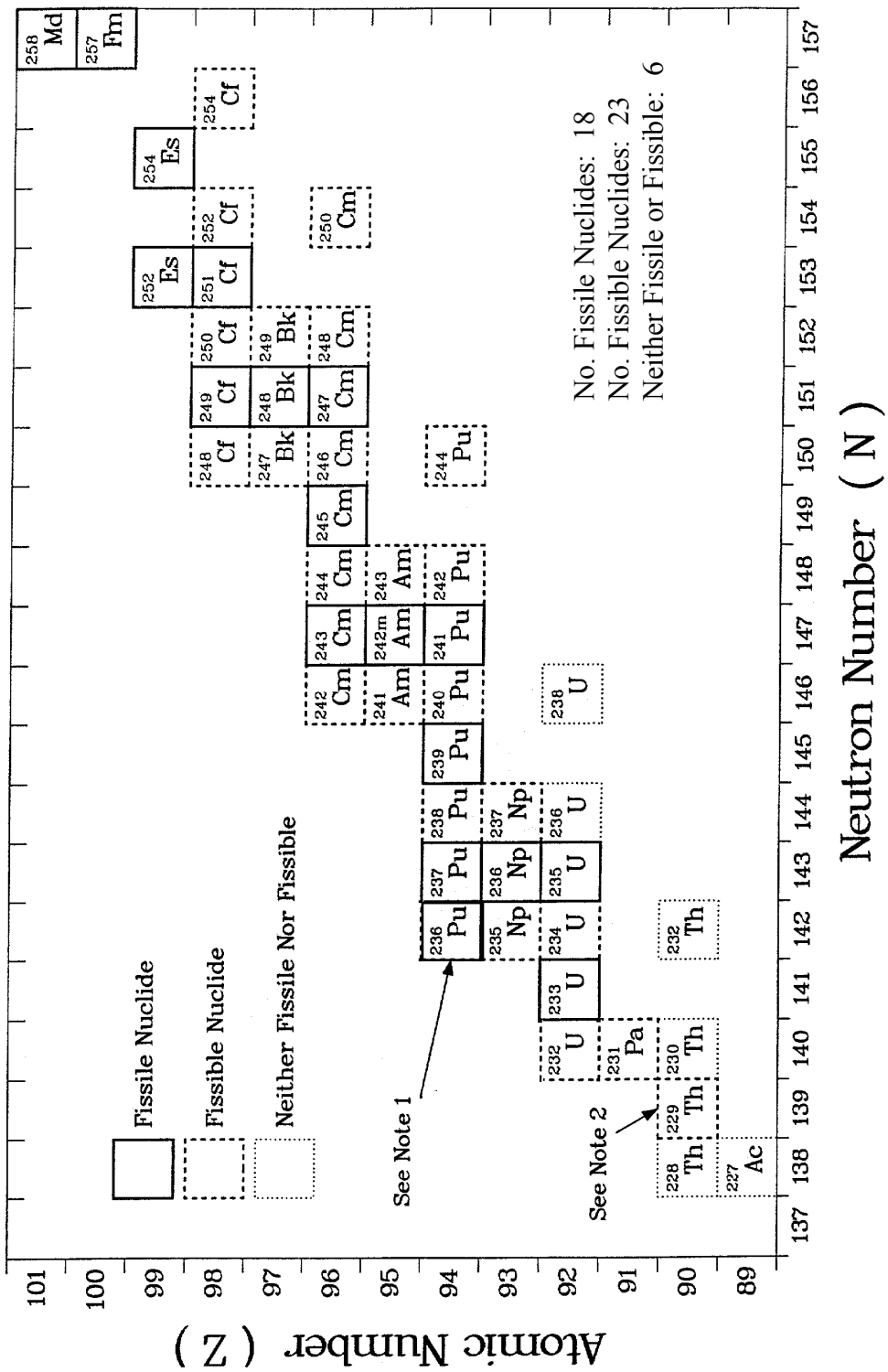


Figure 44. Chart of 46 Actinides with Half-lives Greater than 6 Weeks Identifying Fission Types

[a] as per calculations reported by C. T. Rombough, of CTR Technical Services, in May 2005 on the Fissionability of Plutonium-236.

Neutron fission, a prerequisite for a self-sustaining chain reaction in the actinides, 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.

The atom is said to be made up from "big ideas" about exceedingly small things. An artist's rendition of this concept of the atom is portrayed in Figure 45 by H. E. Krueger of Battelle – Pacific Northwest Laboratories. (Should the reader wish to indulge, he or she may actually locate the different numbers of electrons that appear at various distances from the nucleus in each of the seven shells for uranium and will note that they add up to 92, beginning with two in the innermost shell and ending with two in the outermost shell; [see Figure 46]). Since the nucleus of the atom occupies an extremely small portion of the atom's volume, the atom is mostly space. This space, however, is pervaded by powerful fields of electric force, as a consequence of the charged electrons surrounding the atom. It has been said that an atom is as much larger than the nucleus and the electrons as a large cathedral is larger than a few grains of sand. The subdivision of matter into molecules, and molecules into atoms, with the atoms in turn likened to miniature solar systems with further subdivision into other entities, brings to mind the quotation:

> "Great fleas have little fleas upon their backs do bite em and the little fleas have lesser fleas so on adinfinitem. Now the great fleas themselves in turn have greater fleas to go on while these in turn have greater still and greater still and so on." (Augustus De Morgan (1806-1871, ***The Budget of the Paradoxes***)

Classically, the atom has then been likened to a miniature solar system in which electrons orbit a heavy, dense nucleus composed of neutrons and protons. Quantum theory continues the subdivision, and the neutrons and protons are now known to be composed of quarks. Dr. Murray Gell-Mann, Cal. Tech. Theorist, first proposed quarks as the construction of neutrons and protons in 1964. Each neutron and each proton consists of a combination of 3 quarks having fractional changes of +2/3 and -1/3 that of the electron. Quarks are point entities without structure and occupy virtually no space within the volume of the neutron or proton defined by the rapidly rotating quarks as portrayed schematically in Figure 47. The size of a quark is of the order 10^{-17}cm. No free quark has ever been observed outside the nucleus of an atom nor has a fractional charge ever been observed. Table IX gives data on substructure relevant to size and densities.

Table IX. Data on Substructure Relevant to Size and Densities

Substructure	Size
Molecules	Separation Between Deuterons in D2 Molecule ~ 0.74 x 10^{-8} cm.
Chemical Atom	First Bohr Radius of Hydrogen Atom ~ 0.53 x 10^{-8} cm.
Protons and Neutrons	Radius of Proton ~ 2.8 x 10^{-13} cm.
Quarks (No Structure)	Point Entities Radius of Quarks ~ 0.5 x 10^{-17} cm.
? Higgs Boson (The "God Particle" – As Yet Undiscovered)	
Density of Uranium Nucleus	~ 130 million metric tons/cc.
Density of Quarks	~ 816 million million million metric tons/cc.
Fraction of Space Occupied by 3 Quarks in Proton	F = 0.000000000000017 (f = 1.7 x 10^{-14})
Consistency of Black Holes in space: "Quark Soup"	

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. = 2 n^2), etc. The attraction that one atom has for another is known as the valence force, wherein lies the whole basis for the field of chemistry. The source of chemical energy is the re-arrangement 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 sub-shells 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, and 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.

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.[88] These discontinuities occur [89] 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.[90] 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 sub-shell 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.

Figure 45. The Atom (A "Big Idea" About An Exceedingly Small Thing)

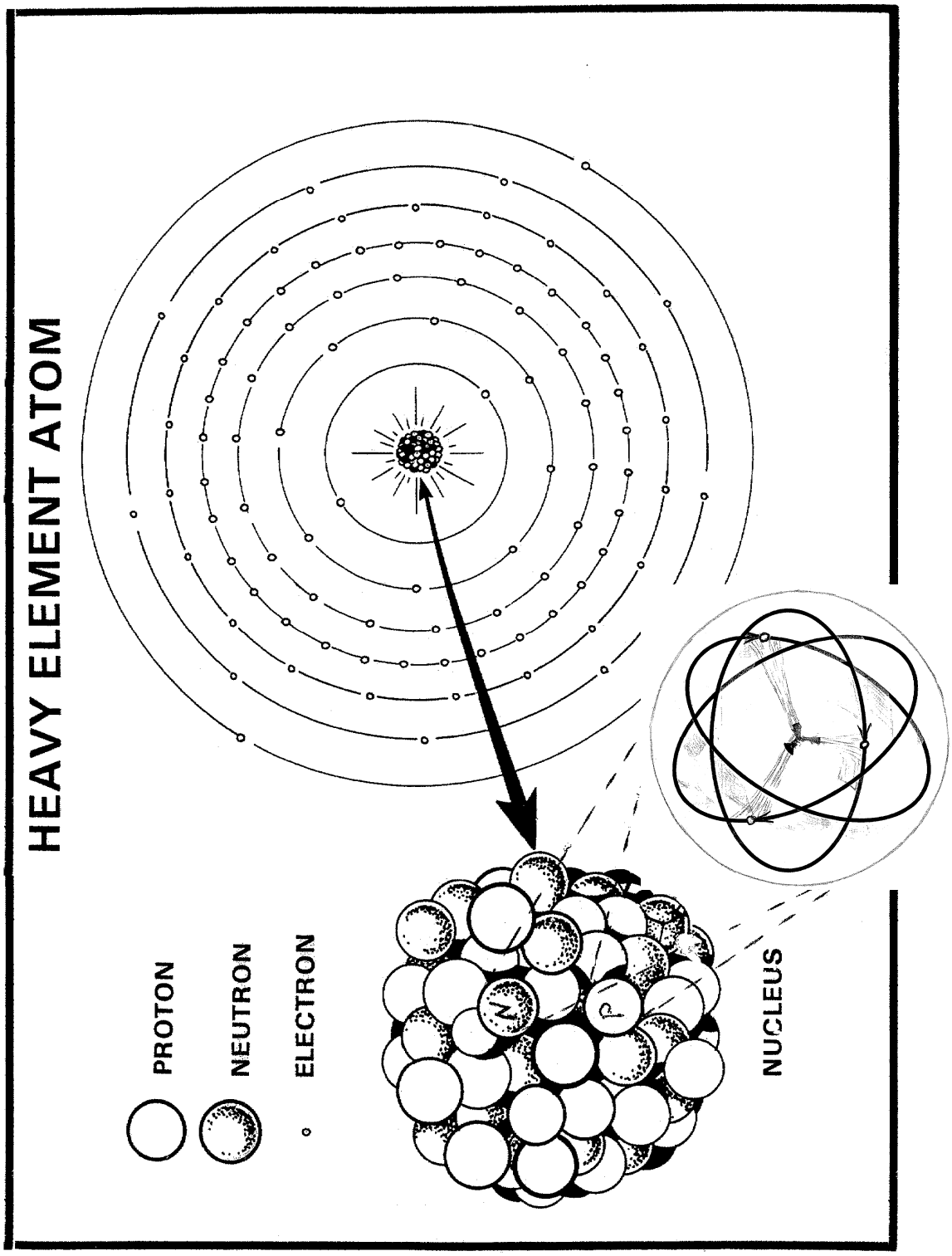


Figure 46. Heavy Element Atom (Quarks and the Nucleon)

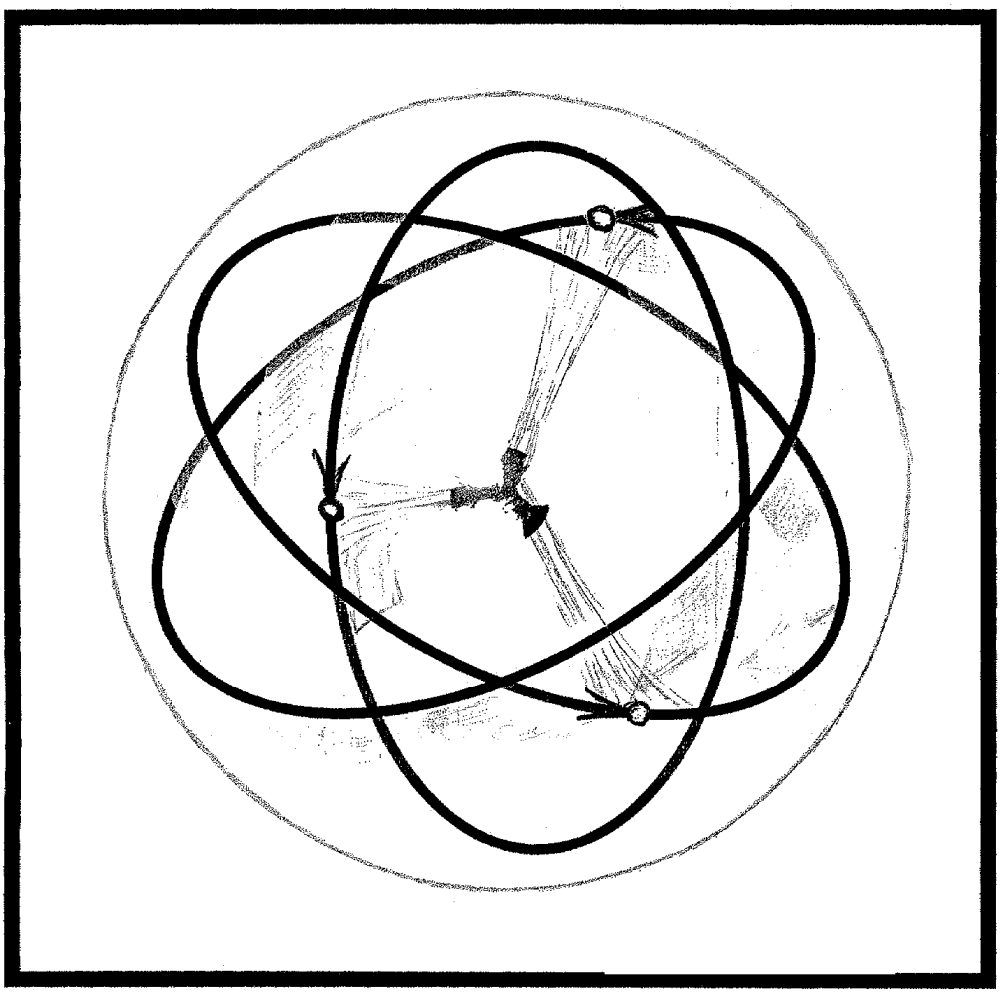


Figure 47. Nucleon (3 Point Entity Quarks)

Needless to say there are orders of magnitude differences in the quantity of energy released from chemical and nuclear reactions such as fission and fusion. On the basis that matter is condensed or frozen energy, and energy evaporated matter, some simple comparisons are given below regarding the fraction of mass actually converted to energy from chemical and nuclear reactions.

Fraction of Mass Converted to Energy
($E = mc^2$)

1.	Chemical Reactions:	One billionth – 0.000000001
2.	Fission:	One thousandth – 0.001
3.	Fusion:	One two-hundredth – 0.005
4.	Matter Annihilation:	One hundred percent – 1.0

The consequence of this is that nuclear reactions generate small quantities of wastes and tiny volumes of pollutants, per energy produced, relative to chemical reactions. A one GW electrical generating nuclear power plant requires about 30 tonnes of 3 - 4% enriched uranium per year.

By comparison, a one GW electrical generating coal fixed plant consumes some 6,050 tonnes of coal per day – but only about 0.013 kg, a mere 1/2 of an ounce, of all of that coal is actually converted into electrical energy on the basis of $E = MC^2$. A one GW electrical coal plant releases some 17,800 tonnes of greenhouse emissions (CO_2) into the atmosphere daily along with the emissions of sulfur dioxide and nitrogen oxides attendant therewith; and depending on the quality of coal, up to 2,700 tonnes of ash is formed daily (up to some one million tonnes yearly). This ash contains hundreds of tonnes of toxic poisonous metals such as, arsenic, cadmium, lead and mercury. These elements are not radioactive and do not decay away!

2. Considerations on Fissioning and Stability of Actinides

In reviewing the *Chart of the Nuclides,*[1] it is noted that stable isotopes with even atomic numbers are about 50% more prevalent than any other kind, and hence, must be more stable. Pertaining to the actinide elements, however, it is noted that even-even actinides have spontaneous fission rates that are greater, on average, by a factor of about 10^3 over spontaneous fission rates in even-odd and odd-even isotopes, and greater by a factor of some 10^5 above spontaneous fission rates in odd-odd isotopes.

Based on the observed spontaneous fission rates, an empirical formula was developed some years ago by R. Vandenbosch and G. T. Seaborg that can be used to calculate the neutron activation energy for fission.[90] Using this formula, activation energies for fission for the various classes of actinides have been calculated vs. Z^2/A (the fissionability parameter) and are shown in Figure 48.[20] Once energy has been imparted to the nucleus, competition sets for various modes of de-excitation. The fission barrier height represents a fission time of 10^{-21} seconds. Because of the barrier penetration, nature of the fission process induced fission will, however, be observed at an energy below the barrier. The activation energy is taken to be 0.9 MeV less than the barrier height wherein the time for fission becomes comparable to the time for de-excitation by gamma emission, or in a time of 10^{-14} seconds.

Taken as a class, the activation energy is the least for the even-even nuclides and greatest for the odd-odd nuclides. Note that the "activation" energy required for fission is <u>less</u> for ^{238}U than for ^{235}U. It is also less for fission of ^{240}Pu than for ^{239}Pu, etc., but in this case, it is the two odd-N nuclides that will support thermal neutron chain reactions.

In the case of fission induced by neutrons, it is the compound nucleus (nucleus plus neutron) that undergoes fission. The binding energy available on capture of a neutron into an even-N or odd-N nuclide must be considered. Adding a neutron to an odd-N actinide isotope releases, on average, more than 1 MeV of energy than if the same neutron were added to an even-N actinide. For example, if a neutron is added to ^{239}Pu, which has an odd number of neutrons, the nuclide becomes ^{240}Pu, which requires less energy for fission as the compound nucleus and then has an even number of protons and neutrons. Note that the activation energies given in Table X are for the fissioning of the compound nucleus that is formed on absorption of the neutrons (for example: activation energy listed for ^{239}Pu in Table X is that for ^{240}Pu, and the neutron binding energy is the separation energy from ^{240}Pu).

