

**$k_{\infty}$  OF THREE WEIGHT PER CENT  
 $U^{235}$  ENRICHED  $UO_3$  AND  $UO_2(NO_3)_2$   
HYDROGENOUS SYSTEMS**

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**SEPTEMBER, 1961**

**HANFORD LABORATORIES**

**HANFORD ATOMIC PRODUCTS OPERATION  
RICHLAND, WASHINGTON**

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HW-66882

UC-46, Criticality Studies  
(TID-4500, 17th Ed.)

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AND UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub> HYDROGENOUS SYSTEMS

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September, 1961

HANFORD ATOMIC PRODUCTS OPERATION  
RICHLAND, WASHINGTON

Work performed under Contract No. AT(45-1)-1350 between the  
Atomic Energy Commission and General Electric Company

Printed by/for the U. S. Atomic Energy Commission

Printed in USA. Price \$1.25. Available from the  
Office of Technical Services  
Department of Commerce  
Washington 25, D. C.

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ABSTRACT

The value of the infinite multiplication constant ( $k_{\infty}$ ) has been determined as a function of the hydrogen-to-uranium (H:U) atomic ratio for 3.04 weight per cent  $U^{235}$  enriched  $UO_3$  and  $UO_2(NO_3)_2$  hydrogen moderated homogeneous systems. The work was done to evaluate nuclear safety of and establish operational limits for slightly enriched homogeneous systems and was performed in the Hanford Physical Constants Testing Reactor.

The amount of thermal neutron absorber, commonly referred to as poison, necessary to reduce the value of  $k_{\infty}$  of these systems to unity was also obtained directly from these studies. This value is of interest for processing reactor fuels in which soluble poisons would be used for criticality control.

Curves of the value of  $k_{\infty}$  as a function of the H:U atomic ratio for 3.04 weight per cent  $U^{235}$  enriched  $UO_3$  and  $UO_2(NO_3)_2$  are shown. The curve of  $k_{\infty}$  versus H:U is determined by a least squares fit to the experimental data.

Values of the amount of thermal poison necessary to reduce  $k_{\infty}$  to unity as a function of the H:U ratio are tabulated.

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$k_{\infty}$  OF THREE WEIGHT PER CENT  $U^{235}$  ENRICHED  $UO_3$   
AND  $UO_2(NO_3)_2$  HYDROGENOUS SYSTEMS

### INTRODUCTION

In order to evaluate nuclear safety in operations involving the handling of enriched  $UO_3$  and  $UO_2(NO_3)_2$ , a knowledge is needed of the criticality parameters which characterize these systems. Restrictions can then be placed on either the mass, volume, or concentration of the fissile material. The infinite multiplication factor ( $k_{\infty}$ ) of the material in question is a very useful parameter in that the critical conditions can be determined if the value of  $k_{\infty}$  is known. For example; since a chain reaction cannot be sustained for any amount of the material if  $k_{\infty} < 1$ , a knowledge of the limiting concentration at which  $k_{\infty} = 1$  allows restrictions to be placed on the concentration; no criticality hazard will exist when the concentration is maintained below this limiting value.

The Physical Constants Testing Reactor (PCTR) of Hanford Atomic Products Operation, Richland, Washington, is a very useful tool for determining the value of  $k_{\infty}$  for the type of systems of interest here. (1-4)

A series of experiments was undertaken in the PCTR to determine the value of  $k_{\infty}$  for 3.04 weight per cent enriched  $UO_3$  and  $UO_2(NO_3)_2$  hydrogen moderated systems as a function of the hydrogen-to-uranium (H:U) atomic ratio. The determination of the curves of  $k_{\infty}$  versus H:U atomic ratio will permit the specification of critically safe conditions for all operations involving 3 per cent enriched  $UO_3$  and  $UO_2(NO_3)_2$  hydrogen moderated homogeneous systems.

### DISCUSSION

The amount of nonfissile thermal neutron absorber necessary to reduce  $k_{\infty}$  of a fissile material having  $k_{\infty} > 1$  to unity can be determined experimentally. Also, the hydrogen-to-uranium ratio at which  $k_{\infty}$  is

unity for the unpoisoned material may be measured; however, the value of  $k_{\infty}$  itself is a derived value and is hence dependent upon some formalism. A description of the formalism used in these studies is discussed here. Other formalisms are discussed in previous works. (1-4)

The infinite medium neutron multiplication factor of a multiplying material can be determined from the quantity of thermal neutron absorber necessary to reduce  $k_{\infty}$  of the multiplying medium to unity.

The quantity of thermal neutron absorber necessary may be obtained from the principle that the  $k_{\infty}$  of an infinite just critical medium and the  $k_{\infty}$  of a vacuum are the same, namely, unity, and the substitution of a void for a finite region of the infinite poisoned multiplying material produces no perturbation in the system. The amount of thermal neutron absorber necessary to produce this condition can thus be determined.

The infinite multiplication factor for the unpoisoned system is given by

$$k_{\infty} = \eta \epsilon p f$$

where the terms have their usual meaning. (5) Then  $k_{\infty}$  for the poisoned system is

$$k_{\infty} = \eta' \epsilon' p' f' = 1.$$

It is assumed that the addition of the thermal neutron poison does not effect either  $\epsilon$  or  $p$  so that

$$\epsilon = \epsilon' \text{ and } p = p'$$

and thus,

$$k_{\infty} = \frac{k_{\infty}}{1} = \frac{k_{\infty}}{k_{\infty}'} = \frac{\eta f}{\eta' f'}$$

The  $\eta f$  for a homogeneous medium is defined as

$$\eta f = \frac{(\nu \Sigma_f \varphi v)_{\text{fuel}}}{\Sigma_i (\Sigma_a \varphi v)}$$

where "fuel" refers to uranium and "i" refers to all materials except the poison, and the other terms have their usual meanings. <sup>(5)</sup>

Then,

$$\eta' f' = \frac{(\nu' \Sigma'_a \phi' \nu)_{\text{fuel}}}{\Sigma_i (\Sigma'_a \phi' \nu)_i + (\Sigma'_a \phi' \nu)_{\text{poison}}}$$

The fluxes in these terms will cancel out because the medium is homogeneous. If the medium is homogeneous, but the poison is of a heterogenous nature, the same formalism may be kept with the introduction of the "disadvantage" factor for the poison which is defined as <sup>(5)</sup>

$$F = \frac{\bar{\phi}_{\text{fuel}}}{\bar{\phi}_{\text{poison}}}$$

where  $\bar{\phi}_{\text{fuel}}$  refers to the average flux in the fuel and  $\bar{\phi}_{\text{poison}}$  refers to the average flux in the poison.

With the following substitutions

$$(\Sigma'_a \nu)_i = (N\sigma)_i,$$

$$F = \frac{\bar{\phi}_{\text{fuel}}}{\bar{\phi}_{\text{poison}}},$$

and

$N$  = number of atoms/cm<sup>3</sup>

$\sigma$  = microscopic cross section,

then

$$k_{\infty} = \frac{nf}{n'f'} = \left[ \frac{\sigma_f}{\sigma_f} \frac{235}{235'} \right] \left[ \frac{\Sigma_i (N\sigma'_a)_i + \left( \frac{N\sigma'_a}{F} \right)_{\text{poison}}}{\Sigma_i (N\sigma'_a)_i} \right].$$

It has been assumed that  $\nu$  is a constant so that <sup>(6)</sup>

$$\nu = \nu'.$$

The term in the second set of brackets is  $f/f'$ . However, as written, the equation refers to the condition in which exactly the right amount of

poison has been added; whereas in actual practice, this right amount of thermal poison necessary to reduce  $k_{\infty}$  to unity is not used because of the difficulty in adding exactly the right amount. Instead, the test sample is poisoned quite close to the correct amount. Then, a small amount of poison is added and an extrapolation is made to determine the correct amount. This extrapolation is made as follows.

By methods of perturbation theory, it can be shown that<sup>(7)</sup>

$$\Delta\rho = \alpha M_{\text{poison}} \varphi_{1/v}$$

where

$M_{\text{poison}}$  = mass of poison;  
 $\alpha$  = proportionality constant;

$\varphi_{1/v}$  = thermal (1/v) flux in the system; and

$$\Delta\rho = \rho_{\text{poison}} - \rho_{\text{void}}$$

where  $\rho$  is defined as the reactivity of the reactor.

Then since  $N_{\text{poison}}$  (number of atoms of poison/cm<sup>3</sup>) is proportional to  $M_{\text{poison}}$ ,

$$N_{\text{poison}_0} = N_{\text{poison}} \left( \frac{\rho_{\text{unpoisoned}} - \rho_{\text{void}}}{\rho_{\text{unpoisoned}} - \rho_{\text{poisoned}}} \right) \left( \frac{\varphi_{1/v \text{ unpoisoned}}}{\varphi_{1/v \text{ void}}} \right)$$

where

$N_{\text{poison}_0}$  = the correct number of poison atoms.

If the poison is added in two batches as mentioned above, then

$$N_{\text{poison}_0} = N_{\text{poison}_1} + N_{\text{poison}_2} \left( \frac{\rho_{\text{poison}_1} - \rho_{\text{void}}}{\rho_{\text{poison}_1} - \rho_{\text{poison}_2}} \right) \left( \frac{\varphi_{1/v \text{ poison}_1}}{\varphi_{1/v \text{ void}}} \right)$$

Then substituting this term for  $N_{\text{poison}_0}$  into the equation for  $k_{\infty}$ ;

$$k_{\infty} = \left[ \frac{\sigma_f^{235}}{\sigma_f^{235'}} \right]$$

$$\left[ \frac{\sum_i (N\sigma_a') + \left(\frac{N\sigma_a'}{F}\right)_{\text{poison}_1} + \left(\frac{N\sigma_a'}{F}\right)_{\text{poison}_2} \left( \frac{\rho_{\text{poison}_1} - \rho_{\text{void}}}{\rho_{\text{poison}_1} - \rho_{\text{poison}_2}} \right) \left( \frac{\varphi_{1/v \text{ poison}}}{\varphi_{1/v \text{ void}}} \right)}{\sum_i (N\sigma_a)_i} \right]$$

where

$N$  = number of atoms/cm<sup>3</sup>,

$\sigma$  = microscopic cross section in the unpoisoned system, and

$\sigma'$  = microscopic cross section in the poisoned system.

The term  $\varphi_{1/v}$  is determined from cadmium ratio measurements.<sup>(8)</sup> When a thermal poison is added to the system, there will be a change in the neutron energy spectrum as a result of the increased absorptions in the thermal region. If all the absorption cross sections of the different materials had a "1/v" dependence, this change in the spectrum would not effect the value of  $k_{\infty}$  since  $k_{\infty}$  is determined from the ratio of these different cross sections; however, the absorption and fission cross sections of  $U^{235}$  do not have a 1/v dependence, and thus any change in the spectrum will cause a change in the ratio of these values.

In the analysis of these measurements, this effect is taken into account by using Winger-Wilkens "effective" cross sections which show the "non-1/v" dependence of the  $U^{235}$  cross section as a function of the amount of 1/v poison present in the system. A table of these "effective" cross sections have been tabulated for mixtures of  $U^{235}$ , hydrogen, and a 1/v absorber.<sup>(9)</sup> The only discrepancy in this method is that the depression in the energy spectrum caused by the  $U^{238}$  resonances is not taken into account. The use of the Wigner-Wilkens "effective" cross sections is to account for the non-1/v dependence of the  $U^{235}$  fission and absorption cross sections in these systems and the change in the non-1/v cross sections of  $U^{235}$  caused by the shift in the neutron energy spectrum from the addition of a thermal neutron poison.

