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IN GRAPHITE

*AEC Research and Development Report*



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Page ii, Acknowledgment, line 5.

Change "AEC" to read AE-6

Page 19, Reference 14, line 2.

Delete "Resonance in the Calculation of"

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# EXponential Experiment With A Thorium-Uranium Fuel In Graphite

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## ABSTRACT

An exponential experiment with multirod fuel elements in graphite moderator is described. The fuel is a thorium-uranium alloy. The buckling, thermal utilization, and resonance escape probability for one lattice configuration have been measured. In the analysis, emphasis is placed on determining the effective resonance integral of the fuel element. From the measured resonance escape probability,  $0.856 \pm 0.008$ , an integral (including  $1/v$  capture) of  $9.7 \pm 0.8$  barns is obtained. From the measured buckling ( $10.89 \pm 0.05 \text{ m}^{-2}$ ) and thermal utilization ( $0.9445 \pm 0.0009$ ) a value of  $11 \pm 2$  barns is obtained for the integral. Although in agreement with one another, these values are not in general agreement with either calculated values or the results inferred from other measurements.



## I. INTRODUCTION

Because thorium alloyed with uranium has promising properties for high temperature use and for breeding, it is being considered as a feasible reactor fuel. Experimental information on the nuclear properties of lattices containing such fuels is practically nonexistent. The neutron absorption properties particularly in the resonance energy region are a source of uncertainty in reactor design calculations. There is, therefore, an urgent need for obtaining such information through the performance of exponential experiments. An opportunity to do some initial work was provided when the decision was made to prepare a thorium-uranium alloy fuel loading for the Sodium Reactor Experiment (SRE). Prior to loading the fuel into the SRE, it was made available for a short time for use in exponential experiments.

Although time permitted the investigation of only one lattice configuration, the results have been of value in that the resonance absorption properties have been studied by two independent methods. In one method, the resonance escape probability is measured directly. In the other, it is determined from the measured lattice buckling and neutron flux profile in a lattice cell, together with the age-diffusion critical equation. These methods lead to results that are in agreement.

In addition to the resonance properties of the thorium, the material buckling and thermal utilization of the lattice were measured. Some measurements were also made of the neutron flux surrounding a single fuel element placed in the center of the block of solid graphite moderator. The application of small source theory<sup>1</sup> to the results of these latter measurements permits the determination of the parameters of a lattice of the elements at any spacing. Although this information serves to extend the experimental results to other lattice configurations, it is not yet considered to have the same degree of certainty as the information from the exponential experiment itself. Under a separate project a thorough quantitative study is in progress aimed at evaluating the applicability and accuracy of the single element method.



## II. DIFFUSION LENGTH IN GRAPHITE

Although a diffusion length measurement for the AGOT graphite used in this experiment had been made previously,<sup>2</sup> it has subsequently been moved several times during a long storage period. It was, therefore, necessary to check the purity of the graphite again. In addition, with the aid of the scintillation counting equipment now available, a measurement of greater accuracy was attainable.

The graphite block was assembled on the AE-6 water boiler reactor thermal column which served as a source of neutrons for the measurements. The thermal neutron distribution was measured along the vertical ( $z$  direction) and horizontal ( $x$  and  $y$  directions) axes of the block. The distribution along the vertical axis was fitted<sup>3</sup> to a function of the type  $A_z = A \sinh \nu(z - h)$ . The fit was best for a value of  $\nu = 0.04083 \pm 0.00008 \text{ cm}^{-1}$ . The horizontal distribution was measured at a height where higher harmonics were not influencing the results. This distribution was fitted by a least squares procedure to a function of the form  $B_x = B \cos \mu_o(x - c)$ . The value obtained for  $\mu_o$  is  $0.02512 \pm 0.00003 \text{ cm}^{-1}$ . A calculated correction has to be applied to  $\mu_o$ . It arises from the perturbation of the thermal neutron distribution by the vertical foil holder and is described in detail elsewhere.<sup>4</sup>\* The resulting value for the thermal neutron diffusion length for this graphite is  $49.9 \pm 0.2 \text{ cm}$  which is in agreement with the previously measured<sup>2</sup> value,  $50.8 \pm 0.8 \text{ cm}$ .

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\* The calculation in this reference was done for a system with cylindrical symmetry. However, it may be shown that this is accurate enough for this system.



### III. THE MATERIAL BUCKLING OF THE LATTICE

A diagram of the lattice in which the measurements were made is shown in Figure 1. It consisted of a 5-cell by 5-cell square array of elements in a graphite moderator at a 9.5 in. lattice spacing. Each element consisted of seven cylindrical rods, each 3/4 in. in diameter. A fuel element is shown in cross section in Figure 2. Each of the rods was made from 10 slugs, 6 in. long, of the thorium-uranium alloy fuel stacked one on top of the other. Uranium enriched to 93.13% by weight makes up 7.6% of the fuel weight. They were held in place by cylindrical segments of 2S aluminum, each segment being 6 in. long. The aluminum was used to mock-up the sodium coolant of the SRE.

