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C-46 Criticality Hazards
M-3679 (21st Ed.)

Plutonium Graphite Assemblies

by
A. Goodwin, Jr.
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THE DOW CHEMICAL COMPANY

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ROCKY FLATS PLANT DENVER, COLORADO

U.S. ATOMIC ENERGY COMMISSION CONTRACT AT (29-1)-1106

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ABSTRACT

Neutron multiplication measurements and theoretical calculations were made on cylindrical assemblies of graphite and plutonium discs.

ACKNOWLEDGMENTS

These tests were made possible by the cooperation of Mr. I. B. Venable and staff. The S_n calculations were performed at the Los Alamos Scientific Laboratories with the cooperation of Dr. Hugh Paxton, Dr. Gordon Hansen, and Mr. Bengt Carlson.

We would also like to thank Mr. H. V. Duba for his assistance in illustrating the report and M. G. Arthur for his review of this report.

1. INTRODUCTION

Neutron multiplication measurements were made on cylindrical disc assemblies constructed of alternate layers of graphite 1/2 in. thick and plutonium metal sheet. The number of individual sheets of metal per layer of graphite was varied from one to six.

Solid cylindrical slabs of metal, graphite end reflected, were also constructed from multiple layers of the metal sheet. The end reflector thicknesses ranged from 1/2 in. to 2 in.

Multigroup age theory (40 to 50 groups) and the LASL S_4 transport code using 16 energy groups were employed to check the experimental mass extrapolations of several arrays.

2. EXPERIMENTAL MATERIALS

The measuring equipment used in these experiments included scalers, Atomic Model 1050-A, coupled to G.E. B^{10}-lined counters encased in 8-in. diameter polystyrene moderators.

2.1 Materials

2.1.1 Graphite (Moderator and Tamper) National Carbon Grade CS-312

A) Moderator dimensions:

Discs 14 in. in diameter and 1/2 in. thick

B) Moderator and tamper density:

1.76 g/cm^3

2.1.2 Fuel

A) Plutonium density approximately 15.8 g/cm^3

B) Size of fuel pieces

1. 13.5 x 0.056 in.
2. 13.25 x 0.054 in.

C) Average weight of fuel piece

1. 2100 g
2. 1940 g

3. THEORETICAL

Multigroup age-diffusion calculations were made on homogeneous mixtures of plutonium and carbon. The Pu-C ratios used in the calculations are those corresponding to the experiments with one and two sheets of plutonium per 1/2 in. of graphite.

These calculations yield a critical buckling, spectrum and extrapolation length. The method of calculation used is given in the Appendix. The cross sections used were read from the curves in BNL-325[(1)] and its supplement with rough averaging in the resonance region. The spectra are fast enough that this averaging is not too important.

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(1) Donald J. Hughes, John A. Harvey, "Neutron Cross Sections," USAEC Report BNL-325, July 1, 1955.

Donald J. Hughes, Robert B. Schwartz, Supplement No. 1, USAEC Report BNL-325, January 1, 1957.

Figure 1 shows the results of these calculations for C/Pu ratios of 20 and 10 corresponding to one and two sheets of Pu per 1/2 in. of graphite respectively.

These curves were drawn using the formula

$$B^2 = \left(\frac{2.4048}{R + \delta}\right)^2 + \left(\frac{\pi}{H + 2\delta}\right)^2$$

where B^2 is the calculated buckling and δ is the extrapolation length. Table I gives these calculated parameters.

TABLE I

	C/Pu = 10	C/Pu = 20
B^2	0.025	0.0135
δ	2.5 cm	2.6 cm

The extrapolation lengths were calculated by taking 0.71 times the average transport mean free path. The transport mean free path was averaged over the calculated spectrum. The curve in Figure 1 for C/Pu = 10 was calculated using a buckling of 0.023, derived from a 40-group treatment; however, by carrying the calculation out 10 more groups the buckling was increased to 0.025. This decreases the calculated sizes and increases the discrepancy between the calculated and experimental values. The correct calculated buckling is listed in Table I.

An experimental point has been plotted in Figure 1 for comparison. The calculated size is too small by approximately 10% on the radius or 15% on the height. This results in a calculated critical mass which is 15 to 20% low. Improvements could perhaps be made by a more careful selection of group cross sections. For example, self shielding was not taken into consideration.

The calculational procedure used here has some limitations besides the usual age theory approximations. The lethargy intervals must be chosen so that they are larger than ξ, the average logarithmic energy decrement, but not so large that the trapezoidal formula is not adequate. Also, age theory applies to gradual slowing down and is not applicable to inelastic energy transfers. For these reasons it was not used on the pure metal calculations.

S_n calculations were done on delta-phase plutonium metal and on a plutonium-carbon mixture corresponding to two sheets of plutonium per half inch of graphite. The S_n method is a numerical method for solving the transport equation.[(2)] A multigroup S_n calculation has been coded for the IBM-704 and is called the SNG code. It is applicable to sphere, infinite cylinder, and infinite plane problems.

Six-group cross sections were used in solving the metal problems whereas 16-group cross sections were used for the plutonium-graphite mixtures. Both sets of cross sections ~~and the carbon cross sections~~ were obtained from Los Alamos

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(2) Bengt G. Carlson, "Solution of the Transport Equation by S_n Approximations" USAEC report LA-1891, February, 1955.