The values of  $k_{\infty}$  which are quoted in this report were found by use of these effective cross sections.

Because of the lack of an established error in these effective cross sections, the error in the final value of  $k_{\infty}$  includes an additional term which is the difference between  $k_{\infty}$  as determined from standard 1/v, 2200 meter/sec cross sections and  $k_{\infty}$  as determined from the effective cross sections. This amounts to approximately  $\pm 0.013$  for most cases. The derived value of  $k_{\infty}$  is higher when the effective cross sections are used.

As the systems become more thermal (large H:U atomic ratios) and less poison is needed, the difference between the value of  $k_{\infty}$  derived by use of 2200 meter cross sections and effective cross sections becomes smaller.

The amount of thermal poison necessary to reduce  $k_{\infty}$  to unity is a measured rather than a derived value; as a result these values do not depend upon any formalism which is used and hence the error in this value is smaller.

The error analysis of these values is fully discussed in Appendix IV.

#### EXPERIMENTAL PROCEDURES

The H:U atomic ratio for these materials, 3.04 w/o  $U^{235}$  enriched  $UO_3$  and  $UO_2(NO_3)_2$ , was varied by the addition or removal of water or polyethylene. The use of polyethylene as the moderator has a distinct advantage in that a mixture of uranium and polyethylene may be prepared for any H:U atomic ratio whereas water can be used only where soluble compounds are available or the water of hydration and absorbed water can be used as the moderator. From the theoretical point of view, water and polyethylene should be nearly equivalent in an infinite system except for slight differences in the carbon and oxygen cross sections and possible differences in the chemical binding of the hydrogen in the two molecules.

A number of experiments were conducted in which systems were moderated with both water and polyethylene. The value of  $k_{\infty}$  obtained for the two moderators was in agreement for all experiments. However, in some earlier experiments, the error in analysis for water was quite large so that the agreement was not conclusive. One experiment was also conducted in which the moderator was approximately 50 per cent water and 50 per cent polyethylene. The value of  $k_{\infty}$  obtained for this case was also in agreement with the value of  $k_{\infty}$  when all polyethylene moderation was used.

The preparation, loading, unloading, and sampling of the materials used in these experiments was carried out by the Chemical Research and Development Operation of Hanford Laboratories Operation and Critical Mass Physics personnel at HAPO.

The materials used in the  $\text{UO}_3$  experiments were  $\text{UO}_3$  powder and polyethylene pellets or powder. Small pieces ( $1/4 \times 1/4 \times 0.005$  inch) of borated polyethylene sheet were used as the poison.

The  $\text{UO}_3$  powder was used both in dry and wet (water of hydration and absorbed water) states. The uranium contained 3.04 w/o  $\text{U}^{235}$  and the dry  $\text{UO}_3$  powder usually contained approximately 1 per cent water by weight, depending somewhat upon its exposure to the atmosphere. This material was free of any significant neutron poisons. It had an approximate particle size of 100 microns. Originally 317 pounds of this material having a density of approximately  $1.7 \text{ gm/cm}^3$  were available.

For the  $\text{UO}_2(\text{NO}_3)_2$  experiments, the original  $\text{UO}_3$  material was converted to  $\text{UO}_2(\text{NO}_3)_2$ . The particle size of the  $\text{UO}_2(\text{NO}_3)_2$  was approximately the same as the  $\text{UO}_3$ . The water content of the  $\text{UO}_2(\text{NO}_3)_2$  was kept as low as possible upon conversion so that all the  $\text{UO}_2(\text{NO}_3)_2$  experiments could be run by simply adding polyethylene to obtain higher H:U atomic ratios. After conversion, the  $\text{UO}_2(\text{NO}_3)_2$  had the following properties:

53.18 w/o uranium

26.94 w/o  $\text{NO}_3$

11.47 w/o water.

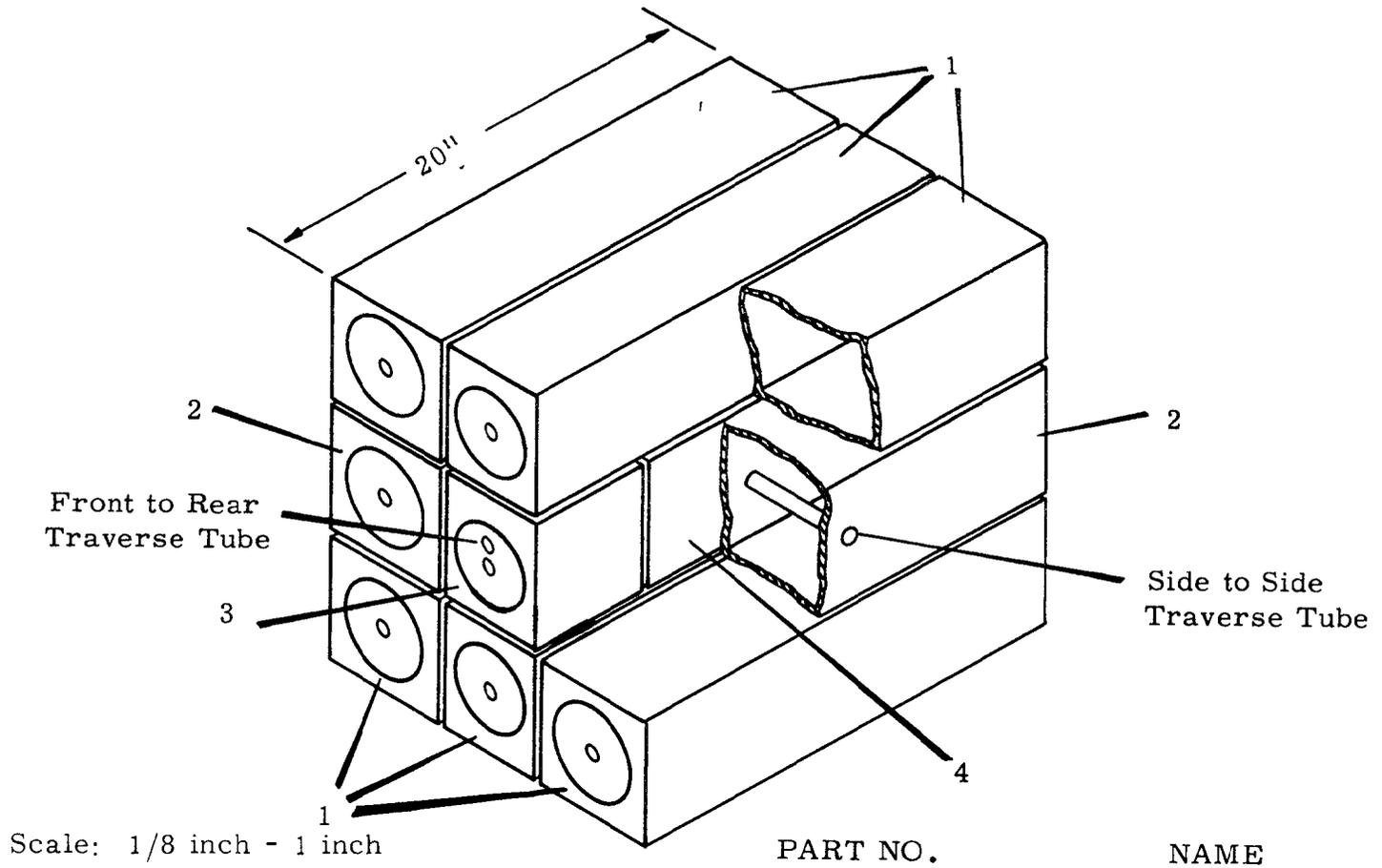
Polyethylene was used in these experiments because measurements had to be made for H:U atomic ratios for which no homogeneous and stable solution of uranium and water could be obtained. A polyethylene with the trade name Tenite\* was used in all work. In all experiments where the H:U atomic ratio was  $< 30$ , the polyethylene was in the form of pellets (1/8-inch spheres) and for H:U atomic ratios  $\geq 30$ , powdered polyethylene was used. The H:C atomic ratio for commercial polyethylene of this type is within 99 per cent of 2.<sup>(10)</sup> This was a linear polyethylene having a density of 0.916 gms/cm<sup>3</sup>. The use of the polyethylene pellets was prompted by the ease with which these pellets could be separated from the other constituents by a simple screening process. When powdered polyethylene was used, the experiments were arranged such that the moderator could always be added and hence no separation of the materials was necessary until all the experiments were completed.

The thermal neutron absorber was a specially prepared material; its preparation and properties are fully discussed in Appendix II. Again the choice of the form of this material was for ease in separation by screening.

The size of the containment tanks was limited by the limited amount of dry UO<sub>3</sub> powder initially available. Drawings of these tanks are given in Appendix V. Basically, this system was designed to be a "heterogeneous-homogeneous" system for ease in handling and preparation. Figure 1 is a cutaway drawing of the assembled system. Figure 2 is a picture of the assembled system in the PCTR. These tanks were assembled in an aluminum framework which supported the rear end of the tanks by having a diamond-shaped extension to the tank slip into the rear framework. The front and sides of the tank were "shimmed" into place. Traverse tubes were provided through these tanks in order to obtain necessary measurements of the neutron flux and spectrum by foil activation techniques.

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\* Produced by Eastman Chemical Products, Inc.