It is somewhat of an anomaly, then, that in terms of fission, the heavy even-even actinide elements make up the least stable isotopes, but it is the odd-N nuclides that present criticality problems with thermal neutrons.

It is solely due to the fact that the neutron serves as the chain carrier that thermal neutron fission can occur at all in odd-N isotopes. The neutron not only provides the activation

energy needed to induce fission (from the binding energy on capture), but in the process converts the odd-N isotope to an even-N isotope that requires less energy for fission.

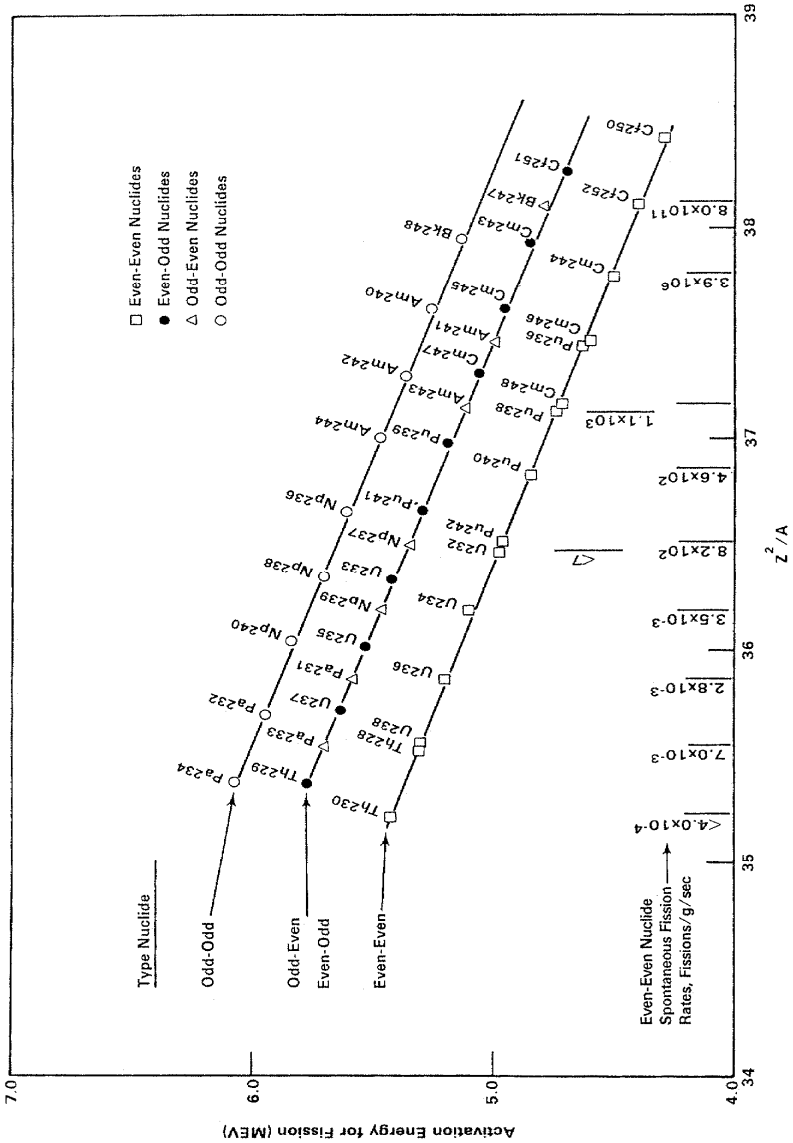


Figure 48. Calculated Activation Energy for Fission vs. Z^2/A

Calculated or measured critical masses, now available for a number of actinides, also are tabulated in Table X, and the sources for these critical mass values identified.

Table X. Neutron Fissionability and Criticality

Nuclide	Type [A)]	Z^2/A	E_a [B)] (MeV)	B_n [C)] (MeV)	B_n-E_a) (MeV)	Projected Criticality Aspects		Computed or Measured Critical Masses of Solution Spheres[D)]; Thermal Systems at Optimum Moderation (General Ref. a, c, d, g, h, p-s)		Computed or Measured Critical Masses of Metal Spheres; Fast, Unmoderated Systems (General Ref. a, c, d, g, h, p-s)		
						Slow Neutron Chain Reaction	Fast Neutron Chain Reaction	Bare (kg)	Water Reflected (kg)	Bare (kg)	Water Reflected (kg)	Steel Reflected (kg)
$^{227}_{89}Ac$	Odd-Even	34.89	6.29	5.04	-1.25	No	No	-----	-----	---	-----	-----
$^{227}_{90}Th$	Even-Odd	35.68	5.21	7.13	1.92	Yes	Yes	-----	-----	-----	-----	-----
$^{228}_{90}Th$	Even-Even	35.53	5.77	5.24	-0.53	No	No	-----	-----	-----	-----	-----
$^{229}_{90}Th$	Even-Odd	35.37	5.42	6.8	1.37	No	Yes	-----	-----	-----	-----	-----
$^{230}_{90}Th$	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	-----	-----	-----	-----	-----
$^{230}_{91}Pa$	Odd-Odd	36.00	5.595	6.818	1.22	Yes	Yes	-----	-----	-----	-----	-----
$^{231}_{91}Pa$	Odd-Even	35.85	5.94	5.6	-0.34	No	Yes	-----	-----	~189±50[(a)]	-----	65.9[(n)]
$^{232}_{91}Pa$	Odd-Odd	35.49	5.7	6.5	0.8	Yes	Yes	-----	-----	-----	-----	-----
$^{233}_{91}Pa$	Odd-Even	35.54	6.06	5.2	-0.86	No	No	-----	-----	-----	-----	-----
$^{231}_{92}U$	Even-Odd	36.64	4.97	7.27	2.30	Yes	Yes	-----	-----	-----	-----	-----
$^{232}_{92}U$	Even-Even	36.48	5.4226	5.743	0.3204	Yes	Yes	-----	4.6[(b2)]	3.70[(b1)]	2.25[(b2)]	-----
$^{233}_{92}U$	Even-Odd	36.33	5.1	6.8	1.7	Yes	Yes	~1.2[(c)]	0.55[(c)]	15.4[(d)]	9.2[(d)]	-----
$^{234}_{92}U$	Even-Even	36.17	5.53	5.31	-0.23	No	Yes	-----	-----	148[(a)]	137[(a)]	89[(a)]
$^{235}_{92}U$	Even-Odd	36.02	5.2	6.5	1.3	Yes	Yes	1.5[(c)]	0.8[(c)]	49.0[(d)]	22.8[(d)]	-----

Nuclide	Type [A)]	Z^2/A	E_a [B)] (MeV)	B_n [C)] (MeV)	B_n-E_a) (MeV)	Projected Criticality Aspects		Computed or Measured Critical Masses of Solution Spheres[D)]; Thermal Systems at Optimum Moderation			Computed or Measured Critical Masses of Metal Spheres; Fast, Unmoderated Systems	
								(General Ref. a, c, d, g, h, p-s)				
						Slow Neutron Chain Reaction	Fast Neutron Chain Reaction	Bare (kg)	Water Reflected (kg)	Bare (kg)	Water Reflected (kg)	Steel Reflected (kg)
$^{236}_{92}U$	Even-Even	35.86	5.64	5.13	-0.52	No	No	(Calc. k_∞ for metal ~0.8)				
$^{237}_{92}U$	Even-Odd	35.71	5.297	6.143	0.846	No	Yes	-----	-----	193[e]	-----	-----
$^{238}_{92}U$	Even-Even	35.56	5.7	4.8	-0.9	No	No	-----	-----	-----	-----	-----
$^{235}_{93}Np$	Odd-Even	36.804	5.607	5.69	0.084	No	Yes	-----	-----	65.8[a]	59.7[a]	39.5[a]
$^{236}_{93}Np$	Odd-Odd	36.648	5.362	6.62	1.257	Yes	Yes	-----	-----	-----	-----	-----
$^{237}_{93}Np$	Odd-Even	36.49	5.7	5.5	-0.2	No	Yes	-----	-----	57.0[a]	53.0[a]	35.3[a]
$^{238}_{93}Np$	Odd-Odd	36.34	5.47	6.23	0.755	Yes	Yes	-----	-----	-----	-----	-----
$^{239}_{93}Np$	Odd-Even	36.19	5.83	5.17	-0.66	No	Yes	~105[f] (k_∞ for metal near unity)				-----
$^{236}_{94}Pu$	Even-Even	37.44	5.0216	5.859	0.837	Yes	Yes	-----	~1.2[b]	8.21[b1]	3.3[b]	-----
$^{237}_{94}Pu$	Even-Odd	37.28	4.74	7.0	2.26	Yes	Yes	-----	-----	-----	-----	-----
$^{238}_{94}Pu$	Even-Even	37.13	5.1904	5.656	0.523	No	Yes	-----	-----	7.1[g]	6.1[g]	4.2[g]
$^{239}_{94}Pu$	Even-Odd	36.97	4.8459	6.4	1.554	Yes	Yes	0.90[c]	0.50[c]	10.0[d]	5.24[d]	-----
$^{240}_{94}Pu$	Even-Even	36.82	5.3010	5.24	-0.005	No	Yes	-----	-----	33[g]	29[g]	19[g]
$^{241}_{94}Pu$	Even-Odd	36.66	5.0	6.3	1.3	Yes	Yes	-----	0.244[h]	12.4[i]	5.8[i]	5.9[i]
$^{242}_{94}Pu$	Even-Even	36.51	5.4	5.0	-0.37	No	Yes	-----	-----	85[a] (80±1)[E)]	77.6[a]	49.8[a]
$^{244}_{94}Pu$	Even-Even	36.21	5.52	4.72	-0.797	No	Yes	-----	-----	~360[j]	-----	-----
$^{239}_{95}Am$	Odd-Even	37.76	5.27	5.94	0.67	No	Yes	-----	-----	-----	-----	-----

Nuclide	Type [A)]	Z^2/A	E_a [B)] (MeV)	B_n [C)] (MeV)	B_n-E_a) (MeV)	Projected Criticality Aspects		Computed or Measured Critical Masses of Solution Spheres[D)]; Thermal Systems at Optimum Moderation (General Ref. a, c, d, g, h, p-s)			Computed or Measured Critical Masses of Metal Spheres; Fast, Unmoderated Systems (General Ref. a, c, d, g, h, p-s)	
						Slow Neutron Chain Reaction	Fast Neutron Chain Reaction	Bare (kg)	Water Reflected (kg)	Bare (kg)	Water Reflected (kg)	Steel Reflected (kg)
$^{240}_{95}Am$	Odd-Odd	37.60	5.02	6.66	1.64	Yes	Yes	-----	-----	~12[(j)]	-----	-----
$^{241}_{95}Am$	Odd-Even	37.54	5.4	5.5	0.1	No	Yes	-----	-----	107.6[(a)]	97[(a)]	63[(a)]
$^{242}_{95}Am$	Odd-Odd	37.29	5.13	6.4	1.3	Yes	Yes	-----	0.019[(h)]	8.88[(k)]	3.21[(k)]	3.4[(k)]
$^{243}_{95}Am$	Odd-Even	37.14	5.5	5.4	-0.1	No	Yes	-----	-----	153[(g)]	~138[(l)]	96[(g)]
$^{244}_{95}Am$	Odd-Odd	36.99	5.24	6.05	0.808	Yes	Yes	-----	-----	-----	-----	-----
$^{242}_{96}Cm$	Even-Even	38.08	4.85	5.70	0.86	No	Yes	-----	-----	15.4[(l)]	10.4[(l)]	-----
$^{243}_{96}Cm$	Even-Odd	37.93	4.5	6.8	2.3	Yes	Yes	-----	0.122[(h)]	9.72[(l)]	3.35[(l)]	3.28[n4)]
$^{244}_{96}Cm$	Even-Even	37.77	5.0	5.51	0.51	No	Yes	-----	-----	13.5[(g)]	11.5[(g)]	7.6[(g)]
$^{245}_{96}Cm$	Even-Odd	37.62	4.6	6.5	1.9	Yes	Yes	-----	0.041[(g)]	12.4[(m)]	3.59[(m)]	4.8[(m)]
$^{246}_{96}Cm$	Even-Even	37.46	5.1	5.16	0.06	No	Yes	-----	-----	70[(m)]	58.6[(m)]	38.7[(m)]
$^{247}_{96}Cm$	Even-Odd	37.31	4.7	6.2	1.5	Yes	Yes	-----	2.05[(h)]	7.25[(m)]	3.01[(m)]	3.15[(m)]
$^{248}_{96}Cm$	Even-Even	37.16	5.18	4.71	-0.463	No	Yes	-----	-----	-----	-----	-----
$^{250}_{96}Cm$	Even-Even	36.86	5.28	~4.3	~-0.96	No	Yes	-----	-----	-----	-----	-----
$^{252}_{96}Cm$	Even-Even	36.57	5.39	~3.9	~-1.47	No	Yes	-----	-----	-----	-----	-----
$^{246}_{97}Bk$	Odd-Odd	38.25	4.79	6.81	2.02	Yes	Yes	-----	-----	-----	-----	-----
$^{247}_{97}Bk$	Odd-Even	38.09	5.14	5.56	0.418	No	Yes	-----	-----	-----	-----	-----
$^{248}_{97}Bk$	Odd-Odd	37.94	4.90	6.22	1.32	Yes	Yes	-----	-----	-----	-----	-----

Nuclide	Type [A)]	Z^2/A	E_a [B)] (MeV)	B_n [C)] (MeV)	B_n-E_a) (MeV)	Projected Criticality Aspects		Computed or Measured Critical Masses of Solution Spheres[D)]; Thermal Systems at Optimum Moderation (General Ref. a, c, d, g, h, p-s)			Computed or Measured Critical Masses of Metal Spheres; Fast, Unmoderated Systems (General Ref. a, c, d, g, h, p-s)	
						Slow Neutron Chain Reaction	Fast Neutron Chain Reaction	Bare (kg)	Water Reflected (kg)	Bare (kg)	Water Reflected (kg)	Steel Reflected (kg)
$^{249}_{97}Bk$	Odd-Even	37.79	5.25	4.97	-0.282	No	Yes	-----	-----	-----	-----	-----
$^{249}_{98}Cf$	Even-Odd	38.57	4.3	6.6	2.3	Yes	Yes	-----	0.070[(h)]	7.8[(n)]	-----	-----
$^{250}_{98}Cf$	Even-Even	38.42	4.73	5.114	0.38	No	Yes	-----	-----	-----	6.6[(o)]	3.73[(o)]
$^{251}_{98}Cf$	Even-Odd	38.26	4.4	6.17	1.77	Yes	Yes	-----	0.022[(h)]	8.3[(n)]	-----	-----
$^{252}_{98}Cf$	Even-Even	38.11	4.83	4.793	-0.04	Yes	Yes	-----	-----	-----	5.1[(o)]	3.50[(o)]
$^{254}_{98}Cf$	Even-Even	37.66	4.96	4.46	-0.50	No	Yes	-----	-----	-----	-----	-----
$^{252}_{99}Es$	Odd-Odd	38.89	4.55	6.22	1.67	Yes	Yes	-----	-----	-----	-----	-----
$^{254}_{99}Es$	Odd-Odd	38.59	4.66	5.98	1.32	Yes	Yes	-----	~0.029[(a)]	5.7[(f)]	-----	-----
$^{257}_{100}Fm$	Even-Odd	38.91	4.146	~5.90	~1.85	Yes	Yes	-----	-----	-----	-----	-----
$^{258}_{101}Md$	Odd-Odd	39.54	4.321	~5.90	~1.58	Yes	Yes	-----	-----	-----	-----	-----

Footnotes:

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. p). Activation energy is for the fissioning of the compound nucleus that is formed on absorption of the neutrons. (Example: activation energy listed for ^{239}Pu is that for ^{240}Pu, and the neutron binding energy is the separation energy of the neutron from ^{240}Pu.)
C) B_n = neutron binding energy for nuclide of mass A + 1 (Ref. p).
D) Minimum mass for homogeneous aqueous solutions.
E) Measured value for ^{242}Pu metal at LANL; private communication from Roger Brewer at ANS-8.15 meeting, Chelan, Washington, September 6, 1997; (density of ^{242}Pu: 20.059 g/cc).

References for Table X

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(b) C. T. Rombough, CTR Technical Services Inc., "Fissionability of Plutonium-236," (May 2005); (b1) Water reflected values for Pu-236 and U-232 from July 28, 1997 letter; and (b2) bare metal values for Pu-236 and U-232 from October 1997 letter.
(c) N. L. Pruvost and H. C. Paxton, *Nuclear Safety Guide*, LA-12808, Los Alamos National Laboratory, Los Alamos, New Mexico (1996),
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(h) H. K. Clark, "Subcritical Limits for Special Fissile Actinides," Nuclear Technology, Vol. 48, p. 164 (1980).
(i) E. D. Clayton, Critical Masses for Actinide Elements, letter to W. C. Cliff from E. D. Clayton, Battelle PNL, unpublished (March 20, 1995).
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(p) R. Vandenbosch and F. T. Seaborg, Considerations on the Probability of Nuclear Fission, The Physical Review, Vol. 110, p. 507 (1958).
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(r) V. Sviridov and B. Ryazanon, "Systematics of Criticality Actinide Isotopes with Even-N Nuclides," Proceedings of the International Conference on Nuclear Criticality Safety, Oxford, United Kingdom, Vol. III, pp. 25-30 (September 9-13, 1991).
(s) American National Standard for Nuclear Criticality Control of Special Actinide Elements, ANSI-ANS-8.15, American Nuclear Society, La Grange Park, Illinois (1981).

3. Fissile vs. Fissible

Since the criticality aspects for the various actinide isotopes differ significantly from one another, a term was proposed in nuclear engineering to help distinguish differences.[18] Fissible nuclide means:

> "A nuclide that cannot support a slow-neutron chain reaction but is only capable of a fast-neutron chain reaction, provided the 'effective' fast-neutron production cross section $\overline{\gamma\sigma_f}$ exceeds the 'effective' fast-neutron removal cross section."

