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A MEASUREMENT OF THE INFINITE MEDIUM THERMAL NEUTRON MULTIPLICATION FACTOR FOR 2 WT. % U-235 ENRICHED URANIUM TETRA FLORIDE PARAFFIN MODERATED AT A HYDROGEN TO U-235 ATOMIC RATIO OF 195

A Thesis

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by

Victor Isaac Neeley

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The thesis of Victor Isaac Neeley, "A Measurement of the Infinite Medium Thermal Neutron Multiplication Factor of 2 wt. % U-235 Enriched Uranium Tetre Floride Paraffin Moderated at a Hydrogen to U-235 Atomic Ratio of 195", is hereby approved:

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BIOGRAPHICAL SKETCH OF THE AUTHOR

Victor Isaac Neeley was born in Bell, California on November 5, 1930. He attended the St. Ansgar High School, St. Ansgar, Iowa and received a diploma in 1949. That September he enrolled in St. Olaf College, Northfield, Minnesota. Following one year at St. Olaf College he served four years in the Navy. In September 1954 he enrolled in the University of Minnesota and in June 1957 he received the degree of Bachelor of Arts in Physics. In September 1957 he entered the Graduate School of the University of Idaho through the extension plan at the General Electric School of Nuclear Engineering, Richland, Washington and in June 1959 completed the requirements for the Master of Science degree, of which this thesis is a part.

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TABLE OF CONTENTS

A MEASUREMENT OF THE INFINITE MEDIUM THERMAL NEUTRON MULTIPLICATION FACTOR FOR 2 WT. % U-235 ENRICHED URANIUM TETRA FLORIDE PARAFFIN MODERATED AT A HYDROGEN TO U-235 ATOMIC RATIO OF 195

BY

VICTOR ISAAC NEELEY

INTRODUCTION

The processing, fabrication, and storage of fissile materials requires adequate knowledge of the criticality parameters of these materials

One of the most useful of these parameters is the infinite medium thermal neutron multiplication factor which is defined as the ratio of the total number of thermal neutrons absorbed, on the average, in one generation to the number of thermal neutrons absorbed in the preceding generation, in an infinite medium(1).

The knowledge of this factor for a particular fissile material may then enable the calculation of critically safe conditions under which the material may be handled or, enable calculations of the geometry of a reactor made up of this material.

The experimental determination of the infinite medium thermal neutron multiplication factor requires that certain assumptions be made. It is therefore of vital importance that methods be devised by which these assumptions may be verified.

PURPOSE

The research to be described has been carried out with the following definite objectives in view:

1. To measure the infinite medium thermal neutron multiplication factor of 2 weight % U-235 enriched UF$_4$ paraffin moderated at a hydrogen-to-U-235 atomic ratio of 195 as a parameter for criticality specifications for safe handling and storage of this material. This measurement was made by using the Physical Constants Testing Reactor at Hanford Atomic Products Operation, Richland, Washington.
2. To use this experimental measurement as a basis for verifying other measurements of this type made by using the Physical Constants Testing Reactor. Oak Ridge National Laboratory has conducted experiments of a different nature on this same material from which the infinite medium thermal neutron multiplication factor was obtained(2). A comparison of these answers was then made.

DISCUSSION OF THE PROBLEM

The infinite medium thermal neutron multiplication factor, hereafter referred to as k_∞ of a multiplying material may be determined from the quantity of thermal neutron absorber necessary to reduce k_∞ of the multiplying medium to unity(3) (4).

The quantity of thermal neutron absorber necessary may be determined from the principle that k_∞ of the infinite just critical medium and a vacuum are the same, namely, unity. Then the substitution of a void for a finite region of the material will produce no perturbation on the system(3). The necessary amount of thermal neutron absorber, hereafter referred to as the poison, to produce this condition may then be determined. The assumption is made that the addition of the poison to the medium changes only the thermal utilization of the medium(3) (4). The validity of this assumption will be shown in later arguments.

The infinite medium thermal neutron multiplication factor for the unpoisoned system is given by

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

where these terms have their usual meaning(1).

Then k_∞ for the poisoned system is given as

$$k_\infty = \eta \epsilon p f' = 1$$

then defining Δk_∞ as

$$\Delta k_\infty = k_\infty - 1$$

Δk_∞ is then found to be

$$\Delta k_\infty = k_\infty - 1 = \frac{k_\infty - k_\infty'}{k_\infty'} \quad \frac{f - f'}{f'}$$

The thermal utilization, f, for a homogeneous medium is defined as

$$f = \frac{(\Sigma_a \phi V)_{fuel}}{\sum_i (\Sigma_a \phi V)_i}$$

where "fuel" refers to the fissionable isotope U-235 and "i" refers to all the other materials in the system except the poison.

Then

$$\Delta k_\infty = \frac{f - f'}{f'} = \frac{\dfrac{(\Sigma_a \phi V)_{fuel}}{\sum_i (\Sigma_a \phi V)_i} - \dfrac{(\Sigma_a \phi V)_{fuel}}{\sum_i (\Sigma_a \phi V)_i + (\Sigma_a \phi V)_{poison}}}{\dfrac{(\Sigma_a \phi V)_{fuel}}{\sum_i (\Sigma_a \phi V)_i + (\Sigma_a \phi V)_{poison}}}$$

$$= \frac{(\Sigma_a V)_{poison}}{\sum_i (\Sigma_a V)_i}$$

The fluxes in these terms cancel out because the medium is completely homogeneous.

If the medium is homogeneous but the poison is of a heterogeneous nature, the same formalism may be kept with the introduction of the disadvantage factor for the poison which is defined as(1),

$$F = \frac{\phi_s}{\bar{\phi}_{poison}}$$

where ϕ_s refers to the flux at the surface of the poison and $\bar{\phi}$ refers to the average flux in the poison.

Then

(1) ○

$$\Delta k_{\infty} = \frac{\frac{\Sigma_a V}{F}\ \text{poison}}{\sum_i (\Sigma_a V)_i}$$

The quantity of poison necessary to reduce k_{∞} of a test medium to unity may be found in a finite reactor by providing a cavity in the reactor into which a sample of the poisoned medium may be placed and by providing means for adjusting the neutron energy spectrum incident on this cavity such that the incident spectrum is identical to that which would exist in an infinite medium of the test material.

The above is a simple description of the Physical Constants Testing Reactor at Hanford Atomic Products Operation, Richland, Washington. A cavity is provided which is much larger than the test sample° so that a layer of the medium, the buffer region, surrounds the test sample and brings the neutron energy spectrum into equilibrium. Further adjustment of the spectrum is made by changing the loading configuration of the reactor external to the cavity.