Scientific Laboratory ~~Also, the~~ plutonium cross sections were obtained using a ~~procedure~~ developed by Los Alamos Scientific Laboratory.

The plutonium metal S_n calculations were performed on a bare infinite slab and a bare infinite cylinder. The calculations were done for delta-phase plutonium, but can be changed to other densities by changing all dimensions in inverse ratio to the change in density. The calculated dimensions are:

infinite cylinder radius - 4.33 cm

infinite slab thickness - 4.36 cm

for a density of 15.8 g/cm^3.

An interpolation to finite geometries can be made through the use of a constant buckling formula. The buckling and extrapolation length need to be determined and these can be found in several ways. The two dimensions calculated above are sufficient to determine both an extrapolation length and buckling. This method yields the following values:

$B^2 = 0.150$ $\delta = 1.87$ cm

Another method of obtaining the extrapolation length is to graphically extrapolate the calculated flux in each group to zero. This yields six extrapolation lengths which then must be averaged. A better way would seem to be the following The SNG code calculation provides a spatial fissioning distribution. Since a one-group flux is proportional to the number of fissions per cubic centimeter one can extrapolate the fissioning distribution for the slab graphically to zero as shown in Figure 2. The extrapolation length obtained is 1.45 cm. This extrapolation length and the size of the critical slab gives a buckling of 0.187.

The fact that the above extrapolation length is smaller than the one determined from the dimensions is not surprising. The S_n method solves the transport equation and so gives the sum of asymptotic and non-asymptotic fluxes. The correct extrapolation length to be used in the buckling equation is the extrapolation length for the asymptotic flux, and this is larger than the extrapolation length of the asymptotic plus non-asymptotic fluxes.

Table II shows a comparison of critical masses calculated by the above two sets of buckling and extrapolation length. Also listed in Table II are some experimental values obtained by extrapolation of multiplication curves.

The method of obtaining a buckling and extrapolation length from the infinite slab and infinite cylinder sizes is preferable because of the reasons stated above. This is further emphasized when one calculates the critical mass of a bare sphere using the two methods. The values obtained from the dimensional analysis is 16 kg and the values from the graphical extrapolation is 13 kg. The experimental value is slightly more than 16 kg.

TABLE II

A. End Tamper Thickness (in.)	0	0	0.5	1.0	1.5	2.0	3.0
B. Pu Sheet Diameter (in.)	13.25	13.50	13.25	13.25	13.25	13.5	13.5
C. Number of Sheet in Exp. Core ~~including the End Reflector~~	0	0	22	19	17	15	14
D. Total Length of Exp. Core Including the End Reflectors	-	-	2.31	3.12	3.92	-	-
E. Percent Air Void in the Core	-	-	5.20	2.97	1.27	-	-
F. Mass of Pu in the Exp. Core (kg)	-	-	42.7	36.9	33.0	31.5	29.7
G. Experimental Extrapolated Critical Mass (kg)	66	74					
H. Calculated Critical Mass	66.3*	68.7*					
	68.5**	70.7**					

* Values arrived at from extrapolation length $\delta = 1.45$ cm from S_n calculations and B^2 from the S_n value of the ∞ slab.

** Arrived at from equating B^2 and δ as calculated from ∞ slab + ∞ cyl. derived from S_n calculations.

Four S_n calculations on various geometries of Pu-C mixtures were performed using a C/Pu ratio of 10 corresponding to the experiment with two sheets of Pu per half inch of graphite. The average density of Pu in the mixture was 2.81 g/cm^3 and the average density of the graphite was 1.43 g/cm^3. These were obtained using a density of 15.6 g/cm^3 for Pu metal and 1.74 g/cm^3 for the graphite. These density values are actually slightly less than densities found later for the experimental components. The effect on the mass would be small; less than 2%. Three of the calculations were done with a reflector of graphite having the same average density as the graphite in the core. The fourth calculation (spherical geometry) was done without a reflector in order to determine the effect of the reflector. Table III gives the calculated sizes for the various geometries.

TABLE III

Geometry	Radius or Thickness of core (cm)	Reflector Thickness (cm)
Slab	17.10	1.07
Cylinder	14.29	1.06
Sphere	19.77	1.10
Sphere	20.47	0.0

From this table it can be seen that there is only a 2% difference between the total size of the reflected and unreflected spheres. This means that the reflector saving is nearly equal to the reflector thickness for the small reflector thicknesses. As a consequence of this, a constant buckling formula can be used to interpolate to finite cylinders. A buckling and extrapolation length were obtained from the slab and cylinder overall dimensions (core size plus reflector thickness) listed in Table III. The curve in Figure 3 was drawn using this buckling and extrapolation length. As a check on the use of the constant buckling formula, the radius of a sphere was calculated and found to be the same as the overall radius of the reflected sphere.

The experimentally determined (extrapolated) critical height of a 35.6-cm diameter slightly reflected cylinder was found to be 41 cm, whereas the calculated value as shown in Figure 3 is 44 cm. The agreement is quite good, considering the experimental errors. The experimental point shown in Figure 3 is from the same data as the point shown in Figure 1. However, it should be noted that in Figure 1 the cylinder diameter is assumed to be that of the metal graphite core, only 13.25 in. in diameter, whereas in Figure 3 it is assumed to be the outside diameter of the assembly which is 14 in.