**FIGURE 1**  
 Cutaway Drawing of 3 Weight Per Cent  $UO_3$   $k_{\infty}$  Experiments,  
 Assembled System

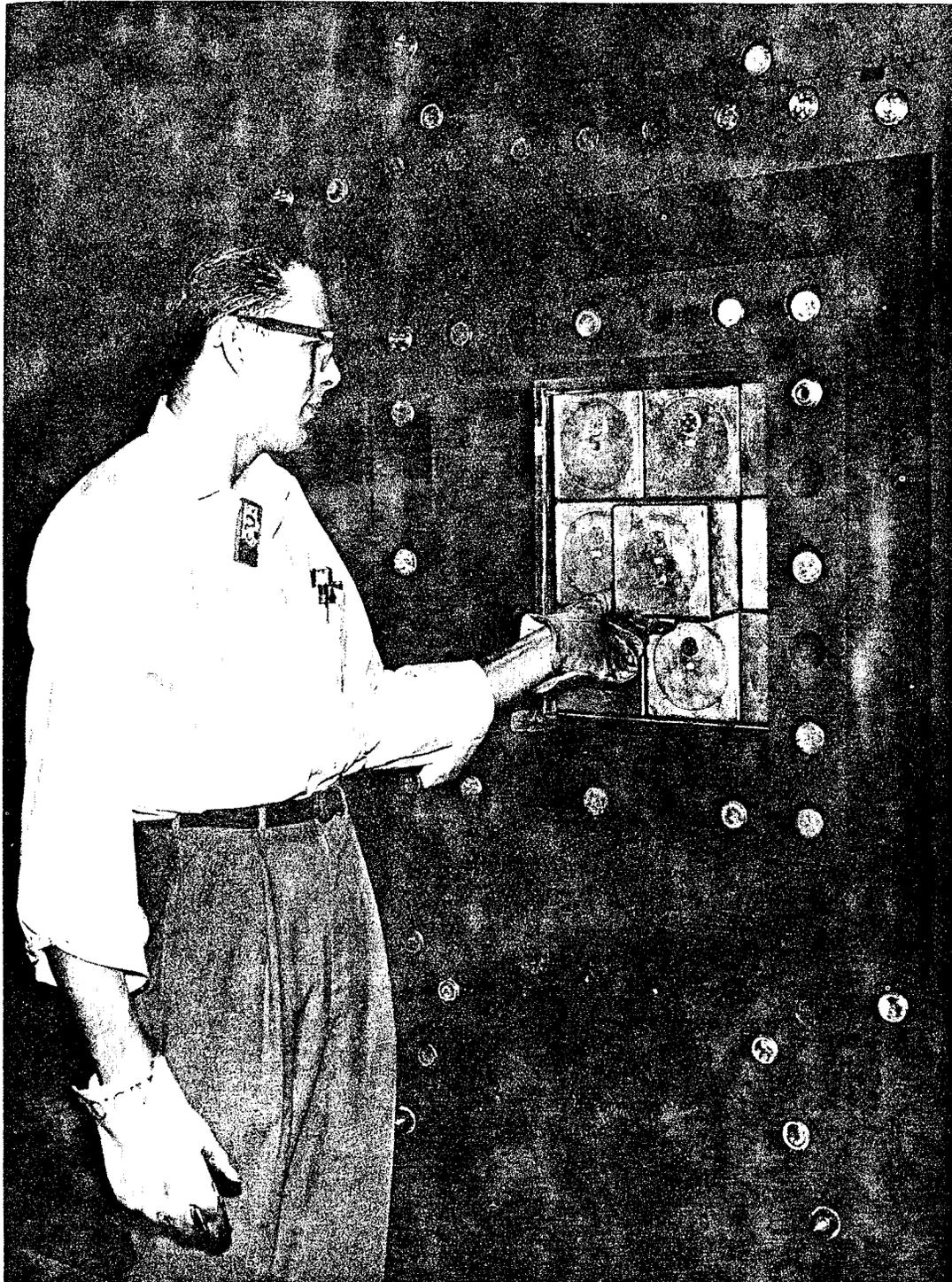


FIGURE 2

Assembled System of 3 Weight Per Cent  $\text{UO}_3$  and  $\text{UO}_2(\text{NO}_3)_2$   
Experimental Vessels in PCTR Cavity

A measurement of  $k_{\infty}$  for a particular H:U atomic ratio consisted of the following steps:

Calculations of  $k_{\infty}$  were conducted to obtain an estimate of the value to be measured. If any previous data were available, estimates or extrapolations were made from them. The buffer material was then poisoned down to approximately  $k_{\infty} = 1$  based on these estimates. Several core tanks were also prepared with varying amounts of poison in them. A "glove box" was kept especially for the purpose of making small changes in the core tanks. The assembled system was placed in the PCTR, and a particular reactor loading was chosen to bring the reactor to criticality. With a particular control rod setting, the reactivities of the various core tanks were compared to the reactivity of a void (helium filled tank). Flux traverse measurements were then made by foil activation techniques. The reactor driver region loading was then changed, and the measurements were repeated. This was continued until the proper driver region loading was determined; then if the original estimate of  $k_{\infty}$ , and hence the amount of poison in the buffer region, was close enough to the correct amount, the experiment was complete. If the original estimate was very wrong, then the buffer poison had to be changed, and the series of measurements repeated. Early experiments were conducted to determine how closely the buffer had to be poisoned. The proper driver region loading was determined from plots of the cadmium ratio measurements in the core and extending out into the buffer region. The shape of these plots would indicate whether the spectrum was too "fast" or too "slow", and then the proper spectrum could be found. The amount of buffer poison was considered to be close enough to the proper amount if the resulting  $k_{\infty}$  in the buffer was within the expected error limit of the experiment.

The weights of the materials in a particular experiment were determined by accurately weighing the amounts of poison and polyethylene before putting them into the tanks. The amount of water was determined

by Karl Fisher techniques for analyzing for water. When comparing the water moderated systems to the polyethylene moderated systems, special precautions had to be taken to avoid either the loss or gain (dependence on water content of the material) of water from the sample during the analyses. In many of the early  $\text{UO}_3$  experiments this problem was so critical that the water analysis could be relied upon only within 10-15 per cent until special techniques were developed for handling these systems.

At the high H:U atomic ratios, the spectrum seems to be a slowly varying function of the H:U ratio, and hence the problem of correctly poisoning the buffer region is greatly reduced. As an example, for the  $\text{UO}_3$  system with the buffer at a  $k_\infty = 1.1$ , the H:U for which  $k_\infty$  equals unity was very accurately predicted with no buffer poison; however, as the H:U ratio was reduced, the system became much more sensitive to small spectral changes, and hence the buffer poison had to be more carefully controlled.

Extensive studies were conducted to determine the effects, if any, of the aluminum containment tanks on the resulting experimental value of  $k_\infty$ . Any effect these containment vessels might have would result from slight spectral effects caused by neutrons being absorbed in the aluminum containment walls and tending to "harden" the spectrum incident on the sample material.

Containment materials having larger macroscopic absorption cross sections were used to show that this effect was present; however, for these aluminum containment vessels the magnitude of this effect was less than the error of a reactivity measurement in the PCTR.

#### CORRELATION OF THEORY AND EXPERIMENT

The specification of nuclear safety parameters for any operation involving the handling of enriched  $\text{UO}_3$  and  $\text{UO}_2(\text{NO}_3)_2$  requires accurate formulas and constants for calculation of criticality conditions for these systems. One of the more basic values in these calculations is  $k_\infty$ ; however,

most efforts to calculate the theoretical value of  $k_{\infty}$  for  $\text{UO}_3$  and  $\text{UO}_2(\text{NO}_3)_2$  hydrogen moderated systems yield curves in which the maximum value of  $k_{\infty}$  is larger than the experimental value and is displaced toward a higher H:U atomic ratio.

The theoretical value of  $k_{\infty}$  was calculated from the "four-factor" formula,

$$k_{\infty} = \eta \epsilon p f$$

The major discrepancy between the theoretical and experimental values can be attributed to errors in the calculation of the terms  $\epsilon$  and  $p$  since the cross sections available for calculating the terms  $\eta$  and  $f$  are quite adequate for this type of calculation and should yield fairly accurate values. The cross sections used in the calculation of  $\epsilon$ , however, are not very reliable, and the resonance integral formula used in calculations of  $p$  is probably quite poor.

In determining the "best fit" to the experimental values of  $k_{\infty}$ , a least squares fitting technique was used. This method was originally suggested by Handler in his work on low enriched  $\text{UO}_3$  systems.<sup>(2)</sup> In these experiments, the  $\text{UO}_3$  systems were studied in detail; however, the  $\text{UO}_2(\text{NO}_3)_2$  systems were studied only enough to establish the maximum and general shape of the  $k_{\infty}$  versus H:U curve. Thus there were twenty experimental values to be used in fitting the  $\text{UO}_3$  data, but only four values for the  $\text{UO}_2(\text{NO}_3)_2$  data. The experimental values of  $k_{\infty}$  were divided by calculated values of  $\eta f$  and then fitted to the theoretical curve of  $\epsilon p$ . The constants obtained from the least squares fit of the  $\text{UO}_3$  data were then used to obtain a theoretical calculation of  $k_{\infty}$  for  $\text{UO}_2(\text{NO}_3)_2$ .

The authors have derived a formula for calculating  $\epsilon$  which represents a good model of the physics involved and should give fairly good results depending on the accuracy of the cross sections used. The derivation and equations are shown in Appendix I.

The method of making a least squares fit to the  $k_{\infty}$  values is as follows:

The usual method for calculating p is to use the formula,

$$p = \exp. - \frac{N_{238}}{\bar{\xi} \Sigma_s} \int \left( \sigma_{ao_{eff}} \right) \frac{dE}{E}$$

where the resonance integral is (5)

$$\int \left( \sigma_{ao_{eff}} \right) \frac{dE}{E} = 3.9 \left( \frac{\Sigma_s}{N_{238}} \right)^{0.415}$$

so that

$$p = \exp. - \frac{3.9}{\bar{\xi}} \left( \frac{N_{238}}{\Sigma_s} \right)^{0.585}$$

$N_{238}$  = number of  $U^{238}$  atoms/cm<sup>3</sup>,

$$\Sigma_s = \sum_i (N \sigma_s)_i,$$

$$\bar{\xi} = \frac{\sum_i (\xi N \sigma_s)_i}{\sum_i (N \sigma_s)_i},$$

$N_i$  = number of atom/cm<sup>3</sup>,

$\sigma_i$  = microscopic cross section, and

$\xi_i$  = average logarithmic energy decrement/collision.

In this analysis p will be assumed to have the form,

$$p = \exp. - \frac{a}{\bar{\xi}} \left( \frac{N_{238}}{\Sigma_s} \right)^{0.585}$$

where "a" is a constant to be determined from the least squares fit.

As shown in Appendix I,  $\epsilon$  has the form

$$\epsilon = 1 + \beta \left[ \frac{\nu \Sigma_f}{\sum_i \gamma_i \Sigma_i - (\nu - 1) \Sigma_f} \right]$$

which can be written as

$$\epsilon = 1 + \left( \frac{\nu \Sigma_f \beta}{N_u \gamma_H \sigma_H} \right) \left[ \frac{1}{\frac{N_H}{N_u} + \left[ \sum_i \gamma_i \Sigma_i - (\nu - 1) \Sigma_f \right]} \right]$$

Then in this analysis  $\epsilon$  will be assumed to have the form

$$\epsilon \approx \exp. \left[ \frac{b}{\frac{N_H}{N_u} + \left( \sum_i \gamma_i \Sigma_i - (\nu - 1) \Sigma_f \right)} \right]$$

for  $\left[ \frac{b}{\frac{N_H}{N_u} + \left( \sum_i \gamma_i \Sigma_i - (\nu - 1) \Sigma_f \right)} \right] \ll 1$

where  $i$  refers to all materials except hydrogen, and  $b$  is another constant to be determined by the least squares fit.