Almost without exception, all nuclides classified as "fissible" contain even numbers of neutrons. ($^{229}_{90}Th$ and $^{237}_{92}U$, odd-N nuclides are possible exceptions, since a chain reaction appears likely only with fast neutrons.) Fissile nuclides may have even or odd atomic numbers. Fissile nuclides are known to be capable of supporting thermal neutron chain reactions as well as fast. Most fissile nuclides have odd numbers of neutrons, with the possible exception of $^{232}_{92}U$, $^{236}_{94}Pu$ and $^{252}_{98}Cf$ (see note on Figure 44). No fissile nuclide is known, however, to have an odd atomic number and even neutron number. Fissible nuclides characteristically exhibit rather sharp fission thresholds in their fission cross sections in the 500-KeV to 2-MeV neutron energy range, with little or no probability for sub-threshold fission at thermal neutron energies. The fission cross section vs. energy is shown for five fissible nuclides in Figure 49. At thermal neutron energy the capture cross section for the n,γ reaction predominates over that for n, f, and little fission can be expected, but in the region of the fission threshold the reverse occurs.

Figure 50 is a plot of the fission cross sections for two common fissile nuclides, ^{235}U and ^{239}Pu, and two of the fissible nuclides, ^{237}Np and ^{241}Am. Note that the fast fission cross section for ^{241}Am above the fission threshold is substantially higher than that of the fissile nuclide ^{235}U. The fast fission cross section for ^{243}Am, not shown in the figure, also exceeds that of ^{235}U and ^{237}Np above 1 MeV. The effect of moderation is to slow down the neutrons below the fission threshold, where neutron capture predominates. Water, therefore, serves as a strong criticality deterrent when mixed with a fissible nuclide.[82] In the case of fissible nuclides, the minimum critical mass occurs for unmoderated material, whereas, as is well known, the minimum critical mass for fissile nuclides occurs for dilute aqueous solutions, or well moderated systems (except for "small mass" concepts involving special circumstances).[91, 92] Aqueous solutions containing

fissible nuclides present no criticality problems provided there are no highly fissile nuclides present in the solution.

For fissible nuclides, Clayton and Bierman (1971) estimated the hydrogen to actinide atom ratio needed to reduce the infinite neutron multiplication factor (k_∞) to approximately 1.0. Table XI presents the results of this estimation for five selected nuclides. Estimates of this kind were not included in ANS-8.15,[19] due to the expected difficulty on the part of plant personnel in knowing and controlling the H/X ratio at such low values.

Table XI. Estimated Limiting Critical Densities of Five Fissible Nuclides in Uniform Aqueous Mixtures

Nuclide (X)	Density of X (g/cm^3)	Hydrogen-to-Nuclide Atom Ratio (H/X)	K_∞[a]
^{237}Np	~12.7	~0.80	~1.0
^{238}Pu	~5.1	~3.80	~1.0
^{240}Pu	~17.3	~0.18	~1.0
^{241}Am	~7.6	~1.25	~1.0
^{244}Cm	~6.7	~2.00	~1.0

(a) Values larger than the H/X ratios given are expected to result in k_∞ less than unity.

Some of the highly fissile actinides have very large fission cross sections at thermal neutron energies relative to ^{235}U or ^{239}Pu. Consequently, they have minimum critical masses that are much smaller than those for either ^{235}U or ^{239}Pu. For example, the thermal fission cross section for ^{242}Am is ~6600 b, for ^{245}Cm, ~2020 b, and for ^{251}Cf, ~4300 b, whereas the fission cross section for ^{239}Pu at thermal energy is 742 b. A computed minimum critical mass for an aqueous homogeneous solution of ^{245}Cm in a water–reflected sphere is 41 g and that for ^{251}Cf only 22 g.[83] This compares with the minimum critical mass for ^{239}Pu of about 510 g. Perhaps contrary to expectation, a very small minimum critical mass under moderated conditions does not necessarily translate into a relatively small critical mass for the metal or oxide under a fast (unmoderated) condition.

As noted earlier an anomaly is considered as something not in keeping with expected notions of fitness and order. In referring to Figure 44, it should be noted that for the isotopes with half-lives of six weeks or more, the majority of isotopes (some 23) are fissible (principally

fast neutron chain reactions), whereas, only 18 are classed as fissile (fast and/or thermal chain reactions). This is contrary to the “early day” expectations of the past.

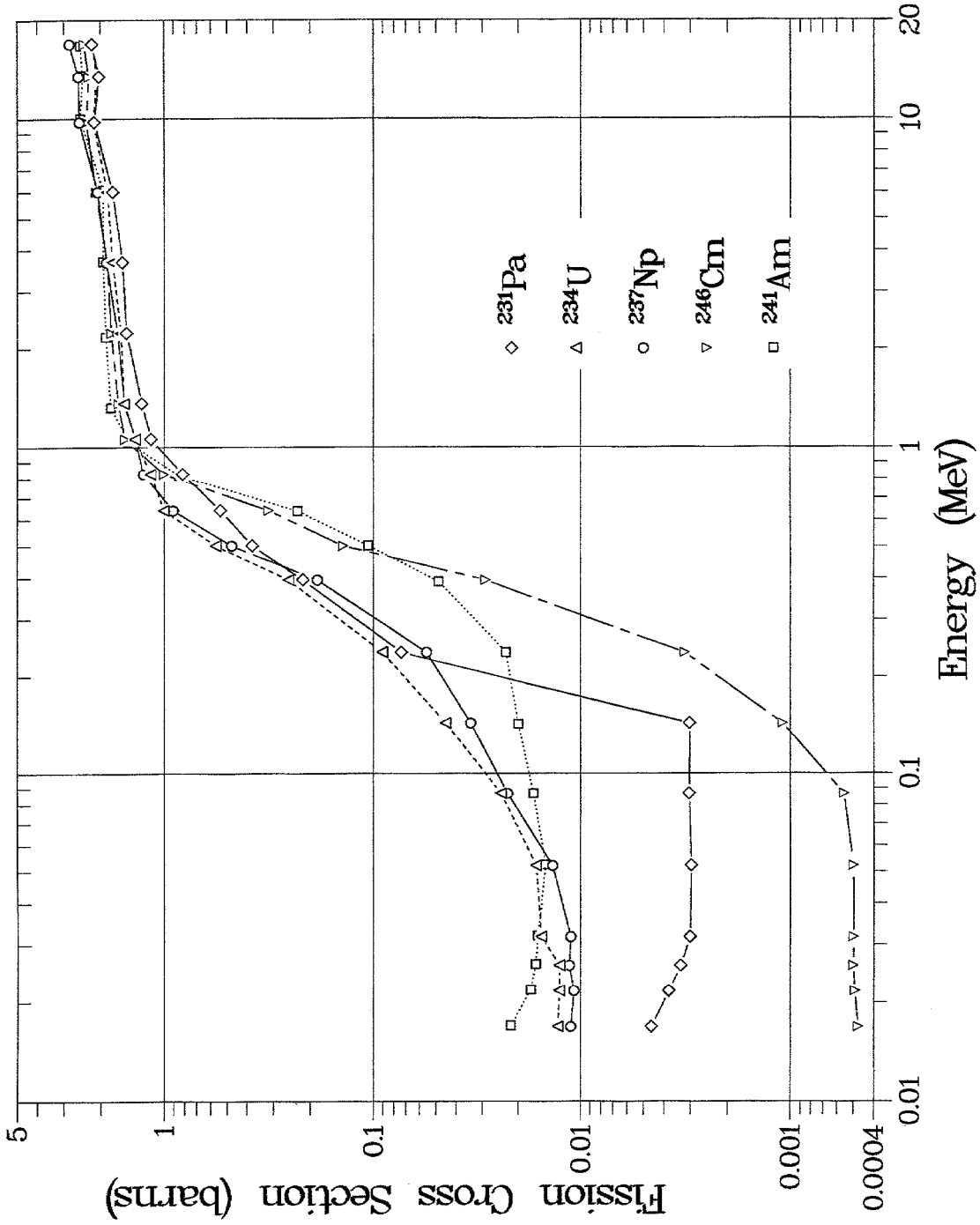


Figure 49. Fission Cross Sections of Five Fissible Nuclides

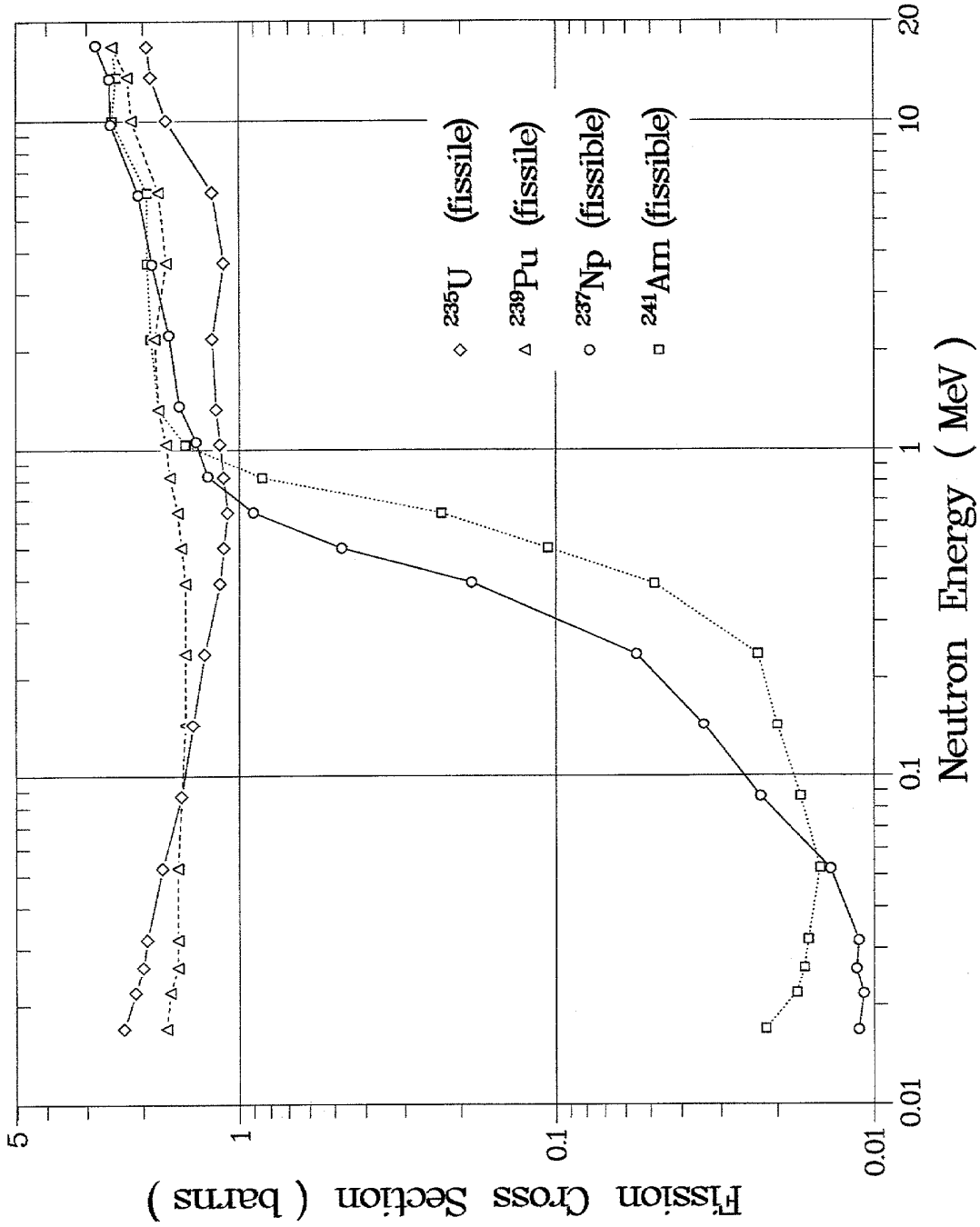


Figure 50. Comparison of Fission Cross Sections for Two Fissile and Two Fissible Nuclides

4. Correlation of Minimum Critical Masses for Fissile Nuclides in Aqueous Solution

Small quantities of actinide elements may be produced from time to time for research purposes in nuclear physics and medicine. The fact that the quantity of the nuclide being handled may seem to be relatively small does not preclude the requirement of a criticality safety review.

For example, Clayton was requested to evaluate the nuclide ^{254}Es in 1987.[93] At that time, evaluated cross sections were not available for calculation of the minimum critical mass for the rare einsteinium nuclide in an aqueous solution. To provide an estimate of its minimum critical mass, Clayton found a reasonable correlation to exist between the "known" minimum critical masses (calculated or measured) of fissile nuclides when plotted against $\nu\sigma_f$ on a log-log scale. This correlation is shown in Figure 51. The equation of the line is $\ln (Mc) = -1.32 \ln (\nu\sigma_f) + 15.8$ M_c is the critical mass in grams, σ_f is the thermal fission cross section, and ν is the neutrons released per fission at thermal energy.

From Figure 51, the critical mass for ^{254}Es, which falls between the calculated critical masses of ^{251}Cf and ^{245}Cm, was estimated to be about 29 grams. Since the quantity of ^{254}Es being handled was more than a thousand times less, the estimate was judged to be fully adequate to assure criticality safety in the handling of the nuclide.

Srinivasan showed[94] that the critical mass for fissile nuclides in aqueous solutions could be predicted from known values of k_∞ at specified hydrogen-to-nuclide atomic ratios. This correlation is less useful, however, because specific values of k_∞ would generally not be known, unless evaluated cross-section data were available to calculate them. There are other techniques that may be used to provide estimates of subcritical limits for highly fissile nuclides, but these are beyond the scope of the discussion here.

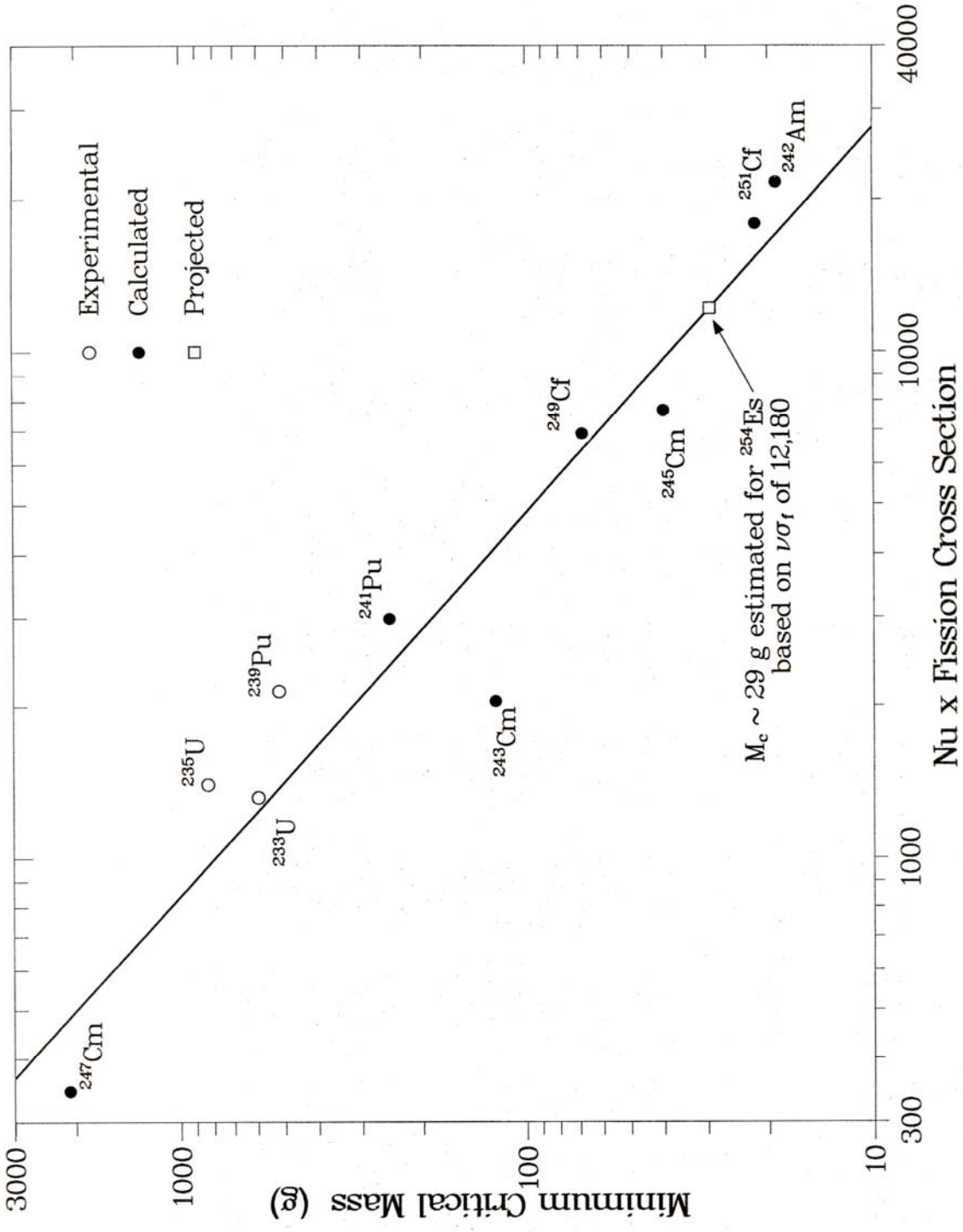


Figure 51. Experimental and Estimated Minimum Critical Masses of some Fissile Actinide Nuclides in Aqueous Solution

5. Fissile-Fissible Isotopic Mixtures – Condition when the Minimum Critical Mass Occurs for the Unmoderated Mixture

With fissile nuclides the minimum critical mass can be expected to occur under conditions of "optimum moderation". With mixtures of fissile and fissible isotopes, a question arises as to whether the minimum critical mass occurs below the mass of the dry oxide. This point is illustrated in calculations, by Clark,[83] shown in Figure 52. The calculations pertain to oxide mixtures of ^{239}Pu and the fissible nuclide ^{238}Pu. The critical mass of the dry oxide mixture is equal to the minimum critical mass of the oxide-water mixture when the ^{239}Pu content is ~37%. Considering the uncertainties in the calculations, the subcritical limit provided in the Standard for ^{239}Pu-^{238}Pu oxide mixtures, regardless of the H/Pu atomic ratio is 8 kg of Pu provided the Pu contains at least 67% ^{238}Pu, and the isotopic concentration of any ^{241}Pu that is present is less than that of ^{240}Pu (ANSI/ANS-8.15).[19] Thus, in considering mixtures of fissile and fissible nuclides, the single mass limit may be either that for the oxide (or metal) without moderator, or that for a mixture with water, depending on the isotopic composition of the nuclear material.