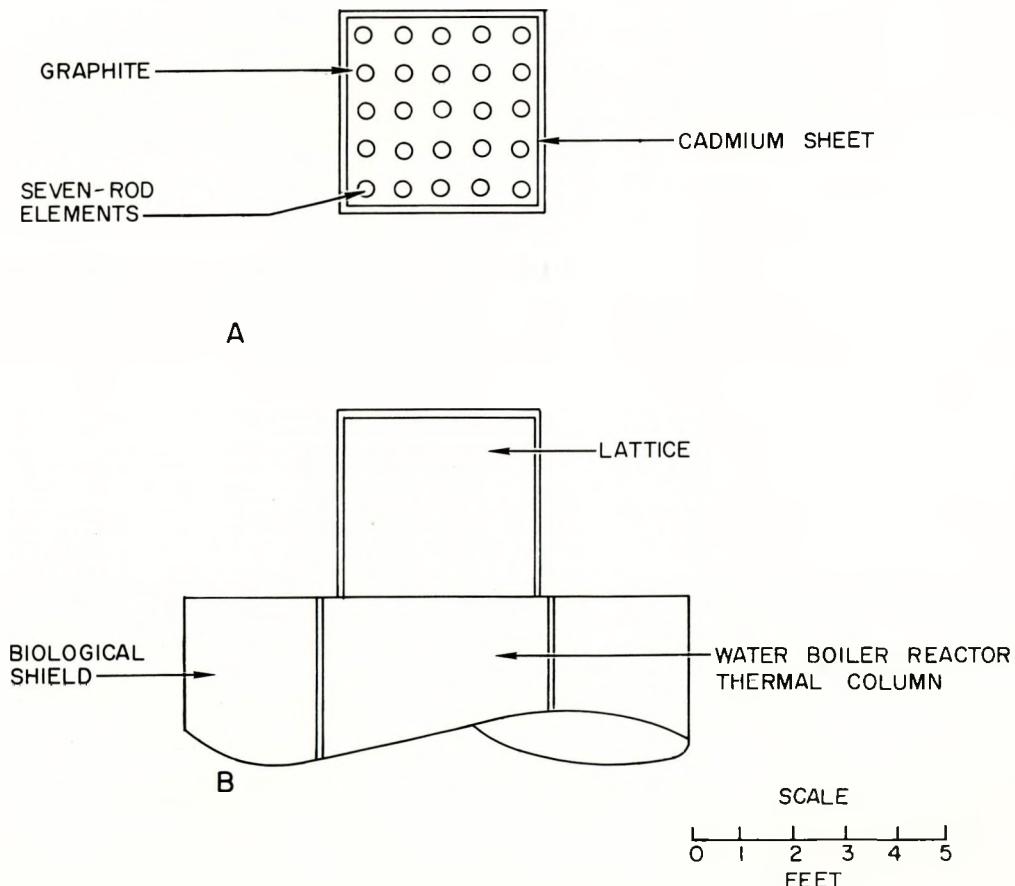


Figure 1. The Lattice and its Location on the Reactor Thermal Column

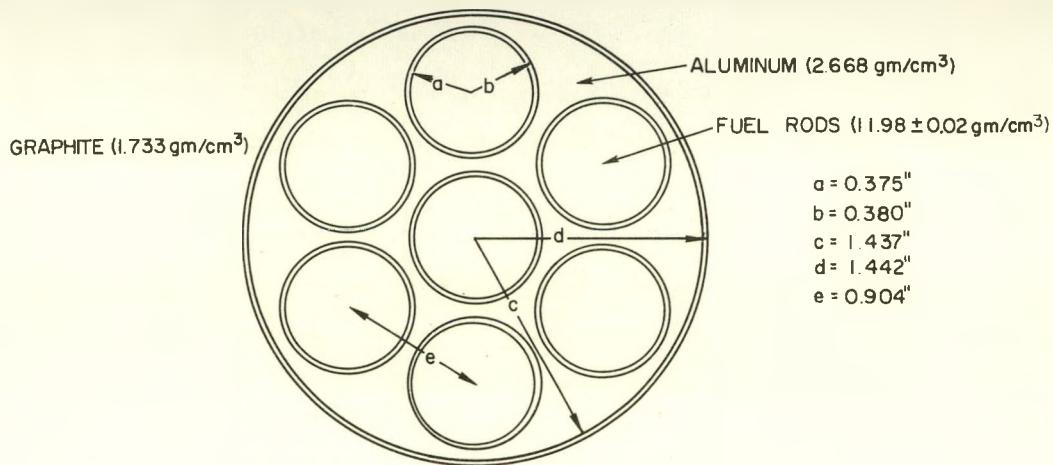


Figure 2. Cross Sectional Diagram of Fuel Element

The material buckling of a lattice according to diffusion theory is given by

$$B_m^2 = 2\mu_o^2 - \nu^2,$$

and applies accurately<sup>5</sup> to the region of a sub-critical lattice far enough away from the source that the neutrons present arise from fissions in the fuel and at least a distance greater than a slowing-down length from the boundaries of the lattice. The quantity  $\nu$  is obtained from a fit<sup>3</sup> of the measured vertical distribution of the thermal neutrons to a function of the form.

$$A_z = A \sinh \nu(z - h).$$

The thermal neutron distribution in this vertical direction was obtained by taking the difference between irradiated sets of bare and cadmium-covered indium foils (75% In, 15% Pb, and 10% Al) placed 2 in. apart in a graphite foil holder. The foil holder was placed in the lattice so that the foils were situated along the corner line of the central cell. The value obtained for  $\nu$  is  $0.01331 \pm 0.00016 \text{ cm}^{-1}$ . In principle, the horizontal buckling could be obtained by measuring the thermal neutron distributions in horizontal directions and fitting them to a function of



the form

$$B_x = B \cos \mu_o (x - c).$$

For the present graphite lattice, this measurement is not practical because of the uncertainty arising from having to separate the detailed intracell flux from the gross cosine variation. Instead, the extrapolated width, and hence the value of  $\mu_o$ , was taken to be the same as that measured for the solid block of graphite in the previous section. After correcting<sup>4</sup> for the perturbation due to the foils in the vertical holder, a value for the material buckling was found to be

$$B_m^2 = 10.89 \pm 0.05 \text{ m}^{-2}.$$

This value of the material buckling may be compared with the value  $9.7 \pm 0.2 \text{ m}^{-2}$  found<sup>2</sup> for a similar lattice loaded with the uranium metal 2.778 at.% enriched fuel. This comparison indicates that, of the two, the Th-U alloy fuel is the more reactive.