Equation (1) refers to the condition in which the test sample has been correctly poisoned. In actual practice this condition is not fulfilled in that it is quite difficult to obtain the exact amount of poison necessary. Instead the test sample is poisoned quite close to the correct amount and then a small amount of poison is added and an extrapolation is made to the correct amount. This extrapolation is made as follows:

By methods of perturbation theory it can be shown that(4)

$$\Delta\rho \propto M_{\text{poison}}\, \phi_{1/v}$$

where M_{poison} = mass of poison

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

and ρ is defined as the reactivity of the reactor.

Then rewriting equation (1)

$$\Delta k_{\infty} = \frac{\frac{(\Sigma_a V)}{F}_{\text{poison}} + \frac{(\Sigma_a V)}{F}_{\text{poison}}\, \gamma}{\sum_i (\Sigma_a V)_i}$$

where

$$= \left(\frac{\Delta\rho}{\phi_{1/v}}\right)_{\text{void}} \left(\frac{\phi_{1/v}}{\Delta\rho}\right)_{\text{poison}},$$

$$= \left(\frac{\rho_{\text{poison}} - \rho_{\text{void}}}{\rho_{\text{poison}} - \rho_{\text{poison}'}}\right) \frac{\phi_{1/v\,\text{poison}'}}{\phi_{1/v\,\text{void}}}$$

This may be rewritten as

$$(2)\quad \Delta k_{\infty} = \frac{\left(\frac{N\sigma_a}{P}\right)_{\text{poison}} + \left(\frac{N\sigma_a}{P}\right)'_{\text{poison}} \left(\frac{\rho-\rho_{\text{void}}}{\rho-\rho'}\right)\left(\frac{\phi_{1/v'}}{\phi_{1/v\ (\text{void})}}\right)}{\sum_i (N\sigma_a)_i}$$

where N_i = total number of gram atoms or molecules of the particular material

$(\sigma_a)_i$ = microscopic absorption cross section of the particular material

EXPERIMENTAL WORK

MATERIALS AND ASSEMBLY

The material, 2 wt. % U-235 enriched UF_4 paraffin moderated at a hydrogen-to-U-235 atomic ratio of 195, was fabricated at the Oak Ridge National Laboratory, Oak Ridge, Tennessee(2). The composition of the material was 92.0% UF_4 and 8% paraffin by weight. This gave a uranium concentration of 0.698 grams uranium per gram of material. The average particle size of the UF_4 was less than 0.020 inches. The material was pressed into blocks of various sizes (4 inches x 4 inches x 4 inches, 1 inch x 4 inches x 4 inches, 2 inches x 2 inches x 4 inches, etc.) for ease of handling and assembly. Each of these blocks was wrapped in a thin piece of aluminum foil to eliminate any contamination problems in handling the material.

This particular material was chosen because of a need for information in this general range of U-235 enrichment and because critical experiments could easily be conducted and thus a comparison of the values of the infinite medium thermal neutron multiplication factor obtained by two different methods could be made.

The poison used in this experiment consisted of boron carbide impregnated polyethylene containing 4 wt. % boron carbide(7). This material was in the form of 0.005 inch thick blown tubing. It was developed in the past for use in experiments of this type. The neutron absorption cross section of this poison was measured with respect to standard copper by danger coefficient methods in the Physical Constants Testing Reactor.

The assembled system was 24 inches x 24 inches x 30 inches with a 6-inch x 6-inch x 12-inch central section used as the test sample. This gave a 9-inch buffer region surrounding the test sample.

A 6 1/2-inch x 6 1/2-inch x 30 1/2-inch rectangular tube constructed from 1/4-inch 61ST aluminum was used to house the test sample and the buffer material on either end of the test sample. The test sample and two end buffers were contained in rectangular tanks. These tanks were constructed of 1/8-inch 61ST aluminum. The tank for the test sample was 6 1/4 inches x 6 1/4 inches x 12 1/4 inches and the two end buffer tanks were 6 1/4 inches x 6 1/4 inches x 9 1/4 inches. Figure (1) shows the partially assembled system and some of the different size blocks and Figure (2) shows the assembled system with the tanks being put in place.

A total of 1321.14 Kg of 2 wt. % U-235 enriched UF_4 paraffin moderated at a hydrogen-to-U-235 atomic ratio of 195 were used. This assembly contained 18.4 Kg of U-235.

Previous to this experiment the bare critical mass of this material had been measured by Oak Ridge National laboratory and was found to contain 25.3 Kg U-235$^{(2)}$. For a graphite reflected system the minimum critical mass was considerably less, and a good deal more than the minimum critical mass would be present when the system was assembled in the cavity of the Physical Constants Testing Reactor which is graphite moderated. For this reason, extreme care had to be exercised while assemblying the system. Each individual block was wrapped with poison before being placed in the cavity. The amount of poison had been previously determined from theoretical calculations of k_∞ and from an estimate of k_∞ based on the work done at Oak Ridge National Laboratory. The theoretical calculation of k_∞ is shown in the Appendix. Then based on these predictions of k_∞ the assembled poisoned system would have k_∞ equal to unity. Tables 1 and 2 give the mass of each material used in the test sample and buffer region as well as the 2200 meter/sec. cross section values and atomic or molecular weights.

Some copper was used to poison down the buffer region in addition to the boron carbide impregnated polyethylene. The reason for using copper was that since the boron carbide impregnated polyethylene was still in somewhat of a developmental stage not enough of it was available to do the complete poisoning.

The test sample tank was provided with holes and the blocks of material in it were drilled so that flux traverse measurements could be made in it. These holes were in the longitudinal and transverse directions to the test sample and extended three inches into the buffer region so that the traverses could be extended out into the buffer region.

EXPERIMENTAL PROCEDURE

The flux traverses were made by irradiating bare and cadmium covered gold foils placed in positions along the traverse holes. Figure 3 shows the traverse bars and the gold foils in place. These foils are 0.25 inch x 0.5 inch x 0.005 inch gold and the cadmium dishes used were 0.040 inch thick. The activity of the foils was obtained by placing them on a 3-inch crystal scintillation counter and reading the gamma from Au^{198} decay. The activities were normalized to unit mass of the foils and to a constant reactor power level by use of a monitor foil placed outside the assembly.

A test sample tank filled with helium was used as a substitute for a void. Helium is very good for this purpose because of its low neutron absorption and scattering cross sections.

The method used to find the correct amount of poison and hence k_{∞} was then to measure the reactivity of the reactor at a particular control rod setting with the test sample in place. The test sample was then removed, and with the control rods positioned at the same previous setting, a reactivity measurement of the test sample tank filled with helium was made. The poison in the test sample was then adjusted towards the correct value. A flux traverse measurement was then made and the poison in the buffer region adjusted to obtain the correct incident flux. These measurements were repeated and adjustments made until the test sample was poisoned to within 1% of the correct value and the correct neutron energy spectrum was incident on the test sample. A small amount of poison was then added to the test sample and the reactivity and flux traverse measurements were again made. Then as shown in previous arguments an extrapolation was then made to the correct value.