The effect of inhomogeneity in the one and two sheet experiments should have a small effect on the comparison of experiment and calculation. This is so because the plutonium sheet was only a fraction of an absorption mean free path thick in the important energy range.

The neutron energy spectra are obtained from both the S_n and age-diffusion calculations. Figure 4 shows the spectra calculated by both methods. The ordinate is the flux per unit energy plotted on an arbitrary scale. The abscissa is the energy in Mev. A related function calculated by S_n is the number of fission neutrons produced by neutrons ~~above 3 kev~~. This function shows that 96% of the fission neutrons are produced by neutrons above 3 kev.

In Figure 4 the S_n calculation shows the maximum at a lower energy than the age-diffusion calculation. This is probably due in part to the fact that inelastic energy transfers are allowed in S_n while only elastic scattering is taken into account by the age theory. Also the elastic transfer cross sections for S_n were calculated assuming a $\frac{1}{E}$ flux and therefore are larger than would be obtained from the non-$\frac{1}{E}$ flux which actually occurs in the system.

3. EXPERIMENTAL RESULTS

The experimental results are tabulated in Tables II and IV.

Table IV and Figure 5 provide values for the critical masses of graphite and metal assemblies.

Table II gives the critical masses of graphite end reflected metal slabs. An extrapolation to the bare critical mass was attempted in Figure 6. The uncertainty of these extrapolations mask the tamping effect of the stainless steel glove box enclosure in which these experiments were performed. The experiments were carried out in glove boxes to prevent the spread of contamination.

Figure 7 and 8 are included as examples of the reciprocal multiplication plots. Figures 5 and 6 were drawn from extrapolations such as these.

Due to the warped nature of the plutonium sheet, C-clamps were needed to keep the air void to a minimum. The air void accounts for between 1.27% and 5.20% of the total assembly volume.

TABLE IV

Number of Metal Sheets* Per Graphite Thickness**	Mass (kg) of Metal in the Experimental Core	Core Height (in.) Experimental Core	Critical Mass (kg) Extrapolated	% Air Void in Experimental Core
2	65.96	11.06	96.0	1.90
3	58.2	7.25	74.9	1.66
4	50.44	5.56	62.7	2.69
6	52.38	-	61.6	-

* Circular plutonium sheets, 13.25-in. diameter, approximately 0.0543-in. thickness.

** Circular graphite discs, 14-in. diameter, 0.50-in. thickness.

APPENDIX

Age-diffusion formulation.

The age equation can be written as:

1) $$\frac{dq(u)}{du} + A(u)q(u) = f(u)$$ and

$$\frac{B^2}{3\Sigma_{tr}^{th}} \Phi_{th} - \Sigma_a^{th} \Phi_{th} = q_{th}$$ thermal flux equation(3)

This was derived using $\frac{q(E)}{\xi E \Sigma_t} = \Phi(E)$.

We have defined

$$f(u) = \nu \chi(u)$$

$$A(u) = \frac{B^2}{3\xi \Sigma_t \Sigma_{tr}} + \frac{\Sigma_a}{\xi \Sigma_t}$$

where

B^2 = buckling

ξ = average logarithmic energy decrement

Σ_t = total macroscopic cross section

Σ_a = macroscopic absorption cross section

Σ_{tr} = macroscopic transport cross section

ν = average number of neutrons per fission

$\chi(u)$ = fission spectrum

The condition for criticality

2) $$\int_0^{u_{th}} \frac{\Sigma_f}{\xi \Sigma_t} q(u)du + \Sigma_f^{th} \Phi_{th} = 1$$

is already in included in equation 1).

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(3) J. W. Webster, "Practical Reactor Theory," USAEC Report AECD-4083, October 21, 1953.

The solution to equation 1) is:

3) $$\sout{q(u)}\ q(u_{th}) = \exp\left[-\int_0^u A(u')du'\right] \int_0^u \exp\left[\int_0^{u'} A(u'')du''\right] f(u')du'$$

The general solution contains another term which has been dropped because we will choose E_o such that $q(o) = 0$. The value of E_o chosen was 10 Mev.

In solving these equations one guesses a value of B^2 and using the trapezoidal rule calculates q(u) from equation 3). This value of u is substituted into the left side of equation 2) again using the trapezoidal rule. If the result of this calculation is unity the correct B^2 is obtained. Actually one uses two or three values of B^2 and plots the left side of equation 2) versus B^2 and graphically finds the correct B^2.

Generally for fast systems one does not need to carry the calculation to thermal energies because q(u) is very small. Therefore the calculations are terminated when the contribution to equation 2) is negligible.

