

[REDACTED]

UNCLASSIFIED

PUBLICLY RELEASABLE  
LANL Classification Group  
*ADam, 9/28/85*

LOS ALAMOS SCIENTIFIC LABORATORY

of the

UNIVERSITY OF CALIFORNIA

Report written:  
May 1953

LA-1548

This document consists of 31 pages

[REDACTED]

Classification changed to UNCLASSIFIED  
by authority of the U. S. Atomic Energy Commission,

Per H. Kab. L10 #16  
5-10-57

By REPORT LIBRARY 9 Feb 10-16-58

A SIMPLE METHOD OF CALCULATING

CRITICAL MASSES OF PROTON MODERATED ASSEMBLIES

Work done by:

George I. Bell  
Numerical computations by  
Max Goldstein and Group T-1

Report written by:

George I. Bell

[REDACTED]

LOS ALAMOS NATL. LAB. LIBS.  
3 9338 00414 7863

[REDACTED]

UNCLASSIFIED

• • • • •  
 • • • • •  
 • • • • •

## REACTORS--RESEARCH AND POWER

Distributed: SEP 21 1953

LA-1548

Los Alamos Report Library	1 - 20
AF Plant Representative, Burbank	21
AF Plant Representative, Seattle	22
ANP Project Office, Fort Worth	23
Argonne National Laboratory	24 - 34
Armed Forces Special Weapons Project (Sandia)	35
Atomic Energy Commission, Washington	36 - 40
Battelle Memorial Institute	41
Bechtel Corporation	42
Brookhaven National Laboratory	43 - 45
Bureau of Ships	46
California Research and Development Company	47 - 48
Carbide and Carbon Chemicals Company (ORNL)	49 - 56
Carbide and Carbon Chemicals Company (Y-12 Plant)	57 - 62
Chicago Patent Group	63
Chief of Naval Research	64
Commonwealth Edison Company	65
Department of the Navy - Op-362	66
Detroit Edison Company	67
duPont Company, Augusta	68 - 71
duPont Company, Wilmington	72
Foster Wheeler Corporation	73
General Electric Company (ANPP)	74 - 76
General Electric Company, Richland	77 - 80
Hanford Operations Office	81
Idaho Operations Office	82 - 88
Iowa State College	89
Knolls Atomic Power Laboratory	90 - 93
Massachusetts Institute of Technology (Kaufmann)	94
Monsanto Chemical Company	95
Mound Laboratory	96
National Advisory Committee for Aeronautics, Cleveland	97
National Advisory Committee for Aeronautics, Washington	98
Naval Research Laboratory	99
New York Operations Office	100 - 101
North American Aviation, Inc.	102 - 103
Nuclear Development Associates, Inc.	104
Patent Branch, Washington	105
Pioneer Service & Engineering Company	106
Powerplant Laboratory (WADG)	107
RAND Corporation	108
San Francisco Operations Office	109
Savannah River Operations Office, Augusta	110
Pratt & Whitney Aircraft Division (Fox Project)	111
U. S. Naval Radiological Defense Laboratory	112
University of California Radiation Laboratory, Berkeley	113 - 114
University of California Radiation Laboratory, Livermore	115 - 116
Vitro Corporation of America	117
Walter Kidde Nuclear Laboratories, Inc.	118
Westinghouse Electric Corporation	119 - 124
Technical Information Service, Oak Ridge	125 - 139
AF Plant Representative, Wood-Ridge	140
USAF-Headquarters	141

SECRET

UNCLASSIFIED

## ABSTRACT

Semi-empirical modifications of age theory have been made to interpret the critical masses of proton moderated assemblies, which may or may not have reflectors of various substances, in particular, water or steel.

Section I shows that critical masses of untamped water moderated systems can be calculated by use of an appropriate age and linear extrapolation length. Assuming that all captures and fissions take place at thermal, a familiar expression can be set up which contains the age and extrapolation length which are not well determined by theory. An age value suggested by theory (Sec. II) is taken, and ORNL criticality data (K343) are used to find the best extrapolation length. With these values the K343 critical masses can be computed to about  $\pm 5\%$ , which is their estimated experimental uncertainty; this for H/U-235 atomic ratios between 43 and 755.

Section II contains a theoretical discussion of effects which the simple "theory" of Sec. I neglects. It is indicated that several of these effects compensate to make a simple theory more useful than one would believe at first sight.

To take into account reflectors of water, water shielded by cadmium, or steel of various thicknesses, it proves sufficient to alter the extrapolation length appropriately. For moderators which contain appreciable  $\text{HNO}_3$ ,  $\text{H}_3\text{PO}_4$ , or steel, parameters are given in Sec. III which enable one to compute the appropriate ages and extrapolation lengths. Critical masses are again calculated to within about  $\pm 5\%$ .

In Sec. IV a simple mode generalization is suggested for large scale inhomogeneities in the fuel region which preserve a constant age throughout. The critical equations and numerical parameters are assembled in the appendix.

SECRET

UNCLASSIFIED

~~CONFIDENTIAL~~  
CONFIDENTIAL

I. CRITICAL EXPRESSION FOR UNTAMPED ASSEMBLIES.


Age theory is generally regarded as inapplicable to the slowing down of fission neutrons in hydrogenous substances. The age theory is invalid primarily because the hydrogen scattering cross section changes by a large amount in one slowing down interval.

One may incline toward more complex theoretical treatments such as multi-group methods. These manifest the disadvantages of complexity, particularly when one wishes to see quickly how conditions depend upon a number of parameters. Also, in the past, they have sometimes yielded results which were not in good agreement with experiment, presumably because the group parameters were not well known.

Our approach has been based on the wealth of experimental information on the critical masses of proton moderated assemblies. We started by constructing the simplest possible age type theory for the reactivity of a proton moderated substance which contains the age and/or extrapolation distance as adjustable parameters. We then found that we were able to fit all the K343 data on water moderated, untamped (or actually tamped by 1/16" stainless steel) assemblies with a single value of age and extrapolation length. Indeed it did not prove possible to improve the theory by comparison with experiment. The theoretical expression allows modifications in age and extrapolation length to account for a reflector and moderation by other hydrogenous substances than water, as will be discussed in Sec. III.