Then the experimental data is fitted to the curve:

$$\frac{k_\infty}{\eta f} = \exp. \left[ -\frac{a}{5} \left( \frac{N_{238}}{\Sigma_s} \right)^{0.585} + \frac{b}{\frac{N_H}{N_u} + \left( \sum_i \gamma_i \Sigma_i - (\nu - 1) \Sigma_f \right)} \right]$$

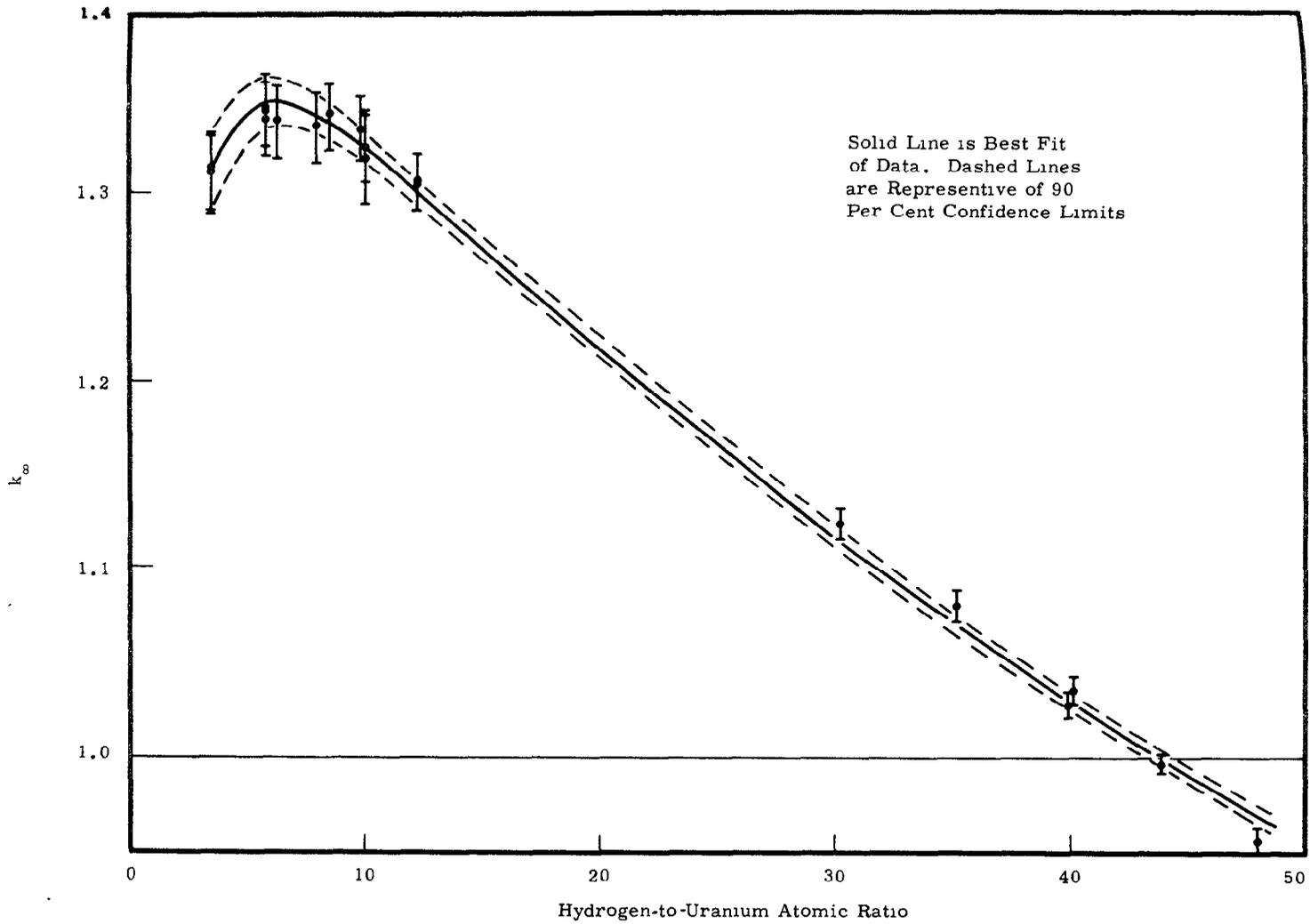
where the experimental data is  $\frac{k_\infty}{\eta f}$ , and where  $k_\infty$  is the experimental value and  $\eta f$  is calculated as shown in Appendix I.

Figure 3 shows the results of the least squares analysis of the  $UO_3$  data where the solid curves are the best fit of the experimental data, and the dashed curve is representative of the 90 per cent confidence limits. Figure 4 shows the  $UO_2(NO_3)_2$  data where the solid curve is the calculation using the constants determined by the least squares fit to the  $UO_3$  data. The points showing error limits are the experimental points. Details of the least squares fitting technique are given in Appendix I.

The values of the constants "a" and "b" obtained from least squares fitting the  $UO_3$  data may then be used in calculations of  $k_\infty$  for other slightly enriched systems. The values of these constants are

$$a = 4.75 \pm 0.18$$

$$b = 11.63 \pm 0.27$$



**FIGURE 3**  
 $k_{\infty}$  Versus Hydrogen-to-Uranium Atomic Ratio  
for 3.04 Weight Per Cent  $U^{235}$  Enriched  $UO_3$  Hydrogen Systems

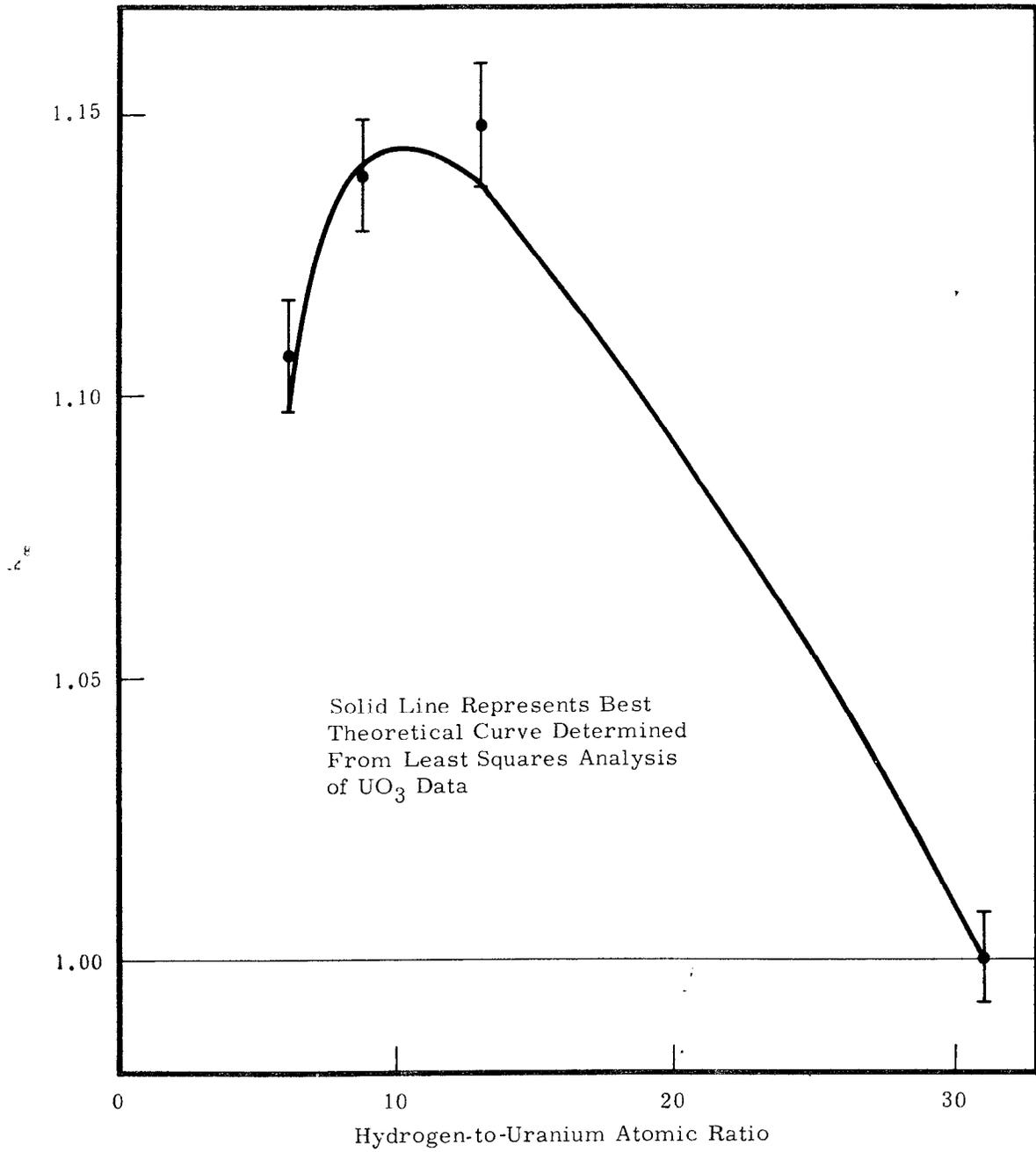


FIGURE 4

$k_{\infty}$  Versus Hydrogen-to-Uranium Atomic Ratio  
for 3.04 Weight Per Cent Enriched  $UO_2(NO_3)_2$  Hydrogen Systems

Another quantity obtained directly from these experiments is the amount of boron necessary to reduce the  $k_{\infty}$  of a material to unity. This value is of interest in the processing of reactor fuels where soluble poisons might be used for criticality control. Table I lists the amount of boron necessary to reduce  $k_{\infty}$  to unity for all experimental measurements.

In these experiments the quantity ( $N\sigma$ ) was measured. In computing the ratio of  $\frac{\text{Boron Atoms}}{\text{Uranium Atoms}}$  required to reduce  $k_{\infty}$  to unity, the microscopic cross section of boron was assumed to be 755 barns.

### CONCLUSIONS

The curves of  $k_{\infty}$  versus the hydrogen-to-uranium atomic ratio for 3.04 w/o  $U^{235}$  enriched  $UO_3$  and  $UO_2(NO_3)_2$  hydrogen moderated homogeneous systems are shown in Figures 3 and 4. These curves are the result of a least squares fitting technique and the dashed lines represent 90 per cent confidence limits.

Values of the amount of thermal poison necessary to reduce  $k_{\infty}$  to unity as a function of the H:U ratio are shown in Table I.

The maximum value of  $k_{\infty}$  for 3.04 w/o  $U^{235}$  enriched  $UO_3$  was found to be  $1.350 \pm 0.013$ . The H:U ratio for which  $k_{\infty} = 1$  was  $43.9 \pm 0.5$ . The amount of boron necessary to poison any 3.04 w/o enriched  $UO_3$ -hydrogenous system was  $0.0113 \pm 0.0003$  atoms of boron per uranium atom.

The maximum value of  $k_{\infty}$  for 3.04 w/o  $U^{235}$  enriched  $UO_2(NO_3)_2$  was found to be  $1.145 \pm 0.010$ . The H:U ratio for which  $k_{\infty} = 1$  was  $31.2 \pm 1.0$ . The amount of boron necessary to poison any 3.04 w/o  $U^{235}$  enriched  $UO_2(NO_3)_2$ -hydrogen system was  $0.0055 \pm 0.0003$  atoms of boron per uranium atom.