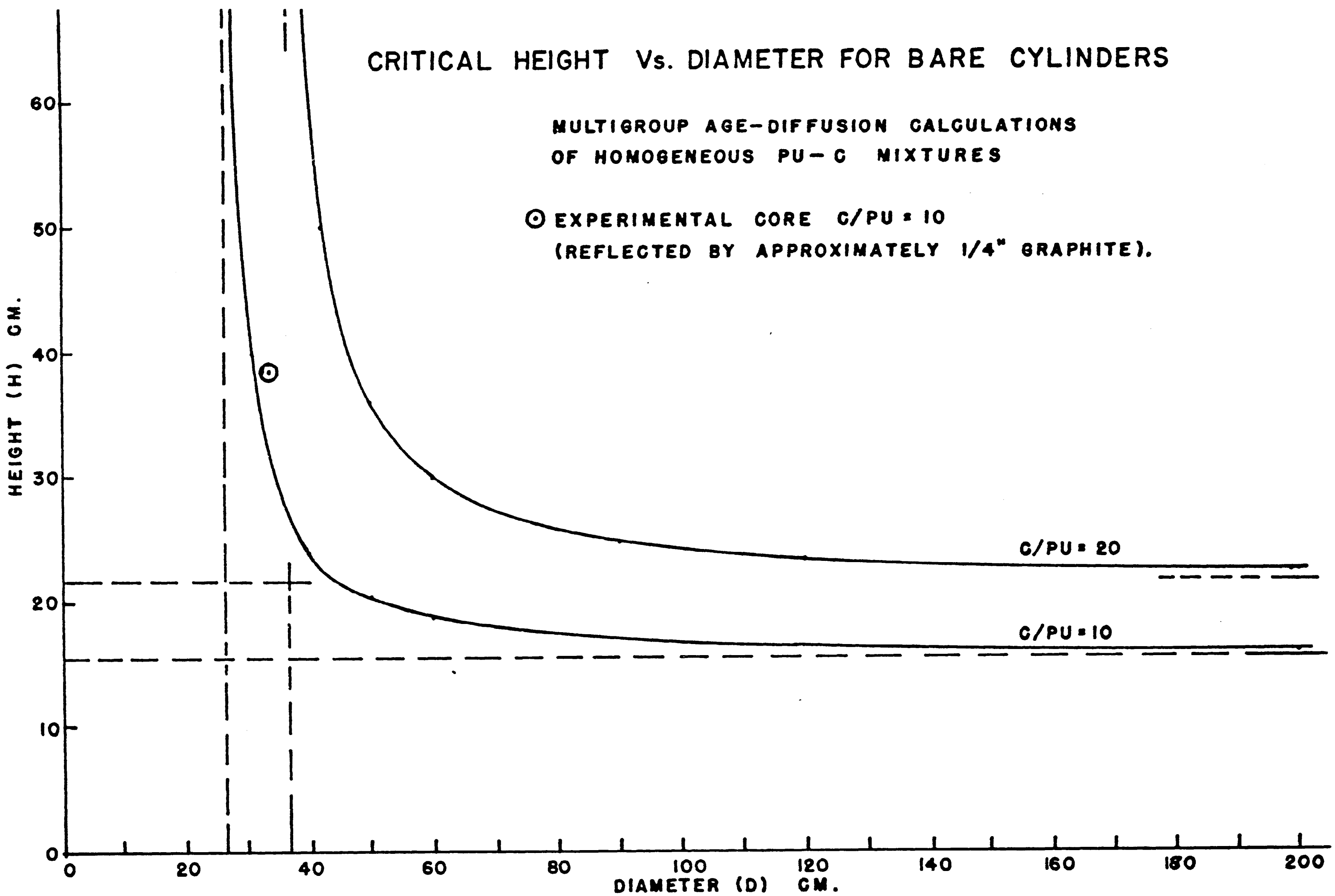
CRITICAL HEIGHT Vs. DIAMETER FOR BARE CYLINDERS
MULTIGROUP AGE-DIFFUSION CALCULATIONS
OF HOMOGENEOUS PU-C MIXTURES
⊙ EXPERIMENTAL CORE C/PU = 10
(REFLECTED BY APPROXIMATELY 1/4" GRAPHITE).
C/PU = 20
C/PU = 10
HEIGHT (H) CM.
0
10
20
30
40
50
60
0
20
40
60
80
100
120
140
160
180
200
DIAMETER (D) CM.

FIGURE 1.