Likewise, there will be a concentration of the fissible nuclide for other fissile-fissible isotopes, wherein the effect of moderation does not reduce the critical mass below that of the dry mixture, i.e., for ^{240}Pu-^{239}Pu, ^{241}Am-^{242}Am, ^{244}Cm-^{245}Cm, etc.

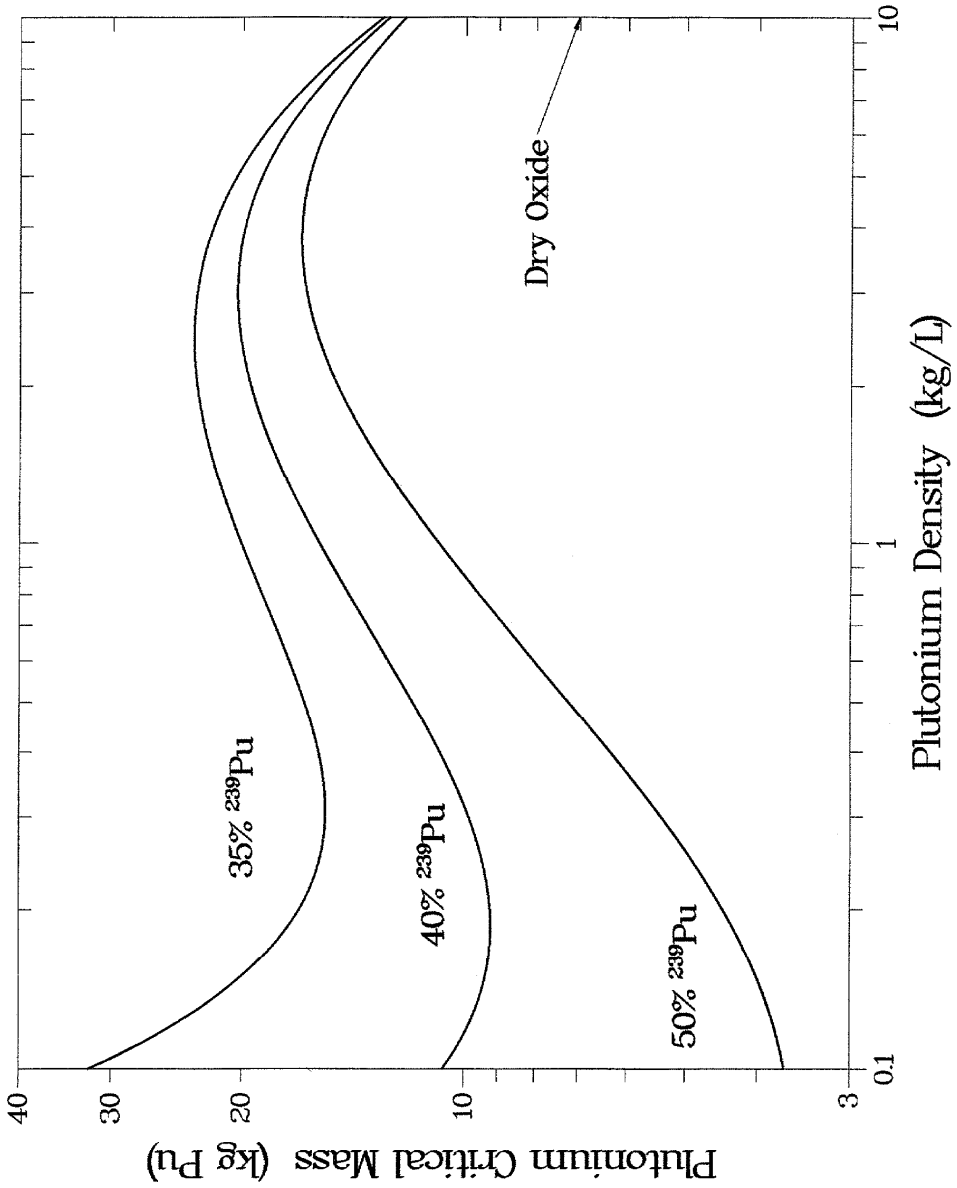


Figure 52. Calculated Critical Mass of Pu for Oxide Mixtures of ^{238}Pu and ^{239}Pu in Water

(adapted from a figure by Clark)[83]

6. The Use of Γ Widths in Survey of Criticality

As noted previously, the apriori condition for criticality in a finite system is that the reproduction factor in the infinite system (k_∞) exceed unity to account for neutron leakage (1-P) from the finite system. Knowledge of k_∞, therefore, provides a quick answer as to whether any nuclide can or cannot support a chain reaction.

In general the cross section for the fission of an excited compound nucleus (nucleus plus neutron) can be written as the product of the cross section for its formation by the factor for competitive disintegration:

$$\sigma(f,n) = \sigma_c(n)\ \Gamma_f / (\Gamma f + \Gamma + \Gamma_n + \ldots)$$

Γ_f is the width for fission of the compound nucleus, Γ_Γ the width for the (n,γ) capture reaction, Γ_n the width for neutron emission, etc. $\sigma_c(n)$ is the cross section for formation of the compound nucleus. $\Gamma_I\ /\hbar$ is the probability per unit time that the compound nucleus will de-excite with process i. Nuclear cross sections (having units of area) are not equal to Γ widths (having units of energy), but the ratios of cross sections are equal to the ratio of widths, i.e.,

$$\sigma_f/\sigma(n,\gamma) = \Gamma_f/\Gamma_\gamma$$

By definition, the expression for k_{inf} is

$$k_\infty = v\sigma_f / \sigma_a$$

with v being the number of neutrons released in fission. The absorption cross section $\sigma_a = \sigma_f + \sigma_\sigma$. Since we are taking ratios of cross sections, k_∞ may also be written in terms of the Γ widths as follows,

$$k_\infty = v\Gamma_f / (\Gamma_f + \Gamma_c) = v/ (1 + \Gamma_c/ \Gamma_f)$$

Thus, if the ratios of the widths can be independently calculated, these can be used to estimate k_∞ in lieu of using cross sections per se, even though calculations of widths cannot be made in detail.

Since fissible nuclides (threshold fissioners) have fission thresholds in the energy range 0.5 – 2 MeV and undergo fast fission with little or no subthreshold fission, another process must also be considered if the above formula is to be used for this type of nuclide. The effect of inelastic scattering must also be considered because if the neutron's energy falls below the fission threshold, it would no longer be available for fission and therefore, would be equivalent to a "capture". If the width for inelastic scattering, Γ_{in} is included, the equation now becomes:

$$K_{inf} = \nu\Gamma_f / (\Gamma_f + \Gamma_c + \Gamma^*_{in})$$

Inelastic scattering leaves the product nucleus in an excited state from which it decays by the emission of one or more gamma rays. For a neutron of energy E_0, the energy E_r of the re-emitted neutron can be obtained from the energy balance equation: $E_0 + S_n = E_i + S_n + E_r$ where S_n is the separation energy of the neutron from the compound nucleus and E_i is the energy emitted by one or more gamma rays. E_i is also the loss in energy of the re-emitted neutrons from that of the incoming neutron. If this decrease in energy results in a neutron with energy below the fission threshold, then the emitted neutron will not have a second chance for fission and the process is, in effect, the equivalent to neutron capture. This consideration is especially important in regard to the criticality of threshold fissioners. A pertinent question is: What fraction of the neutrons released in fission that scatter inelastically will still retain energy above the fission threshold? Thus we have written Γ_{in} with an asterisk to indicate that it is only some fraction of the inelastic scattering that results in loss of neutrons to the fission process. Cross sections for (n, 2n) and (n, 3n) reactions are very small since the average energy of the fission neutrons is well below the neutron energy thresholds for these reactions and so these can be ignored.

In regard to fissile nuclides, the inelastically scattered neutron, being of less energy, can be expected to encounter a larger cross section for fission than before. This is precisely the opposite to that encountered when the nuclide is a threshold fissioner! The fission cross section of a fissile nuclide varies slowly throughout the energy region characterized by the threshold fissioners and becomes very large at thermal neutron energy. Further, the cross section for inelastic scattering, when averaged over the Watt spectrum, is comparable to and in some cases larger than, the fission cross section averaged over the same spectrum.

The fission cross section for some threshold fissioners, such as ^{238}Pu and ^{241}Am, may experience an increase with decreasing neutron energy below the fission threshold and become even a few barns larger than the values above the threshold. The large increase in neutron capture cross section at thermal energy, however, precludes the possibility of a thermal neutron chain reaction. For example, at 2200 m/s, the fission and capture cross sections for ^{238}Pu are σ_f = 18 b and σ_c = 562 b and for ^{241}Am, σ_f = 3.3 b and σ_c = 577 b.

The reason for this lengthy discussion is that there are more fissible nuclides than there are fissile nuclides with half lives greater than six weeks, i.e., the fissible nuclides predominate! Also, an interesting paper is described below that makes use of the Γ widths in a survey on the criticality of threshold fissioners. The paper was presented by Russian engineers at the International Conference on Nuclear Criticality Safety, Oxford, England, in 1991.[21] Sviridov described a method of calculating k_∞ for metal systems of even-N actinide nuclides based on calculated partial widths. The method made use of information on the neutron and fission widths (denoted Γ_n and Γ_f) for heavy nuclei, $90 \leq Z \leq 100$. Using this method, the authors calculated k_∞ for 29 different actinide nuclides with even-N beginning with ^{228}Th and ending with ^{254}Cf. The value of v was calculated at a neutron energy equal to 2 Mev corresponding to the average energy of the neutrons released in fission, which differs only slightly among nuclides. It also was assumed that the probability of fission above the fission threshold in the even-N nuclides could be determined over the energy range of interest at a constant ratio of Γ widths, since Γ_c is expected to vary slowly with energy.

The calculated multiplication factors showed that all even-N nuclides between ^{235}Np and ^{254}Cf would be capable of supporting neutron chain reactions, in addition to the light nuclides, ^{231}Pa, ^{232}U, and ^{234}U.[21]

Sviridov reported that the calculated multiplication factors were in good agreement with earlier results from other authors. The Russian paper represents the most complete survey to date on the criticality aspects of the threshold fissioners, or even-N nuclides.

7. Infinite Multiplication Factors (k_∞) for Metal Systems of Fissile Nuclides

In a recent paper, Srinivasan[23] showed that k_∞ for metal systems of fissile nuclides correlates linearly with the fissionability parameter Z^2/A. This correlation (see Figure 53) can be used to

estimate values of k_∞ for metal systems of fissile nuclides where cross-section data are not available. This correlation, however, is not applicable to the fissible nuclides.

8. Critical Masses of Metal Systems of Fissile and Fissible Nuclides

By definition, as k_∞ approaches unity, the critical size, the critical mass, and the ratio, critical mass/surface area $\left(\frac{M_c}{s}\right)$, will all approach infinity. Consequently, a plot of $\frac{s}{M_c^b}$ will extrapolate to zero when k_∞ = unity $\left(\frac{M_c^b}{s} = \sigma_c^b \text{ in Srinivasan's notation}\right)$.[23] As shown in Figure 54, k_∞ is unity when $\frac{Z^2}{A} = 34.1$. This suggests that $\frac{Z^2}{A}$ must be at least 34.1 for a fast neutron chain reaction to occur with fissile nuclides. If we then plot the quantity $\frac{s}{M_c^b}$ vs. the quantity $\left[\left(\frac{Z^2}{A}\right) - \left(\frac{Z^2}{A}\right)_{\lim}\right]$, where $\left(\frac{Z^2}{A}\right)_{\lim} = 34.1$, we obtain a roughly linear correlation that passes through the origin (see Figure 55). Furthermore, if calculated data for fissible nuclides are used, the data will roughly approximate a linear correlation if $\left(\frac{Z^2}{A}\right)_{\lim}$ is taken to be 34.9. This suggests that $\frac{Z^2}{A}$ must be at least 34.9 for a fast neutron chain reaction to occur with fissible nuclides.

Referring to Figure 55, the equation of the line shown is

$$\frac{S}{M_c^b} = 0.0106\left[\left(\frac{Z^2}{A}\right) - \left(\frac{Z^2}{A}\right)_{\lim}\right] cm^2/g \qquad (2)$$

Several of the calculated points seem to depart significantly from the line; ^{247}Cm, ^{243}Am, and ^{238}Pu are some examples. These departures suggest that the Srinivasan correlation is only a rough approximation.

Nevertheless, by means of the correlation, the bare core critical mass can be roughly estimated for a nuclide at any density, from the simple equation

$$M_c^b = \frac{36\pi}{\rho^2}\left[\frac{M_c^b}{S}\right]^3 \qquad (3)$$

One need only compute $\left[\left(\frac{Z^2}{A}\right)-\left(\frac{Z^2}{A}\right)_{\lim}\right]$, then read $\frac{s}{M_c^b}$ from the figure. No knowledge of cross sections is required.

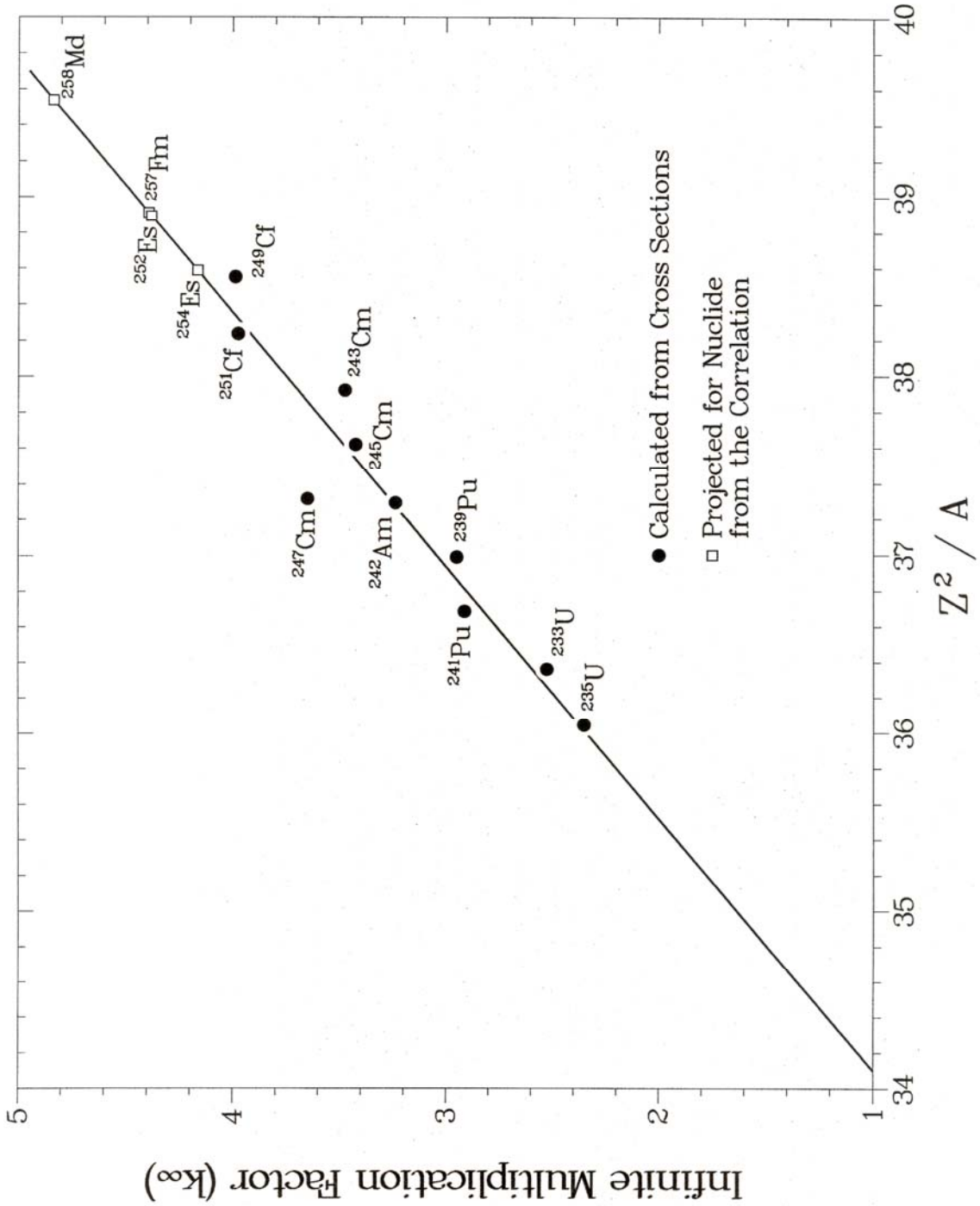


Figure 53. Infinite Multiplication Factor (k_∞) vs. Z^2/A for Metal Fissile Systems Adapted by a figure from Srinivasan[23]