Thermal and epithermal neutron flux distributions have been measured about a single element, like those used in the exponential experiment, in a block of

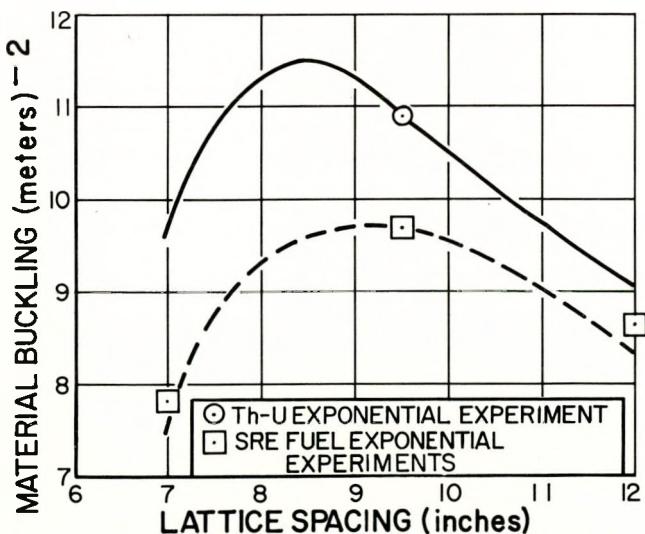


Figure 3. Material Bucklings of Lattices Containing Thorium-Uranium and Uranium Fuels (The curves are determined by the application of small source theory to flux measurements about a single element.)

graphite moderator. These data have been tentatively analyzed by means of small source theory<sup>6</sup> in order to find the material buckling of a regular square lattice of these elements in graphite as a function of the lattice spacing. The results are shown by the solid curve in Figure 3. The experimental point shown on the curve is that determined from the exponential lattice measurements described above. Therefore, within the certainty of the small source theory then, one may regard this solid curve as a measure of the material buckling of a square lattice of these elements, as a function of the lattice spacing when



spaced uniformly in the graphite moderator. To see how the material buckling measured in this manner compared with that obtained by exponential experiments, consider the dashed line curve in this same figure. This curve represents the tentative results for the material buckling deduced from the measurement of the thermal neutron distribution about a single 7-rod uranium metal (2.778 at. % enriched) element in the same moderator. The experimental points indicated by the squares are those obtained from measurements on sub-critical lattices of the same elements in the same moderator. This indicates that the most probable values obtained by these independent methods, may be said to agree to within an amount having an order of magnitude of about 5%.



#### IV. THERMAL UTILIZATION

We consider the thermal utilization to be one of the four factors of the infinite multiplication constant. It is defined as the ratio of the absorption of thermal neutrons in the fuel of a cell, of an infinite array of the elements with the same pitch as the lattice, to the thermal neutron absorption everywhere in such a cell. Algebraically this is given by the equation

$$f = \left( V_f \bar{\phi}_f \Sigma_f^a \right) \cdot \left( V_f \bar{\phi}_f \Sigma_f^a + V_g \bar{\phi}_g \Sigma_g^a + V_{Al} \bar{\phi}_{Al} \Sigma_{Al}^a \right)^{-1}.$$

In this equation, the subscripts  $f$ ,  $g$ , and  $Al$  refer to the fuel, graphite, and aluminum components of the cell respectively, where the small voids about each rod and the element have been included with the aluminum. The  $V$ 's are the volume fractions of these components. The  $\bar{\phi}$ 's and  $\Sigma^a$ 's are the corresponding values of the average thermal neutron fluxes and the macroscopic thermal neutron absorption cross sections. Calculation of the volume fractions is straightforward. The thermal neutron macroscopic absorption cross sections are Maxwell-Boltzmann averaged.

The determination of the average thermal neutron flux will now be discussed. To obtain the thermal neutron distribution of a unit cell, flux traverses were made in the various regions of the central unit cell. The traverses were made at a distance of 18-1/4 in. above the bottom of the assembly. At this vertical level the cadmium ratio had already become independent of the height above the bottom. Dysprosium foils having dimensions of 10 mm by 2 mm by 0.172 mm were used as detectors for the flux measurements. Both bare and cadmium-covered sets of foils were irradiated in the unit cell shown in cross section in Figure 4.

First, sets of bare and sets of cadmium-covered foils were irradiated along the diagonal of the cell so that their 10 mm by 2 mm faces were parallel to the vertical axis of the fuel element and parallel to the cell diagonal. The cadmium-covered activities were adjusted to account for the attenuation of the epithermal neutrons by the 0.020 in. cadmium boxes in which they were placed. These "cadmium covered" activities were then subtracted from the corresponding bare