The original purpose of this work has been accomplished in that this data will permit the specification of nuclear safety parameters for operations

TABLE I  
MEASURED VALUES OF  $k_{\infty}$  AND AMOUNT OF POISON  
REQUIRED TO REDUCE  $k_{\infty}$  TO UNITY

| <u>H:U</u>  | <u><math>k_{\infty}</math></u> | <u>1/v Barns/Total<br/>of Poison*/ U Atoms<br/>Required to Reduce<br/><math>k_{\infty}</math> to Unity</u> | <u>Boron/Total<br/>Atoms/ U Atoms</u> |          |           |
|---|--------------------------------|--|---------------------------------------|----------|-----------|
| <u>3.04 w/o Enriched <math>UO_3</math>-Polyethylene Systems</u> |                                |  |                                       |          |           |
| 3.58  | 117.8                          | 1.309  | 7.250                                 | 0.00960  | 0.3158 ✓  |
| 3.58  |                                | 1.311  | 7.312                                 | 0.00969  | 0.3188 ✓  |
| 5.86  | 192.8                          | 1.345  | 8.426                                 | 0.01116  | 0.3671 ✓  |
| 5.86  |                                | 1.340  | 8.307                                 | 0.01100  | 0.3618 ✓  |
| 5.86  |                                | 1.346  | 8.445                                 | 0.01118  | 0.3678 ✓  |
| 5.86  |                                | 1.346  | 8.454                                 | 0.01119  | 0.3681 ✓  |
| 6.38  | 209.9                          | 1.338  | 8.322                                 | 0.01102  | 0.3625 ✓  |
| 8.01  | 263.5                          | 1.336  | 8.506                                 | 0.01126  | 0.3704 ✓  |
| 8.60  | 282.9                          | 1.342  | 8.411                                 | 0.01114  | 0.3664 ✓  |
| 9.94  | 327.0                          | 1.335  | 8.363                                 | 0.01107  | 0.3641 ✓  |
| 10.08   | 331.6                          | 1.318  | 8.242                                 | 0.01091  | 0.3589 ✓  |
| 10.12   | 332.3                          | 1.324  | 8.436                                 | 0.01117  | 0.3674 ✓  |
| 12.36   | 406.6                          | 1.306  | 8.108                                 | 0.01073  | 0.3530 ✓  |
| 12.37   | 406.9                          | 1.306  | 8.100                                 | 0.01072  | 0.3526 ✓  |
| 30.20   | 493                            | 1.123  | 4.151                                 | 0.00550  | 0.1809 ✓  |
| 35.26   | 1160                           | 1.080  | 2.844                                 | 0.00376  | 0.1237 ✓  |
| 39.77   | 1308                           | 1.028  | 1.041                                 | 0.00138  | 0.0454 ✓  |
| 40.05   | 1317                           | 1.035  | 1.292                                 | 0.00171  | 0.0562 ✓  |
| 43.85   | 1442                           | 0.996  | -0.163                                | -0.00022 | -0.0072 ✓ |
| 47.98   | 1578                           | 0.955  | -0.774                                | -0.00235 | -0.0773 ✓ |
| <u>3.04 w/o Enriched <math>UO_2(NO_3)_2</math> Systems</u>      |                                |  |                                       |          |           |
| 6.10  |                                | 1.114  | 3.307                                 | 0.00438  |           |
| 8.81  |                                | 1.139  | 4.014                                 | 0.00532  |           |
| 13.02   |                                | 1.148  | 4.091                                 | 0.00542  |           |
| 31.00   |                                | 1.000  | 0.000                                 | 0.00000  |           |

\*  $\frac{1/v \text{ Barns of Poison}}{\text{Total U Atoms}} = \frac{(N\sigma) \text{ Poison}}{N \text{ Uranium}}$

where N = number of atom/cm<sup>3</sup>

$\sigma$  = 2200 meter/sec "1/v" microscopic cross section.

involving the handling of 3 per cent enriched  $\text{UO}_3$  and  $\text{UO}_2(\text{NO}_3)_2$ . In addition the correlation of theory and experiment has been accomplished and a set of empirical formulas developed which yield good results for theoretical calculation of  $k_\infty$  for  $\text{UO}_3$  and  $\text{UO}_2(\text{NO}_3)_2$  hydrogen moderated systems at low  $\text{U}^{235}$  enrichments.

#### ACKNOWLEDGEMENTS

The authors wish to acknowledge the contributions of M. D. Alford of the Chemical Research and Development Operation for his assistance in the preparation of the uranium mixtures, of Mrs. J. J. Woody, who performed many of the machine calculations necessary to analyze the experimental data and who did most of the counting of the neutron activated foils necessary for the analysis of these experiments, and of the personnel of the Experimental Reactors Operation for the handling of materials and operation of the Physical Constants Testing Reactor.

APPENDIX I

THEORETICAL CALCULATIONS OF  $k_{\infty}$  AND LEAST SQUARES  
FITTING TECHNIQUES FOR ANALYZING  $k_{\infty}$  DATA

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FITTING TECHNIQUES FOR ANALYZING  $k_{\infty}$  DATA

The purpose of this section is to develop a set of formulas for calculating  $k_{\infty}$  from the four-factor formula which give theoretical results that are in agreement with experimental values.

The method, as pointed out in the main text, is to calculate the values of  $\eta$  and  $f$ , using the best cross sections, nuclear constants and formulas presently available, and then force the product of all four factors to be in agreement with the 3 per cent  $\text{UO}_3$   $k_{\infty}$  data by adjustment of constants in the formulas  $\epsilon$  and  $p$ .

The resonance escape probability for a homogeneous system has the formula

$$p = \exp. - \left( \frac{N_{238}}{\bar{\xi} \Sigma_s} \right) \int \left( \sigma_{ao\text{eff}} \right) \frac{dE}{E}$$

where

$N_{238}$  = number of  $\text{U}^{238}$  atoms/cm<sup>3</sup>,

$$\Sigma_s = \sum_i (N \sigma_s)_i$$

and

$$\bar{\xi} = \frac{\sum_i (\xi N \sigma_s)_i}{\sum_i (N \sigma_s)_i}$$

$N_i$  = number of atoms of the  $i^{\text{th}}$  material/cm<sup>3</sup>

$\sigma_s$  = microscopic scattering cross section of the  $i^{\text{th}}$  material

$\xi_i$  = average logarithmic energy decrement per collision for the  $i^{\text{th}}$  material.

The resonance integral  $\int \sigma_{ao\text{eff.}} \frac{dE}{E}$  is assumed to have the form

$$\int \sigma_{ao\text{eff.}} \frac{dE}{E} = a \frac{\Sigma_s}{N_{238}} 0.415$$

where the constant is usually assumed to have the value

$$a = 3.9. \quad (5)$$

Formulas for  $\eta f$

The theoretical formulas for  $\eta$  and  $f$  were the usual ones with the exception that Wigner-Wilkins "effective" cross sections were used.

The theoretical formula for  $\eta f$  is then,

$$\eta f = \frac{\nu N_{235} \sigma_f^{235}}{N_{235} \sigma_a^{235} + N_{238} \sigma_a^{238} + N_{\text{oxygen}} \sigma_a^{\text{oxygen}} + N_{\text{H}_2\text{O}} \sigma_a^{\text{H}_2\text{O}}}$$

$\nu$  = number of fast neutrons/fission in  $U^{235}$

$N_i$  = number of atoms of the  $i^{\text{th}}$  material/cm<sup>3</sup>

$\sigma_a$  = microscopic Wigner-Wilkins "effective" absorption cross section for the  $i^{\text{th}}$  material

$\sigma_f^{235}$  = microscopic Wigner-Wilkins "effective" fission cross section for  $U^{235}$ .

The Wigner-Wilkins cross sections are tabulated in Reference 9.

The 2200 meter/sec cross sections needed for Reference 6 are listed below. (11)

$$\begin{aligned} \nu &= 2.47 \\ \sigma_a^{\text{oxygen}} &= 0.0 \text{ barns} \\ \sigma_a^{\text{H}_2\text{O}} &= 0.664 \text{ barns} \\ \sigma_a^{238} &= 2.71 \text{ barns} \end{aligned}$$

Derivation of  $\epsilon$

Consider a system of  $\eta$  virgin fission neutrons, fast neutrons, having a fission energy distribution. A fraction of these ( $\beta$ ) have energies greater

than the  $U^{238}$  fission threshold and may possibly cause fission in  $U^{238}$ . After each of these  $\beta\eta$  neutrons has had an interaction, the number of neutrons having energies above the  $U^{238}$  fission threshold is,

$$\eta\beta \left[ \frac{\nu\Sigma_f + \sum_i (1 - \gamma_i) \Sigma_i}{\Sigma_f + \sum_i \Sigma_i} \right] = \eta\beta Z$$

where

- $\nu$  = number of fast neutrons, having energies above the  $U^{238}$  fission threshold, produced per fast fission in  $U^{238}$
- $\Sigma_f$  = macroscopic fission cross section of  $U^{238}$
- $\Sigma_i$  = macroscopic cross section for the  $i^{\text{th}}$  interaction, exclusive of  $U^{238}$  fission energy range
- $\gamma_i$  = probability of a neutron, having energy in the  $U^{238}$  fission range, of being reduced to an energy below the  $U^{238}$  fission threshold
- $\beta$  = fraction of virgin fission neutrons having energies above  $U^{238}$  fission threshold.

The number of neutrons removed from the  $U^{238}$  fast fission resonance region is

$$\eta\beta \left[ \frac{\Sigma_f + \sum_i (\gamma_i \Sigma_i)}{\Sigma_f + \sum_i \Sigma_i} \right]$$

Consider the next condition after all neutrons above the  $U^{238}$  fission threshold have had a second collision. The number of neutrons having energy above the  $U^{238}$  fission threshold is,

$$\eta\beta Z \left[ \frac{\nu\Sigma_f + \sum_i (1 - \gamma_i) \Sigma_i}{\Sigma_f + \sum_i \Sigma_i} \right] = \eta\beta Z^2$$

and the number of neutrons removed from the fast fission region is

$$\eta\beta Z \left[ \frac{\Sigma_f + \sum_i (\gamma_i \Sigma_i)}{\Sigma_f + \sum_i \Sigma_i} \right]$$

and for the  $n + 1^{\text{th}}$  collision, the number of neutrons having energies in the  $U^{238}$  fission energy range after the  $N^{\text{th}}$  collision is,

$$\eta\beta Z^n$$

The number of neutrons having energy above the  $U^{238}$  fission threshold is,

$$\eta\beta Z^n \left[ \frac{\nu\Sigma_f + \sum_i (1 - \gamma_i) \Sigma_i}{\Sigma_f + \sum_i \Sigma_i} \right] = \eta\beta Z^{n+1}$$

The number of neutrons removed from the fast fission energy region in the  $n + 1$  collision is,

$$\eta\beta Z^n \left[ \frac{\Sigma_f + \sum_i (\gamma_i \Sigma_i)}{\Sigma_f + \sum_i \Sigma_i} \right]$$

The fast effect ( $\epsilon$ ) is just the total number of neutrons removed from the fast energy region per primary fission neutron, thus

$$\begin{aligned} \epsilon &= \frac{\eta(1 - \beta) + \eta\beta \left[ \frac{\Sigma_f + \sum_i (\gamma_i \Sigma_i)}{\Sigma_f + \sum_i \Sigma_i} \right] (1 + Z + Z^2 + \dots + Z^n)}{\eta} \\ &= (1 - \beta) + \beta \left[ \frac{\Sigma_f + \sum_i (\gamma_i \Sigma_i)}{\Sigma_f + \sum_i \Sigma_i} \right] \left( \frac{1}{1 - Z} \right) \\ &= 1 + \beta \left[ \frac{\nu\Sigma_f}{\sum_i (\gamma_i \Sigma_i) - (\nu - 1)\Sigma_f} \right] \end{aligned}$$

The  $U^{238}$  fission cutoff energy is taken as  $E_c = 1.2$  Mev. The average energy,  $\bar{E}$ , of fission neutrons above this cutoff energy is

$$\bar{E} = 2.25 \text{ Mev,}$$

and the fraction of fission neutrons,  $\beta$ , above 1.2 Mev is<sup>(12)</sup>

$$\beta = 0.608.$$

For elastic scattering type of collisions:

$\gamma_i = \frac{1}{\text{Average number of collisions a neutron will make in the } i^{\text{th}} \text{ medium in being slowed down from } \bar{E} \text{ to } E_c}$

$$\gamma_i = \frac{1}{\frac{\xi_i}{\ln \left( \frac{\bar{E}}{E_c} \right)}} = \frac{\xi_i}{0.629}$$

Then

$$\gamma_{s \text{ U}^{238}} = 0.0133$$

$$\gamma_{s \text{ U}^{235}} = 0.0135$$

$$\gamma_{s \text{ Oxygen}} = 0.192$$

$$\gamma_{\text{U}^{238}} (\text{Capture} + \text{Inelastic}) = 1$$

$$\gamma_{\text{U}^{235}} (\text{Capture} + \text{Inelastic}) = 1$$

In the case of hydrogen,  $\gamma_s$  was determined as follows: The average energy of virgin neutrons above the  $U^{238}$  fission threshold of 1.20 Mev was determined to be 2.25 Mev. After the first scatter of these neutrons, the number of neutrons falling into an energy band  $\Delta E$  is the same for all energies. Thus the fraction of neutrons falling below 1.20 Mev upon first collision is simply  $\frac{1.20}{2.25} = 0.533$ . The average energy of neutrons above the 1.2 Mev threshold after this first collision is  $\frac{2.25 + 1.20}{2} = 1.73$ . We thus proceed as formerly for the second collision and etc.

| <u>Collision</u> | <u>Average Energy</u> | <u>Fraction (of Original Number) Falling Below 1.7 Mev</u> | <u>Fraction of Original Number Remaining</u> |
|------------------|-----------------------|--|--|
| 1st              | 2.25                  | 0.533  | 0.467  |
| 2nd              | 1.73                  | 0.324  | 0.143  |
| 3rd              | 1.47                  | 0.117  | 0.026  |
| 4th              | 1.34                  | 0.023  | 0.003  |
| 5th              | 1.27                  | 0.0028   | 0.0003                                       |
| 6th              | 1.23                  | 0.000293   | 0.000017                                     |

Then the average number of collisions a neutron would make with a hydrogen atom is given by,

$$\begin{aligned} \frac{\text{Avg. No. Coll.}}{\text{neutron}} &\cong \frac{(1)(0.533) + 2(0.324) + 3(0.117) + (0.023)}{(0.533) + (0.324) + (0.117) + (0.023)} \\ &= 0.533 + 0.648 + 0.351 + 0.092 + 0.014 + 0.0018 \\ &= 1.64 \end{aligned}$$

then

$$\gamma_s^{\text{Hydrogen}} \cong \frac{1}{1.64} = 0.61$$

This calculation is rather crude, but actually is very likely not to be greatly in error.

Then

$$\begin{aligned} \sum_i (\gamma_i \Sigma_i) &= N_{238} \left[ \gamma_{(c+in.)}^{238} (\sigma_a^{238} + \sigma_{in.}^{238}) + \gamma_s^{238} \sigma_s^{238} \right] \\ &+ N_{235} \left[ \gamma_{(c+in.)}^{235} (\sigma_a^{235} + \sigma_{in.}^{235}) + \gamma_s^{235} \sigma_s^{235} \right] \\ &+ N_{\text{Hydrogen}} \gamma_s^{\text{Hydrogen}} \sigma_s^{\text{Hydrogen}} + N_{\text{oxygen}} \gamma_s^{\text{Oxygen}} \sigma_s^{\text{Oxygen}} \end{aligned}$$

where

(c+in.) = capture + inelastic scattering, and

$\sigma_s$  = fast neutron microscopic scattering cross section.

The following values were used in these calculations

$$\nu_{238} = 2.62$$

$$\sigma_f^{238} = 0.55 \text{ barns}$$

$$\sigma_s^{238} = 3 \text{ barns}$$

$$\sigma_s^{235} = 3 \text{ barns}$$

$$\sigma_s^{\text{Oxygen}} = 1.5 \text{ barns}$$

$$\sigma_s^{\text{Hydrogen}} = 2.3 \text{ barns}$$

$$\sigma_{(c+in.)}^{238} = 3.5 \text{ barns}$$

$$\sigma_{(c+in.)}^{238} = 3.5 \text{ barns}$$

### Least Squares Fitting Technique

The least squares fitting technique was originally suggested by H. E. Handler and used by him in fitting some similar slightly enriched (1 per cent  $U^{235}$ )  $UO_3$   $k_{\infty}$  data.

The following form is taken for  $\epsilon$

$$\epsilon = \frac{b}{\frac{N_H}{N_U} + \sum_{i'} (\gamma_{i'} \Sigma_{i'}) - (\nu - 1) \Sigma_f}$$

where  $i'$  refers to all materials except hydrogen.

$$\epsilon \cong \exp. \frac{b}{\frac{N_H}{N_U} + \sum_{i'} (\gamma_{i'} \Sigma_{i'}) - (\nu - 1) \Sigma_f}$$

$$\text{for } 1 \gg \frac{b}{\frac{N_H}{N_U} + \sum_{i'} (\gamma_{i'} \Sigma_{i'}) - (\nu - 1) \Sigma_f}$$

and p has the form

$$p = \exp. - \frac{-a \left[ \frac{N_H}{N_u} + \frac{\Sigma'_s}{N_u \sigma_s \text{Hydrogen}} \right]^{0.415} (N_{238})^{0.585}}{\Sigma_{i'} \left( \frac{N_H}{N_u} \xi_{\text{Hydrogen}} + \xi_{i'} \frac{N_{i'}}{N_u} \frac{\sigma_s^{i'}}{\sigma_s \text{Hydrogen}} \right)}$$

Then the experimental data is fitted to the curve  $\epsilon p$ , where the experimental points are

$$\frac{k_{\infty} (\text{experimental})}{\eta f (\text{calculated})}$$

Twenty points were used in fitting the  $\text{UO}_3$  data, and four points were used in the  $\text{UO}_2(\text{NO}_3)_2$  data. The actual calculation was performed by a "Generalized Least Squares Fitting Code" on the Hanford IBM 7090 computer. (12,

Figures 3 and 4 show the results of the least squares fit.

PREPARATION AND PROPERTIES  
OF BORATED POLYETHYLENE  
AS A THERMAL NEUTRON ABSORBER

During the planning stages of this set of  $k_{\infty}$  experiments, it was decided that copper, which had been used as the usual neutron poison in the past, could not be used as the poison for these experiments. This was because that amount of copper which would be required would be so excessive that other perturbations might occur to the system. The choice of a borated polyethylene as the poison resulted from a number of requirements:

1. The  $1/v$  dependence of the boron absorption cross section made it very desirable for use as a thermal poison.
2. Both boron carbide and polyethylene are essentially inert to most acids and it was hoped that this poison could be used to study uranyl nitrate systems where acids would be present.
3. Since polyethylene was to be used as the moderator for these systems, it was an obvious choice for the plastic used to hold the poison.

The initial batch of poison was prepared at HAPO by the authors. The initial poison consisted of a mixture of boron carbide in 20 per cent paraffin and 80 per cent Epolene-C (trade name of a low-melt polyethylene wax produced by Eastman Chemical Products, Inc., Kingsport, Tennessee)

An ultrasonic generator was used to "beat" the liquid in order to break up any of the boron carbide particles which tended to cling together. The material, which contained approximately 3.5 per cent boron carbide by weight, was then solidified in the form of a cylinder. Chips  $1/4 \times 1/4 \times 0.005$  inch were machined from this cylinder. Although this initial material was uniform enough so that it was used quite satisfactorily for the first experiments, it was brittle and broke up quite easily. For this reason the search for a better product was continued.

The Product Development Division of Phillips Chemical Company, Bartlesville, Oklahoma, was contacted. They very kindly agreed to try to improve this material by mixing boron carbide and polyethylene on a Banbury rubber mill and extruding and blowing the material to obtain a blown tubing approximately 9 inches in diameter and 0.005-inch thick.<sup>(14)</sup> They made 20 pounds of the material in this manner. The resulting film proved to have a very uniform dispersion of boron carbide and uniform thickness. It was made using Marlex Type 9 polyethylene and boron carbide having a particle size of 2-5 microns. The material contained very close to 4 per cent boron carbide by weight and was designed to have the same neutron absorption properties as 0.020-inch thick copper. Extensive tests on the material showed no loss of boron carbide from the material and it was inert to concentrated nitric acid.

The error in any  $k_{\infty}$  measurement made in the PCTR using this material as a poison would be directly proportional to the error in the known value of its absorption cross section; hence, very accurate measurements of the absorption cross section were necessary. These measurements were made in the PCTR by comparing the material to another standard material for which the absorption cross section was accurately known.

Measurements were made in the PCTR to determine the cross section of both the 3.5 weight per cent and the 4 weight per cent materials using copper as a standard as measured by Bennett, Lanning, and Donahue.<sup>(15)</sup> The formalism used in determining the copper cross section was used throughout these experiments.

The value of  $\left(\frac{\sigma_a}{A}\right)_p$  (cross section of  $B_4C$  per gram molecular weight of borated polyethylene material) was then determined using the equation,<sup>(15)</sup>

$$\left(\frac{\sigma_{ao}}{A}\right)_p \left(\frac{A}{\sigma_{ao}}\right)_{cu} \left(1 - \frac{\alpha}{CdR}\right)_{cu} = \left[\frac{\rho_T}{M} \left(1 - \frac{1}{CdR}\right)\right]_p \left(\frac{M}{\rho_T}\right)_{cu} (P)(D) = K$$

where the subscripts "p" and "cu" refer to the poison (borated polyethylene) and copper standard, respectively, and

$\sigma_{ao}$  = 220 meter/sec value for the microscopic cross section,

A = atomic or molecular weight,

D = ratio of the disadvantage factors of the copper and borated polyethylene,

$\rho_T$  = reactivity of the reactor,

P = ratio of the products of the flux and favorableness factor at the surface of the samples,

M = mass of the sample,

$\alpha$  = ratio of the favorableness factors for neutrons absorbed in bare copper and cadmium covered copper, and

CdR = cadmium ratio.

The value of K was then plotted against  $\frac{1}{\text{CdR}_{\text{Cu}}}$  and extrapolated to  $\frac{1}{\text{CdR}_{\text{Cu}}} = 0$  to obtain  $\left(\frac{\sigma_{ao}}{A}\right)_f$ .

Measurements were made for  $\text{CdR}_{\text{Cu}}$  values of 102 and 7 for the 4 weight per cent material and for values of 197, 95.5 and 6.6 in the case of the 3.5 weight per cent. Figure 5 shows the results of these measurements.

The value of  $\left(\frac{\sigma_{ao}}{A}\right)_f$  for 3.5 weight per cent boron carbide impregnated polyethylene was found to be  $1.899 \pm 0.026$  and for the 4 weight per cent material,  $2.086 \pm 0.029$ .

An error of 1.5 per cent in K was assumed rather than the approximate 1 per cent observed in Figure 5 to allow for possible errors in  $\text{CdR}_{\text{Cu}}$ , since in two cases CdR for 0.005 copper was measured and then converted to  $\text{CdR}_{\text{Cu}}$  of 0.010 and 0.020 inch. Measurements of  $(\text{CdR})_p$  were made using 1/4-inch diameter  $\text{BF}_3$  tubes rather than the theoretical thin foil. This may have also contributed small errors.