---

CONFIDENTIAL  
~~CONFIDENTIAL~~  
CONFIDENTIAL

  
 5110

Let us assume that no neutrons react during slowing down so that all capture and fission occur at thermal energies. Then we may write the multiplication factor,  $K$ , as

$$K = K_{\infty} P_f P_s \quad (1)$$

where  $K_{\infty}$  is the number of neutrons produced per neutron absorbed in the region containing active material,  $P_f$  is the probability that the neutron does not escape from the vessel during slowing down, and  $P_s$  is the probability that it does not escape while thermal. For a homogenous assembly with one fissionable element we have

$$K_{\infty} = \frac{\nu}{1 + \sum_i N(i) \sigma_c(i) / N_f \sigma_f} \quad (2)$$


where  $\nu$  is the number of neutrons per fission,  $N_f \sigma_f$  the macroscopic thermal fission cross section and  $\sum_i N(i) \sigma_c(i)$  the macroscopic thermal cross section for capture without fission. If the fissionable atoms are U-235 then

$$K_{\infty} = \frac{2.12}{1 + \sum_i N(i) \sigma_c(i) / 650N(U-235)} \quad (2')$$

where the summation does not here include U-235.

Suppose next that neutrons of all energies are distributed in the same spatial normal mode of the assembly.  $P_s$  is then immediately given by one velocity diffusion theory and is

$$P_s = \frac{1}{1 + L_{th}^2 B^2} \quad (3)$$



SECRET

where  $L_{th}$  is the thermal diffusion length, namely  $\sqrt{\frac{l_t l_c}{3}}$  with  $l_c$  and  $l_t$  the capture and transport mean free paths of the solution at thermal, and  $B^2$  is the buckling or negative Laplacian of the neutron distribution. For a distribution of neutrons in the first normal mode of a cylinder

$$B^2 = \frac{(2.405)^2}{(R+x)^2} + \frac{\pi^2}{(H+2x')^2} \quad (4)$$

where  $R$  and  $H$  are the physical radius and height of the cylinder and  $x$  and  $x'$  are extrapolation distances. We assume  $x' = x$ , which is a good approximation for large cylinders. Presumably  $x \approx .71 l_t$  but we shall leave it unspecified.

For solutions of U-235 in water,  $P_s$  is usually close to unity and the least important factor in (1). As one varies the H/U-235 ratio of such solutions from 30 to 750,  $P_s$  changes from about .995 to .960.

For most non-hydrogenous moderators, the probability,  $P_f$ , that a neutron does not escape during slowing down is given by an expression

$$P_f = e^{-\tau B^2} \quad (5)$$

where the age  $\tau$  is given by

$$\tau = \frac{1}{3} \int_0^u l_t(u) l_{sl}(u) du. \quad (6)$$

Here  $u = \ln(E_0/E)$  with  $E_0$  the source energy,  $l_{tr}$  and  $l_{sl}$  the transport and slowing down mean free paths. In Sec. II we show that (6) is invalid for proton moderation and derive the correct expression for an energy independent space distribution. The correct  $\tau$  is a complicated

SECRET

SECRET

function of  $B^2$ . However for a large range of H/U ratios  $\tau$  may be expected to be essentially constant. Thus as one varies the H/U-235 ratio from 30 to 750,  $B^2$  changes by about a factor two, and  $\tau$  by no more than a few percent, as will be seen in Sec. II.  $\tau$  is expected to be about  $27 \text{ cm}^2$ .

Because the transport mean free path for such moderators as water changes rapidly above around 10 kev neutron energy, the spatial distribution cannot in fact be independent of energy. This means that the extrapolation length is undetermined. We discuss this matter briefly in Sec. II but propose to regard  $x$  as a parameter to be determined by experiment. To test our expression and, if possible, fix  $x$  and  $\tau$ , we first, for a few values of  $x$ , check the constancy of  $\tau$  by using critical data from K343. In Table 13 of K343 are listed critical conditions for nineteen cylindrical stainless steel reactors without water reflectors. The steel was 1/16" thick, the H/U-235 atomic ratio varied from 44 to 755, and the ratio of height to diameter varied from 1.84 to 0.28. Reactors of four different diameters were used, namely 10, 12, 15, and 20 inches.

For three values of  $x$  which for simplicity we applied as equal for thermal and fast leakage, we computed from eqns. (1), (2'), (3), (4), and (5), and for each of the nineteen reactors, that value of  $\tau$  for which  $K = 1$ . For a given  $x$ , the relative rms deviation of  $\tau$  from its average value is about 2%. This means that when we use such an average  $\tau$ , the computed bucklings for the 19 cases have a relative rms devia-

SECRET

SECRET

tion of about 2% from the experimental ones. If we assume the same ratio of height to diameter for the calculated and experimental critical dimensions, we thus compute the critical masses to within about +3%.

Calculations were made for three values of the extrapolation length and results are indicated in Table I and Figure I.

TABLE I

Ages and extrapolation lengths for 19 untamped water moderated reactors

Extrapolation length, cm	2.5	3.5	4.5
Average age, $\bar{\tau}$ , cm <sup>2</sup>	21.7	24.5	27.7
$(\bar{\tau} - \tau)^2$ <sup>1/2</sup> cm <sup>2</sup>	.72	.49	.38
Maximum value of $ \bar{\tau} - \tau $	1.72	1.00	1.08

It appears that an extrapolation length near 4.5 cm is a good one. Larger extrapolation lengths do not give improvement. We have taken  $\tau = 27 \text{ cm}^2$ , as suggested by the theory of Sec. II, and the corresponding  $x = 4.3 \text{ cm}$ . Using these values we can compute critical masses to within better than 3% rms or about 6% at worst. In quoting this accuracy we imply that the experimental and calculated ratios of diameter/height are taken essentially equal. If instead either the height or radius is fixed, then the uncertainty in the critical mass should be computed from the uncertainty in buckling, namely 1.5% rms or 4% max.

Considerably better agreement between computed and experimental critical masses can be obtained by allowing  $\tau$  to vary slightly with



•• •• •• •• ••

reactor size. However since already we have agreement to within approximately the estimated experimental uncertainty (+5% L.E.) it is unclear that such improvement would be real and not merely a reflection of systematic experimental deviations with size or H/U ratio. We merely note that  $\tau$  should probably be taken larger for H/U ratios  $< 75$ . If we eliminate the three cases of such solutions, the remaining critical masses can be computed to about 2% L.E. with one set  $(x, \tau)$ .

## II. THEORETICAL DISCUSSION.

In writing down our simple expressions of Sec. I, we made a number of assumptions which are difficult to justify. In this section we shall attempt to show why some, due to compensating effects, may be better in practice than in principle.

First of all we assumed that capture and fission occur only at thermal energies. This will evidently be a good approximation for sufficiently large H/U ratios. An estimate of the probability of a neutrons being captured during slowing down is given by

$$1 - \exp \left\{ - \int_0^u \frac{\sum_c(u')}{\sum_H(u') + \sum_c(u')} du' \right\}$$

where  $\sum_c$  and  $\sum_H$  are the macroscopic capture and hydrogen scattering cross sections. For an H/U-235 ratio of 200 the probability of capture during slowing down in water is about 5 or 10%, and proportionally larger for smaller H/U ratios.