Further measurements were made to observe the effect of overpoisoning the buffer and poisoning the test sample completely heterogeneously. In the latter experiment the poison was placed outside the test sample rather than around the individual blocks. The purpose of these experiments was to verify some work done previously on low U-235 enrichments for which the heterogeneous type poisoning was used(8). Perturbation theory would predict that each of the two methods give the correct answer.

Figures 4 and 5 are examples of the traverses made with the bare and cadmium covered gold foils. The cadmium ratio plots were used to insure the incident neutron energy spectrum was the correct spectrum for the material.

Table (3) shows the values of k_∞ and other results obtained from experimental data before correction factors were introduced.

DISCUSSION OF ERROR ANALYSIS

It can be shown by two group analysis that the error in $k_\infty - 1$ can be minimized by requiring that the cadmium ratio be constant in the test sample and out into the buffer region$^{(4)}$. The error involved here results from spectral mismatching in the test sample. An expression for this error may be written

$$\sigma \Delta k_\infty = \frac{\left(\frac{\phi_1'}{\phi_2'} - \frac{\phi_1}{\phi_2}\right)\left(\frac{m_1'}{m_2'} - \frac{m_1}{m_2}\right)}{\left(\frac{\phi_1}{\phi_2}\right)\left(\frac{m_1}{m_2}\right)}$$

where ϕ_1/ϕ_2 and m_1/m_2 are the fast-to-slow flux and adjoint flux ratios which would be found in the infinite critical poisoned medium and ϕ_1'/ϕ_2' and m_1'/m_2' are the similar flux ratios in the test sample. Thus if either the flux spectrum or the adjoint flux spectrum is matched the spectral error in k_∞ will vanish.

The assumption has been made that the addition of poison to the system changes only the thermal utilization of the system. The addition of the poison has two effects. First, it causes a slight shift in the neutron temperature, and second, because of the polyethylene contained in the poison the hydrogen-to-U-235 ratio is slightly increased. The second of these is the largest and can be corrected for by observing the effect of the addition of a small amount of polyethylene and extrapolating this effect back to the correct value. This correction amounts to a 2 per cent decrease in the observed value of Δk_{∞}. The first effect is more difficult to determine; however, by varying the cadmium ratio over the range involved it was determined that the combined effect of this and any possible mismatch in the spectrum was no greater than 1 per cent in Δk_{∞}. This is included as an error in the error analysis.

The latter effect results from slight changes in η and p which are dependent on neutron temperature; from their basic definitions, it can be seen no other change occurs in η, ϵ or p because of the addition of the poison$^{(1)}$.

One of the most difficult problems in an experiment of this type is the determination of the proper absorption cross section values for the different materials. Because of the low moderation and high enrichment the effective neutron temperature was considerably higher than for most common thermal reactor systems. The use of standard 2200 meter/sec. cross section values for the different materials in the system was introducing an error because of the higher neutron temperature and in addition the neutron energy spectrum was not the common Maxwellian with 1/E tail for which these cross sections were designed(10). The magnitude of this effect can be determined by use of some fairly recent developments in this field. A table of effective cross section values has been tabulated for systems containing a mixture of hydrogen, U-235 and a 1/v absorber(9). These cross sections were obtained by calculating the Wigner-Wilkens spectrum for different mixtures of these ingredients and averaging the cross sections over the spectrum by dividing the spectrum into 30 groups. All the materials in this experiment except U-235 and hydrogen were then classified as the 1/v absorber. Any non 1/v effect of carbon and flourine are negligible due to their low cross sections and boron is strictly a 1/v absorber. Then the only discrepancy in this method is the effect the U-238 resonances have on the spectrum. In spite of this fact it is believed that this is a better approximation than the 2200 meter/sec. values since most of the U-238 resonance effect is accounted for in the resonance escape probability term.

Using this Wigner-Wilkens method a 2.8% increase in the value of k_∞ is obtained over the value obtained from 1/v

2200 meter cross section values. Because of the inexactness of this correction the value of the increase is also included as an error in the final answer.

This then results in a value of k_∞ of 1.216 ± 0.013. The rest of the error analysis is shown in the Appendix.

SUMMARY AND CONCLUSIONS

Values of k_∞ obtained from experimental data are listed in Table 3. Six different measurements of k_∞ were made with the sixth measurement being made by a different method of poisoning the test sample. All of these measurements are in good agreement, and a simple average value is taken as the final value. This final value was then corrected for a slight change in hydrogen to U-235 ratio caused by the hydrogen content of the material used as the thermal neutron absorber. A further correction was made to account for the abnormal neutron energy spectrum which was present in this system. The final corrected value of k_∞ was then found to be 1.216 ± 0.013.

A theoretical calculation of k_∞ is shown in the Appendix and gives a k_∞ value of 1.23. This is in good agreement with the experimental value.

Critical experiments done on this material at Oak Ridge National Laboratory show that the minimum critical mass for a bare "square" cylinder contains 25.8 kg of U-235[2]. A calculated value of the amount of U-235 in a just critical bare square cylinder was obtained using the experimental k_{∞} value. Again there was good agreement with the experimental results. This calculation is shown in the Appendix.

A rather unique atmospheric contamination problem was encountered and is also discussed in the Appendix.

In conclusion, the value of k_{∞} for 2 wt. % U-235 enriched Uranium Tetra Floride paraffin moderated at a hydrogen to U-235 atomic ratio of 125 was found to be 1.216 ± 0.013.

As a result of this experiment, the Physical Constants Testing Reactor was found to be a valid instrument for determining criticality parameters for specifications of safe handling and storage conditions of fissile materials.

APPENDIX

THEORETICAL CALCULATION OF k_{∞}

The value of k_{∞} may be calculated by use of standard 2200 meter/sec cross sections and the knowledge of the materials in question. The method used was to calculate k_{∞} from the four factor formula

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

where η, ϵ, p and f have their usual meaning and may be written as follows(1).

$$\eta = \frac{\nu N^{235} \sigma_f^{235}}{N^{235} \sigma_a^{235} + N^{238} \sigma_a^{238}}$$

$$p = e^{-\frac{3.9}{\bar{\xi}}} \left[\frac{N_a^{238}}{\Sigma_s}\right]^{.585}$$

$$f = \frac{N^{235} \sigma_a^{235} + N^{238} \sigma_a^{238}}{N^{235} \sigma_a^{235} + N^{238} \sigma_a^{238} + N^{C25H52} \sigma_a^{C25H52} + N_F \sigma_a^{F}}$$

where

$\nu = 2.47$ $\sigma_f^{235} = 571$

$N^{235} = .0203$ $\sigma_a^{235} = 681$ $\sigma_s^{235} = 10$ $\xi^{235} = .00838$