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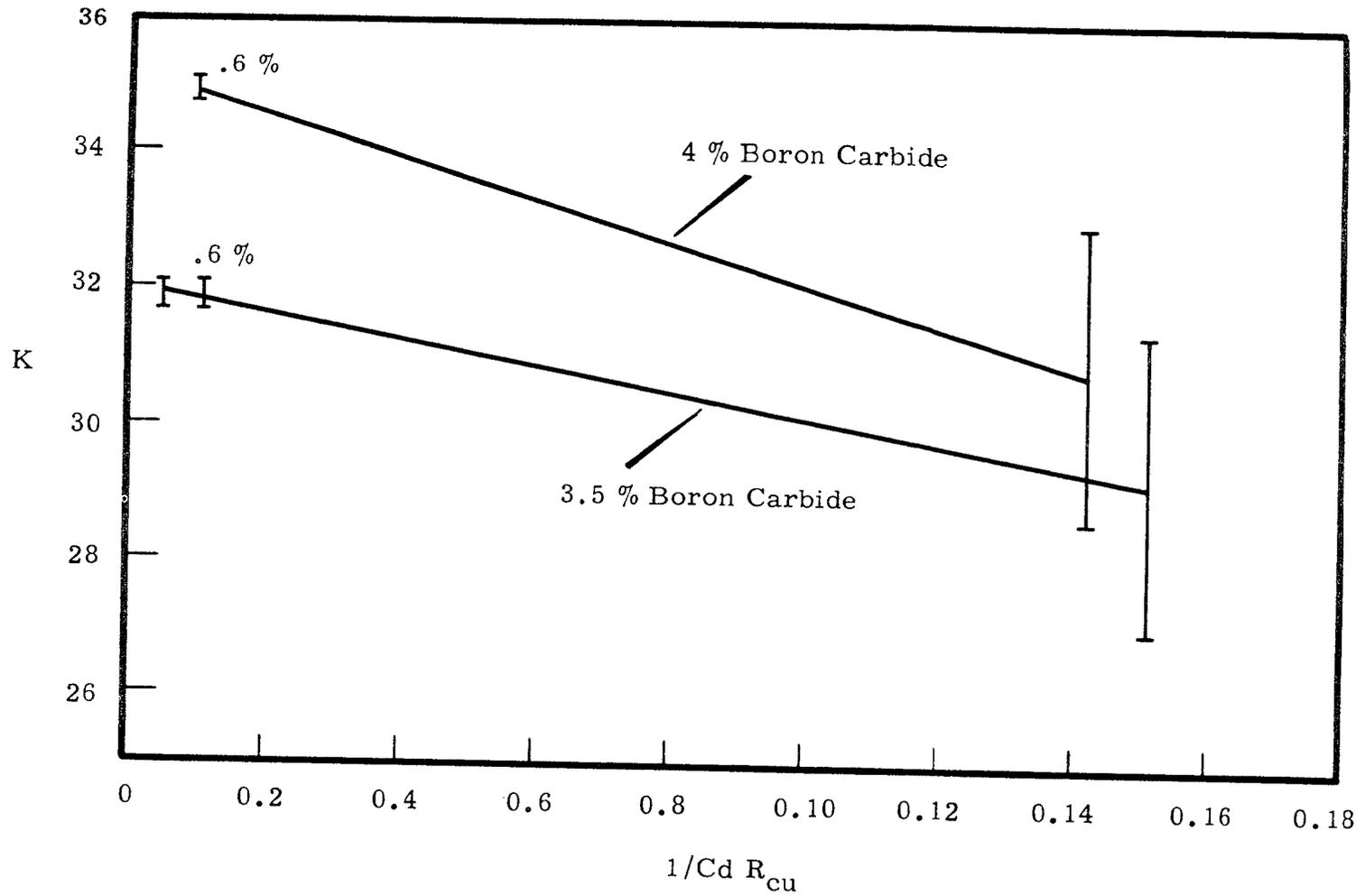


FIGURE 5  
Measurements of the Cross Section of Borated Polyethylene  
Used as a Thermal Poison

The method for obtaining the disadvantage factor of the poison material was to use the standard blackness approach which yields the formula<sup>(16)</sup>

$$F = \left(\frac{2V}{S}\right) \Sigma_a \left[ \frac{Z - Z \left(\frac{\Sigma_s}{\Sigma_T}\right) P_\infty}{\Sigma_a \left(\frac{4V}{S}\right) 1 - P_\infty} - 1 \right]$$

where

V = foil volume,

S = foil surface, and

$P_\infty$  = collision probability for a neutron in the foil.

The poison used in these experiments consisted of boron carbide impregnated polyethylene film cut into foils  $1/4 \times 1/4 \times 0.005$  inch. For the purpose of calculating the disadvantage factor of one of these small pieces, the foil was considered to be homogeneous; however, a correction was made for the finite size of the boron carbide particles (2 - 5 microns) by including the disadvantage factor of the particles in the calculation of the foil disadvantage factor. The disadvantage factor of a  $1/4 \times 1/4 \times 0.005$ -inch foil of 4 weight per cent boron carbide impregnated polyethylene was then found to be 1.020. An error of 0.005 was assumed in order to account for any grouping of the foils in the test cell.

APPENDIX III  
DISCUSSION OF ERRORS

APPENDIX III  
DISCUSSION OF ERRORS

Where it was possible, errors were analyzed by propagation-of-error techniques. Weights of different materials, 2200 meter/sec cross-section values, and flux and reactivity measurements could be treated in this manner. Errors because of mismatching of the incident neutron spectrum on the core tank were estimated from experimental data. Errors resulting from the use of "effective" cross sections could only be crudely estimated.

Table II is a summary of the values which were used in the calculations.

A number of weighings were made to establish that 1 per cent was a reasonable error in the weights.

Errors because of any mismatching of the incident neutron spectrum were estimated from measured values of  $k_{\infty}$ . These errors were estimated from the cadmium ratio measurements made with different driver loadings. This error was usually  $\pm 0.003$  in the value of  $k_{\infty}$ . For high H:U atomic ratios this error is reduced because of the more slowly changing spectrum and the more "effective" or thicker buffer region.

The error due to use of the "effective" cross sections was only an estimate. No reliable value for this error is available; however, a limiting value has been established.<sup>(3)</sup> The value results from the difference in  $k_{\infty}$ , as calculated from 2200 meter/sec cross sections, and from the "effective" cross sections. It is undoubtedly too large; however, it does represent a limiting factor.

TABLE II  
SUMMARY OF VALUES

| <u>Weight</u><br><u>Material Error</u>                           |         | <u>Molecular</u><br><u>Weight</u> | <u><math>\sigma_a</math> (at 2200</u><br><u>Meter/Sec) in Barns</u> | <u><math>(\sigma_a/A)^{(a)}A =</math></u><br><u>Atomic Weight</u> | <u>Error in</u><br><u><math>(\sigma_a/A)</math></u> |
|--|---------|-----------------------------------|---|---|---|
| U <sup>238</sup>   | 1%      | 238.125                           | 2.71  | 0.01138   | ±0.00008  |
| U <sup>235</sup>   | 1%      | 235.117                           | (b)   | (b)   | (b)   |
| H <sub>2</sub> O<br>(Water)                                      | (c)     | 18.021                            | 0.332   | 0.03685   | ±0.00022  |
| CH <sub>2</sub><br>(Polyethylene)                                | 1%      | 14.031                            | 0.6674  | 0.04756   | ±0.00028 <sup>(d)</sup>                             |
| Poison<br>(4 w/o Boron Carbide<br>Impregnated Poly-<br>ethylene) | 0.001 g | -                                 | -   | 2.086   | ±0.022  |

- (a) All values of  $\sigma_a$  (2200 meter/sec) and its standard deviation were taken from BNL-325.<sup>(11)</sup>
- (b) Effective cross sections were used for  $\sigma_f^{235}$  and  $\sigma_a^{235}$  (see Reference 9).
- (c) No error is quoted for the mass of water because of the large deviation in cases where water was used as the moderator; however, for the "dry" UO<sub>3</sub> powder, the error in the mass was 1 per cent.
- (d) Measured value (see Appendix II).