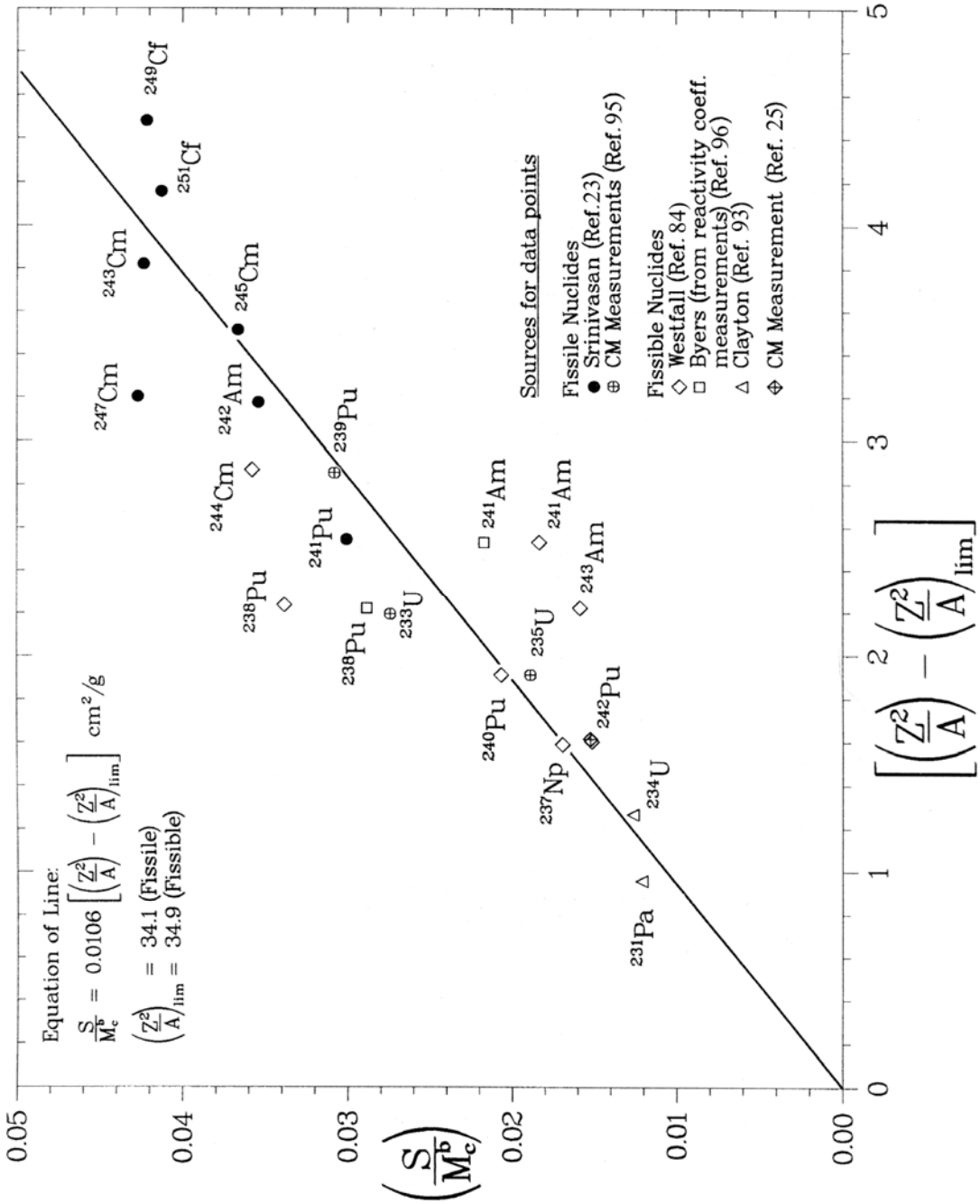


Figure 54. Plot of (S/M^b_c) vs. $[(Z^2/A) - (Z^2/A)_{lim}]$
Adapted from a figure by Srinivasan[23]

Estimated critical masses of bare metal spheres for a few selected nuclides are provided in Table XII below, illustrating the use of Srinivasan's correlation.[23]

Table XII. Estimated Critical Masses of Bare Metal Spheres using Correlation of Srinivasan

Nuclide	Density (g/cm^3)	Critical Mass (kg)
^{233}U	18.9	23.8
^{235}U	18.9	37.3
^{237}Np	20.45	55.5
^{239}Pu	19.6	10.3
^{242}Pu	20.05	56.4
^{240}Am	13.65	11.7
^{241}Am	13.65	31.1
^{242m}Am	13.65	15.6
^{243}Am	13.78	45.2
^{245}Cm	13.5	11.9
^{252}Es	13.5	4.7
^{254}Es	13.5	5.7
^{257}Fm	13.5	4.7
^{258}Md	13.5	3.2

The correlation results in a critical mass for ^{239}Pu that agrees well with the known experimental value. The correlation, however, does not agree nearly as well with the known experimental values for ^{233}U and ^{235}U.

9. <u>The Case of Fissile and Fissible Isotopic Mixtures</u>

With mixtures of fissile and fissible isotopes, a question arises as to whether the minimum critical mass of the oxide-water mixture occurs below the mass of the dry oxide at theoretical density. This point is illustrated in calculations, again by Clark,[83] shown in Figure 52. The calculations pertain to oxide mixtures of fissile ^{239}Pu and the fissible ^{238}Pu. The critical mass of the dry oxide mixture is equal to the minimum critical mass of the oxide-water mixture when the ^{239}Pu content is ~37%. Considering the uncertainties in the calculations, the subcritical limit given in ANS-8.15 (1981) for ^{239}Pu-^{238}Pu oxide mixtures, regardless of the H/Pu atomic ratio is 8 kg of Pu provided the Pu contains at least 67% ^{238}Pu, and that the isotopic concentration of any ^{241}Pu that is present is less than that of ^{240}Pu. Thus, in considering mixtures of fissile and fissible isotopes, the smallest critical mass may be either that for the unmoderated material

(oxide or metal), or that for a mixture with water, depending on the isotope composition of the nuclear material.

Overall the mixture may then be considered to behave either as a “fissible” or “fissile” system, depending on which of the critical masses is the smaller, that for the case of no moderator, or that for the system when moderated.

10. Infinite Multiplication Factors for Metal Systems of Fissible Nuclides

In 1973, Clayton[82] reported an empirical correlation between k_∞ for a nuclide and the difference between its neutron binding energy (Bn) and the activation energy for fission (Ea) of the compound nucleus. This correlation suggested the possibility of providing estimates of k_∞ for other even-N (fissible) nuclides. Calculated values of the activation energy for fission (Ea)[90] and the binding energies of the neutron (Bn), for nuclides ranging from ^{232}Th to ^{254}Es were tabulated together with the known critical (fissionability) parameters.

The evidence suggested that it would be possible to obtain criticality with the even –N protactinium nuclide, ^{231}Pa, a nuclide lighter than uranium. The empirical correlation by Clayton that applies only to even-N actinides is shown in Figure 55. The fact that ^{231}Pa could support a chain reaction was also confirmed by a report given by Russian engineers at the International Conference on Nuclear Criticality, Oxford, England in 1991.[21]

The criticality and fissionability aspects of the fissile and fissible actinides nuclides also were reviewed by Clayton.[20]

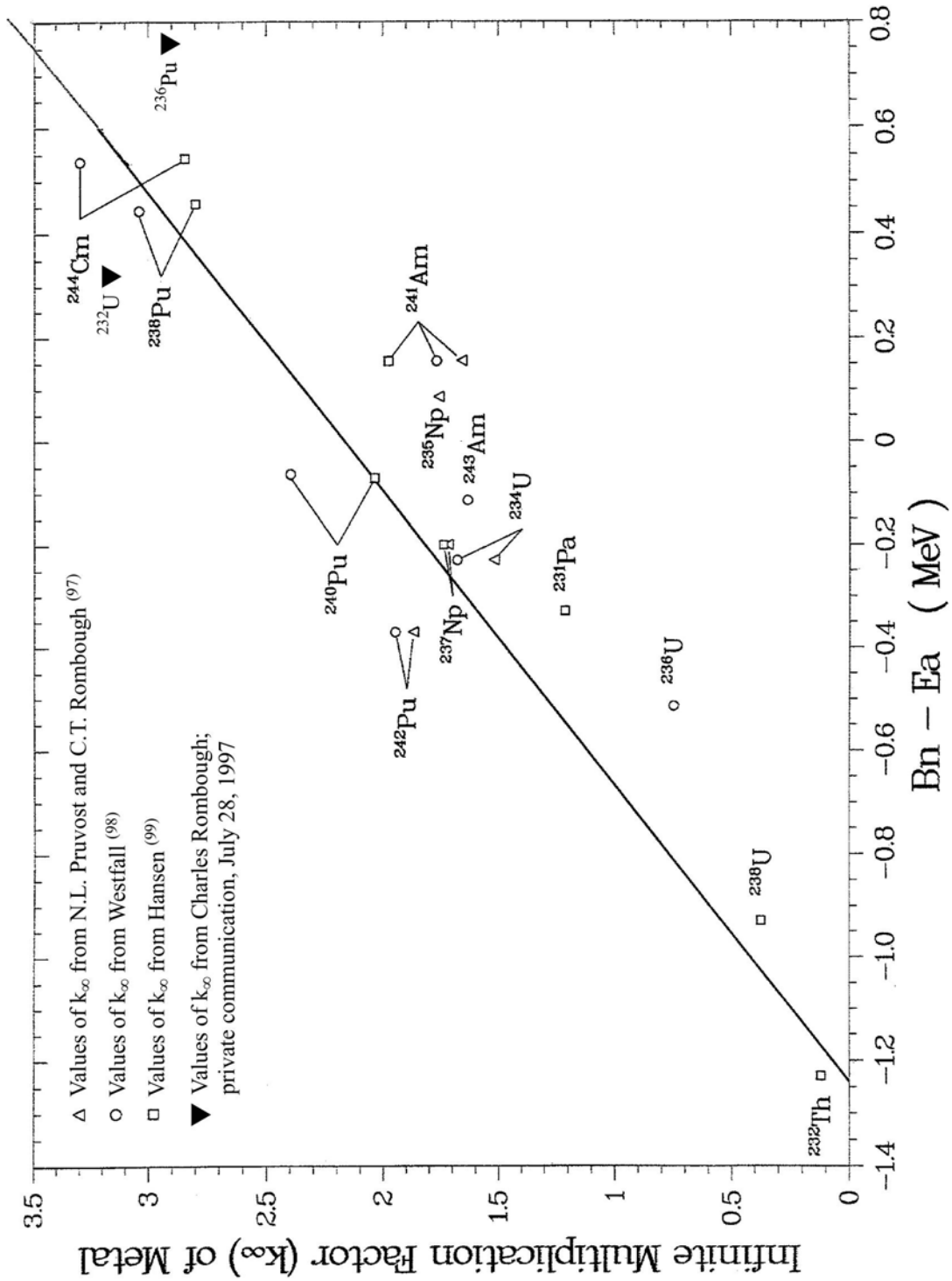


Figure 55. Calculated Infinite Multiplication Factors (k_∞) of Metal Systems of Even-N Nuclides vs. (Bn-Ea)

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 υ (neutrons per fission,) σ_f/σ_a (ratio of the fission cross section to the absorption cross section), and σ1 (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 something 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 = 0.2 or ~ 7%. If the empirical correlation is correct, Figure 55 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 ~ 2.84; for $^{252}_{98}$Cf (B_n-E_a ~ 0) ~ 2.17, etc.

S. THE "CASE OF THE VANISHING DOLLAR"

1. Even-N Nuclides

Dr. W. Seifritz of the Swiss Federal Institute for Reactor Research, brought the following anomaly called, "The Case of the Vanishing Dollar," to light.

Calculations and analysis show that criticality would be possible for a number of even-n actinide isotopes including, $^{231}_{91}$Pa, $^{237}_{93}$Np, $^{236}_{94}$Pu, $^{238}_{94}$Pu, $^{240}_{94}$Pu, $^{241}_{95}$Am, $^{243}_{95}$Am, $^{244}_{96}$Cm, $^{246}_{96}$Cm, and $^{252}_{98}$Cf. The results of known calculations were summarized in Table X. Although criticality now appears possible for each of the above even-n nuclides, it may only be achieved under essentially unmoderated conditions ($^{236}_{94}$Pu is an exception). 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:

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

When k_{eff} exceeds unity, the reactivity in terms of dollars becomes infinite if $ß_{eff}$ (the effective delayed neutron fraction) approaches zero.

The mean energy of the delayed neutrons that are released after fission is of the order of 300-400 Kev, which is considerably less than the energy of the prompt neutrons emitted at the time of fission. Although the fraction of neutrons that are delayed (β) differs from zero, the effective delayed neutron fraction (β_{eff}) becomes very small, though not zero under these circumstances. Because of the fission cross-section thresholds of the even-n actinide isotopes, the "worth of the dollar" can become very small. Under these circumstances the difference between delayed and prompt criticality can become very small and delayed and prompt criticality are in practice one and the same, i.e., delayed criticality ceases to exist. (Hence, the "Case of the Vanishing Dollar".)

Due to the near impossibility of controlling a prompt critical assembly, any future criticality measurements on these types of isotopes will by necessity be made from subcritical extrapolations only.

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.[100] 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. An 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 rated capacity of the solution pump ranges up to 1150 gpm.) If the returning precursors were 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 mechanical means via circulation of the reactor's fuel.

T. THE INFINITE SEA CRITICAL CONCENTRATION

The infinite sea critical concentration may be referred to in the sense that if there is a large volume of water, and if fissile atoms are added uniformly to form a homogenous mixture with water, then at a certain concentration of these fissile atoms, criticality will occur. (This is the "infinite sea" or limiting critical concentration.) As 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 reabsorbed in the fissile nuclide and one half in the water, or diluent. This will be 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 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 6 for the case of Pu in water.)

Critical masses have been calculated for bare spherical reactors containing homogenous mixtures of the fissile atoms ^{233}U, ^{235}U and ^{239}Pu, in each of the above diluents or moderators.[41] 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 infinite 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 hydrogenous 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 from the viewpoint of inadvertent criticality with these fissile nuclides. The water-Pu mixture after all, has the smallest critical mass.

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

The ANSI/ANS-8.1-1998, "Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors," provides in its Table II, limits for aqueous solutions.[15] The infinite sea concentrations for ^{235}U and ^{239}Pu are given as 11.6 and 7.3 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 homogenous aqueous solution, which is given as 1.00 wt.% ^{235}U. At this enrichment, the maximum value of k_∞ will be less than unity at 11.6 g/ℓ ^{235}U. This limit is also valid for all lesser enrichments, as the presence of ^{238}U will require ^{235}U concentrations > 11.6 g/ℓ if criticality is to be achieved. It should be recalled that an aqueous homogenous solution of uranium having enrichment below the limiting critical value of 1 wt.% ^{235}U, would have a k_∞ less than unity for any concentration whatsoever.

The limit given for plutonium (7.3 g Pu/ℓ) will not necessarily be subcritical if the plutonium is mixed with natural uranium. The computed value of k_∞ is given in Figure 56 as a function of the weight fraction of plutonium in the Pu + U for homogenous aqueous solutions.[13] Moving from the right (weight fraction of one) to the left, the value of k_∞ is initially seen to decrease from unity until the weight fraction, Pu/(Pu + U), is decreased to ~ 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.

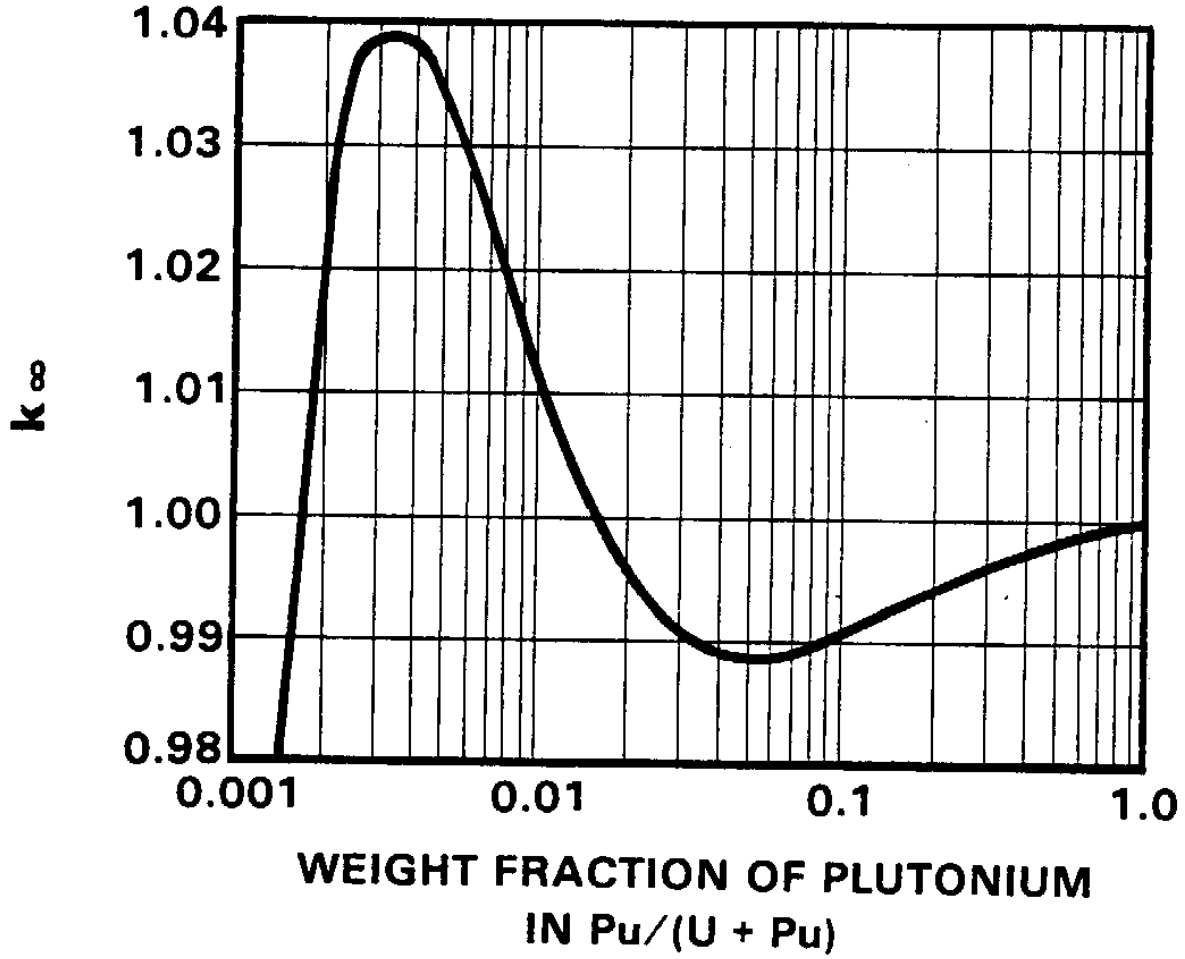


Figure 56. Computed k_∞ vs. Weight Fraction of Plutonium in Pu + U Homogeneous Aqueous Solutions of ^{239}Pu + U (nat) O_2 (Plutonium Concentration Held Fixed at 7 g Pu/ℓ)

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. At a weight fraction of only 0.0035, it is estimated that the value of k_∞ might be as high as ~ 1.04 for a plutonium concentration of 7.0 g Pu/ℓ. To ensure subcriticality in the solution, the Pu/(Pu + U) fraction for mixed oxides in water is 0.0013. At this weight fraction, the ^{239}Pu concentration will be ~ 5 g ^{239}Pu/ℓ; the H/fissile atom ratio giving the highest value of k_∞ is ~ 500.