$N^{238} = .9797$ $\sigma_a^{238} = 2.71$ $\sigma_s^{238} = 8.3$ $\xi^{238} = .00838$

$N^{F} = 4$ $\sigma_a^{F} = .010$ $\sigma_s^{F} = 3.9$ $\xi^{F} = .1000$

$N^{H} = 3.935$ $\sigma_a^{H} = .332$ $\sigma_s^{H} = 20$ $\xi^{H} = 1.00$

$N^{C} = 1.999$ $\sigma_a^{C} = .0034$ $\sigma_s^{C} = 4.9$ $\xi^{C} = .158$

$$\bar{\xi} = \frac{\sum_i \left(\xi_i N_i \sigma_{si}\right)}{\sum_i \left(N_i \sigma_{si}\right)}$$

$$\Sigma_s = \sum_i \left(N_i \sigma_{si}\right)$$

The calculation of ϵ is taken from a partly theoretical partly empirical formula whose only justification is that from past experience it seems to work.

Then

$$\epsilon = 1 + \left(\frac{\nu \Sigma_f^{238}}{\Sigma_T} - \frac{\Sigma_a^{238}}{\Sigma_T} \right) \left[\left(\frac{\Sigma_e + \nu \Sigma_f^{238}}{\Sigma_T} \right) (\bar{\gamma} - 1) + 1 \right]$$

ν = 2.55 neutrons per fission

$\Sigma_f = .29 \text{ cm}^{-1}$

$\Sigma_a = .39 \text{ cm}^{-1}$

$\Sigma_e = \sum_i (\alpha \, \sigma_e)_i = 36.24 \text{ cm}^{-1}$

α = number of atoms per cm.^{3}

σ_e = microscopic elastic scattering cross section

$\sum_i$ = sum on all materials

$\Sigma_T = \Sigma_e + \Sigma_f^{238} + \Sigma_I^{238} + \Sigma_a^{238} = 39.04 \text{ cm}^{-1}$

Σ_I = macroscopic inelastic scattering cross section of U^{238}

$$\bar{\gamma} = \frac{\sum_i (\gamma \, \alpha \, \sigma_e)_i}{i \sum_i (\sigma_e \alpha)_i}$$

$$\gamma_i = \frac{\ln \frac{E_0}{E_1}}{\xi_i}$$

E_0 = 2 mev.

E_1 = 1.1 mev. (U-238 fission threshold)

$\bar{\gamma}$ was found to be 1.67 collisions.

$\epsilon - 1$ has to be reduced by approximately 3% to account for the inadequate treatment of the energies of fission neutrons. Then from the above formulas the following values were obtained:

$$p = .747$$

$$\eta f = 1.60$$

$$\epsilon = 1.03$$

Then

$$k_{\infty} = 1.23$$

The error in a calculation of this type has normally been assumed to be about 5% of k_{∞}. The error was mainly due to inadequate theoretical treatment and errors in cross section values.

Then the value of k_{∞} from a theoretical calculation was in good agreement with the experimental value of k_{∞} for this material.

COMPARISON OF RESULTS FROM P.C.T.R. k_∞ MEASUREMENT AND OAK RIDGE NATIONAL LABORATORY CRITICALITY EXPERIMENTS

The bare critical size of a reactor may be obtained from the critical buckling of the reactor. The critical buckling may be obtained from the following equation if values for k_∞, L^2 and τ are known.

$$1 = \frac{k_\infty e^{-B^2\tau}}{1 + L^2B^2}$$

Where B^2 is the geometrical buckling, L^2 is diffusion length squared, and τ is the age to thermal, and k_∞ is the experimental value (1.216).

The following formulas were used to obtain these quantities;(1,11)

$$L^2_{(H/U = 195)} = L^2_{(H_2O)} \frac{(\Sigma_{tr}\,\Sigma_a)_{(H_2O)}}{(\Sigma_{tr}\,\Sigma_a)_{(H/U = 195)}}$$

$$\tau_{(H/U=195)} = \frac{\tau_{(paraffin)}}{\left(\frac{\rho_{(mod.)}}{\rho_{(paraffin)}} + \frac{\Sigma_{sl}^{fuel}}{\Sigma_{sl}^{mod.}}\right)\left(\frac{\rho_{(mod.)}}{\rho_{(paraffin)}} + \frac{\Sigma_{tr}^{fuel}}{\Sigma_{tr}^{mod.}}\right)}$$

$$\tau_{(paraffin)} = \tau_{(H_2O)} \frac{(\Sigma_a\,\Sigma_{tr})_{(H_2O)}}{(\Sigma_a\,\Sigma_{tr})_{(paraffin)}}$$

$$\Sigma_{sl} = \Sigma_{s} + \Sigma_{\text{inelastic}}$$

$$\Sigma_{tr} = \Sigma_{s}(1 - \bar{\mu}_0) \qquad \bar{\mu}_0 = \frac{2}{3A}$$ (A = atomic or molecular weight)

$\rho_{\text{(mod.)}}$ = density of moderator

$\rho_{\text{(paraffin}}$ = density of paraffin

$$B^2 = \left(\frac{2.405}{R + \lambda}\right)^2 + \left(\frac{\pi}{H + 2\lambda}\right)^2$$

λ = 3.4 cm. = bare extrapolation length

$L^2_{(H_2O)}$ = 7.45 cm.^{2}

$\tau_{(H_2O)}$ = 27 to 31.4 cm.^{2} (without actual measurements of τ it is difficult to establish a value better than these limits).

When the above formulas were applied, the amount of U-235 contained in the critical mass of a bare "square" cylinder was found to be from 23.6 to 30.1 Kg. U-235 depending on the value of $\tau_{(H_2O)}$ (27 to 31.4 cm.^{2}). The value obtained from critical experiments performed at the Oak Ridge National Laboratory was 25.8 Kg. U-235$^{(2)}$. Again there is good agreement between the two methods within the restrictions imposed by the $\tau_{(H_2O)}$ values.

ERROR ANALYSIS

The error in the measurement of k_∞ is divided into three main parts.

1. Error due to spectral mismatch and possible changes in γ and p due to the addition of poison.
2. Error due to the use of incorrect cross section for these materials caused by non-Maxwellian 1/v-type spectra and higher effective neutron temperatures.
3. Errors in the standard cross section values and in experimental methods.

The first two have been discussed in the main part of the text and the third will be discussed here.

The method of determining this error was to use standard propagation of error technique.

$$\sigma^2(T) = \sum_i \left[\frac{\partial T}{\partial x_i} \, \sigma(xi) \right]^2$$

Then from the equation

$$\Delta k_\infty = \frac{(\Sigma_a V)_{poison}}{\sum_i (\Sigma_a V)_i}$$

where $(\Sigma_a V)_{poison}$ is the correct poison.