For such H/U ratios, it is unclear whether the effect of these

•• •• •• •• ••

0 1 1 1 1 1 1

epi-thermal captures is to enhance or to reduce the reactivity, as there are two partially compensating effects present. If the average number of fission neutrons produced per capture were independent of the energy of the captured neutron, then epi-thermal capture would perforce enhance the reactivity. However the ratio of radiative capture to fission (in U-235 anyway) is known to be substantially larger for epi-thermal neutrons than for neutrons of .025 ev. The ratio for thermal neutrons is about 0.18, and it appears to be on the average larger for all higher neutron energies up to of the order of 100 kev. This effect per se reduces the reactivity. However, the average number of fission neutrons per capture probably does not change so strongly with energy as the change in the ratio of radiative capture to fission in U-235 would suggest. This is because the U-235 capture and fission cross sections decrease considerably less rapidly than  $1/v$  for energies above thermal while the capture cross section of hydrogen and probably the capture cross sections of other likely elements (e.g., Al, Fe) decrease about as  $1/v$  up to the kilovolt region anyway.

An accurate calculation of the effect on the reactivity of the capture of neutrons while slowing down in reactors of relatively low H/U ratios is difficult. It would have to take into account the resonance structure of U-235, allowing for self shielding and for the variation of the capture to fission ratio with energy. We have not done such a calculation, but the rather good agreement of our simple formulae with experiment even for H/U ratios  $\lesssim 50$  tends to indicate

0 1 1 1 1 1 1

0 1 1 1 1 1 1

SECRET

that capture of epi-thermals does not much change reactivity, presumably due to the above-mentioned compensation.

-----

Secondly, let us investigate the extent to which the probability of a neutrons not leaking out while slowing down can be represented by the form  $e^{-\tau B^2}$  with  $\tau$  a constant.

Consider a homogeneous reactor. To begin with we assume that the neutrons are in a normal mode of the reactor independent of their energy. To date we have found it necessary to make this assumption to achieve simple analytic results. Next we assume that neutrons are slowed down only by collisions with hydrogen. We thus ignore slowing down by elastic and inelastic collisions with heavier atoms. The importance of the elastic collisions has been investigated by Marshak<sup>1</sup> and shown to be small--of the order a 2% decrease in  $\tau$  for water. The inelastic collisions are presumably unimportant for water and difficult to take accurately into account for any solution as inelastic cross-sections for fission energy neutrons are not very well known... Finally we ignore capture and fission while slowing down--this is not necessary, but a considerable simplification.

Consider first a monoenergetic source of neutrons. With the above assumptions, the slowing down equation in differential form is:

$$\left( \sum_H(u) - D(u) \nabla^2 \right) \phi(u, \vec{r}) = e^{-u} \int_0^u \phi(u', \vec{r}) \sum_H(u') e^{+u'} du' + \delta(u) \quad (7)$$

<sup>1</sup>R. E. Marshak, Reviews of Modern Physics 19, (1947) p. 238.

SECRET

SECRET

SECRET

Here  $\phi$  is the flux per logarithmic energy interval,  $u = \ln E_0/E$  with  $E_0$  the source energy,  $\sum_H$  the macroscopic hydrogen scattering cross section, and  $D$  the transport mean free path divided by three. Since we have assumed that the neutron spatial distribution is independent of energy, we can separate variables and solve eqn. (7) at once.

We write  $\nabla^2 \phi = -B^2 \phi$ , so that

$$\left( \sum_H(u) + D(u)B^2 \right) \phi(u) = e^{-u} \int_0^u \phi(u') \sum_H(u') e^{u'} du' + \delta(u).$$

Substituting  $\left( \sum_H(u) + D(u)B^2 \right) \phi(u) = f(u) + \delta(u)$ , and differentiating we see that the integral equation is equivalent to the differential equation

$$f'(u) = -f(u) \left( \frac{D(u)B^2}{\sum_H(u) + D(u)B^2} \right)$$

plus boundary condition

$$f(0) = \left( 1 + \frac{D(0)B^2}{\sum_H(0)} \right)^{-1}$$

$$\text{Thus } f(u) = \left( 1 + \frac{D(0)B^2}{\sum_H(0)} \right)^{-1} e^{-\int_0^u \frac{D(u')B^2}{\sum_H(u') + D(u')B^2} du'}$$

Now the probability of a neutron not diffusing out while slowing down to energy  $E$  is  $\sum_H(u)\phi(u)$ , or

$$\sum_H(u)\phi(u) = \left( 1 + \frac{D(0)B^2}{\sum_H(0)} \right)^{-1} \left( 1 + \frac{D(u)B^2}{\sum_H(u)} \right)^{-1} e^{-\int_0^u \frac{D(u')B^2}{\sum_H(u') + D(u')B^2} du'} \quad (8).$$

SECRET

If age theory were strictly valid for slowing down in hydrogen, instead of eqn. (8) we would have had

$$\sum_H(u) \phi(u) = e^{-\int_0^u \frac{D(u')B^2}{\sum_H(u')} du'}$$

Equation (8) tells us that more neutrons will leak out during slowing down than predicted by age theory.

Now we can equate equation (8) to

$$e^{-\tau_B(u)B^2}$$

and see to what extent  $\tau_B$  is independent of B. One finds that  $\tau_B$  varies by only a few percent while B varies by a factor 2 (from 0.015 to 0.030), this for slowing down from a few Mev to 1.4 ev. One finds also that  $\tau_B$  is quite close to  $L_s^2 = \langle r^2 \rangle / 6$  where the slowing length has been computed essentially on the same basis as  $\tau_B$  by Marshak et al (ref. (1)). That is, Marshak ignored only slowing down by heavy nuclei.

These results may be generalized from a monoenergetic source to a fission spectrum. Thus for slowing down from a fission source to 1.4 ev we may define  $\tau_B$  through:

$$e^{-\tau_B B^2} = \sum_H \phi(1.4 \text{ ev}) = \int_0^\infty \left(1 + \frac{D(1.4)B^2}{\sum_H(1.4)}\right)^{-1} \left(1 + \frac{D(E)B^2}{\sum_H(E)}\right)^{-1} f(E) \exp \left\{ - \int_{1.4}^E \frac{D(u')B^2}{\sum_H(u') + D(u')B^2} du' \right\} dE \quad (9).$$

SECRET

SECRET

displace water and decrease the hydrogen density.