APPENDIX IV  
COMPILED DATA

TABLE III  
TABLE OF COMPILED DATA

| H:U Atomic Ratio                                       | Moderator                                    | Net Weight grams | Weight of Polyethylene grams | w/o of Water of Mixtures | w/o of Impurities of Mixture | $\left( \frac{1 - \text{void}/\%}{1 - \text{void}/\%} \right)^2$ | Average Bare Cd [CdR] | Average Bare Cd [CdR]   | Average Bare Cd [CdR] | Average Bare Cd [CdR] |
|--|--|------------------|------------------------------|--------------------------|------------------------------|--|-----------------------|-------------------------|-----------------------|-----------------------|
| <u>UO<sub>3</sub> Data</u>                             |  |                  |                              |                          |                              |  |                       |                         |                       |                       |
| 5.86   | CH <sub>2</sub>                              | 5754.7           | 688.1                        | 0.80                     | 0.34                         | 0.909  | Bare Cd [CdR]         | 0.172<br>0.232<br>2.822 | 1.304                 | ±0.020                |
| 5.86   | CH <sub>2</sub>                              | 5754.7           | 688.1                        | 0.80                     | 0.34                         | 1.124  | Bare Cd [CdR]         | 0.412<br>0.232<br>2.822 | 1.304                 | ±0.020                |
| 5.86   | CH <sub>2</sub>                              | 5754.7           | 688.1                        | 0.80                     | 0.34                         | 1.138  | Bare Cd [CdR]         | 0.420<br>0.242<br>2.822 | 1.304                 | ±0.020                |
| 3.58   | CH <sub>2</sub>                              | 6056.6           | 444.6                        | 0.94                     | 0.34                         | 1.028  | Bare Cd [CdR]         | 0.551<br>0.288<br>1.910 | 1.304                 | ±0.020                |
| 3.58   | CH <sub>2</sub>                              | 6056.6           | 444.6                        | 0.94                     | 0.34                         | 1.244  |                       |                         | 1.311                 | ±0.020                |
| 8.01   | CH <sub>2</sub>                              | 5168.2           | 807.3                        | 1.08                     | 0.34                         | 4.475  | Bare Cd [CdR]         | 0.722<br>0.268<br>2.686 | 1.334                 | ±0.020                |
| 10.09  | CH <sub>2</sub>                              | 5360.2           | 1030.9                       | 0.81                     | 0.34                         | 5.867  | Bare Cd [CdR]         | 0.813<br>0.276<br>2.938 | 1.318                 | ±0.024                |
| 10.12  | CH <sub>2</sub>                              | 5360.2           | 1034.1                       | 0.81                     | 0.34                         | 1.778  |                       |                         | 1.324                 | ±0.019                |
| 8.60   | CH <sub>2</sub>                              | 5027.5           | 843.5                        | 0.85                     | 0.34                         | 0.446  | Bare Cd [CdR]         | 0.721<br>0.268<br>2.689 | 1.342                 | ±0.020                |
| 9.94   | CH <sub>2</sub>                              | 5142.0           | 997.4                        | 0.85                     | 0.34                         | 0.130  | Bare Cd [CdR]         | 0.773<br>0.272<br>2.841 | 1.335                 | ±0.018                |
| 12.36  | CH <sub>2</sub> Pellets                      | 5313.0           | 1203.0                       | 0.86                     | 0.34                         | 2.20   | Bare Cd [CdR]         | 0.809<br>0.269<br>3.004 | 1.306                 | ±0.016                |
| 12.37  | CH <sub>2</sub> Pellets                      | 5313.0           | 1204.0                       | 0.86                     | 0.34                         | 1.074  | Bare Cd [CdR]         | 0.834<br>0.280<br>2.980 | 1.306                 | ±0.016                |
| 6.38   | CH <sub>2</sub> Pellets                      | 5480.0           | 706.2                        | 0.85                     | 0.34                         | -0.612   | Bare Cd [CdR]         | 0.659<br>0.293<br>2.850 | 1.338                 | ±0.020                |
| 5.86   | CH <sub>2</sub> Pellets                      | 5879.2           | 704.6                        | 0.75                     | 0.34                         | 0.897  | Bare Cd [CdR]         | 0.596<br>0.263<br>2.269 | 1.345                 | ±0.020                |
| 30.20  | CH <sub>2</sub> Powder                       | 3455.0           | 1450.8                       | 1.02                     | 0.34                         | 3.200  |                       |                         | 1.123                 | ±0.008                |
| 35.26  | CH <sub>2</sub> Powder                       | 3191.0           | 1461.9                       | 1.11                     | 0.34                         | 1.890  |                       |                         | 1.080                 | ±0.008                |
| 40.05  | CH <sub>2</sub> Powder                       | 3252.0           | 1598.5                       | 0.82                     | 0.34                         | 0.824  | Bare Cd [CdR]         | 1.307<br>0.175<br>7.460 | 1.035                 | ±0.007                |
| 39.77  | CH <sub>2</sub> Powder                       | 2943.7           | 1446.0                       | 0.51                     | 0.34                         | 0.603  | Bare Cd [CdR]         | 1.225<br>0.174<br>7.624 | 1.028                 | ±0.007                |
| 47.88  | CH <sub>2</sub> Powder                       | 2873.0           | 1547.1                       | 0.47                     | 0.34                         | -0.911   |                       |                         | 0.955                 | ±0.007                |
| 43.85  | CH <sub>2</sub> Powder                       | 2971.0           | 1532.9                       | 0.47                     | 0.34                         | -0.091   | Bare Cd [CdR]         | 1.304<br>0.172<br>7.602 | 0.996                 | ±0.006                |
| <u>UO<sub>2</sub>(NO<sub>3</sub>)<sub>2</sub> Data</u> |  |                  |                              |                          |                              |  |                       |                         |                       |                       |
| 6.10   | CH <sub>2</sub> Pellets and H <sub>2</sub> O | 5230.7           | 36.2                         | 11.47                    | 0.34                         | 1.201  | Bare Cd [CdR]         | 0.798<br>0.342<br>2.334 | 1.114                 | ±0.010                |
| 8.81   | CH <sub>2</sub> Pellets and H <sub>2</sub> O | 6079.1           | 293.1                        | 11.47                    | 0.34                         | 0.608  | Bare Cd [CdR]         | 0.991<br>0.361<br>2.757 | 1.139                 | ±0.010                |
| 13.02  | CH <sub>2</sub> Pellets and H <sub>2</sub> O | 5799.4           | 590.6                        | 11.47                    | 0.34                         | 0.431  | Bare Cd [CdR]         | 1.076<br>0.348<br>3.288 | 1.148                 | ±0.012                |
| 31.0   | CH <sub>2</sub> Powder and H <sub>2</sub> O  | 3712.0           | 1167.0                       | 11.47                    | 0.34                         |  |                       |                         | 1.000                 | ±0.008                |

APPENDIX V  
DRAWINGS OF THE CONTAINMENT VESSELS  
AND THEIR FITTINGS

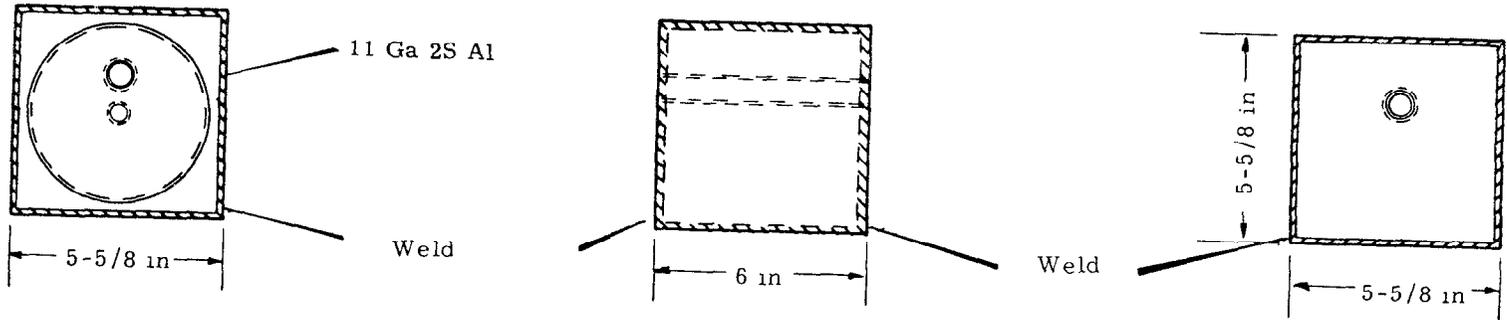


FIGURE 6  
Buffer Tank, Type A, Assembly

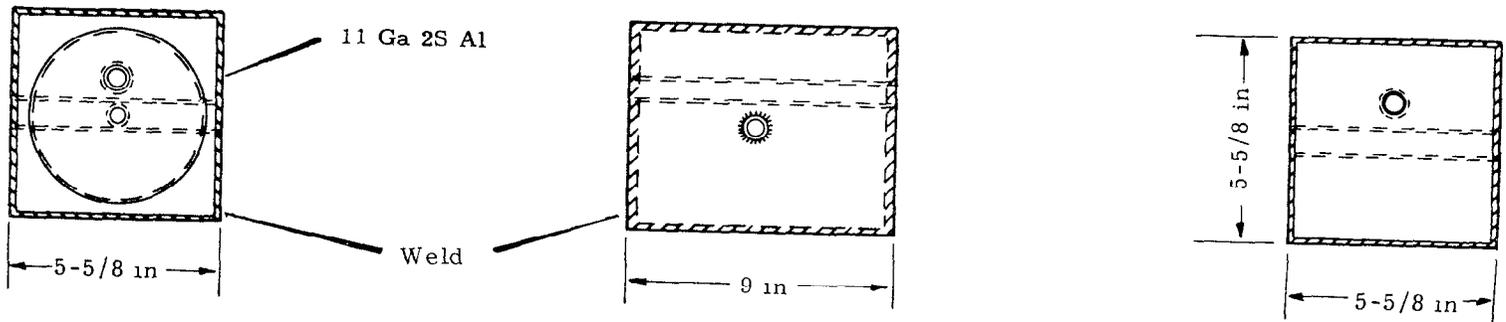
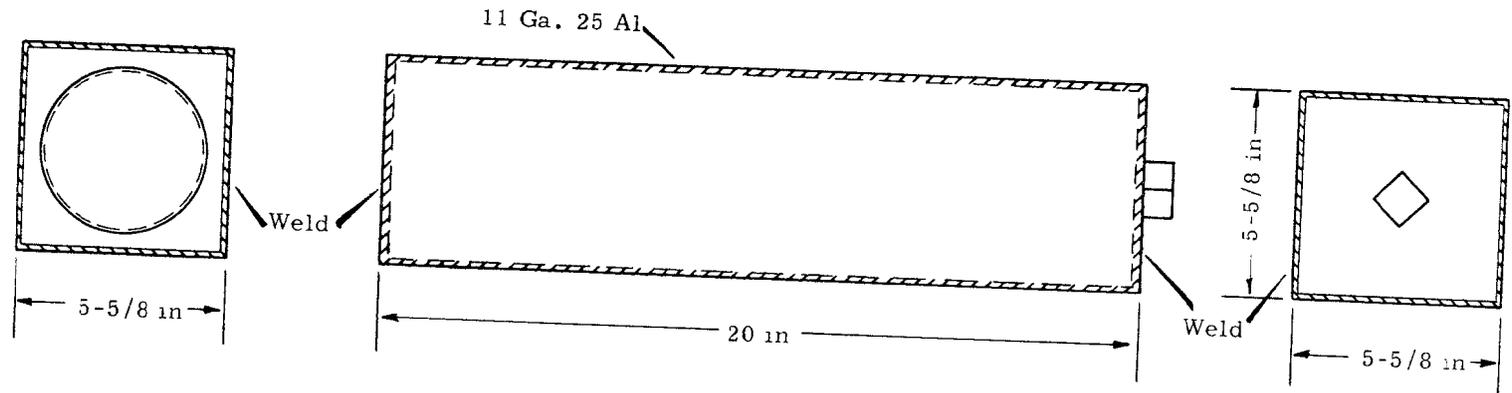


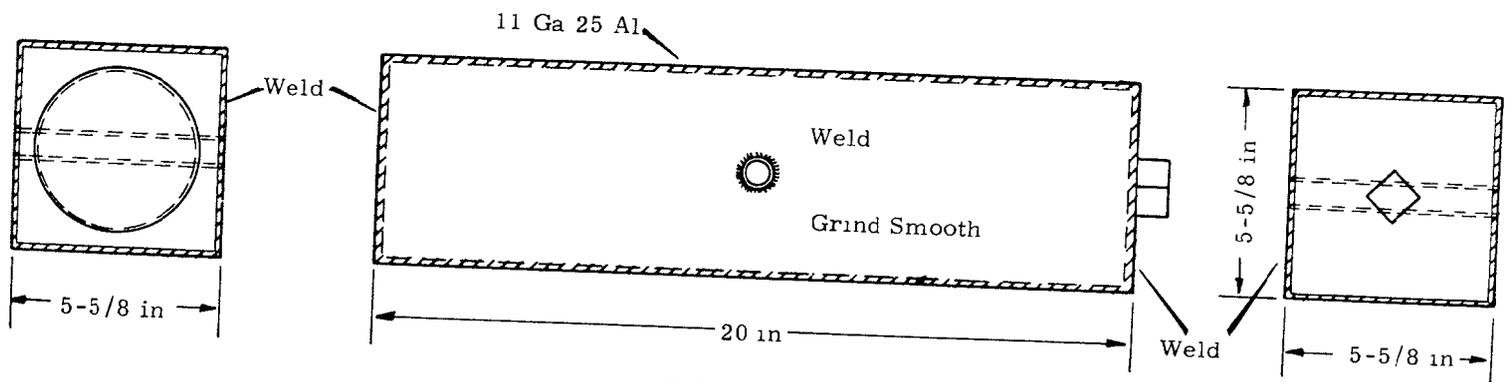
FIGURE 7  
Buffer Tank, Type B, Assembly

Scale: 1/4" = 1"



**FIGURE 8**  
Buffer Tank, Type C, Assembly

Scale: 1/4" = 1"



**FIGURE 9**  
Core Tank Assembly

Scale: 1/4" = 1"

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