There are several reasons for the anomalous behavior of the k_∞ curve. First, 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 are also 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. This is 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.

In other words, 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 $\simeq$ 3680.)[10] At this point, about half of the neutrons released in fission are absorbed in the diluent (H_2O) since

$$k_\infty = 2.08 \bullet f = 1, \text{ where 2.08 is the } \eta \text{ for Pu}$$

It is not the concentration that is important, but rather the ratio of absorbing atoms to fissile atoms that determines this limit and is the controlling factor.

Consider the discharge of dilute plutonium solutions to earth, such as 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.

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

This 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, 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. As Pu builds up in the soil, perhaps from solutions containing only milligram/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 also be noted that under the conditions given, k_{eff} could be reduced by the re-addition of water to the system, or the system would be made further subcritical on flooding.

The bases for these conclusions are presented in Figures 57 and 58 and come from a series of calculations by K. R. Ridgway and R. D. Carter on "Criticality Prevention Parameters of Plutonium in Soils."[101]

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 in Table XIII.

Table XIII. Hanford Soil Compositions, Weight Percent

Components	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	

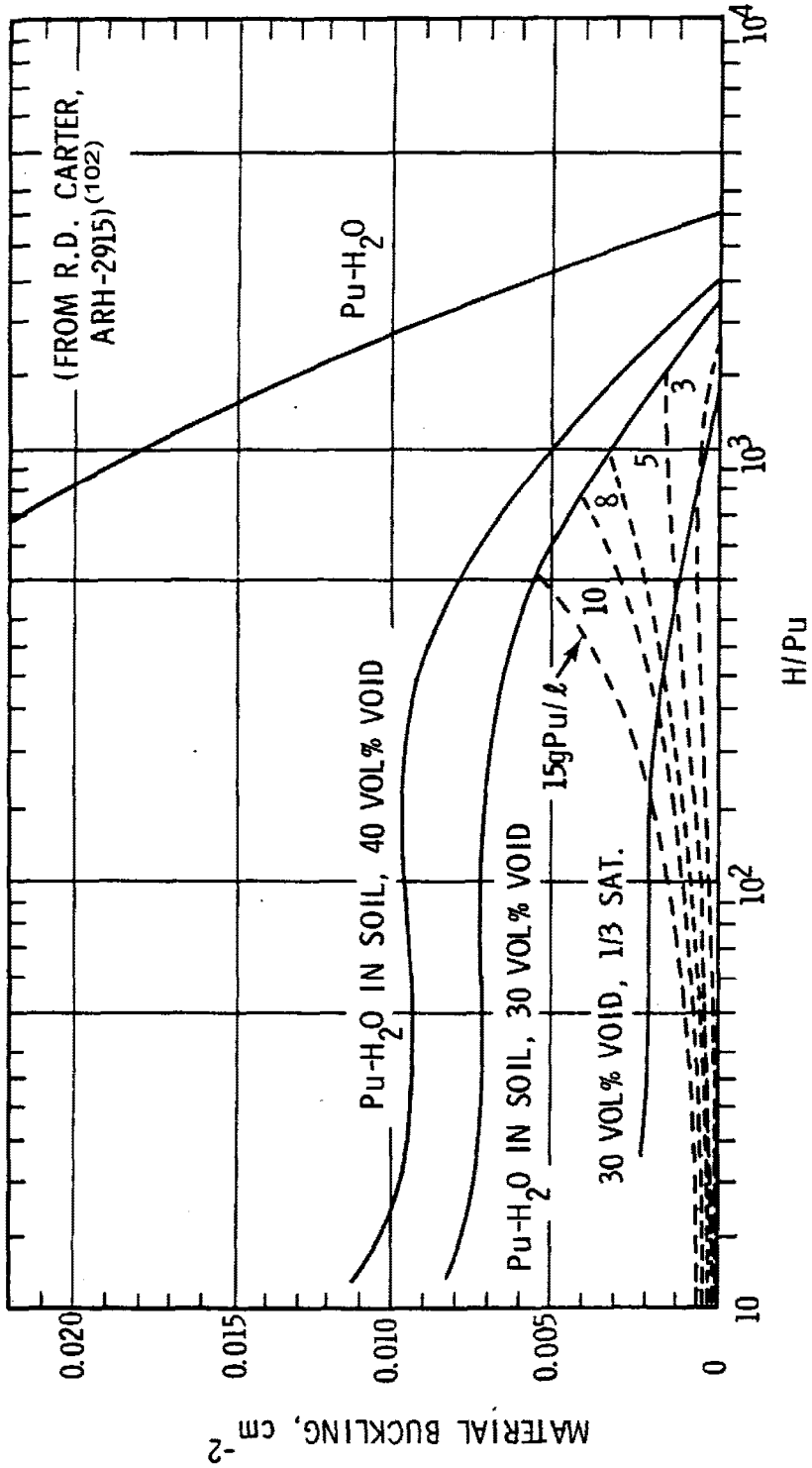


Figure 57. Material Buckling vs. H/Pu (Pu-H_2O in Soils; 3 wt.% ^{240}Pu in Pu, GAMTEC-II Calculation)

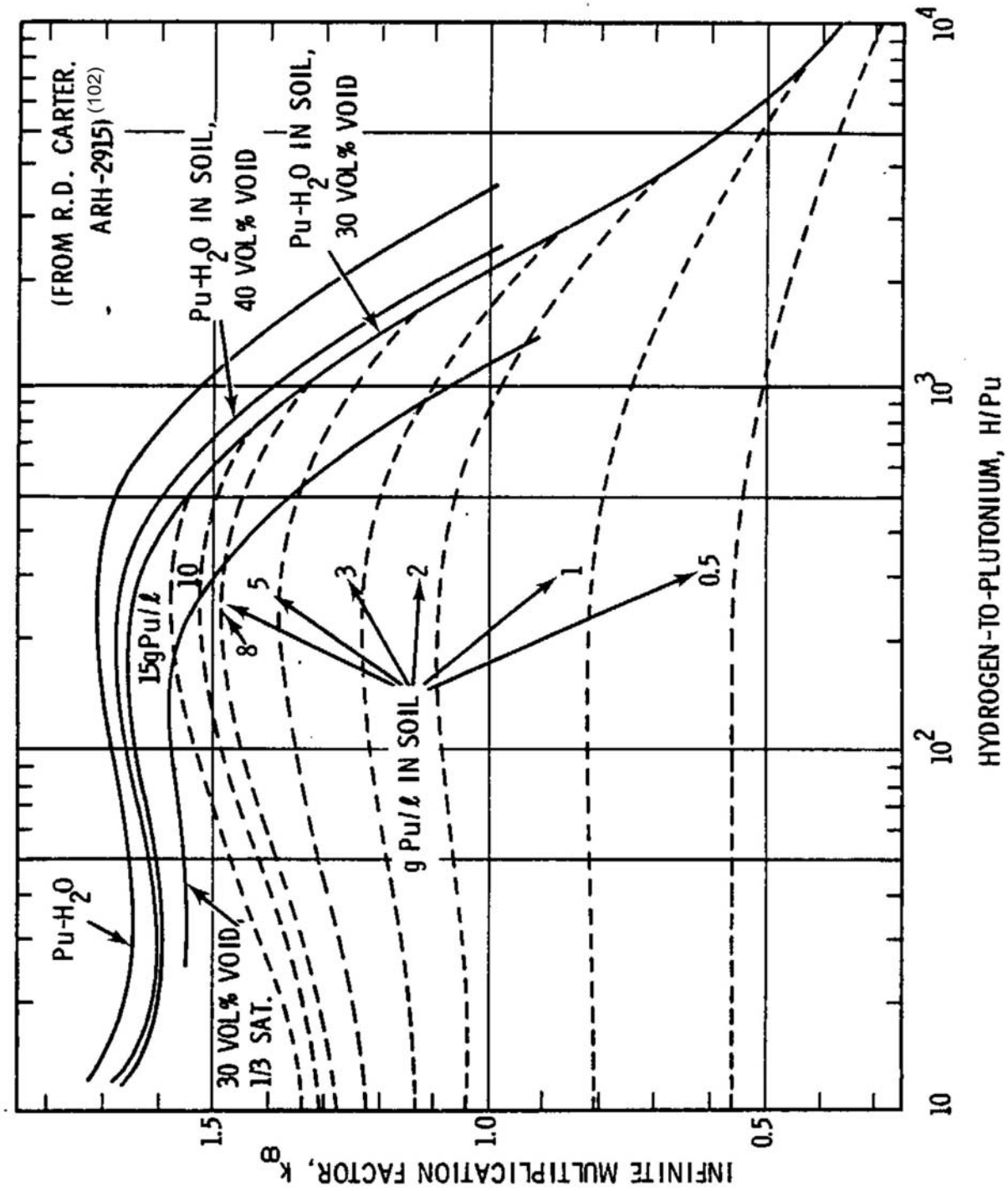


Figure 58. k_∞ vs. H/Pu (Pu-H_2O in Soils; 3 wt.% ^{240}Pu in Pu; GAMTEC-II Calculation)

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 through the process of evaporation, for instance, k_∞ would subsequently increase and go through a maximum value $\geq$ 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 ~ 200.)

4. The Universe – The Beginning

" . . . The universe is not only queerer than we suppose, but queerer than we can suppose."[b]

The ^{235}U atomic percentage in U, as found on earth, is remarkably constant, 0.7200 $\pm$ 0.0006%, with the exception of that found in the Oklo mine (see Section U). The well-known half-life for ^{235}U is 7.13 x 10^8 years, whereas that for ^{238}U is 4.507 x 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 ~ 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 speculate as to whether there is any lower limit on the critical concentration in the absence of 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 ß-decay and absorption within a critical system with a lower limit of nuclei density determined by the radioactive decay constant of the neutron. Under these conditions, the equation for estimating the minimum critical concentration for

[b] J. B. S. Haldane, late British Scientist.

criticality in infinite ether, assuming the neutrons are at the average energy of fission, ~ 2 MeV, can be deduced.

<u>Equation for Criticality in Infinite Ether</u>

$$n\, v\, N\, \sigma_a\, \eta = n\, v\, N\, \sigma_a + \lambda_ß\, n$$

The losses (on the right) come from absorption as well as ß-decay of the neutron, since the neutron is radioactive – half-life about 12 minutes – with a mean life of about 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 equation, $\lambda_ß$ is the decay constant for ß emission by the neutron, taken as 9.6×10^{-4} sec^{-1}. The relativistic 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 ^{235}U, and taking n as 2.94, the mean free path, $\lambda = 1/N\sigma$, 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 59.

The density of the fissile atoms, or infinite ether concentration, under these circumstances would 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. The quantity of material contained within a sphere the radius of which is equal to the above mean free path, even at the nuclei density of 0.054×10^{-9} g/cc, would be 200 times the lunar mass, or about 2 – ½ times the mass of Earth.

It should be considered that some of the fast neutrons would 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. So the effect of inelastic scattering is to increase the number of neutrons lost by ß-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.

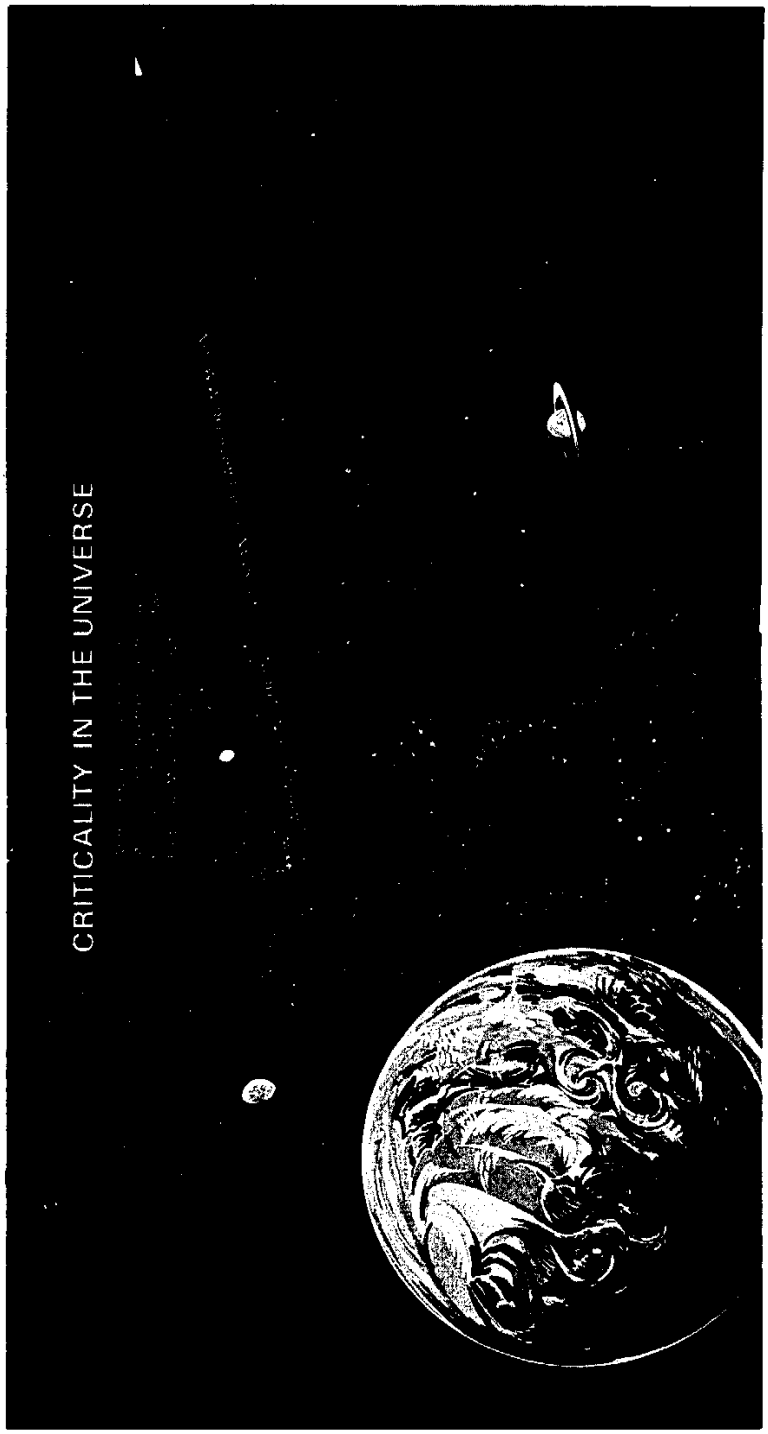


Figure 59. Criticality in Universe (At the Critical Concentration of ^{235}U Atoms in "Infinite Ether," Neutron would Travel some 100 times Distance between Earth and Moon before Absorption)

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

The permissible limit for occupation exposure covering a 50-year time span, for airborne contamination in the case of the naturally occurring ^{235}U isotope ($T_{½}$ = 7.13 x 10^8 years) is set by Federal Regulation (10 CFR 20, Appendix B) in activity concentration units (μC/cm^3). The value expressed in terms of g/cm^3 for ^{235}U in insoluble form is 4.67 x 10^{-11}. This amounts to 3.7 x 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 x 10^{11} atoms/cc in the case of ^{235}U, the number of α-decays from ^{235}U would only be about 7.9 x 10^{-6} d/sec per cc, which only differs by a factor of two from the above 50-year earthbound limit for airborne contamination.

U. NATURE'S ANOMALY IN WEST AFRICA

One of the strangest occurrences since the first manmade criticality (December 2, 1942 by E. Fermi and co-workers) was the discovery of nature's criticality in the Republique of Gabonaise.[103] 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 60 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. The isotopic composition of natural uranium is known to be remarkably constant throughout the world. The ^{235}U atomic percentage is 0.7200 =- 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 naturally occurring elements and was strikingly representative of fission product yields.

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 on 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 akin to today's light water reactor with a burn-up on the order of 20,000 MWd/T. The evidence for the event is most convincing as a result of the well-designed analysis made, and there can be little doubt that the reaction took place, a billion years or so prior to man's achievement of criticality.

V. "SMALL MASS" CONCEPTS

1. Thin Foils and Non-absorbing Low Temperature Moderating Reflectors

In principle, it is now known how to obtain a critical configuration with less than an ounce of ^{239}Pu, or with only about one ounce of ^{235}U. 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[91] was predicated on the 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°K 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 $\sim$ 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.”[92] These concepts are illustrated in Figure 61.