Now decreasing the hydrogen density increases the age. The net effect of this displacement of hydrogen is that as one increases the uranium concentration and hence  $B^2$ ,  $\tau_B$  will increase. The displacement of hydrogen tends to compensate for the intrinsic variation of  $\tau$  with  $B^2$ . If one assumes that an atom of oxygen or fluorine associated with uranium displaces one hydrogen atom, which is approximately the case, then the expected variation in  $\tau_B$  for the reactors of Sec. I is reduced from about 10% to 5%.

Evidently for the reactors of Sec. I, a value  $\tau = 27 \text{ cm}^2$  for slowing down to thermal is reasonable. This was the value we used in Sec. I, and it arises from a value of about  $25 \text{ cm}^2$  for slowing to 1.4 ev plus about  $2 \text{ cm}^2$  for slowing from there to thermal.

In discussing the fast leakage we have assumed that the spatial distribution of neutrons is independent of energy, or that the extrapolation length is independent of energy. Since the transport mean free path varies so strongly with energy for neutrons above a few tens of kilovolts, this is not a very good approximation. However, for every reactor there is of course some extrapolation distance which when used in  $B^2$  correctly gives the probability of escape during slowing down. If we can show that the extrapolation distance does not vary much for a wide range of reactors, this will lend further plausibility to the results of Sec. I.

If the transport mean free path varied by only a small fraction of itself in a slowing down interval, then the spatial distribution of

SECRET



neutrons at any energy would be essentially determined by the transport mean free path at that energy. If we defined an average transport mean free path by

$$\bar{\lambda} = \frac{\int_{E_{th}}^{E_0} \lambda(E) \phi(E) dE}{\int_{E_{th}}^{E_0} \phi(E) dE}$$

with  $\phi$  the flux, then the extrapolation distance would be  $0.71 \bar{\lambda}$ .

Group T-1 has computed  $\bar{\lambda}$  for various reactors with an approximate expression for  $\phi(E)$ , namely:

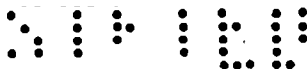
$$\phi(E) = \frac{e^{-B^2 L_s^2(E)}}{\sigma_H(E)E} \int_E^{\infty} e^{-B^2 L_s^2(E')} f(E') dE' . \quad (9')$$

Here  $L_s$  is the slowing down length given by Marshak, ref. (1) and  $f(E)$  is the normalized fission spectrum. For the reactors of Sec. I,  $\bar{\lambda}$  as so computed varies by only 2% or for all practical purposes is constant.

For water, the transport mean free path varies too rapidly to allow any such simple computation of the extrapolation distance. However, the above computations for slowly varying mean free path do not suggest that an extrapolation distance independent of reactor size (for a constant moderator) is a bad approximation.

-----

The above discussion can by no means be taken as demonstrating the validity of the simple expressions of Sec. I for predicting critical masses. This demonstration must come from a comparison with experiment.



The "theoretical discussion" is supposed only to make the agreement somewhat more plausible.

### III. EXTENSION TO VARIOUS TAMPERS AND MODERATORS.

Let us combine the equations of Sec. I to get the critical equation for a cylindrical reactor. At criticality,  $K=1$ , and we have

$$1 = \frac{\nu}{\frac{\sum_c(th)}{\sum_f(th)} + 1} \frac{1}{1 + L_{th}^2 B^2} e^{-\tau B^2} \quad (10)$$

with

$$B^2 = \frac{(2.405)^2}{(R+x)^2} + \frac{\pi^2}{(H+2x)^2} \quad (11)$$

For our reference solution--UO<sub>2</sub>F<sub>2</sub> in water at room temperature--the following constants were used:

$$\sigma_c (\text{U-235}) = 650 \text{ barns}$$

$$\sigma_f (\text{U-235}) = 550 \text{ b}$$

$$\sigma_c (\text{H}) = 0.330 \text{ b}$$

$$\sigma_c (\text{O}) = 0.00 \text{ b}$$

$$\sigma_c (\text{F}) = 0.00 \text{ b}$$

$$\nu = 2.50$$

$$x = 4.3 \text{ cm}$$

$$\tau = 27 \text{ cm}^2$$


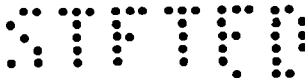
We found: 
$$\frac{\nu}{1 + \sum_c / \sum_f} = \frac{2.12}{1 + 0.00051C}$$

$$L_{th}^2 = \frac{8.20}{1 + 1970/C}$$

17





with  $C$  the atomic ratio of H to U-235. The other uranium isotopes in the alloy were ignored.


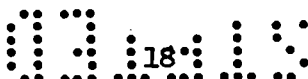
Hence for the reference solution we have the critical equation:

$$1 = \frac{2.12}{1+.00051C} \frac{1}{1+\frac{8.20B^2}{1+1970/C}} e^{-27B^2}. \quad (12)$$

If the reactor is operating at a temperature other than about  $20^\circ\text{C}$ , different thermal constants must be used. Thus  $L_{th}^2$  and  $\sum_c^{(th)} / \sum_f^{(th)}$  must be changed to correspond to the new thermal temperature. A different thermal temperature will per se produce essentially no change in  $x$ , and its small effect on  $\tau$  can be computed by subtracting  $\int_{.025}^{E(th)} \frac{D(E)}{\sum_H(E)} \frac{dE}{E}$  from 27. However, normally a change in temperature will produce a change in density. Evidently  $\tau$  and  $L^2$  change as  $(\text{density})^{-2}$  and  $x$  changes as  $(\text{density})^{-1}$ .

We now shall see that by changing  $x$  it is possible to use eqn. 12 to compute critical masses of suitably tamped reactors of the reference solution. We have found the appropriate values of  $x$  for reflectors of water, and water shielded by Cd by fitting data in K3<sup>43</sup>.

First we considered six water tamped stainless steel (1/16") reactors which had a cadmium sheet between the steel and reflector (viz. Table 9 of K3<sup>43</sup>). For the six cases,  $C$  was between 44 and 226. With  $\tau = 27$  and  $x = 5.6$  we fit the six critical masses to within  $\pm 2\%$  which is very likely fortuitous.

•• : •• : •• : •• : •• : •• : •• : •• : •• : ••

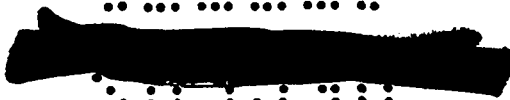
Twenty representative cases of stainless steel reactors with effectively infinite water reflectors were taken from Table 7 of K343. C varied between 24.4 and 755. For these cases,  $x = 7.0$  and  $\tau = 27.0$  gave the critical masses to within about  $\pm 5\%$  rms or  $7\%$  maximum, indicating that the formalism is somewhat less appropriate for the water tamper. This might well be anticipated, and one has of course better methods available for computing critical masses in such cases<sup>2</sup>, making use of the experimental distribution of slowed down neutrons from a point source.