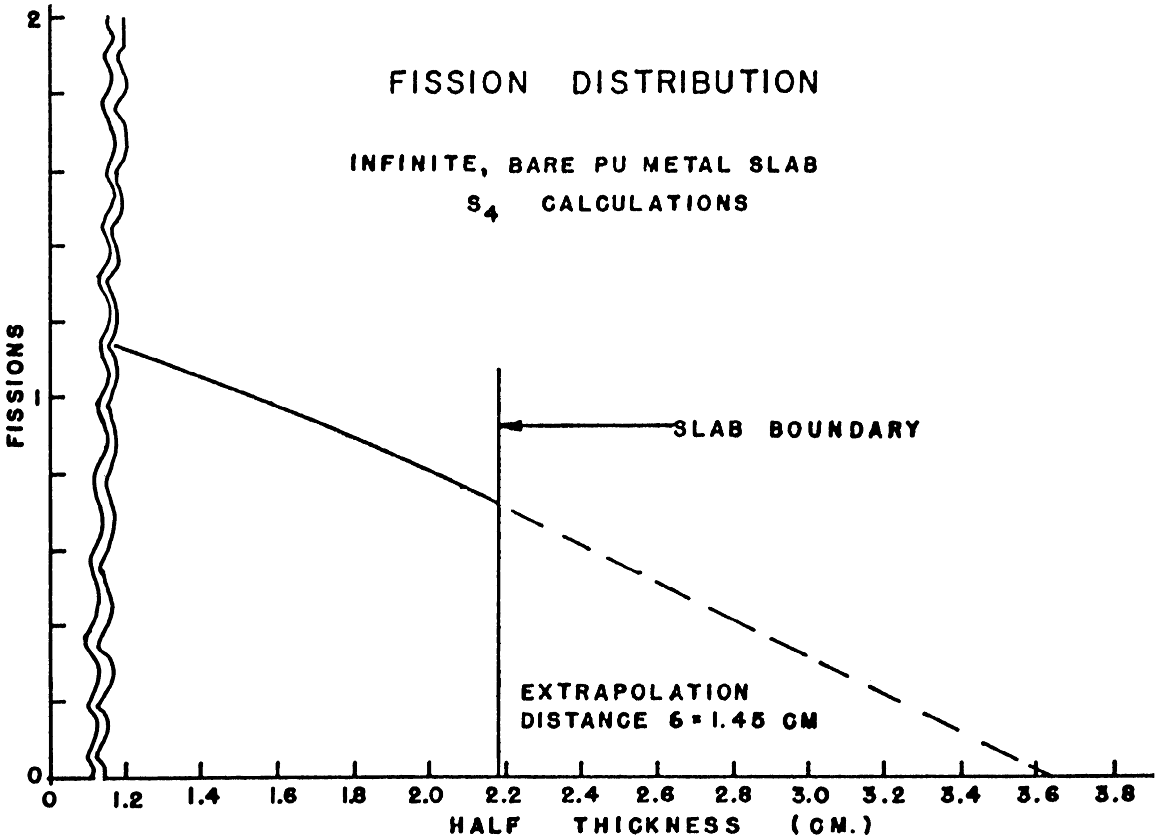

FIGURE 2

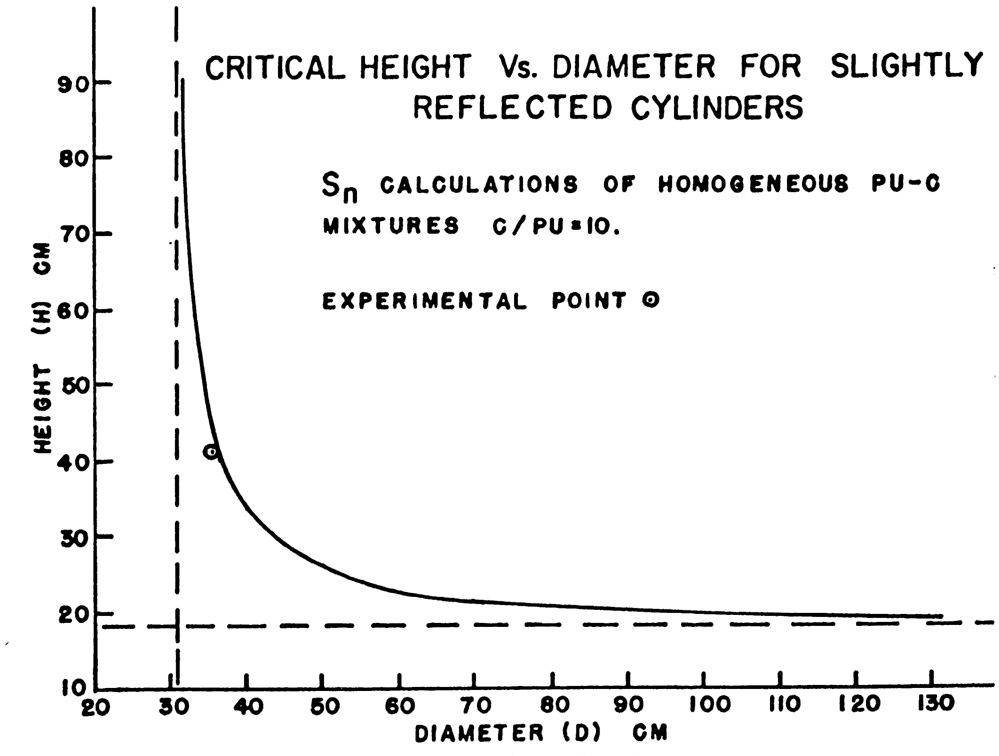

FIGURE 3

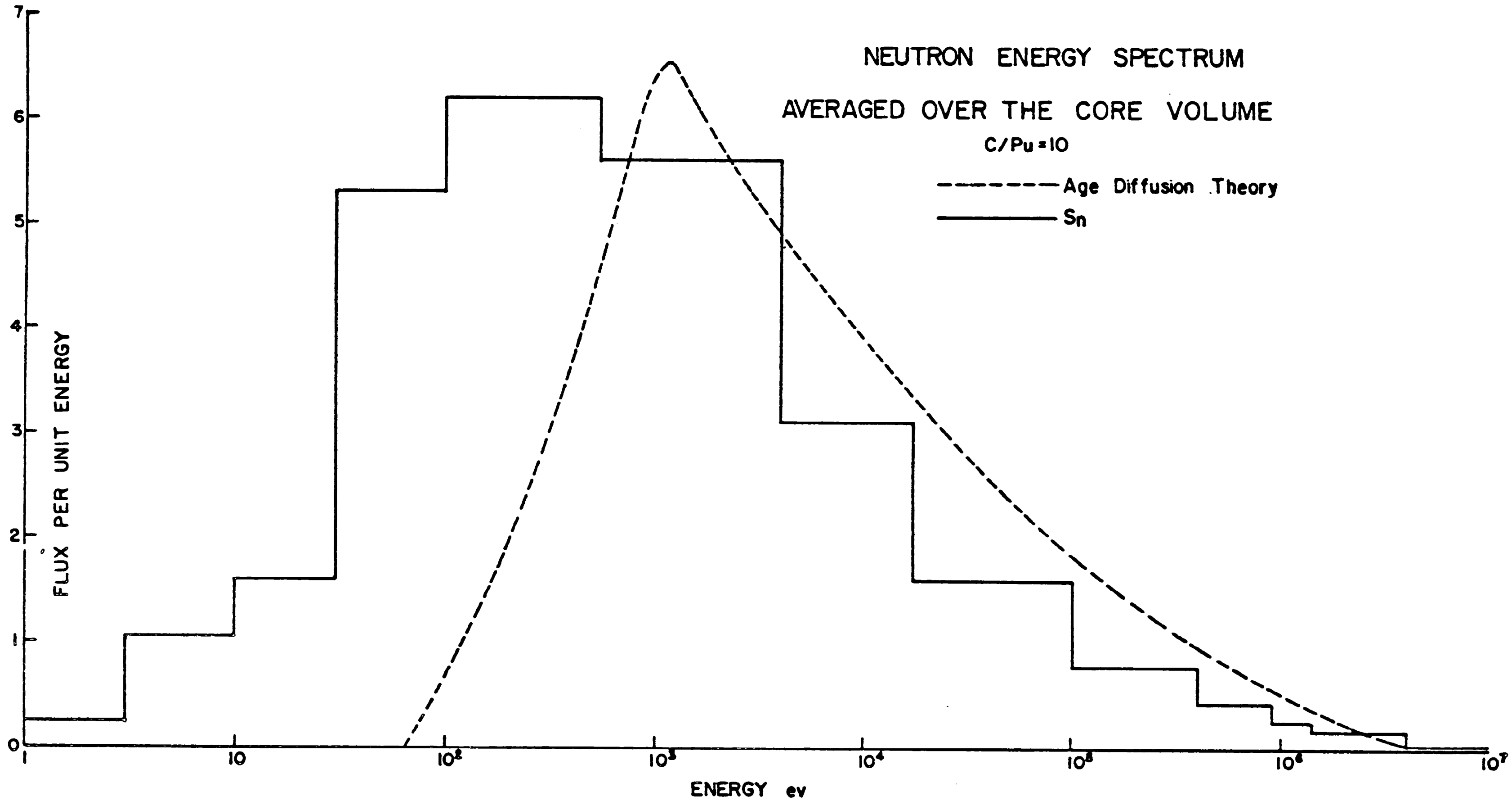
NEUTRON ENERGY SPECTRUM
AVERAGED OVER THE CORE VOLUME
C/Pu = 10
Age Diffusion Theory
Sn
FLUX PER UNIT ENERGY
ENERGY ev
0
1
2
3
4
5
6
7
1
10
10²
10³
10⁴
10⁵
10⁶
10⁷

FIGURE 4.