One can write

$$\Delta k_\infty = \frac{\left(\frac{\Sigma_a V}{F}\right)_{poison} + \left(\frac{\Sigma_a V}{F}\right)'_{poison} \left(\frac{\rho_1 - \rho_{He}}{\rho_1 - \rho_2}\right) \frac{\phi_2 \, 1/v}{\phi_{He} \, 1/v}}{\sum_i (\Sigma_a V)_i}$$

and further

$$\Delta k_{\infty} = \frac{M \frac{\sqrt{a}}{FW}_{\text{poison}} + M \frac{\sqrt{a}}{FW}_{\text{poison}} \left(\frac{\rho_1 - \rho_{He}}{\rho_1 - \rho_2}\right)\left(\frac{\phi_{1/v\,2}}{\phi_{1/v\,He}}\right)}{M \frac{\sqrt{a}}{W}_{U-238} + M \frac{\sqrt{af}}{W}_{U-235} + M \frac{\sqrt{a}}{W}_{C_{25}H_{52}} + M \frac{\sqrt{a}}{W}_{CH_2} + M \frac{\sqrt{a}}{W}_{F_{19}}}$$

where

M = Mass of material in test sample

W = atomic or molecular weight

Let

$$a = \left(M \frac{\sqrt{a}}{WF}\right)^2$$

$$b = \left(M \frac{\sqrt{a}}{WF}\right)^2 \left(\frac{\rho_1 - \rho_{He}}{\rho_1 - \rho_2}\right)^2 \left(\frac{\phi_{1/v2}}{\phi_{1/vHe}}\right)^2$$

$$c = \left[\frac{1}{\left(M \frac{f\sqrt{a}}{W}\right)_{U-235} + \left(M \frac{\sqrt{a}}{W}\right)_{238} + \left(M \frac{\sqrt{a}}{W}\right)_{C_{25}H_{52}} + \left(M \frac{\sqrt{a}}{W}\right)_{CH_2} + \left(M \frac{\sqrt{a}}{W}\right)_{F\,19}}\right]^2$$

$$d = \left(M \frac{\sqrt{af}}{W}\right)^2_{U-235}$$

$$e = \left(M \frac{\sqrt{a}}{W}\right)^2_{U-238}$$

$$f = \left(M \frac{\sqrt{a}}{W}\right)^2_{C_{25}H_{52}}$$

$$g = \left(M \frac{\Sigma a}{W} \right)^{2}_{CF}$$

$$h = \left(M \frac{\Sigma a}{W} \right)^{2}_{F-19}$$

then

$$\sigma^{2} (A k_{\infty}) = C a \left[\frac{\sigma^{2}_{M_{poison}}}{M^{2}_{poison}} + \frac{\sigma^{2}_{\left(\frac{\Sigma}{W}\right)_{poison}}}{\left(\frac{\Sigma}{W}\right)^{2}_{poison}} + \frac{\sigma^{2}_{F_{poison}}}{F^{2}_{poison}} \right]$$

$$+ Cb \left[\frac{\sigma^{2}_{M'_{poison}}}{M^{2}_{poison}} + \frac{\sigma^{2}_{\left(\frac{\Sigma}{W}\right)_{poison}}}{\left(\frac{\Sigma}{W}\right)^{2}_{poison}} + \frac{\sigma^{2}_{F_{poison}}}{F^{2}_{poison}} + \frac{\sigma_{\rho_1}^{2} + \sigma_{\rho_{He}}^{2}}{(\rho_{1} - \rho_{He})^{2}} \right.$$

$$\left. + \frac{\sigma_{\rho_1}^{2} + \sigma_{\rho_2}^{2}}{(\rho_{1} - \rho_{2})^{2}} + \frac{\sigma^{2}_{\phi_{1/v\,2}}}{\phi_{1/v_2}^{2}} + \frac{\sigma^{2}_{\phi\,1/v\,He}}{\phi_{1/v\,He}^{2}} \right]$$

$$+ c^{2} (\sqrt{a} + \sqrt{b})^{2} \left[d \left(\frac{\sigma^{2}_{M_{235}}}{M_{235}^{2}} + \frac{\sigma^{2}_{\left(\frac{\Sigma}{W}\right)_{235}}}{\left(\frac{\Sigma a}{W}\right)^{2}_{235}} + \frac{\sigma_{f_{235}}^{2}}{f_{235}^{2}} \right) \right.$$

$$E \left(\frac{\sigma^{2}_{M_{238}}}{M_{238}^{2}} + \frac{\sigma^{2}_{\left(\frac{\Sigma}{W}\right)_{238}}}{\left(\frac{\Sigma a}{W}\right)^{2}_{238}} \right) + f \left(\frac{\sigma^{2}_{M_{C_{25}H_{52}}}}{M^{2}_{C_{25}H_{52}}} + \frac{\sigma^{2}_{\left(\frac{\Sigma}{W}\right)_{C_{25}H_{52}}}}{\left(\frac{\Sigma a}{W}\right)^{2}_{C_{25}H_{52}}} \right)$$

$$g\left(\frac{\sigma^2_{M_{CH_2}}}{M^2_{CH_2}} + \frac{\sigma^2\left(\frac{\Sigma}{v}\right)_{CH_2}}{\left(\frac{\Sigma_a}{v}\right)^2_{CH_2}}\right) + h\left(\frac{\sigma^2_{M_{F-19}}}{M^2_{F-19}} + \frac{\sigma^2\left(\frac{\Sigma}{v}\right)_{F-19}}{\left(\frac{\Sigma_a}{v}\right)^2_{F-19}}\right)\Bigg]$$

Values of $\frac{\sigma_{Mi}}{Mi}$ and $\frac{\sigma\left(\frac{\Sigma}{v}\right)_i}{\left(\frac{\Sigma}{v}\right)_i}$ have been given in Table 1.