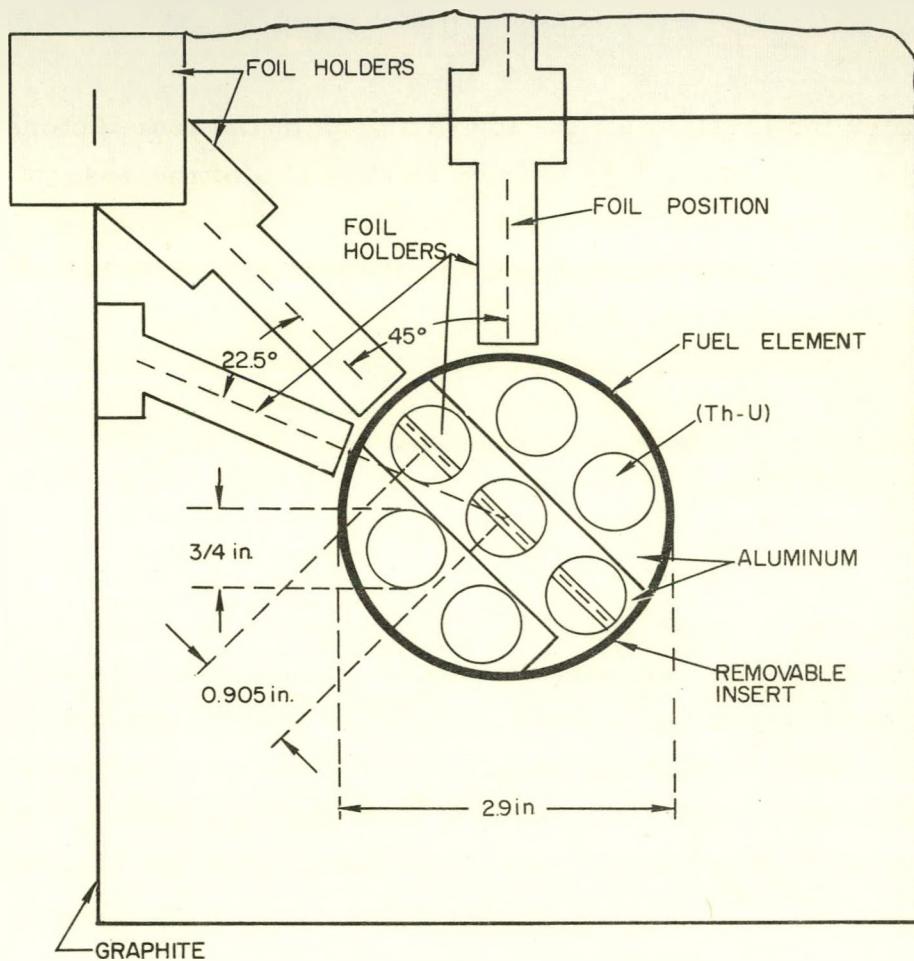


Figure 4. Unit Cell Diagram Showing Position of Foils

foil activities. The resulting activities were then divided by the factor  $\cos \mu_o x \cdot \cos \mu_o y$ , where  $\mu_o = 0.02512 \text{ cm}^{-1}$  and  $x$  and  $y$  are the rectangular cartesian coordinates of the foil centers as measured from the center of the center rod of the element along lines which are at right angles to adjacent faces of the cell. A plot of the thermal activities obtained vs the distance from the center of the cell is shown in Figure 5.

It appeared that the thermal flux in the center rod is symmetrical about the vertical axis of the rod. To check this in detail, flux traverses were made along several diameters of this rod and found to agree to within the experimental uncertainty of the measurements. To obtain the average thermal neutron flux in the central rod, these activities were weighted as a function of their radial distance from the axis of the rod. The results were then graphically integrated to find the average thermal neutron activity in the central rod  $\bar{\phi}_{cr}$ .

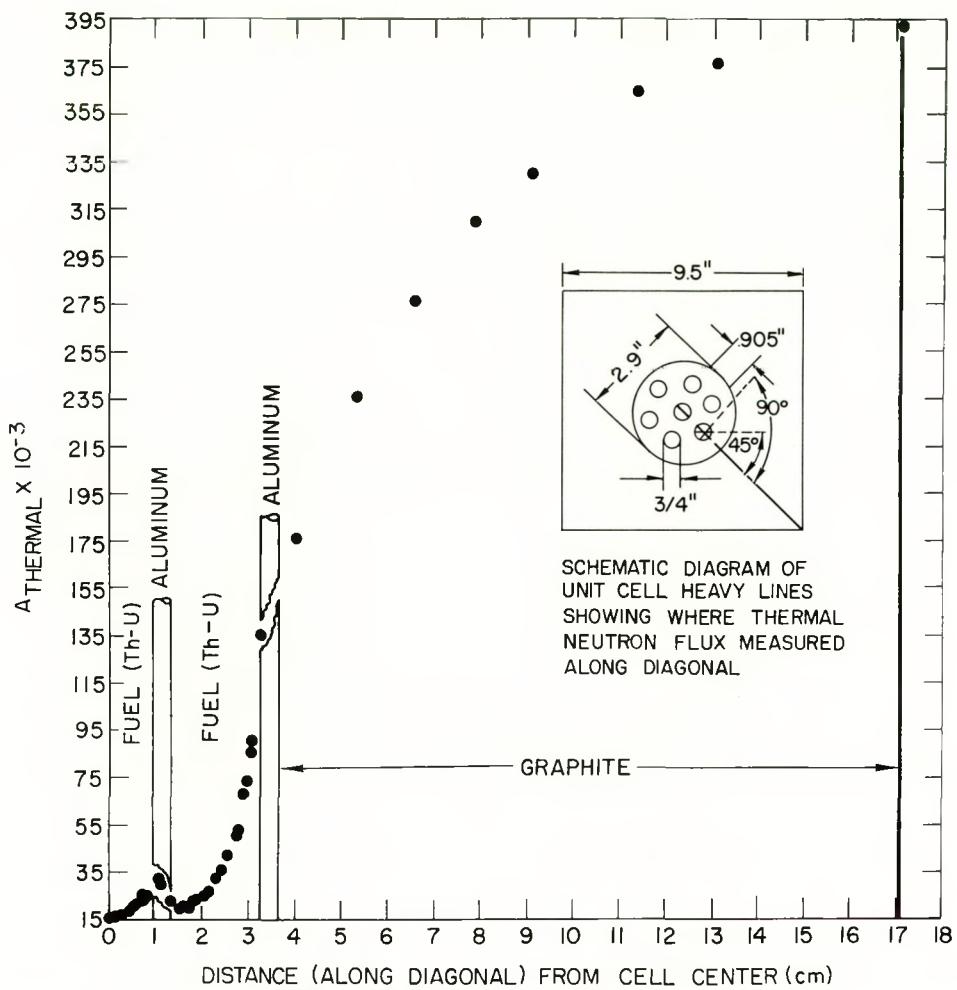


Figure 5. Thermal Neutron Flux Along Diagonal of Unit Cell

Clearly, the thermal neutron flux was asymmetric about the vertical axis of an external rod. This made it necessary to make flux traverses along at least the three directions indicated by the solid lines in the outside rod in Figure 5. The thermal neutron activities obtained for these three directions are shown plotted as a function of the radial distance from the axis of the rod in Figure 6. For a given value of this radial distance, these activities were graphically integrated over the angle. This gave the angle averaged activity as a function of this radial distance. These activities were graphically integrated to obtain the average thermal neutron flux in the outer rod,  $\bar{\phi}_{or}^0$ . The average thermal neutron flux for the fuel is then obtained from the relation