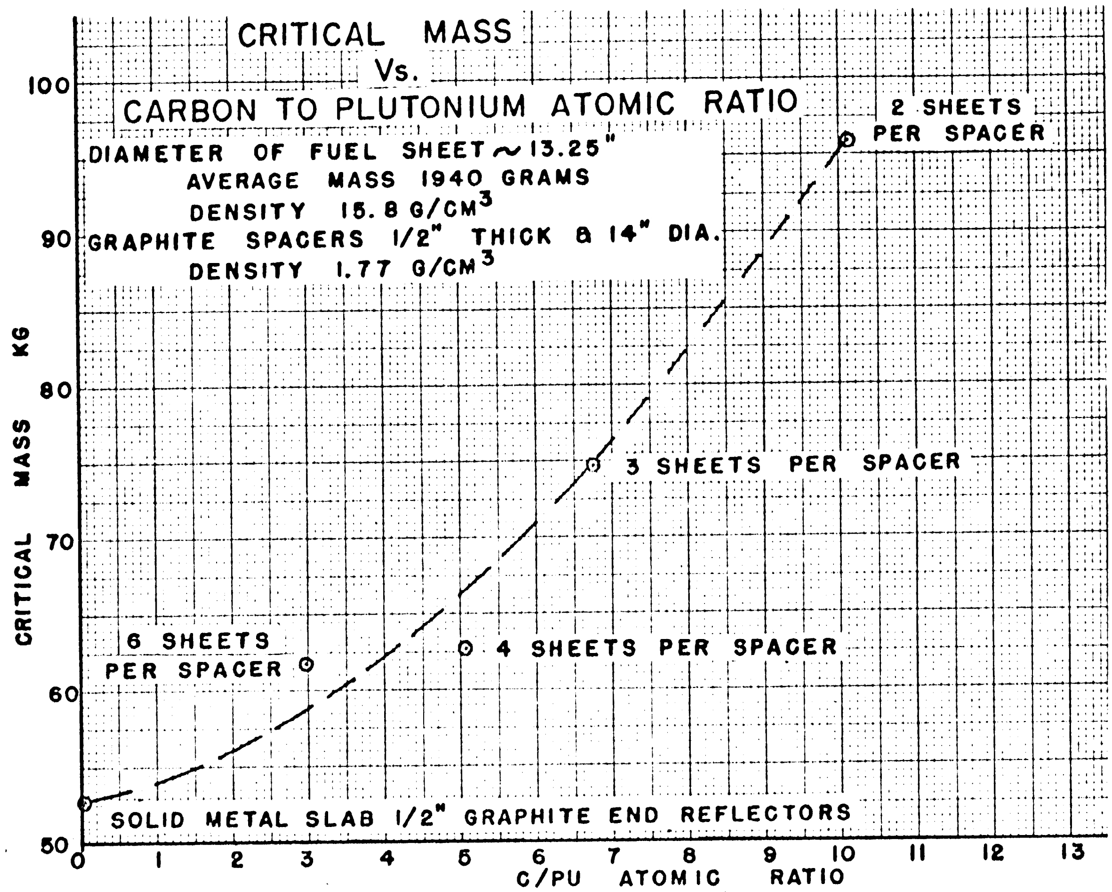

FIGURE 5

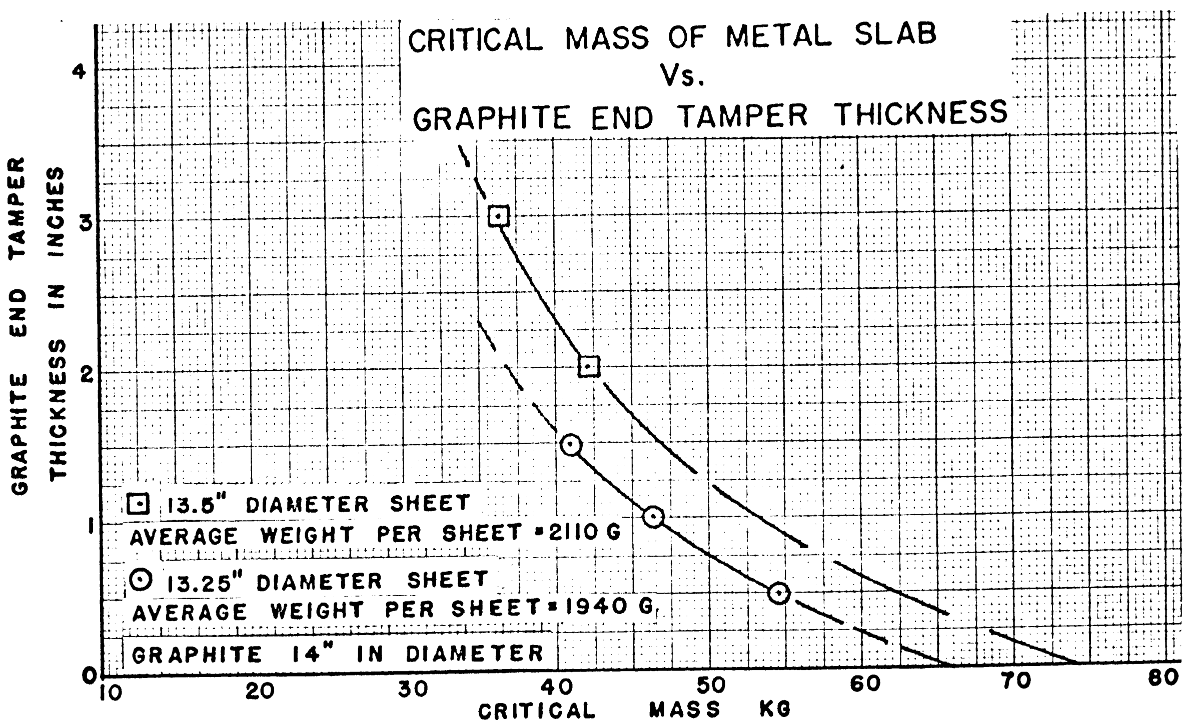

FIGURE 6