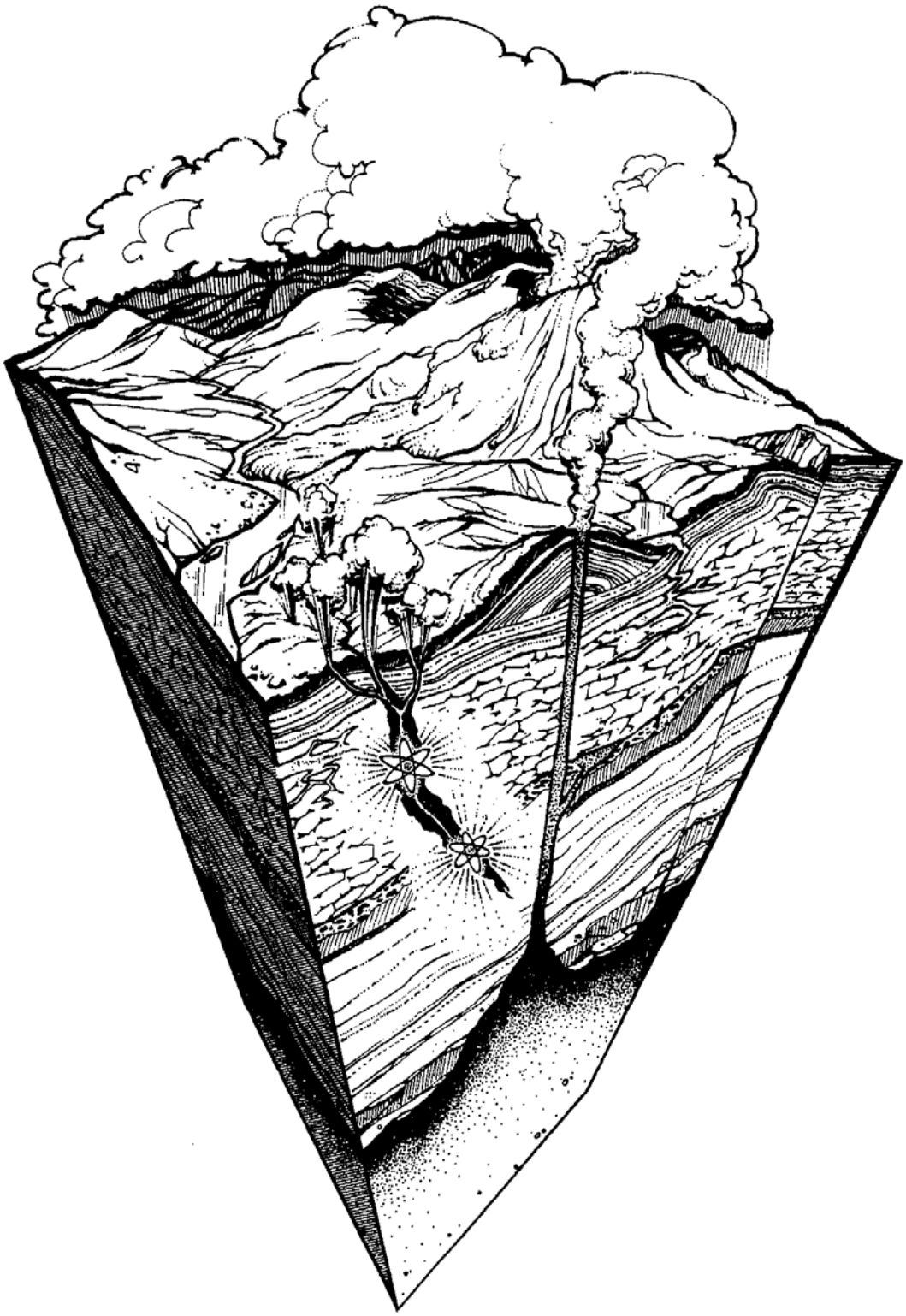


Figure 60. Criticality in Earth – Site of OKLO Mine Circa Two Billion BC

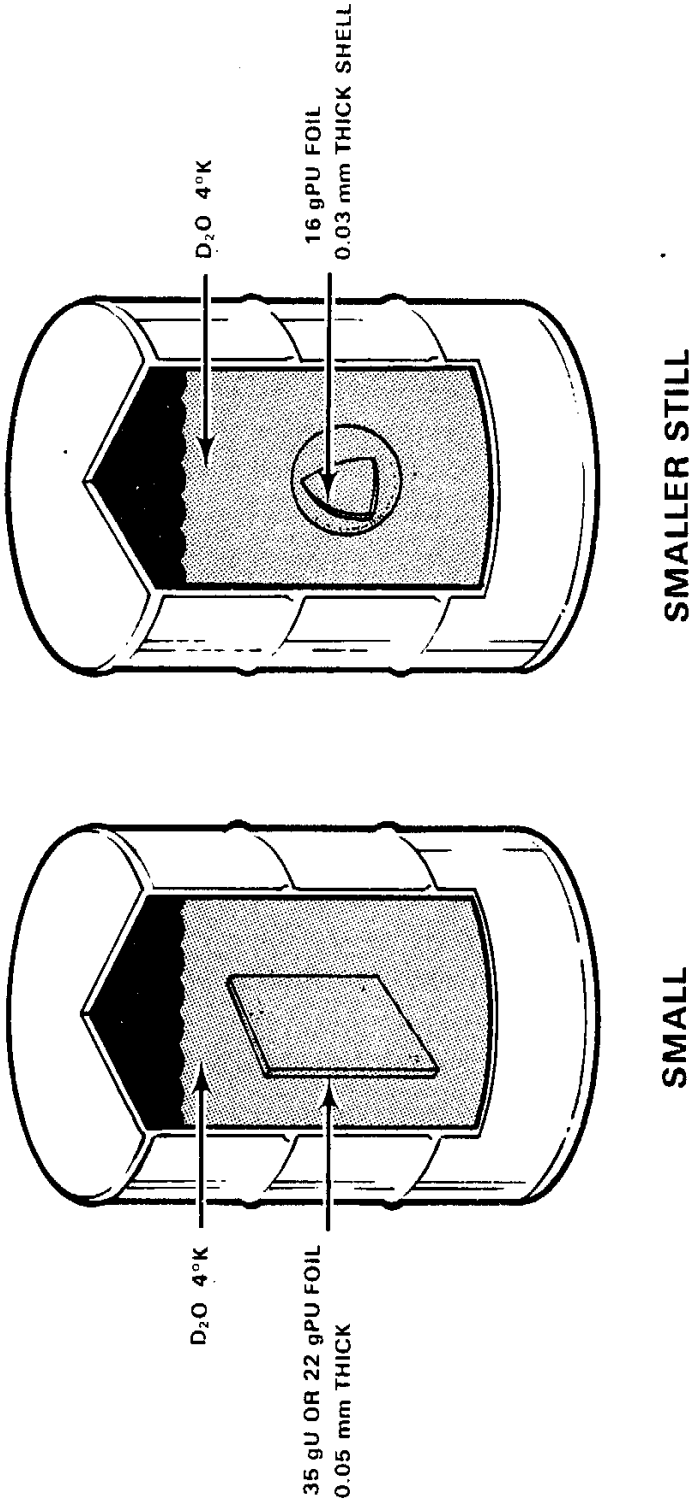


Figure 61. Small Mass Concepts (Thin Foils and Nonabsorbing Low Temperature Moderating Reflectors)

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 there was no provision 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 390g 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 22g x 390/550 = 15.6g of ^{239}Pu would be necessary for criticality.

2. <u>The Laser-Induced Micro-Explosion</u>

As noted previously, the critical mass of an unreflected sphere will vary inversely as the square of the density, $M_c \sim {}_o^{-2}$. For example, if two systems, differing only in density, were critical (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 may be possible by means of powerful laser beams or intense relativistic electron beams, to compress a small fissionable sphere, on the order of a mm, into a highly supercritical assembly.[104, 105] Pressures up to 10^{12} atm, comparable to 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[105] that a pellet containing 0.2g 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 $k_{eff} \simeq 1.25$ if a laser pulse-energy of some 4.7 MJ were absorbed in the outer ablative layer. The number of fissions that would occur during the 0.8 ηsec 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). In the case of the T-D reflector, the initial fissionable pellet diameter could be even smaller, containing only 10^{-2} g Pu.) See Figure 62 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. Should it become feasible in the near future, it would be of great interest 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.

3. The Beryllium Reflector: Polyethylene and BeH_2 Moderation and Spatial 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 and BeH_2 moderation and beryllium reflection. Experiments performed at Los Alamos show the minimum critical mass of ^{235}U in a hydrogenous core with a thick beryllium reflector (about 32 cm. thick) can be reduced to 250 - 300 gm.[106] 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 inches 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, e.g., with a 20% of the fuel moved to the outer 0.4 cm, a 3% decrease in critical mass could be affected.

Calculations on criticality parameters for ^{239}Pu in organic media show the minimum critical mass for a homogeneous mixture of ^{239}Pu in polyethylene – but with a water reflector, to be 0.370 kg.[107,108] This may be compared to .524 kg 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 0.370 kg value would undoubtedly have been substantially smaller.

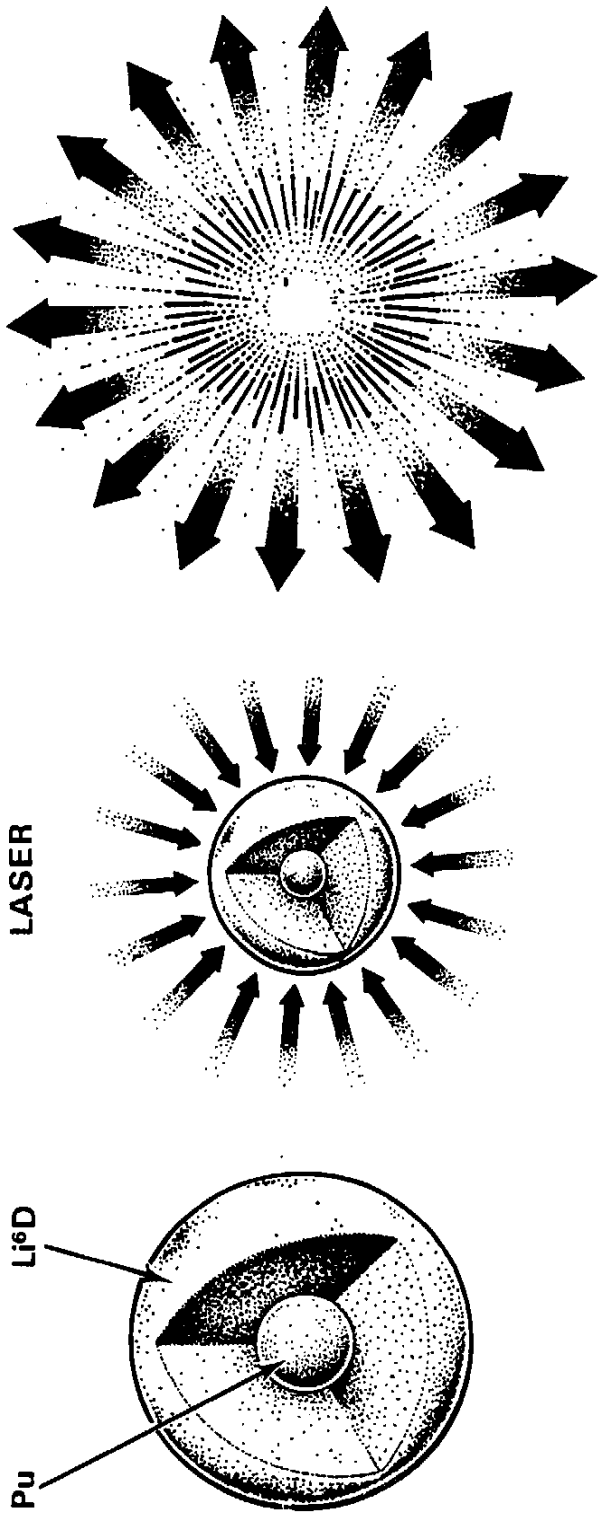


Figure 62. Laser Implosion of Fissionable Pellet Metal Spheres

Calculations have been made with BeH_2 as a moderating material which results in even smaller critical masses than the case of polyethylene.[108] The reasons for this further reduction in minimum critical mass may be accounted for in part from (n, 2n) reactions in beryllium and the possible higher hydrogen atomic density, as well as the better scattering properties of beryllium, especially in the crystalline form of BeH_2. The smallest critical masses were obtained with a BeH_2 moderator at full crystalline density of 780 kg/m^3.

Data from some of the calculations by K. S. Rao and M. Srinivasan are tabulated below.[109]

CRITICAL MASS IN KG

BeH_2 Moderator at Crystalline Density	Bare Critical Radius (m)	Bare Critical Mass (kg)	Reflected Critical Mass		
			H_2O	BeH_2	BeO
U-235	0.1080	0.454	0.275	0.251	0.153
^{233}U	0.0915	0.327	0.213	0.178	0.106
^{239}Pu	0.1089	0.269	0.163	0.150	0.087

It is concluded that for BeH_2 of density 780 kg/m^3, critical masses are lower by about 15% as compared to the minimum critical masses for CH_2-moderated assemblies. Most of the reduction in the critical mass obtained with BeH_2 moderation is due to the superior nuclear properties of beryllium as compared to carbon in CH_2.

The most effective reflector, however, is BeO, not BeH_2. With a 0.2 meter thick BeO reflector in lieu of BeH_2, the critical mass of Pu is only about 0.087 kg. The latter value is some six times smaller than the water moderated and reflected minimum critical mass for ^{239}Pu, at about 0.524 kg.

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.[110] 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 5.24 kg).

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.

W. BEYOND CALIFORNIUM – AN ISLAND OF STABILITY – THE SUPERHEAVY ELEMENT "X"

Increased binding energy at closed or near-closed, shells of nucleons results in increased stability of the nucleus.[88, 89] Shell closures beyond lead are predicted at Z = 114 and N = 184 and possibly, at Z = 164 and N = 318.[111] 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 63. This particular portrayal was adopted from a paper by G. T. Seaborg.[111]

Several new "Superheavy" elements have now been reported, *Element* 114 (Vug), VNUNQUADIUM, in 1998 by scientists at Dubna (Joint Institute for Nuclear Research in Russia). The half-life is about 30 sec. Elements no. 113 (Vut), UNUNTRIUM, No. 115 (Vup), UNUNPENTIUM, and No. 116 (Vuh), UNUNHEXIUM, have also been produced. The half life for Element 113 (Reported in August 2003) is 50-70 ms. The half-lives of the two isotopes of Element 115 (Reported in February 2004) are $^{288}_{115}$Vup (87.5ms) and $^{287}_{115}$Vup (32ms). The half life for element 116, $^{293}_{116}$Vun, reported in July 2000 is 63 ms.

J. R. Nix has predicted some of the properties associated with the fission of the hypothetical Superheavy nuclei.[89] A few of these properties are included in Table XIV.

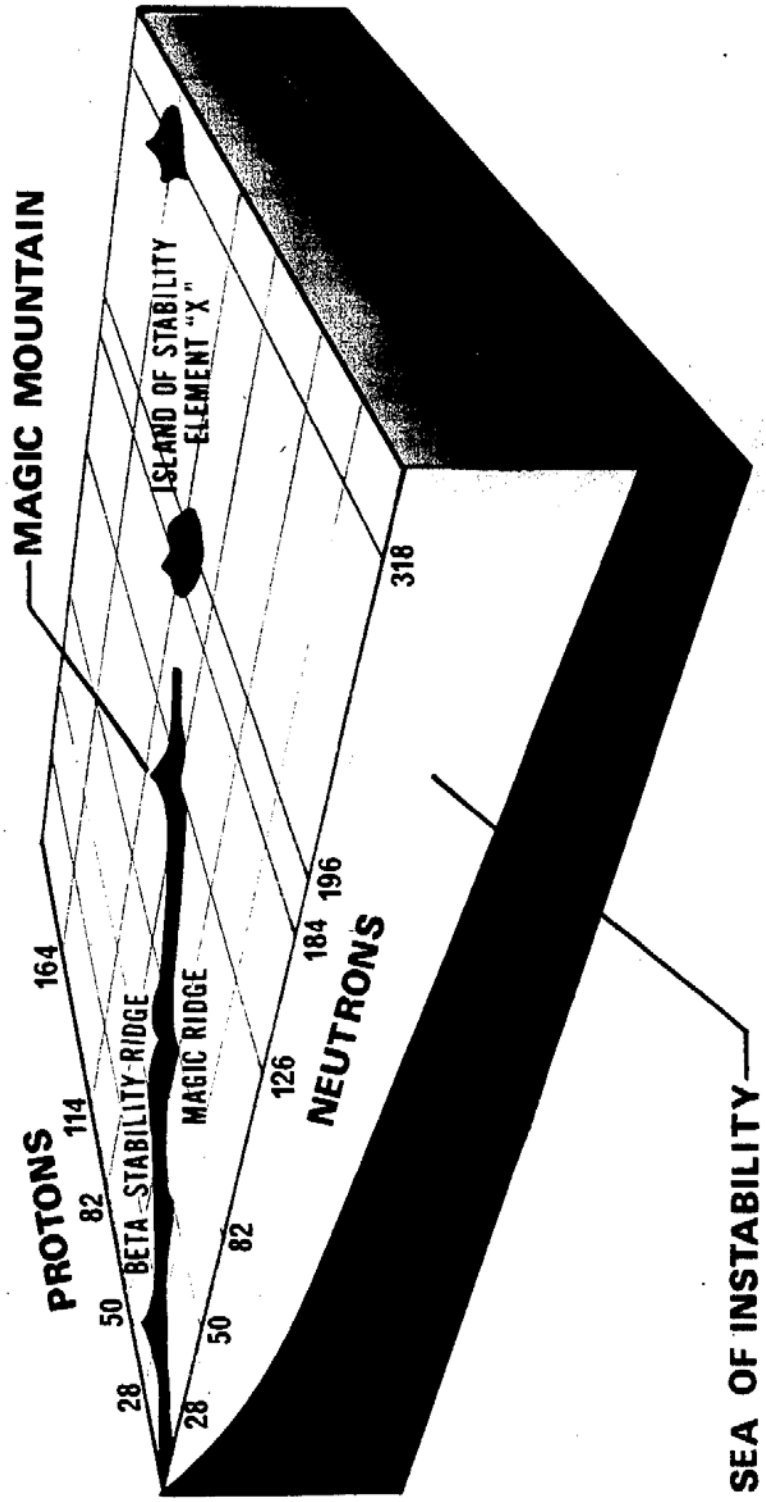


Figure 63. Known and Predicted Regions of Nuclear Stability, surrounding by a Sea of Instability

Table XIV. Properties of Superheavy Nuclei

Fissioning Nucleus	Energy Release per Fission	Number of Neutrons per Fission	Lifetime[(a)]
$^{298}_{114}X$	317 MeV	10.5	~ 10^3 years[(b)]
$^{294}_{110}X$	290 MeV	10.6	~ 10^9 years

(a) Total half-life from decay by spontaneous fission, α-decay, ß-decay, etc.
(b) G. T. Seaborg estimate for this nuclide.[(111)]

Two papers were published in Science, 26 December 1975, Vol. 190, No. 4221, wherein the authors give new evidence for the prior existence of extinct Superheavy elements.[(112, 113)] 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.

