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MEASUREMENT OF NEUTRONIC PARAMETERS
FOR A PLUTONIUM FUELED HTGR LATTICE

MAY 1972

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FOR A PLUTONIUM FUELED HTGR LATTICE

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ABSTRACT

Neutronic parameters have been measured in the High Temperature Lattice Test Reactor (HTLTR) for a graphite moderated ThO_2 - PuO_2 fueled lattice to serve as a benchmark for evaluating computational methods and cross sections used in the design of a plutonium-fueled High Temperature Gas-Cooled Reactor (HTGR). Measurements made as a function of temperature from 20°C to 1000°C included k_∞ , the reactivity perturbations of a number of different fuel and poison samples, and the thorium Doppler coefficient. The calculated and measured changes of k_∞ with temperature are in agreement. An experiment-theory correlation provides a basis for assessing the uncertainties in neutronics calculations.

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MEASUREMENT OF NEUTRONIC PARAMETERS FOR A PLUTONIUM FUELED HTGR LATTICE

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I. INTRODUCTION

The High Temperature Gas-Cooled Reactor (HTGR) concept utilizes thorium for the fertile fuel material and highly enriched uranium for the initial fissile fuel. The ^{233}U bred from thorium is recycled, but some make-up fissile fuel must be added since the conversion ratio is less than unity. A joint Edison Electric Institute--Gulf General Atomic study of plutonium utilization in the HTGR⁽¹⁾ indicated that the fabrication penalty for plutonium as a substitute for ^{235}U make-up would be relatively smaller for the HTGR than for Light Water Reactors (LWRs) since the HTGR fuel fabrication already requires the ability to handle ^{233}U with its radioactivity from ^{232}U decay and toxicity. The probable future availability of plutonium at an attractive price has made it desirable to consider the use of plutonium in the HTGR fuel cycle. An Oak Ridge National Laboratory evaluation of the use of plutonium in the HTGR⁽²⁾ concludes: (a) plutonium make-up in the HTGR is economically preferred over the plutonium-fueled LWR during the period 1970 to 2015, and (b) as the price of plutonium decreases, it will eventually be necessary for the HTGR to operate with plutonium as the initial fissile fuel if it is to compete with the LWR fueled with depleted uranium and plutonium enrichment.

Sophisticated analytical methods, which handle both the complexities of heterogeneity and the effects of low energy resonances in plutonium, must be used to predict the neutronic characteristics of plutonium fuel in the HTGR. Coated PuO_2 particles are mixed with coated ThO_2 and graphite particles to form fuel rods which are components of a heterogeneous lattice array in graphite. Self-shielding factors for both the particles and the

fuel rods as a function of temperature are required in the neutronic analysis. Since the temperature of the moderator in the HTGR at operating conditions is about 1050°K, neutrons can scatter up in energy into the 1.06 eV capture resonance of ^{240}Pu as well as slow down into it. By comparison, the temperature of the moderator in LWRs is only about 570°K. The neutron absorption rate in plutonium also varies with temperature due to Doppler broadening of the resonance cross sections, and due to hardening of the thermal neutron spectrum. These basic complexities lead to larger uncertainties in the neutronic characteristics predicted by calculational methods.

II. SUMMARY

Neutronic parameters for a plutonium-enriched HTGR lattice were measured at the Battelle Pacific Northwest Laboratories (BNWL) in the High Temperature Lattice Test Reactor (HTLTR) to serve as a benchmark for evaluating calculational methods. Measurements of the infinite medium neutron multiplication factor, k_{∞} , were made as a function of temperature from 20° to 1000°C. The thorium Doppler coefficient and reactivity perturbations of a number of fuel and poison samples were also measured as a function of temperature.