$$\bar{\phi}_f = \frac{\bar{\phi}_{cr} + 6\bar{\phi}_{or}}{7}.$$

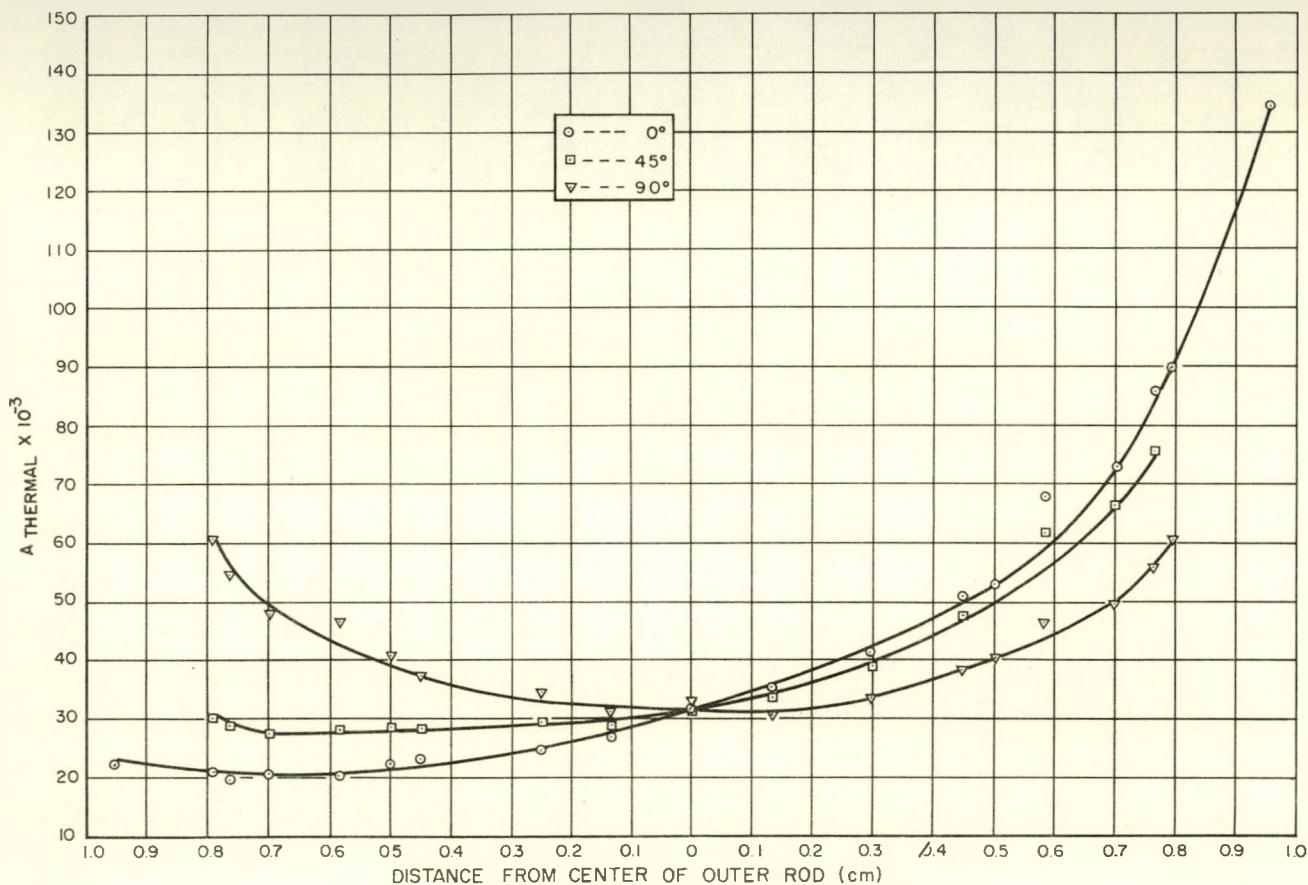


Figure 6. Thermal Neutron Flux Along Diametral Traverses Through the Outer Fuel Rod (The traverses make angles of  $0^\circ$ ,  $45^\circ$ , and  $90^\circ$  with the cell diagonal.)

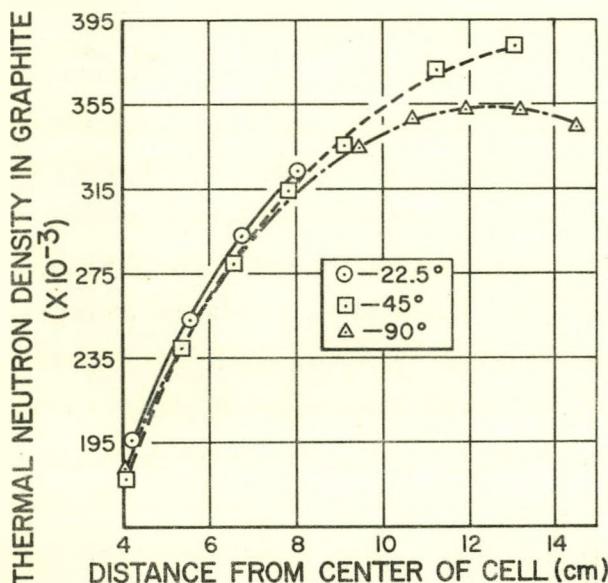


Figure 7. Thermal Neutron Flux in Graphite Along Traverses Through Cell Center

The thermal neutron flux shown for the graphite in Figure 5 was determined from the flux traverses made along the cell diagonal. The analysis of the data for all the graphite was made compatible with that of the cylindrized model<sup>7</sup> for the cell. As far as the graphite is concerned, the integration is carried out to a radial distance  $R$  from the center of the cell where  $R$  has the value which makes  $\pi R^2$  equal to the cross-sectional area of the square cell. This simplification introduces a negligibly small error in the volume of graphite