Other values were

$$\frac{\sigma_{\rho_1}^2 + \sigma_{\rho_2}^2}{\left(\rho_1 - \rho_2\right)^2} = 1.51 \times 10^{-2} \qquad \frac{\sigma_{\rho_1}^2 + \sigma_{\rho_{He}}^2}{\left(\rho_1 - \rho_{He}\right)^2} = 1.194 \times 10^{-2}$$

$$\frac{\sigma^2_{\phi_{1/v_1}}}{\phi^2_{1/v\ 1}} = 4.66 \times 10^{-3}$$

Then

$$\sigma^2 (\Delta k_\infty = 13.44 \times 10^{-6}$$

Then the error introduced because of incorrect 2200 meter/sec. cross sections and because of experimental methods was found to be

$$\sigma(\Delta k_\infty) = 3.7 \times 10^{-3}$$

ATMOSPHERIC CONTAMINATION

A rather unique air contamination problem was encountered during the course of the experiments. Personnel entering the reactor room were found to have parts of their clothing contaminated. The parts were located around the knees, elbows, and waist. The contamination was of quite low activity and had a half life of approximately 15 minutes so presented no serious clothing contamination problem. It was found that fission product gases were being released from the assembly. These gases were then decaying and producing some ionized particles. The ionized particles were then being picked up on the particular parts of clothing where electrostatic charge was built up through friction. This assumption was checked by placing charged and uncharged plastic rods in the reactor room. No activity was present on the uncharged rods while the charged rod activity was easily measurable. Air samples were taken by forcing reactor room air through blotter paper. The isotopes present were then identified by gamma spectrographic methods. The following isotopes were found to be present: Rb^{88}, Te^{133}, Te^{132}, I^{133}, I^{132}, Ce^{141} and Ce^{143}. Rb^{88} was found in the greatest concentration. As would be suspected these isotopes are mainly the daughters of fission product gases which were escaping from the assembly.

The calculation which follows gives an order of magnitude idea of the depth from which these gases escaped through the material.

From gold foil irradiations, the flux in the system was obtained using the formula

$$\phi = \frac{A}{\Sigma_a V\left(1 - e^{-\frac{0.6931\,t}{T}}\right)}$$

where

A = activity of the gold foil

V = volume of the foil

τ = half-life of gold

A ten minute irradiation at 10 watts power level in this system gave a count rate of approximately 5×10^3 counts per minute on the three-inch scintillation counter. Assuming the scintillation counter subtends half the total solid angle this gives

$$\phi = 10^8 \text{ neutrons cm.}^{-2} \text{ sec.}^{-1}.$$

The total number of fissions that occur in a ten-minute irradiation at ten watts was then $\Sigma_f \phi V t$, where Σ_f was the macroscopic fission cross section for the assembly and V is the volumn. Σ_f was found to be 0.0907 cm.$^{-1}$. V was found to be 2.93×10^5 cm.^{3}. Then $\Sigma_f V \phi t = 1.5 \times 10^{15}$ fissions. Assuming that only the fission product from the Z^{88} chain escapes into the atmosphere, and that the fission product yield is 3.57 per cent, then the total number of Br^{88} atoms created was $Br^{88}_{total} = (0.0357)(1.5 \times 10^{15}) = 5.4 \times 10^{13}$ atoms. The Z^{88} fission product chain is

$$Br^{88}_{total}\ (15.5 \text{ sec.}) \begin{cases} \rightarrow (4\%)\ Kr^{87}\ (37 \text{ min.}) + \text{neutron} \\ \rightarrow (96\%)\ Kr^{88}\ (2.8 \text{ hr}) \rightarrow Rb^{88}\ (17.8 \text{ min.}) \end{cases}$$

$$\longrightarrow Sr^{88} (\text{stable})$$

Where the times are the half-lives of these isotopes.

Then the amount of Rb^{88} present at time t after shutdown of the pile was

$$Rb^{88}(t) = No \left(1 - e^{-\frac{.693\,t}{\tau_{Br}}}\right)\left(1 - e^{-\frac{.693\,t}{\tau_{Kr}}}\right) e^{-\frac{.693\,t}{\tau_{Rb}}}$$

The assumption was made that at time $t = 0$ all the Br^{88} was formed. This assumption will hold for calculating the amount of Rb^{88} since $\tau_{Kr^{88}} \gg t$.

t (minutes)	Rb Atoms
0	0.0
30	2.0×10^{12}
60	1.1×10^{12}
120	1.9×10^{11}
240	3.2×10^{9}

Then assuming 4.7% of the reactor room atmosphere was sampled in a period of ten minutes one hour after shutdown and that the filter paper collects 30% of the Rb^{88} atoms passing through it, then the number of Rb^{88} atoms on the filter paper was

$$\int_{60}^{70} (.047)\,(.30)\,\underline{Rb^{88}(t)}\,dt = 1.42 \times 10^{11} \text{ atoms of } Rb^{88}$$

Assuming the Geiger-Müller counter sees 30% of the disintegrations on the filter paper, the count rate per minute was

$$(.30)\,(1.42 \times 10^{11})\left(1 - e^{-\frac{.693}{\tau_{Rb^{88}}}}\right) = 1.6 \times 10^{9} \text{ counts/min.}$$

The actual count rate observed with a Geiger-Müller counter was approximately 8×10^5 counts/minute. It was then assumed that $\frac{8 \times 10^5}{1.6 \times 10^9} = .05\%$ of the fission product gases were escaping. Assuming the assembly to be made up of 4-inch x 4-inch x 4-inch blocks, the total volume was $24 \times 24 \times 30 = 17280$ inches^3 and the total surface area was $(270)(96) = 25920$ inches^2. Then all the gases were escaping from a depth of $\frac{(17280)(.0005)}{25920} = .003$ inches into the material. This does not seem too unreasonable for this tightly bound paraffin material.

TABLE (1)

MATERIALS IN TEST SAMPLE

Material	**Weight in gm.**	**Molecular Weight**	σ_a (at 2200 meters/sec.) in barns	$(\sigma_a/w)^+$ w = atomic or molecular wt.
U-238	21762 ± 62	238.125	2.71	.01138 ± .00008
U-235	440 ± 4	235.117	691	2.396 ± .036
Paraffin $C_{25}H_{52}$	2522 ± 9	352.796	17.349	.04919 ± .00030
Fluorine F-19	6009 ± 25	19.005	<.010	<.0005
Polyethylene CH_2	167.1 ± .4	14.031	.6674	.04756 ± .00028
Poison (4 wt. % Boron carbide impregnated polyethylene)	178.0 ± .4	--	--	2.036 ± .022*

+ All errors are standard deviations.

* Measured value.(3) All other values of σ_a (2200 meters/sec.) taken from BNL-325, Second Edition 1958 (See reference 10).

TABLE (2)

MATERIALS IN BUFFER REGION

Material	Weight in Kg
U-238	876.67
U-235	17.90
Paraffin $C_{25}H_{52}$	102.60
Fluorine F-19	284.66
Polyethylene CH_2	3.22
Poison (1) Borated polyethylene	3.36
(2) Copper	102.60

Because of the lack of the boron carbide impregnated polyethylene some copper had to be used in poisoning down the buffer region. To avoid any possible complications the copper was placed toward the outside and the boron impregnated polyethylene placed toward the central region.