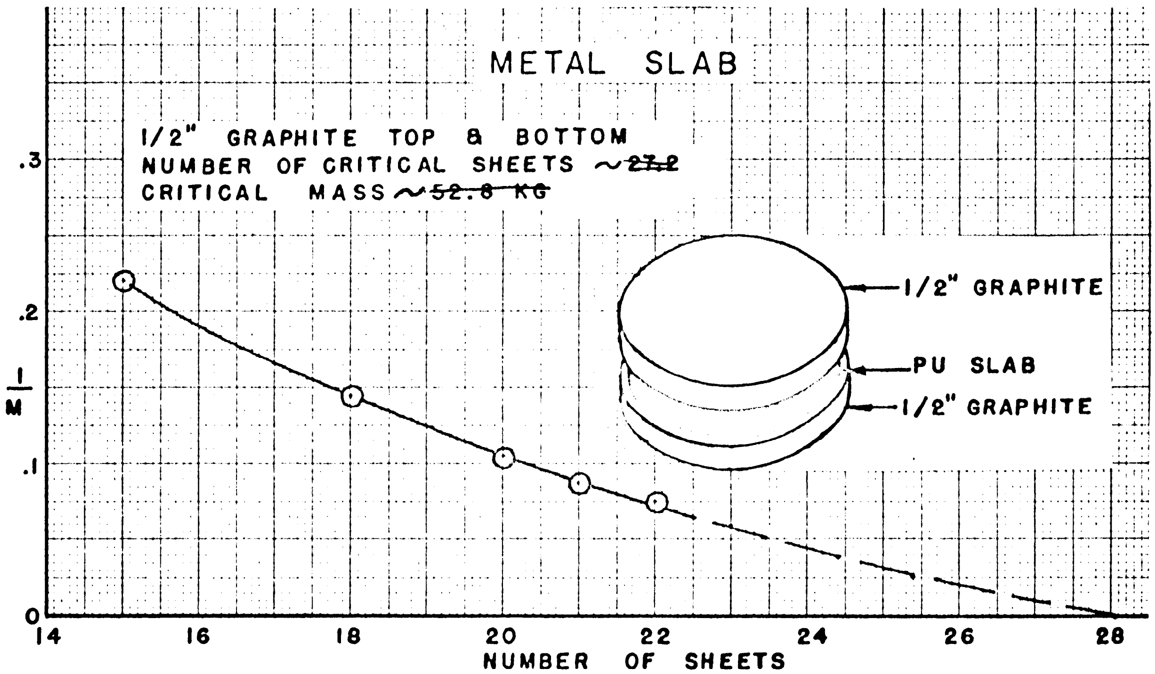

FIGURE 7

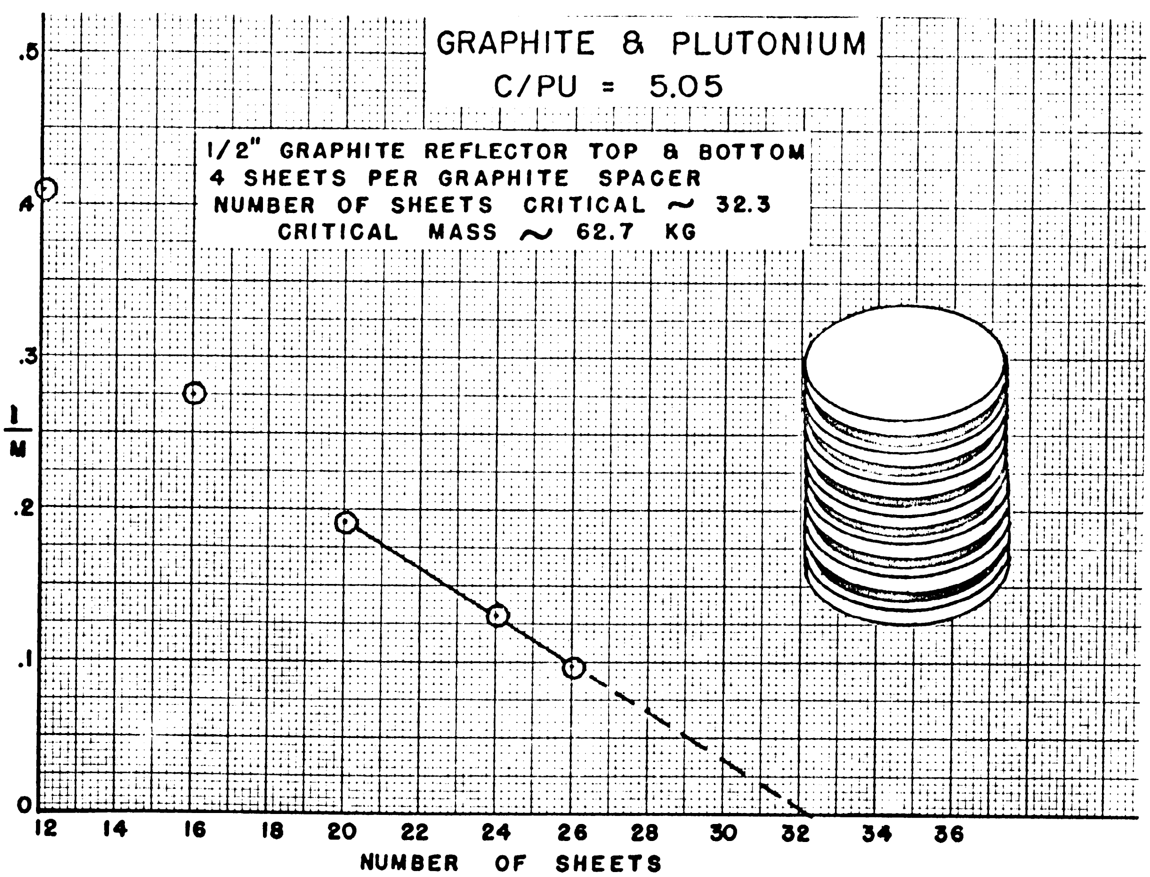

FIGURE 8