Experimental values for k_{∞} were measured at five equilibrium temperatures: 20°, 300°, 500°, 750°, and 1000°C. Values for k_{∞} of this $\text{ThO}_2\text{-PuO}_2$ fueled HTGR lattice were also calculated as a function of temperature using computer codes GRANIT for the thermal region, and EGGNIT for the epithermal region. A comparison of these theoretical and experimental values for k_{∞} of the $\text{ThO}_2\text{-PuO}_2$ HTGR lattice shows that the calculated values are about 0.8% higher than the measured values. This difference is larger than the 0.3% uncertainty in the experimental values. A potential source of error in the calculations is the thorium resonance capture rate. The calculated epithermal-to-thermal capture rate in thorium is about 2% lower than the measured value. It is estimated that a 2% increase in the calculated epithermal capture rate in thorium would reduce the calculated values for k_{∞} by about 0.5%. Another potential source of error in the calculations is the uncertainty in the exact loading of the fuel blocks. A 1% uncertainty in the amount of plutonium loaded in the fuel leads to an uncertainty in k_{∞} of about 0.2%. A much smaller uncertainty exists for the amount of thorium in the fuel. The calculated temperature-dependent changes in k_{∞} are not affected by these small normalizations of reaction rates, and the calculated changes of k_{∞} with temperature are in agreement with the measured data for the $\text{ThO}_2\text{-PuO}_2$ HTGR lattice.

When plutonium is used as the initial fissile fuel, the temperature coefficient of reactivity is changed favorably from that of an HTGR lattice fueled initially with ^{235}U . The reactivity decrease associated with elevating plutonium-thorium fuel to operating temperature is about 30% less than that obtained with ^{235}U -thorium fuel, so the excess reactivity for which the control system must compensate is less with plutonium fueling.

Above the operating temperature range, plutonium tends to make the temperature coefficient of the HTGR more negative due to increased neutron capture in the 1.06 eV resonance of ^{240}Pu . This effect will improve the self-limiting characteristics of the reactor during a power excursion.

III. PRINCIPLE OF THE MEASUREMENT

The primary neutronic characteristic of a test lattice that is measured in the High Temperature Lattice Test Reactor (HTLTR), shown in Figure 1, is the infinite medium neutron multiplication factor, k_∞ , as a function of temperature. The excess neutron production in the test lattice, $k_\infty - 1$, is determined from the ratio of reactivity perturbations of a test lattice sample and a normalizing neutron absorber, using the unpoisoned technique.⁽³⁾ A small sample of the test lattice (referred to as the central cell) is inserted to fill the void in the center of the HTLTR. Surrounding this void is a region of test lattice identical to the central cell, and the neutron spectrum in this region is adjusted to be as close as possible to the fundamental mode condition.⁽⁴⁾ The reactivity perturbation caused by insertion of the central cell is normalized to the perturbation produced when a sample of copper is placed in the central void of the HTLTR. Neutron absorption rates in the central cell constituents and the copper are measured by foil activation methods.⁽⁵⁾

Using first order perturbation theory and the two-energy-group approximation, the relationship between k_∞ and the measured quantities can be derived:⁽⁶⁾

$$\begin{aligned}
 k_\infty = 1 - & \left(\frac{\Delta \rho_{\text{cell}}}{\Delta \rho_{\text{Cu}}} \right) \frac{(\Sigma_a \phi V)_2^{\text{Cu}}}{(\Sigma_a \phi V)_2^{\text{cell}}} \left[1 + \left(\frac{\phi_1^+}{\phi_2^+} \right) \frac{(\Sigma_a \phi)_1^{\text{Cu}}}{(\Sigma_a \phi)_2^{\text{Cu}}} \right] \\
 & - \frac{(1 + \tau B^2)(1 + L^2 B^2)(1 - p)L^2 B^2}{1 + (1 - p)L^2 B^2} + \frac{\tau B^2(1 + L^2 B^2) [\tau B^2 - \eta_1 f_1(1 - p)]}{1 + \tau B^2 - \eta_1 f_1(1 - p)} \\
 & - \frac{\tau B^2(1 + L^2 B^2)}{1 + \tau B^2 - \eta_1 f_1(1 - p)} \left(\frac{\Delta \phi}{\phi} + \frac{\Delta \phi^+}{\phi^+} \right) \\
 & + \frac{(1 + L^2 B^2)[1 - \eta_1 f_1(1 - p)]}{1 + \tau B^2 - \eta_1 f_1(1 - p)} \left(\frac{\Delta \phi}{\phi} - \frac{\Delta \phi^+}{\phi^+} \right). \quad (1)
 \end{aligned}$$

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FIGURE 1. High Temperature Lattice Test Reactor and Supporting Facilities