TABLE (3)

EXPERIMENTAL DATA AND VALUES OBTAINED

Type of Incident Flux *	$\frac{\rho_1 - \rho_{He}}{\rho_1 - \rho_2}$	$\frac{\phi_{1/v_2}}{\phi_{1/v_0}}$ **	$\left(\frac{\Sigma\sigma_a}{v}\right)'_{poison} \left(\frac{\rho_1 - \rho_{He}}{\rho_1 - \rho_2}\right)\frac{\phi_{1/v_2}}{\phi_{1/v_0}}$	k_∞ +
Thermal Loading	1.126	1	5.60	1.2120
Intermediate Loading	1.041	1	5.27	1.2117
Fast Loading	1.196	1	5.94	1.2122
Intermediate Loading with addition of 5959 gm of Cu on front face	1.203	1	6.43	1.2124
Intermediate Loading with addition of 5959 gm of Cu on front face plus 19.3 gm poison on inside of rectangular tube at test cell	1.344	1	6.69	1.2126

(Continued on page 33)

TABLE (3) (continued)

Type of Incident Flux *	$\frac{\rho_1 - \rho_{Be}}{\rho_1 - \rho_2}$	$\frac{\phi_{1/v_2}}{\phi_{1/v_0}}$ **	$\left(\frac{N \Gamma_a}{\gamma}\right)_{poison} \Big/ \left(\frac{\rho_1 - \rho_{Be}}{\rho_1 - \rho_2}\right) \frac{\phi_{1/v_2}}{\phi_{1/v_0}}$	k_∞ +
Intermediate Loading with poison placed on outside of test sample instead of around the individual blocks.	1.506	1	32.51	1.2103

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*These descriptions of the incident flux are only relative to one another.

**The actual experimental values of $\phi_{1/v_2} / \phi_{1/v_0}$ were not exactly unity; however, they are unity within the experimental error.

+ These values have not been corrected for the increase due to polyethylene addition nor for the Wigner-Wilkins spectrum technique.