that one must consider. Generally, in this model one tacitly assumes that the thermal neutron flux in the graphite has cylindrical symmetry about the vertical axis of the cell. To check the validity of this assumption, flux traverses were taken along the three directions indicated in Figure 4. The results for the thermal neutron flux as a function of the distance from the element center are shown in Figure 7. One might expect in going toward the corner of the cell along the diagonal, that the thermal neutron flux would increase somewhat more rapidly than when making the traverse perpendicular to the face of the cell, since there is more moderator between two elements along the diagonal than along the perpendicular line. Even along the  $22-1/2^\circ$  direction, one might expect that the thermal neutron density would increase more rapidly than the other two directions when considering the average distance from points along this line to the adjacent elements. One can see that a small error would be induced in  $V_g \bar{\phi}_g \Sigma_g^a$  if cylindrical symmetry in the thermal neutron flux was assumed, and a single traverse made and then averaged over this traverse only. The situation is improved by averaging the thermal flux in the graphite over the angle for a given value of the radial distance from the center of the element and then graphically integrating these results. There may yet be a small error in  $\bar{\phi}_g$  due to the fact that the thermal neutron flux was not mapped in more detail in this region. This error would add to the error in  $V_g \bar{\phi}_g \Sigma_g^a$  that has been contributed by the error in the volume. However, the net effect on the thermal utilization of the cell should be negligibly small since the term  $V_g \bar{\phi}_g \Sigma_g^a$  is small to begin with in comparison to  $V_f \bar{\phi}_f \Sigma_f^a$ .

The data in Figure 5 give some idea of how the thermal neutron flux varies in the aluminum. The flux in the aluminum outside of a circle centered at the element center and tangent to the outer edges of the outer rod does not vary by a large amount. For integration purposes, it was, therefore, assumed that the flux varied linearly with radial distance in this region, ranging from the measured "angle" averaged value at the surface of the outer rod to the average value obtained from extrapolating the measured values in the graphite to the inner edge of the graphite. The variation of the flux in the aluminum in the "small cylinder" between the inner edges of the outer rods and the center rod is more complex. Some foils were therefore placed in this region in order to map the thermal flux distribution. The thermal neutron flux in the region between the outer rods was



assumed to vary only with radial distance from the cell center. At a given radial distance, it was assumed to be equal to the measured average value where it touches the surface of an outer fuel rod. Under these assumptions the flux in the aluminum was graphically integrated.

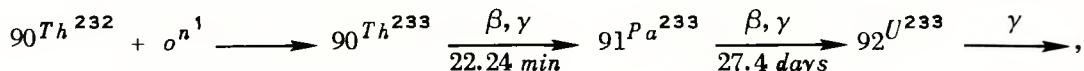
From the values of the average fluxes given in Table I and the known cross sections of the materials, the value of the thermal utilization is found to be  $0.9445 \pm 0.0009$ . This can be compared<sup>2</sup> to a value of  $0.931 \pm 0.008$  for the same lattice with the 2.778 at. % enriched uranium metal fuel present.



## V. RESONANCE CAPTURE FOR THE TH-U ALLOY FUEL

### A. METHOD AND PROCEDURE

The probability,  $p$  that neutrons escape capture in thorium while slowing down to the cadmium cut-off energies was measured directly. The details of the method and procedure closely parallels that discussed elsewhere.<sup>8</sup> The activities of both bare and cadmium-covered Th<sup>232</sup> foils (10 by 2 by 0.127 mm), after exposure in the fuel at a level 18-1/4 in. from the bottom of the lattice, were measured. The measured activities arise from the Th<sup>232</sup> ( $\eta, \gamma$ ) reaction which proceeds as follows:



Pa<sup>233</sup> has an internal conversion X-ray of energy 89.5 kev. The scintillation counting equipment was calibrated, by means of the 89 kev X-ray from a standard Cd<sup>109</sup> source, to detect this radiation. The activities of the bare and cadmium-covered thorium foils in the center rod are shown in Figure 8. The activities of the bare and cadmium-covered foils are plotted as a function of the radial distance of the foil center from the axis of the outer rod in which they were placed in Figures 9 and 10 respectively.

The activities of the bare and cadmium-covered thorium foils were averaged

over the cross-sectional area of the fuel to obtain the values,  $C$  and  $B$ , respectively. The ratio of the activities

$$r = \frac{C}{B - C}$$

is related to the probability,  $p$ , by the equation

$$r = \frac{1 - p}{pfF},$$

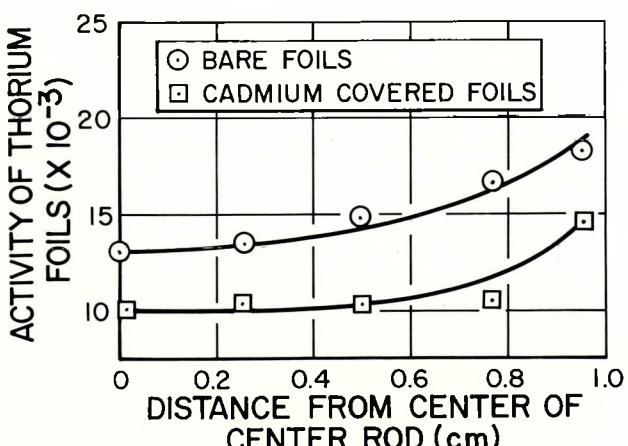


Figure 8. Bare and Cadmium-Covered Thorium Foil Activities in the Center Fuel Rod