Let us next consider how to treat other moderators than water, plus  $\text{UO}_2\text{F}_2$ , which has been our reference solution. In particular suppose we have  $\text{HNO}_3$ ,  $\text{H}_3\text{PO}_4$ , or stainless steel in our solution. First of all the thermal constants which determined  $K_\infty$  and  $L_{th}$  will be changed. The change in  $K_\infty$  is often important and can be calculated quite well from known thermal absorption cross sections. We used the following capture cross sections:  $\sigma_c(\text{N}) = 1.80\text{b}$ ,  $\sigma_c(\text{P}) = 0.20\text{b}$ , and  $\sigma_c(\text{stainless steel}) = 2.70\text{b}$ . The change in  $L^2$  is usually unimportant but easily estimated.

In addition both the extrapolation distance,  $x$ , and the age will change. The change in  $\tau$  could be estimated by using equation 9 with the appropriate cross sections for all elements in the moderator. In such a calculation the transport cross sections of the heavier nuclei for fission energy and slightly degraded neutrons are of cardinal importance. These, however, are poorly known and instead of going through

<sup>2</sup>Greuling, LA-399.

1955



•• : •• : •• : •• : •• : •• : •• : •• : •• : ••

SECRET

a full calculation for each solution with uncertain cross sections, we have used a simpler method for estimating the relative ages of an unknown and the reference solution.

We note that the age for slowing down from a monoenergetic source may be written

$$\tau = \int_0^u \frac{l_{tr} l_{sl}}{3} f(u') du'$$

where  $f(u')$  is for most of the range of integration close to unity while  $f(0)$  and  $f(u)$  are appropriate  $\delta$  functions to reproduce eqn. 8. The important thing is that  $f(u)$  does not depend much on reactor size. Thus one can define relative ages

$$\frac{\tau}{\tau'} = \frac{l_{tr} l_{sl}}{l_{tr} l_{sl}}$$

which are practically independent of buckling. Now for most hydrogenous moderators, the slowing down is essentially due to hydrogen and  $l_{tr}$  is inversely proportional to the hydrogen density. The slowing down by elastic scattering with oxygen in water has been considered by Marshak (loc. cit. p. 238) and shown to give about a 2% reduction in  $L_s^2$  or  $\tau$ . Hence we take the average slowing down cross section of an oxygen nucleus to be .04 that of a proton. For heavier nuclei such as Fe or Ni, there will be a slowing down contribution due to inelastic scattering of fission energy neutrons. Some information on the inelastic cross section for Fe is available<sup>3-4</sup>, and for fission energy neutrons it is of the order of .5 barn. We took the average slowing down cross section for

<sup>3</sup>Preston and Stelson, P.R. 86 132 (1952).

<sup>4</sup>R. Beyster--private communication.

SECRET

~~SECRET~~  
SECRET

Fe or stainless steel to be .2 that of a proton, which is very likely too large but in conjunction with other constants has given good agreement with experiment, as shall be seen.

In estimating the contributions of various nuclei to the average transport mean free path, we must note that the weighting is  $\sim(\sigma_H)^{-1}$  per logarithmic energy interval. This means that the high energy cross sections are weighted heavily. We have used the hydrogen and oxygen cross sections of Fig. II and the spectra of eqn. (9') for several values of  $B^2$ . We concluded that the contributions to the transport mfp per atom of oxygen and hydrogen are essentially equal. The contributions per atom of nitrogen and fluorine are presumably about the same, while those of phosphorus and iron are somewhat larger. The extrapolation length for a given reflector has been taken proportional to the above average transport mean free path.

We have had some success in computing critical masses with the following parameters:

$$\tau = 27 \frac{113.3}{\sum_i \gamma_i^{M_i + 4M_u}} \frac{166.7}{\sum_i \rho_i M_i} \quad (13)$$

$$x = x_0 \frac{166.7}{\sum_i \rho_i M_i}$$

with:

element	H	N	O	F	P	Cr, Fe, Ni	U
$\gamma$	1.0	0.04	0.04	0.04	0.02	.20	--
$\rho$	1.0	1.0	.0	1.0	1.3	1.5	2.0

21 ~~SECRET~~  
SECRET

SECRET

Here  $\eta_i$  is the slowing down efficiency of nucleus  $i$ ;  $\rho_i$  is its transport efficiency; and  $M_i$  is the molarity of the  $i^{\text{th}}$  constituent in moles per liter of solution. The middle factor in (13) represents the ratio of average slowing down mean free paths and the last factor the ratio of average transport mean free paths. The term  ${}^4M_{\text{U-235}}$  is supposed to represent the dilution of H by O and F in the reference solution. No such term occurs in the transport ratio since the transport mfp of the reference solution is essentially the same as for  $\text{H}_2\text{O}$ . Slowing down and epi-thermal capture and fission have been ignored for the uranium.  $x_0$  is the extrapolation distance for any reflector with the reference solution as moderator.

Equation 13 has been tested by comparison with critical experiments reported in K643 and some recent critical assemblies constructed by group W-2 at Los Alamos.

(1) In Table XI of K643 are listed four critical data for the solution  $\text{UH}_{316}^{\text{N}2.9}\text{P}_{53.1}^{\text{O}298}$  in aluminum containers (1/16"), three tamped by  $\text{H}_2\text{O}$  and one bare. Taking an extrapolation distance of 7.50 cm for the reference solution in aluminum containers with a water reflector, (viz. Table 5-U343) we compute the critical masses to within 5%.

(2) In Table IV of K643 are reported sixteen critical reactors containing relatively large amounts of nitric acid. Six are for Al cans without reflector ( $x_0=4.2$ ) and ten for aluminum cans with water reflectors ( $x_0=7.50$ ). Computed critical masses were within 6% of the experimental. The solutions were  $\text{UH}_{88}^{\text{O}65.7}\text{N}_{7.5}$ ,  $\text{UH}_{230}^{\text{O}136.7}\text{N}_{7.5}$ , and

SECRET

: : : : :  
: : : : :  
: : : : :  
: : : : :

${}^{235}_{92}\text{U}$   ${}^{185}_{80}\text{Br}$   ${}^{75}_{33}\text{As}$ . (Here and in (1), U refers to U-235 only.)