The first term in this expression is the preponderant term and is evaluated experimentally. The next two terms are small calculated corrections which account for fundamental mode neutron leakage from the central cell. The last two terms in Equation (1) are calculated two-group corrections for neutron spectrum mismatch from fundamental mode values, where ϕ is defined as ϕ_1/ϕ_2 and $\Delta\phi$ is the difference between ϕ for the fundamental mode and the ϕ actually present with the central cell inserted; $\phi^\dagger$ is defined similarly for the adjoint fluxes except $\Delta\phi^\dagger$ is evaluated at the surface of the void. The remaining calculated parameters are defined as follows:

$$\tau = \frac{D_1}{\Sigma_1^a + \Sigma_{1 \rightarrow 2}} \quad (2)$$

$$L^2 = \frac{D_2}{\Sigma_2^a} \quad (3)$$

$$\eta_1 f_1 = \frac{\nu \Sigma_1^f}{\Sigma_1^a} \quad (4)$$

$$p = \frac{\Sigma_{1 \rightarrow 2}}{\Sigma_{1 \rightarrow 2} + \Sigma_1^a} \quad (5)$$

Since the fundamental mode buckling, B^2 , is not measured experimentally and cannot be calculated to the desired accuracy, B^2 is evaluated using the experimental value of k_∞ from Equation (1) via the relation

$$k_\infty = \frac{(1 + \tau B^2)(1 + L^2 B^2)}{1 + (1 - p)L^2 B^2} \quad (6)$$

It is then necessary either to iterate on the buckling in Equation (1) or to eliminate the buckling and re-solve for k_{∞} as in Reference (6). These methods eliminate errors in the buckling due to uncertainties in the fuel block loading, heterogeneous geometry, and limitations of calculational methods.

The HTLTR experiments are designed to make use of the tremendous power of Equation (1) to determine $k_{\infty} - 1$ and therefore k_{∞} to a high degree of accuracy. In the plutonium-fueled HTGR experiments, the calculated terms contribute about 10% to $k_{\infty} - 1$. Therefore if the sum of these terms are estimated within 10%, they can contribute 1% error in $k_{\infty} - 1$. Reaction rate ratios can be measured within a 2% uncertainty, and the uncertainties in the neutron spectrum mismatch for k_{∞} near unity limit the minimum error to about 0.001. So, a reasonable overall error in $k_{\infty} - 1$ might be 4%. For a typical k_{∞} of 1.08 the error is only 0.003, or 0.3%. No other method utilizing only a small portion of a critical mass can claim this range of accuracy.

Another advantage of the formulation in Equation (1) is that only thermal absorption rates in the central cell constituents must be evaluated from foil activation measurements to determine k_{∞} . The uncertainties involved in evaluating epithermal resonance absorption rates in the fuel and epithermal capture-to-fission ratios in fissionable materials are eliminated. In addition it should be noted that except for the errors in measuring reactivity perturbations, $\Delta\rho$, all other uncertainties in $k_{\infty} - 1$ can be expected to be systematic with changes in temperature. Therefore the error in $[k_{\infty}(T_2) - k_{\infty}(T_1)]$ should also be about 4%, and in fact may be less than the overall error in k_{∞} .

The formulation in Equation (1) has general limitations which must be considered to determine its applicability to particular situations. For most thermal spectrum test lattices the thermal and epithermal neutron leakage correction terms are an order of magnitude smaller than the

experimentally evaluated term which is normalized by thermal neutron absorption rates. If the test lattice is nonthermal, $p \rightarrow 0$ and $\eta_1 f_1 \rightarrow k_\infty$, the epithermal leakage correction becomes prohibitively large and a different formulation is required.

The evaluation of k_∞ in the fundamental mode has the advantage of unique fluxes regardless of the multi-group neutron energy structure used. If there is insufficient test lattice surrounding the center cell to establish these unique fluxes within the epithermal and thermal groups, or if a dissimilar test lattice surrounds the center cell, the two-group mismatch corrections given in Equation (1) would be inappropriate. For those cases, a more general multi-group equation must be used to evaluate k_∞ , with some sacrifice in accuracy.