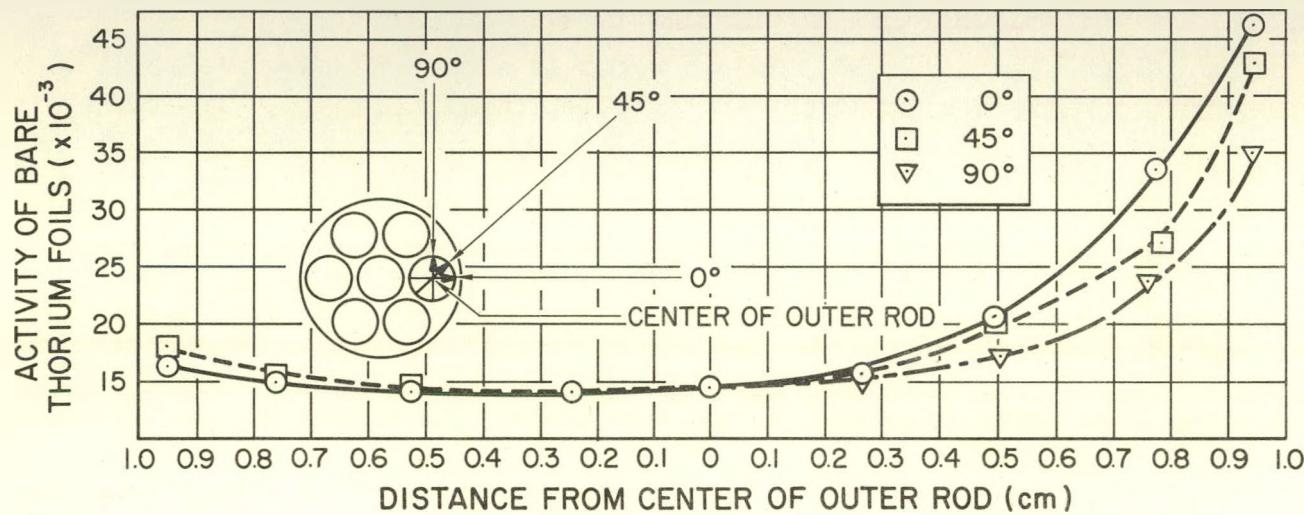


Figure 9. Bare Thorium Foil Activities in the Outer Fuel Rod

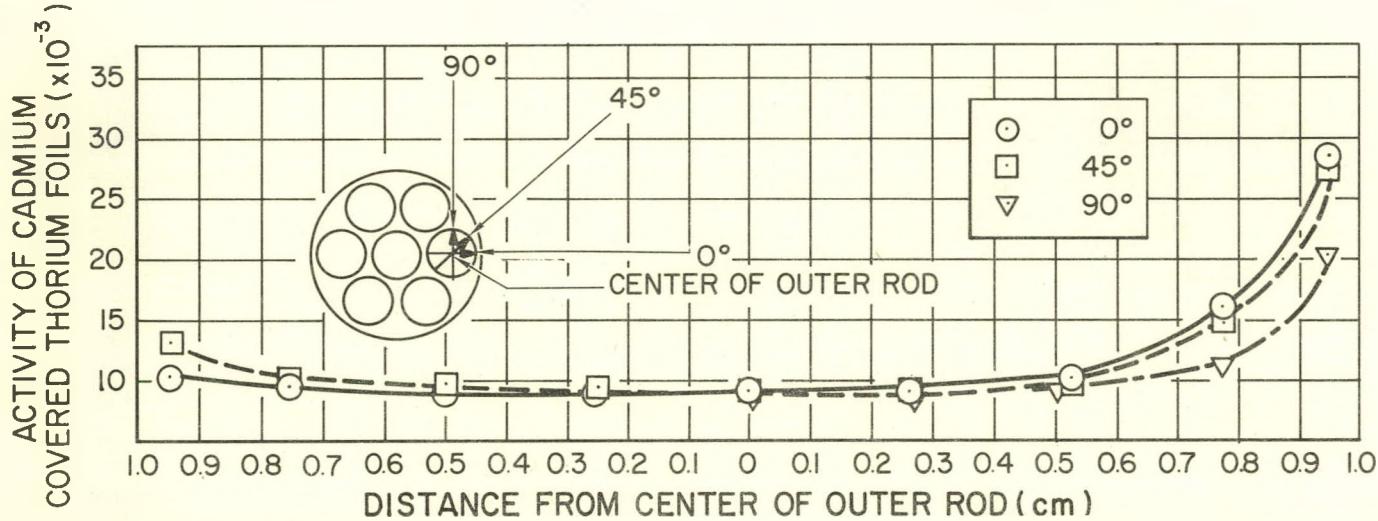


Figure 10. Cadmium-Covered Thorium Foil Activities in the Outer Fuel Rod

where  $f$  is the measured thermal utilization of the lattice,  $0.9445 \pm 0.0009$ , and  $F$  is the fraction of the thermal neutrons absorbed by the fuel that are absorbed by  $\text{Th}^{232}$ . A value of 0.126 for  $F$  is obtained from the known cross sections of the constituents of the fuel material. The measured value of,  $r$  is  $1.413 \pm 0.025$ . With this information the value for the resonance escape probability of  $0.856 \pm 0.008$  is obtained. By the method explained elsewhere,<sup>8</sup> one may use the equation

$$p = \exp(-0.0161 I_r)$$

to find the effective resonance integral (including  $1/v$  capture) for the thorium as



it exists in the fuel element. This value is  $9.7 \pm 0.8$  barns. To find the effective resonance integral for thorium as it stands, in the fuel corrected for one over  $\nu$  capture in the fuel, one must subtract 3.5 barns from this value. The value of  $\sqrt{s/m}$ , where  $s$  is the "rubber band" surface of the thorium, and  $m$  is the mass of thorium per unit of fuel element length, is  $0.293 \text{ cm gm}^{-1/2}$  for this fuel element.

#### B. COMPARISONS WITH OTHER EXPERIMENTS AND CALCULATIONS

Direct measurements of the effective resonance integral as a function of the surface to mass ratio have been made at other laboratories.<sup>9, 10</sup> These measurements did not extend to as low values of surface to mass as used in the present measurement. However, some comparative values have been obtained by a slight extrapolation. Our results are compared with those extrapolated values in the following table.