(3) Three experiments were performed by Group W-2 with 4.26 M phosphoric acid and .508 M  $\text{UO}_3$  in water--with a three inch steel reflector. An extrapolation length of 7.3 cm was used for the reference solution. In one case there was no steel in the solution, in the second case 745 gm/l of stainless steel were distributed uniformly throughout the solution, in the third 1175 gm/l uniformly. The computed critical heights disagreed with the experimental values by < 1%, 6%, and < 1% for the three cases.

A question remains as to how one chooses an extrapolation length for other tampers. We shall suppose that we cannot compute it from first principles. Nevertheless there is in K643 much information on the efficiency of reflectors of various thicknesses of water, stainless steel, and aqueous solutions of natural uranyl nitrate, phosphoric acid, and bismuth subcarbonate as a slurry. If we consider such homogeneous reflectors (with the moderator in an Al can) we find that they are essentially equivalent--equal thicknesses lead to nearly equal reactivities. Hence for all these solutions one may calculate from the experimental data one extrapolation distance as a function of reflector thickness. The calculation is slightly complicated by the fact that thin reflectors were on the lateral cylindrical surfaces only, and these reflectors usually extended far above the upper untamped surface of the reactor. For sufficiently tall reactors this should not introduce much uncertainty in the fast leakage from reactor ends and one can compute  $x$  vs. reflector thickness. The results of such an analysis are given in Fig. III. Most of

: : : : :  
: : : : :  
: : : : :  
: : : : :  
: : : : :  
: : : : :  
: : : : :  
: : : : :

SECRET

the data were taken for stainless steel reflectors (Table V-K643). It is believed that use of such values of  $x$  gives critical bucklings to better than  $\pm 5\%$  but no exhaustive check has been made.

#### IV. MODE METHOD FOR NON-HOMOGENEOUS REACTOR

It has appeared that the equations of age theory with modified constants are appropriate for describing a large class of hydrogen moderated reactors to a fair degree of accuracy. A logical generalization to reactors with large scale inhomogeneities is suggested below.

Suppose that there is a distribution of poisons in the fuel region of the reactor such that the age varies but little as a function of position but  $K_{\infty}$  varies grossly in any manner whatsoever. For example one might have a concentration of cooling pipes in the center of a reactor to flatten the flux and reduce thermal gradients. Then let us assume that an age theory with modified constants holds.

For an axially symmetric poison distribution in a cylinder we may write the slowing down density,  $X$ , at point  $(r, z)$  and age  $\tau$  in terms of the source density  $S(r, z)$  as:

$$X(r, z, \tau) = \sum_{n=1}^{\infty} \sum_{m=1}^{\infty} A_{nm} \cos\left(n \frac{\pi z}{H}\right) J_0\left(\alpha_m \frac{r}{R}\right) e^{-B_{nm} \tau} \int_{-\frac{H}{2}}^{\frac{H}{2}} dz' \int_0^R r' dr' S(r', z') \cos\left(n \frac{\pi z'}{H}\right) J_0\left(\alpha_m \frac{r'}{R}\right). \quad (14)$$

SECRET

Here  $A_{nm}$  is a normalizing factor, namely

$$A_{nm} = \frac{4}{HR^2 J_1^2(\alpha_m)}$$

$$B_{nm} = \frac{n^2 \pi^2}{H^2} + \frac{\alpha_m^2}{R^2},$$

and  $\alpha_m$  is the  $m^{\text{th}}$  root of  $J_0(\alpha_m) = 0$ .  $H$  and  $R$  are extrapolated height and radius. We assume the same extrapolation distance for all modes to get orthogonality. In case higher modes are well populated, this may differ from the unpoisoned extrapolation distance.

We may further express the source density in terms of the age at thermal:

$$S(r,z) = h_{\infty}^{(0)} X(r,z, \tau_{th}) (1+f(r,z)) \quad (15)$$

Here the factor  $f(r,z)$  is to take into account the non-homogeneous poison distribution, so that  $h_{\infty}^{(0)} (f(r,z)+1)$  is the local  $K_{\infty}$ , divided by  $1+L_{th}^2 B^2$ , namely the  $K_{\infty}$  which a uniform reactor of the same composition as at  $(r,z)$  would have.

Substituting (15) in (14) and assuming

$$X(r,z, \tau_{th}) = \sum_{p=1}^{\infty} \sum_{q=1}^{\infty} a_{pq} \cos \frac{p\pi z}{H} J_0 \left( \alpha_q \frac{r}{R} \right)$$

we get a series of linear equations which may be solved for the  $a_{pq}$ .

These equations have the form

$$a_{pq} = h_{\infty}^{(0)} e^{-B_{pq} \tau_{th}} \left( a_{pq} + \sum_{m=1}^{\infty} \sum_{n=1}^{\infty} a_{nm} A_{pq} f_{nm,pq} \right)$$

SECRET



SECRET

where

$$f_{nm,pq} = \int_{-\frac{H}{2}}^{\frac{H}{2}} dz' \int_0^R r' dr' f(z', r') \cos\left(\frac{n\pi z'}{H}\right) \cos\left(\frac{p\pi z'}{H}\right) J_0\left(\alpha_m \frac{r'}{R}\right) J_0\left(\alpha_q \frac{r'}{R}\right)$$

The difficulty of handling this expression depends on the symmetry  $f(r, z)$  and the magnitude of  $\tau_{th}$ .

SECRET

SECRET

APPENDIX.

Formulae for Calculating Critical Masses.

We consider proton moderated solutions of oralloy in cylindrical geometry.

The reactivity is given by

$$K = \frac{2.50}{1 + \frac{\Sigma_c(th)}{\Sigma_f(th)}} \frac{1}{1 + L_{th}^2 B^2} e^{-\tau B^2}$$

with  $B^2 = \frac{(2.405)^2}{(R+x)^2} + \frac{\pi^2}{(H+2x)^2}$

where  $\Sigma_c(th)$  = macroscopic thermal capture cross section

$\Sigma_f(th)$  = macroscopic thermal fission cross section

$L_{th}^2$  = thermal diffusion area

H = physical height

R = physical radius

x = "extrapolation length"

$\tau$  = "age"

In computing  $\Sigma_c(th)$  and  $L_{th}^2$  we used the following capture cross sections for room temperature:

element	H	U-235	O,F	N	P
$\sigma_c$ (barns)	.330	650	0.00	1.80	0.20
	Fe	Cr	Ni	stainless steel	
	2.45	2.90	4.50	2.70.	

SECRET

. . . . .  
 . . . . .  
 . . . . .  
 . . . . .  
 . . . . .

The U-235 fission cross section was taken to be 550b. For uranium in water we took  $L_{th}^2 = \frac{8.20}{1+1970/C}$  with C the atomic ratio of H to U-235.