1. "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 but theoretically predicted (although analyses on the Allende meteorite provide evidence of previous existence). 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 ~ 3.44 cm
critical volume ~ 0.17 ℓ
critical mass ~ 0.5 g
Assuming minimum concentration of ~ 3 g/ℓ
in aqueous solution; water-reflected sphere.
[If a beryllium reflector is used in lieu of
water, make the critical mass ~ 0.2 g (0.007 oz)].

X. THE POWER REACTOR – FOUR BILLION WATTS AND SUBCRITICAL

When the N-Reactor was in operation on the Hanford Reservation, there was a sign that informed the visitor he was 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 was 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 the ^{238}U or ^{240}Pu, etc. The multiplication rate is given approximately by

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

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. To reach the power level, the reactor, however, would have been made super-delayed critical in the beginning.

Y. 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 is involved in connection with the energy crisis and the furor of some over the construction of reactors for production of energy. Gold is involved in connection 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, uranium is a by-product of the mining of gold in South Africa, just as for the most part, silver is a by-product of the mining of copper, zinc, lead and other bare metals.

A series of criticality calculations, until recently forgotten, that had been made on gold-uranium mixtures long before either the energy or monetary crises, has been found again. The only anomaly therein, is perhaps the reason as to why such calculations would have ever been made in the first place. In order that these results be preserved for whatever value they may now have, some data from these calculations are presented below in Table XV.

Table XV. 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)

A U.S. astronaut has been quoted as saying, “We went to sleep counting the light flashes.” In one instance, each of the astronauts on board the spacecraft 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.[114, 115] In 1934, Čerenkov first observed that very high speed electrons, or ß 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 eyes. Yet they claim to have observed a blue flash. Bear in mind that the visible light of the Čerenkov radiation does not transmit through opaque 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. 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 the capture of 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.[116] 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 $v > c/n$, where c is the velocity of light in vacuum and n is the index of refraction of light in the medium.

Čerenkov radiation is somewhat analogous 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 analog 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 miniature 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 $\cos\theta = c/nv$. 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 vacuum (c) that may not be exceeded.) For an excellent description of Čerenkov radiation, refer to Reference 115.

Figure 64 qualitatively illustrates the radiation phenomenon as might be observed from a hypothetical criticality incident. Anyone sufficiently close to the source of radiation might be expected to see a blue flash or to observe some Čerenkov radiation. It would not matter if their eyes were open or closed. To the observer, 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.

3. A Special Case of Criticality Postponed and Fission Power Control
(When the parent nuclide decays into a daughter critical mass)

The calculated critical mass for a bare sphere of ^{236}Pu metal is ~8.2 kg.[117] ^{236}Pu decays ($T_{1/2}$ = 2.85 Y) by α -emission into ^{232}U ($T_{1/2}$ = 68.9 Y), which in turn decays by α-emission to ^{228}Th. The critical mass for ^{232}U is only ~3.7 kg (Prichard, 1997).[118] Thus, the subcritical mass limit for ^{236}Pu should probably be based on the smaller subcritical mass limit for ^{232}U. A quantity of ^{236}Pu that was subcritical to start with, would in several years, decay into ^{232}U, which has the smaller critical mass. The ^{232}U could then become chain reacting as a result of the ^{236}Pu decay. The above constitutes an interesting anomaly, but if the Monte-Carlo calculations of critical masses are essentially correct, the scenario described would be quite possible! What this means is that a subcritical quantity of ^{236}Pu could be placed in isolated storage, and after several years a chain reaction miraculously appears, or in this case, it's an example of "criticality postponed".

A second interesting anomaly follows the first. For example, let us begin with 4.1 kg of ^{236}Pu (1/2 of its bare critical mass). When a critical mass of ^{232}U has been formed, 3.7 kg, the amount of ^{236}Pu remaining will be 4.5 kg. The heat generated from α-decay in the ^{236}Pu will be ~67.3 kW and 9 x 10^{16} atoms of ^{232}U will be formed per second. Once the critical mass is reached for ^{232}U, it is, however, only the surplus ^{232}U atoms produced from α-decay of the parent ^{236}Pu that can be fissioned.

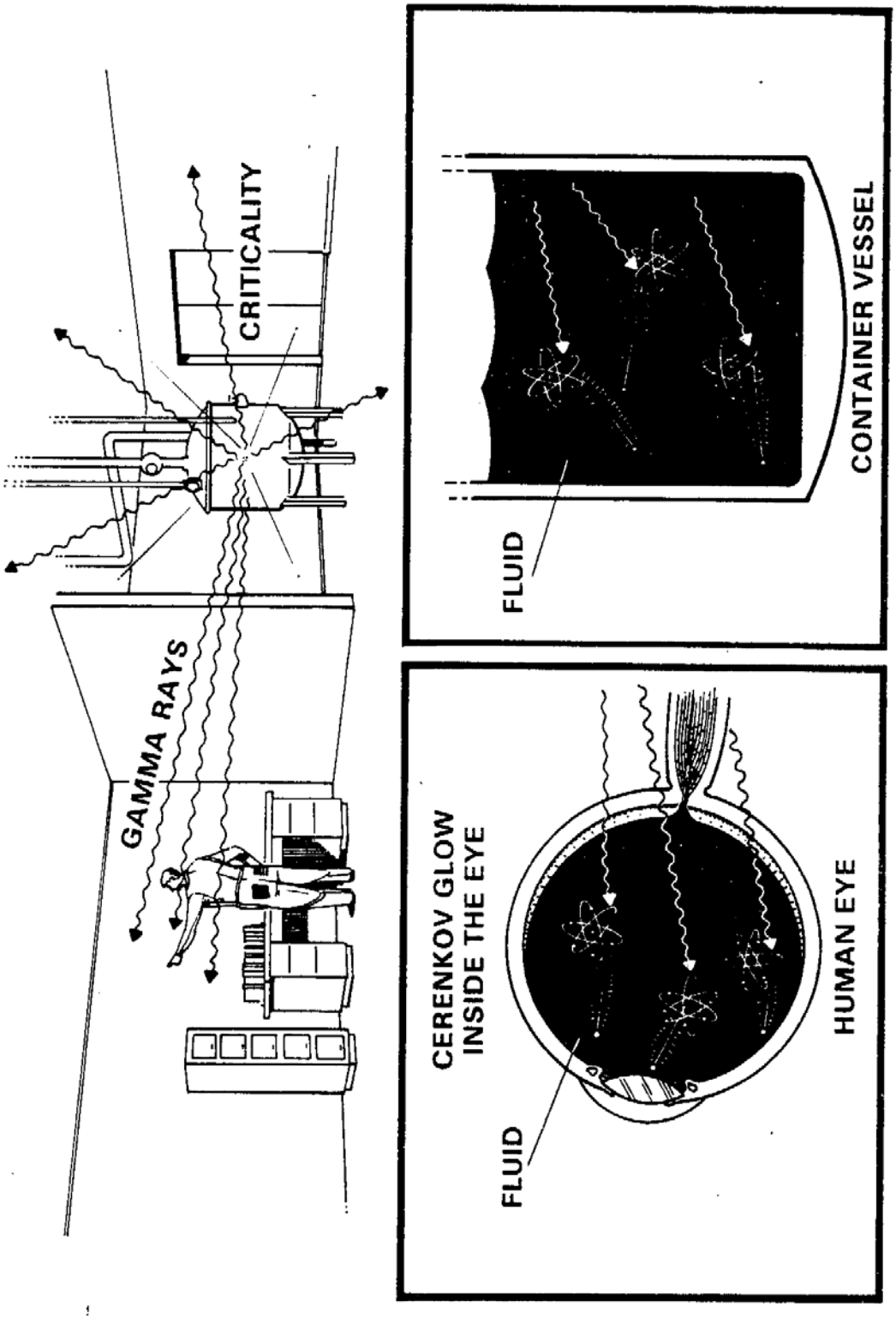


Figure 64. Criticality and the "Blue Flash"

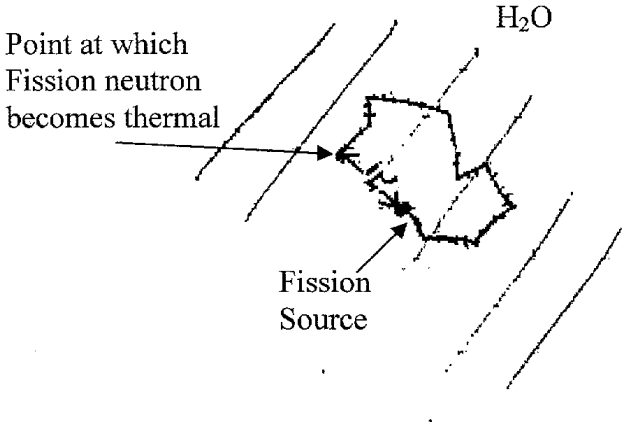
The fissioning of 9 x 10^{16} atoms per second results in a power level of ~3 million watts! Hence the fission power is controlled entirely by the α-decay of the parent, irrespective of the degree of super criticality of the ^{232}U mass that is present.

The power level then "jumps" to ~3 million watts from fission plus 67.3 kW (α-decay) once the critical mass of ^{232}U has been formed.

The tremendous heat generation in this case makes all this rather hypothetical (the ^{236}Pu and ^{232}U would be rapidly melted or vaporized as a consequence of the heat), but otherwise the scenario would be feasible.

Z. Universal Safety Container Size for Subcritical Limits

A question arises as to whether there is a container size, or volume, that would be subcritical for all of the 113 or so fissile isotopes of the actinide group of elements that are believed capable of supporting chain reactions. Provided the container is "isolated" from other nuclear materials, it appears that such a volume limit can be specified. Minimum critical masses for fissile nuclides occur for moderated systems. Irrespective of the fission cross sections of the nuclides in question, the neutrons must slow down and be moderated for the thermal cross section to be effective. The Fermi age is equal to one-sixth the average of the square of the crow-flight distance from the point, where a fission neutron is generated to the point where it becomes thermal, or τ(age) = 1/6 $\bar{r}^2$. Since the age in water is about 33 cm^2, & $\bar{r}^2$ = 14 cm. In the case of minimum critical masses for aqueous solutions of fissile materials, the intention is to slow down the fission neutron (avg. Fission energy ~ 2 MeV) in as small a volume as practical. This can be accomplished only if the characteristic dimensions of the water moderator are significantly larger than the slowing-down length, i.e., the moderator size must be greater than $\bar{r}^2$; the neutrons must be thermalized within the "critical volume" if the large thermal fission cross sections of these fissile nuclides are to be effective.



$$T(age) = \frac{1}{6}\bar{r}^2\,(cm^2)$$

Data on a number of fissile nuclides are provided by M. Srinavasan, et al.[23] These data are reproduced here in Table XVI.

TABLE XVI. Data on a Number of Fissile Nuclides

			20 cm Thick Water Reflected	
	Core (g/l)	Crit R (cm)	Crit V (ℓ)	Crit Mass (g)
$_{92}{}^{233}U$	60	13.22	9.68	581
$_{92}{}^{235}U$	55	16.05	17.32	953
$_{94}{}^{239}Pu$	45	14.36	12.40	558
$_{94}{}^{241}Pu$	30	14.71	13.33	400
$_{95}{}^{242m}Am$	5	12.18	7.56	38
$_{96}{}^{243}Cm$	40	13.16	9.54	382
$_{96}{}^{245}Cm$	12	10.73	5.17	62
$_{96}{}^{247}Cm$	350	12.30	7.79	2728
$_{98}{}^{249}Cf$	12	10.30	4.58	55
$_{98}{}^{251}Cf$	5.5	15.23	14.79	81

Note that the critical radius of the water reflected spheres does not differ all that much from 14 cm, which comes from the age equation. The calculated numbers are used here primarily for purposes of illustration. Note that even though the fission cross section varies between 583 barns (^{235}U) and 6,600 barns (^{242m}Am , the critical volumes are reasonably constant reflecting the fact that a certain volume of water is required to thermalize the neutrons appropriate to these cross sections.

The smallest calculated critical mass, as reported by Srinavasan et al.,[23] for any of the fissile actinides (^{242m}Am-242) is only ~38 g. This amount is about 15 times less than that for ^{239}Pu. The critical volumes of the aqueous solution containing these masses differ, however, by only about 2/3 at 7.56 and 12.4 liters, respectively, though, as noted, the critical masses differ by a factor of nearly 15.

As is well known, the minimum critical mass for a fissile nuclide occurs under conditions of optimum moderation. The critical volume, however, will always be determined by the spherical volume of water needed to thermalize the neutrons. There is no case where this critical volume is less than ~4.6 liters. Based on these data a minimum critical volume of about 4.5 liters might be selected for the fissile nuclides.

1. Universal Containers Limit (~4.5 Liters)

Container volume in which any subcritical mass limit from ANS-8.15[19] might be placed, as based on the water reflected minimum critical masses (and volumes) for aqueous homogeneous fissile solutions is approximately 4.5 liters.

If the container is either greater or smaller than this “universal” volume (see Figure 65), the critical mass for an aqueous-homogeneous solution will be larger than the subcritical limit. There is nothing unusual about the latter statement except that one container size, if suitably chosen, would be suitable for containment of the subcritical mass limits of all the solutions of fissile nuclides, and it would not matter how the nuclear material was distributed within the container.

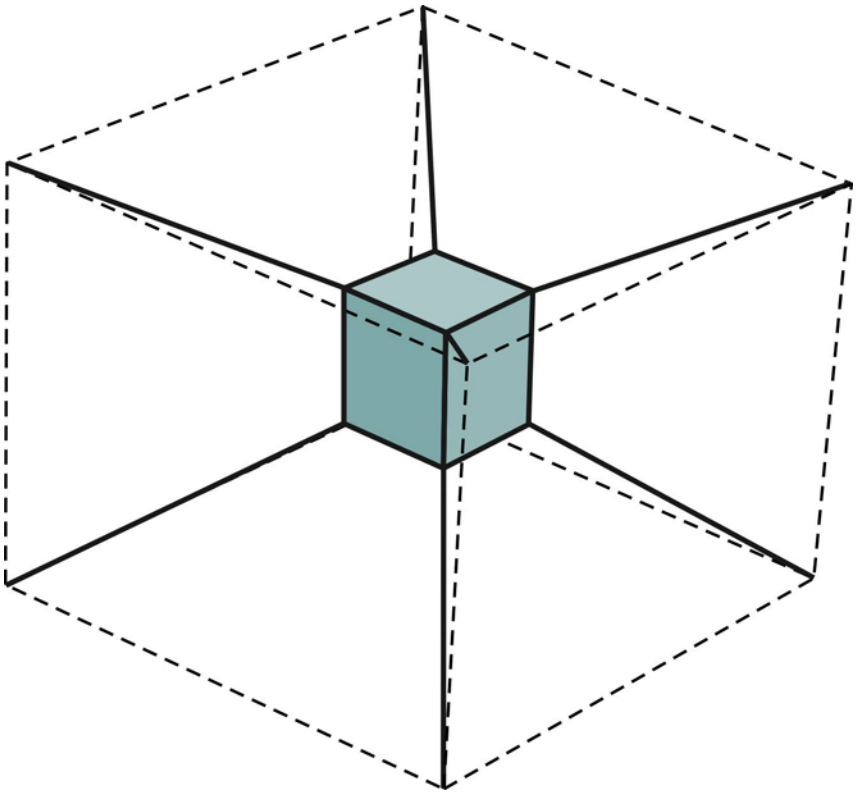


Figure 65. Universal Container Limit (~ 4 ½ liters)

ZA. 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 preceding examples serve 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 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 may not be the configuration of least mass after all; 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. Surprisingly, 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 become very small. Due to the lower energy of the delayed neutrons, the effective delayed neutron fraction can become very small, though not zero. Under

these circumstances, the difference between delayed and prompt critical is very small and delayed and prompt critical are in practice one and the same.

We have seen where a homogenous 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 unity 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 is also 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 that 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.

ZB. 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."[c]

In the early days of aviation, it was recognized that most accidents occurred during takeoff or landing, something still true today. In those times, the statement was made that takeoffs 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 "takeoff" or final stage at which the assembly became chain reacting, was always a point of more than casual interest. Many years ago, experiments were performed on graphite-uranium subcritical piles showing for the first time that a natural uranium, graphite-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 authors of these experiments were also 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 their consternation, 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. The experiments on the small graphite-uranium piles that served as the basis for the lattice design had been carefully 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. Once this was 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 the 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 was achieved. Normally, one thinks of adding fuel to gain reactivity, not removing it; reactors are made critical by adding fuel.

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[c] Omar Khayyam – from the Rubaiyat, 1120 A.C.E.

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 safety and routinely and with as much precision, as the takeoff or landing of a modern jet.

The experiments or calculations, which form the bases of nuclear criticality safety and control, were performed by a special breed of person, criticality experimenters, many of whom 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 stated, there are those individuals who live out their lives in a state of quiet desperation, but for those of us fortunate enough to be involved in the field of nuclear energy during its early stages of development, there have 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 were none.

> "All the knowledge we mortals can acquire is not knowledge positive but knowledge comparative subject to the errors and passions of humanity" (Bulwer Lytton [Edward George Earle, first Barton Lytton], 1803-1873).

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.

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