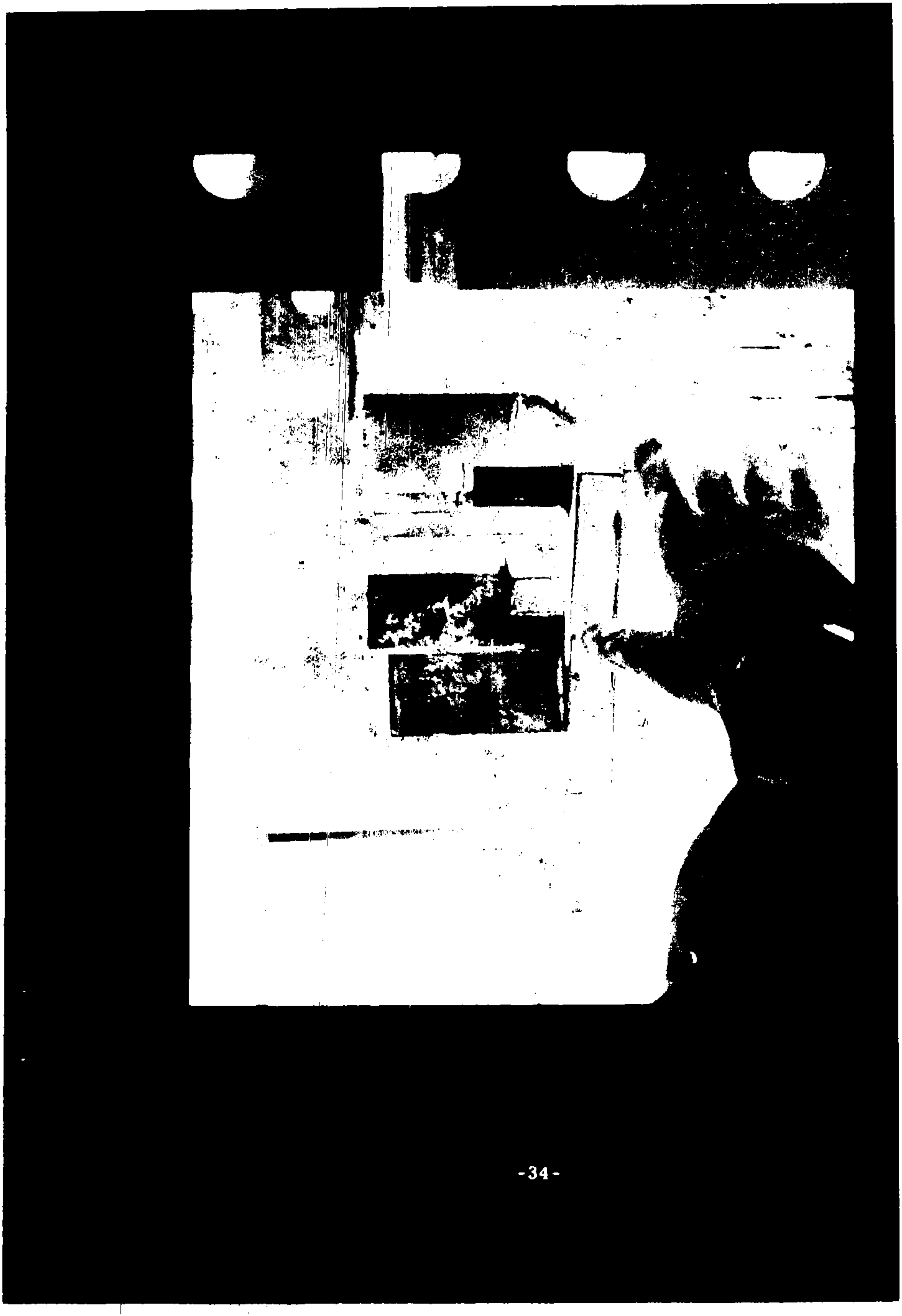

FIGURE 2

Insertion of Test Sample into Assembled System

FIGURE

T[illegible] S[illegible] and [illegible] Materials

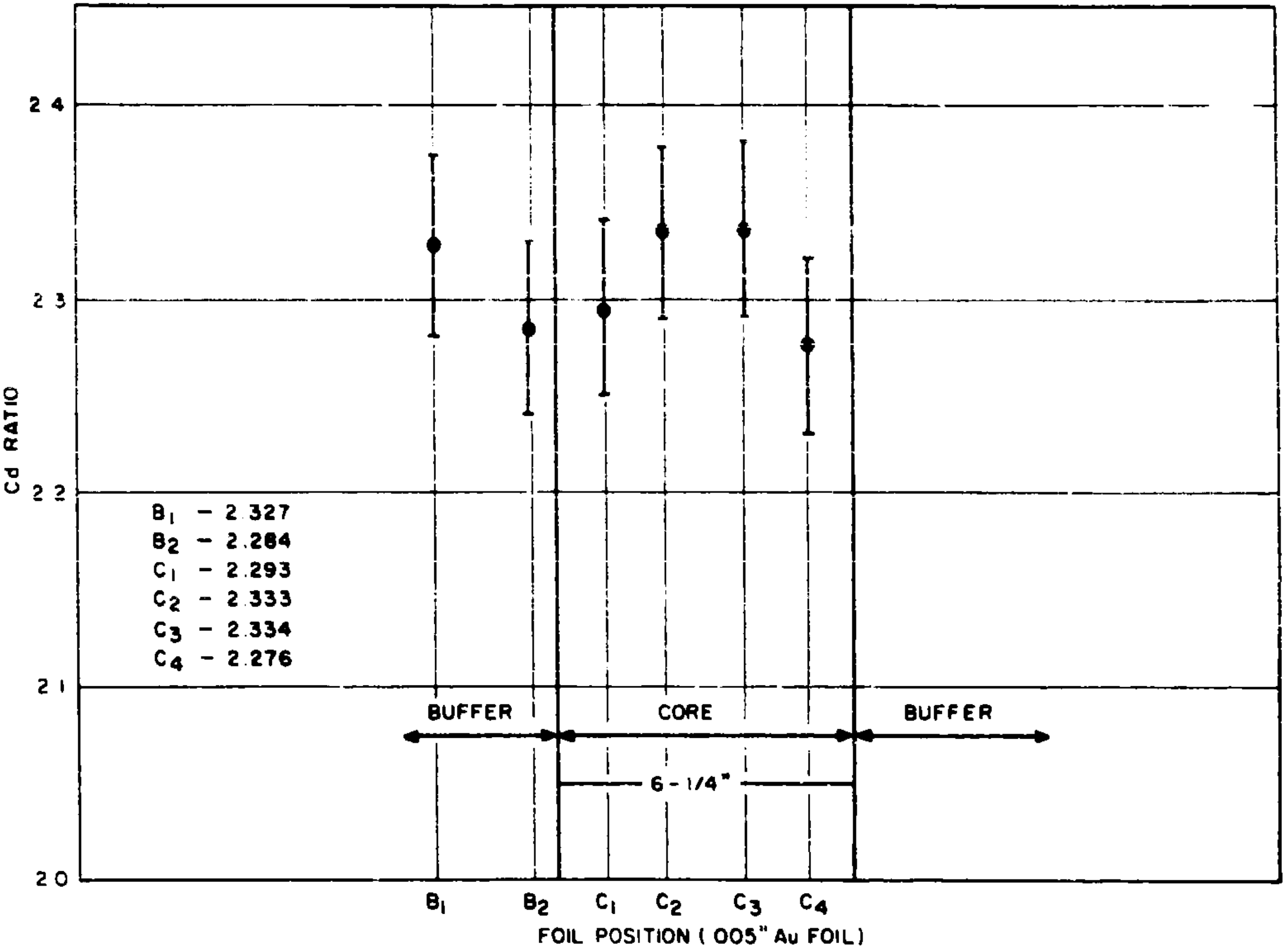

FIGURE 4
RADIAL Cd. RATIOS VS. FOIL POSITION

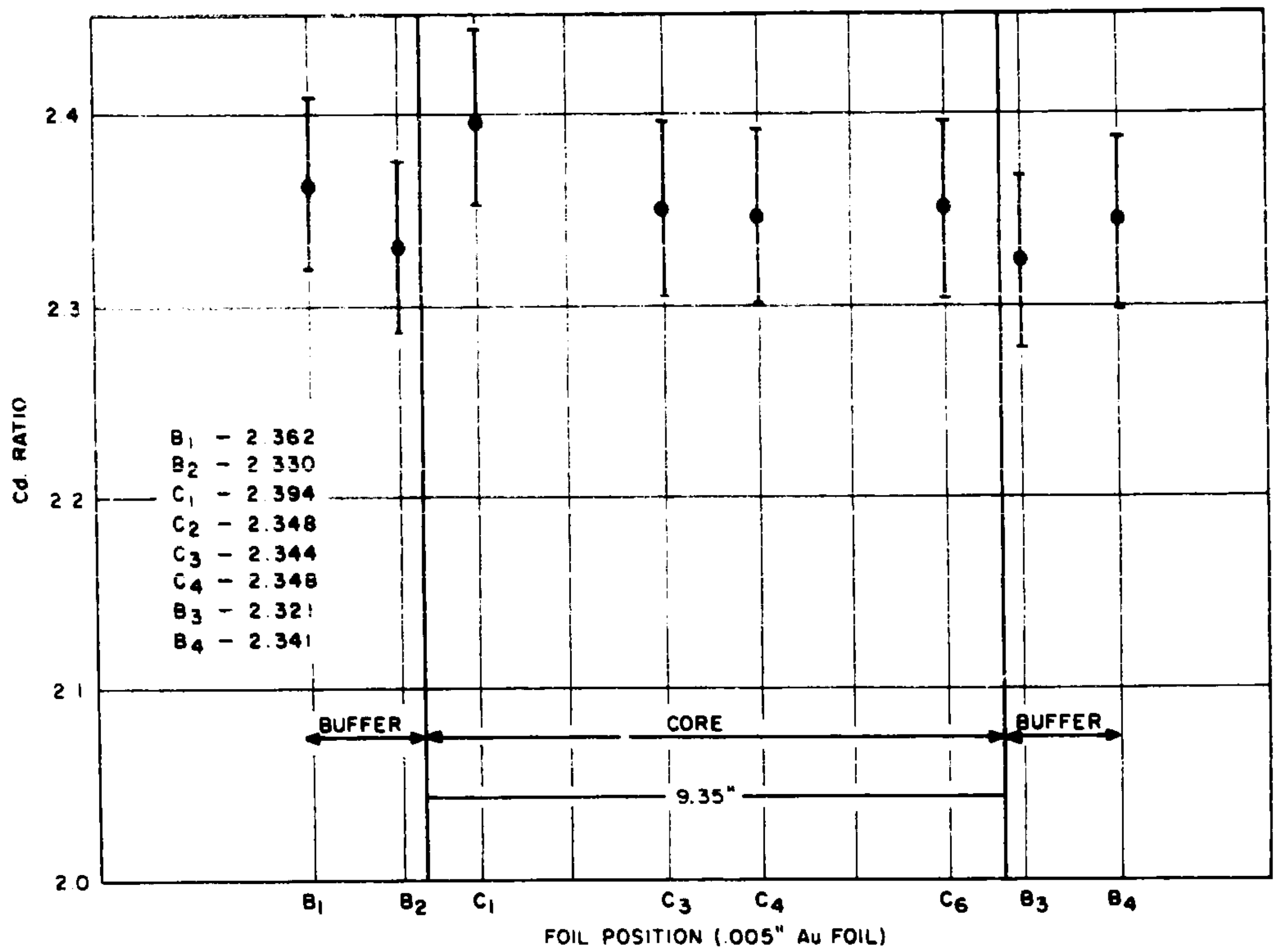

FIGURE 5
LONGITUDINAL Cd. RATIOS VS. FOIL POSITION

7L - SA 83

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