For the reference solution ( $UO_2F_2$  in water with the U or alloy),  $x_0 = 4.3$  cm for solution in 1/16" stainless steel can; 7.0 cm for same with water reflector; 5.6 cm with Cd interposed between water reflector and can. For solution in 1/16" Al can and reflector of water, stainless steel, aqueous solutions of natural uranyl nitrate, or phosphoric acid see Fig. III giving  $x_0$  vs reflector thickness.

$$\tau = 27 \text{ cm}^2 \text{ for reference solution.}$$

For other solutions:

$$\tau = 27 \frac{113.3}{\sum_i \gamma_i M_i - M_u} \frac{166.7}{\sum_i \rho_i M_i}$$

$$x = x_0 \frac{166.7}{\sum_i \rho_i M_i}$$

element	H	N	O	F	P	Cr,Fe,Ni	U
$\rho$	1.0	1.0	1.0	1.0	1.3	1.5	2.0
$\gamma$	1.0	0.04	0.04	0.04	0.02	.20	

$M_i$  = Molarity of  $i^{\text{th}}$  element in moles per liter of solution.

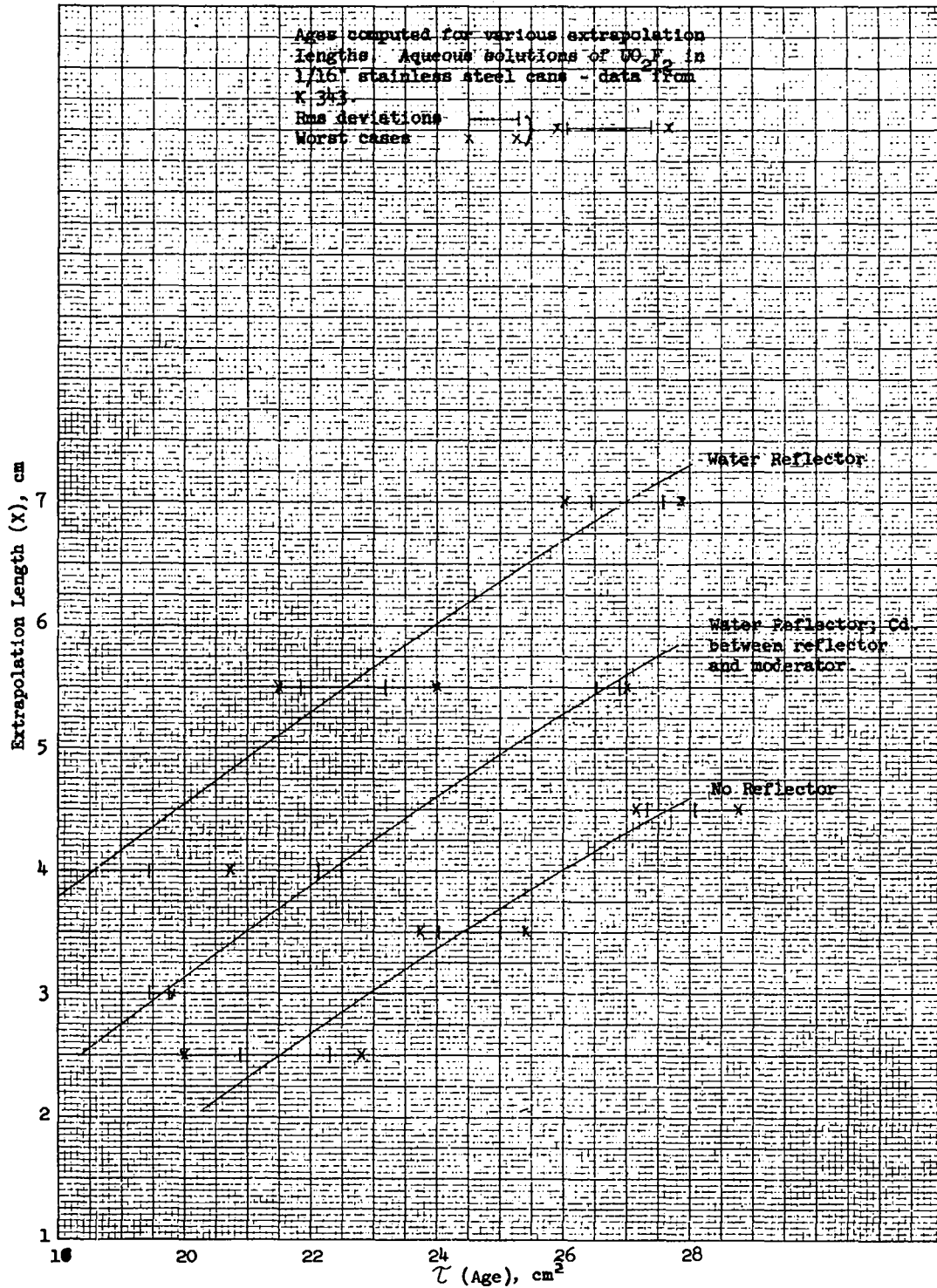
$M_u$  = molarity of uranium.

. . . . .  
 . . . . .  
 . . . . .  
 . . . . .  
 . . . . .