TABLE I

SUMMARY OF RESONANCE CAPTURE INFORMATION FOR THORIUM

$I_r$ (with $1/\nu$ ) barns	Source of information.
$9.7 \pm 0.8$	Direct measure using $\text{Th}^{232}$ foils.
20.9	Single element measurements.
10	Dayton and Pettus. Extrapolation of data from direct measurements.
12.3	Sher. Extrapolation of data from direct measurements.
13.2	M. V. Davis. Extrapolation from direct measurement.
13.6	R. Vernon. Calculation.
11 $\pm$ 2	Using $I = \frac{\eta \rho f e^{-B_m^2 \tau}}{1 + L^2 B_m^2}$ where $B_m^2$ and $f$ were measured experimentally, and the other quantities calculated.
13.5	L. Nordheim, private communication.



One may also use a semi-empirical approach to obtain indirectly a measured value of  $p$ . One may use the age-diffusion theory critical equation<sup>5</sup>

$$1 = \frac{\eta \epsilon p f_e \frac{-B_m^2 \tau}{m}}{1 + L^2 B_m^2} .$$

In this equation  $f$  and  $B_m^2$  are assigned their measured values for this lattice. The parameters,  $\eta$ ,  $\epsilon$ ,  $\tau$ , and  $L^2$  were assigned the values 1.823, 1.0166, 308.6  $\text{cm}^2$ , and  $97.5 \text{ cm}^2$  which were calculated using a semi-empirical procedure.<sup>2</sup> This gives a value of  $0.884 \pm 0.010$  for the resonance escape probability for the lattice. It must be remembered that one must regard the values of  $p$ , obtained using this equation, as being automatically corrected for  $1/v$  capture. These then would be compared with the activation measurement of 0.906 using the  $\text{Th}^{232}$  foils. The value of the effective resonance integral deduced by this analysis with one over  $v$  capture added is shown in the table.

Analyses of the epithermal flux about a single 7-rod Th-U element in a block of graphite moderator also enables one to find the resonance escape probability<sup>6</sup> for the lattice used in the exponential experiment. The equation  $p = \exp(-0.01611_f)$  is used to find the effective resonance integral for the fuel. This tentative result is also included in the table.

Values of the effective resonance integral for the thorium having the same  $s/m$  value as that of the thorium in the element have been calculated.<sup>11, 12</sup> The results of these calculations are shown in the table.

The direct measurement of the effective resonance integral for thorium is in rather good agreement with that predicted from the extrapolation of the results of Dayton and Pettus to small  $s/m$  values. The results of Dayton and Pettus, according to Dresner,<sup>13</sup> could have a large systematic error present for two reasons. First, the mass absorption contributes a negative amount to the thorium effective resonance integral that has been corrected for  $1/v$  capture. Secondly, Dresner's calculations are such that he should be able to predict the value of the effective resonance integrals for the oxide of thorium if he is given the correct value of the thorium metal effective resonance integral. Dresner is unable to make his calculated results agree with the oxide measurements of Dayton and



Pettus. This may indicate that only the oxide data contain a systematic error, and agreement of our directly-measured value with the extrapolation of the Dayton and Pettus metal measurements could be expected if any interference of the thorium resonance capture with that of  $U^{238}$  and  $U^{235}$  can be neglected.

It appears that the effective resonance integrals obtained using the critical equation are more nearly a property of the element than a property of the thorium in the element. If this is the case, then one might expect that the effective resonance integral, using these equations, may be higher than that measured for the thorium present using the thorium foils. It has been estimated<sup>11</sup> that the small amount of  $U^{238}$  present, assuming no interference between the  $U^{238}$ ,  $U^{235}$ , and the  $Th^{232}$  resonances, could contribute 0.8 of a barn to the effective resonance integral of the element. The  $U^{235}$  present may also contribute to the resonance absorption of the element.

One should be careful in concluding that the agreement between the results using the critical equation with those obtained upon the extrapolation of experimental data is anything but fortuitous since it has been pointed out<sup>2</sup> that the results of these two are generally in disagreement with one another.

Even the agreement of the results from the critical equation with the calculations should be viewed with some skepticism since admittedly some of the thorium resonance parameter data used in the computations leave much to be desired at the present time. This skepticism becomes even stronger when one recalls that some simplifying assumptions<sup>14</sup> have been made in deriving the expression for the effective resonance integrals.

The results listed in the table indicate that considerable uncertainty exists regarding the value that one should use for the effective resonance integral of the element. This uncertainty is even more striking when one considers the corresponding values of the effective resonance integrals (subtract about 3.4 barns from those listed in the table) corrected for the  $1/v$  capture of the thorium. These  $1/v$  corrected effective resonance integrals differ in some cases by almost a factor or two. A similar observation was made regarding measurement<sup>15</sup> of the effective resonance integral for thoria-urania systems.

Thus, there is a need for a more precise measurement of the effective resonance integrals for thorium and fuels containing thorium. It also should



serve to point up the fact that the expressions used in the interpretation of the direct and semi-empirical results, even though adequate for systems using fuels of very low enrichment, may be wholly inadequate when applied to systems using moderately enriched fuels. This inadequacy could even be magnified in the thorium-uranium systems where it appears that the  $\text{Th}^{232}$  and  $\text{U}^{235}$  resonances could undergo considerable interaction. If it is a good approximation to neglect both resonance fission and  $1/v$  capture above cadmium cut-off with the SRE fuel as it appeared to be,<sup>2</sup> then it is difficult to understand how it could also be a good approximation for this Th-U alloy fuel where there is only about 1.5 times more  $\text{U}^{235}$  present, and the  $1/v$  portion of the effective resonance integral for  $\text{Th}^{232}$  is about three times as large as that for the  $\text{U}^{238}$  in the SRE fuel. It appears that reactors with fuels enriched to a few percent or greater will require calculations utilizing a greater number of energy groups to be able to accurately describe them theoretically.



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