28



FIGURE I



SECRET

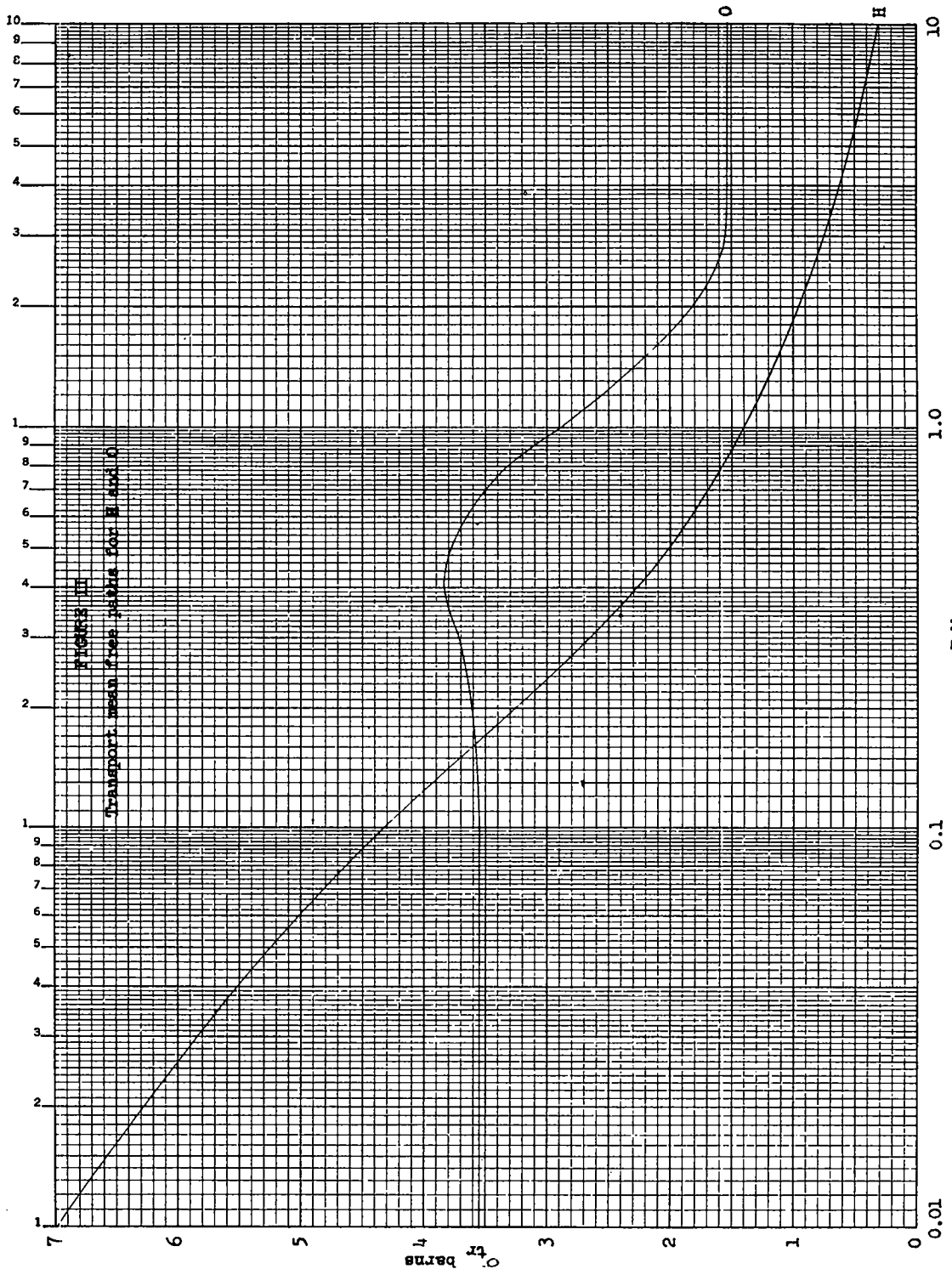


FIGURE 1  
Transport mean free path for H and O

SECRET

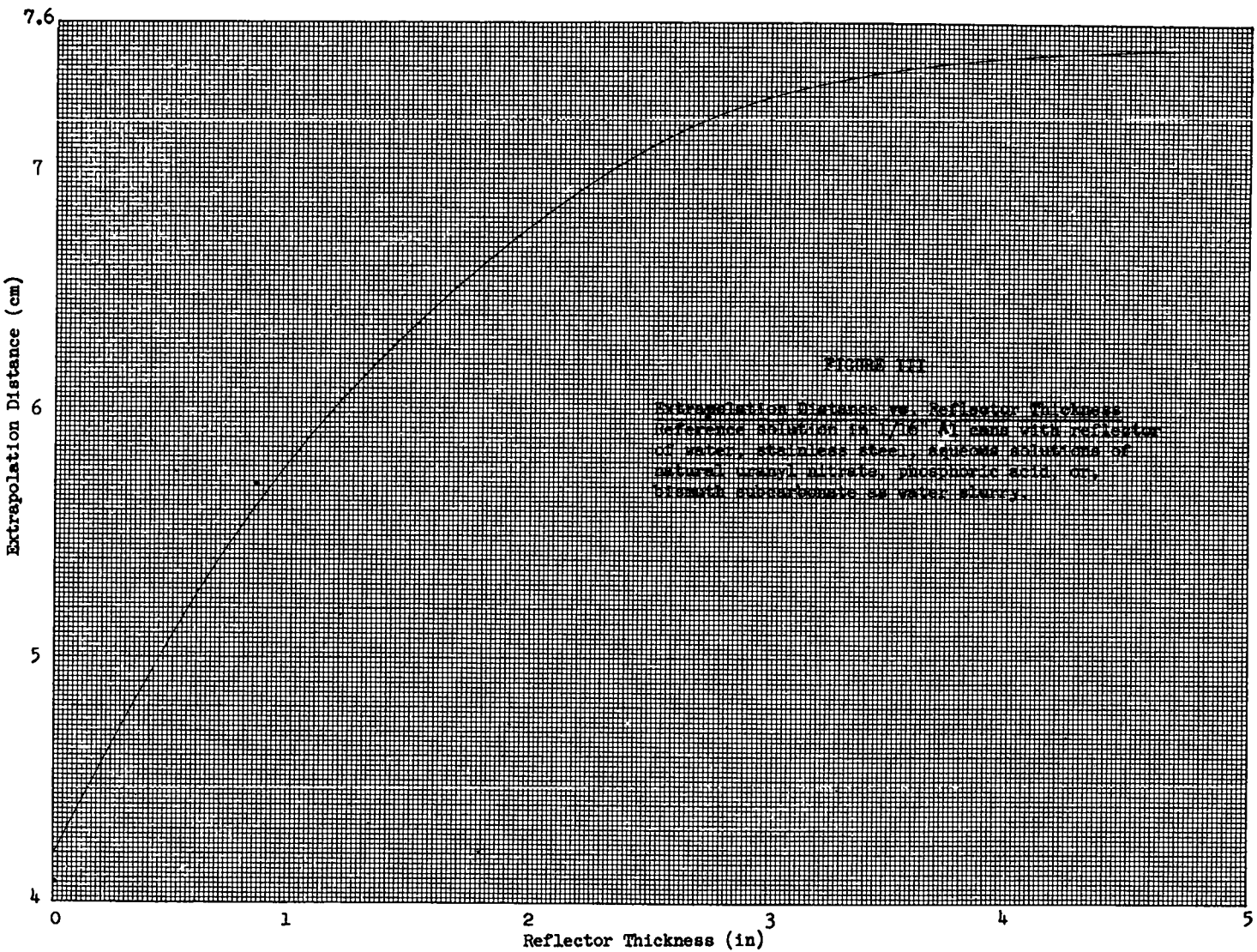


SECRET

UNCLASSIFIED

SECRET

UNCLASSIFIED



SECRET

UNCLASSIFIED

08.11.53  
031700

UNCLASSIFIED

UNCLASSIFIED

REPORT LIBRARY

REC. FROM Ja

DATE 9-21-53

RECEIPT [Signature]

08.11.53  
031700