A third difficulty involves heterogeneities in the lattice. For example, Equation (1) does not provide a means for evaluating effects due to perturbations of central cell ends. The value for k_∞ without end caps has been evaluated by subtracting the measured worth of dummy end caps in the void from the worth of the central cell. The k_∞ derived in this manner has some error due to this treatment (estimated to be small in comparison to other sources of error) but it can be compared directly to the k_∞ calculated with unit cell computer codes. The value for k_∞ can also be evaluated with the end caps from the measured data. However, to obtain k_∞ with end caps from calculations to compare with experiment, either the end caps must be smeared into the moderator when single cell calculations are made, or a two-dimensional multigroup calculation must be carried out for the central cell and end cap regions explicitly. It is apparent, therefore, that k_∞ determined with end caps included is not necessarily much more accurate experimentally and the effect is still present to the same degree in any comparison with calculations. A comparison between experimental and calculated $k_\infty(T)$ evaluated both with and without end caps provides information about the adequacy of the model used to account for the end caps and about the effect of the end caps on the temperature dependence.

IV. DESCRIPTION OF THE LATTICE

A. FUEL BLOCKS

The fuel element for the HTGR uses a graphite fuel body which serves as the structural element to contain the coated particle fuel.⁽⁷⁾ The HTGR fuel blocks used in the HTLTR experiments, shown in Figure 2, are 3 3/4-inch by 3 3/4-inch square by 24-inch long SGBF graphite bars. The 2200 m/sec absorption cross section of this graphite, including all impurities, is about 3.80 millibarns per carbon atom,⁽⁸⁾ and the density is about 1.62 g/cm³. Each graphite block contains 25 channels, 0.470-inches in diameter, on a 0.750-inch square pitch. Each channel was closed at both ends with threaded graphite plugs 0.5-inches in length, resulting in a fuel channel length of 23.0-inches.

The fuel loaded in these channels consisted of a blend of pyrocarbon-coated PuO₂ microspheres, ThO₂ particles and graphite. The PuO₂ microspheres, shown in Figure 3, are actually miniature fuel elements, since the pyrocarbon coating isolates the plutonium from the reactor coolant and also contains the fission products. These microspheres were specially prepared at Oak Ridge National Laboratory (ORNL) from plutonium furnished by Battelle. According to the ORNL characterization report,⁽⁹⁾ the mean diameter of the PuO₂ kernel is 209 microns with a standard deviation of 20 microns, and the mean coating thickness of pyrocarbon is 91 microns. The average PuO₂ kernel density is 11.2 g/cm³ and the coated particles are 45.0% plutonium by weight. Isotopic analyses of the plutonium made both at Battelle and ORNL were in agreement when the ²⁴¹Pu content was corrected for natural decay after the date of the analysis. The isotopic percentages of the plutonium at the time of the HTLTR experiments were: 72.8% ²³⁹Pu, 22.9% ²⁴⁰Pu, 3.3% ²⁴¹Pu, and 1.0% ²⁴²Pu. The ²⁴¹Am content was about 0.20% of the plutonium by weight. Although the total weight of pyrocarbon coated microspheres in each block is accurately known, the amount of plutonium in each block has about 1% uncertainty due to the limitations of the chemical analyses.

FIGURE 2. Typical Fuel Block for HTGR Experiments in the HTLTR

FIGURE 3. Pyrocarbon Coated PuO_2 Microspheres for $\text{ThO}_2\text{-PuO}_2$ HTGR Lattice

The ThO_2 particles consist of irregular shapes of various sizes. The distribution of sizes by weight percent as determined by screening a small sample is given in Table I.

TABLE I. Thoria Particle Size Distribution

Tyler Standard Screen Mesh	Particle Size Range (microns)	Average Particle Size (microns)	Weight Percent
100-65	149-210	180	11.1
150-100	105-149	127	16.3
200-150	74-105	90	13.7
325-200	44-74	59	18.8
-325	0-44	22	40.1

These ThO_2 particles were not coated with pyrocarbon, so the total weight of ThO_2 in each block was determined very accurately. The uncertainty in the ThO_2 loading is estimated to be less than 0.1%. Spectrochemical analyses of the $\text{ThO}_2^{(10)}$ confirmed the absence of significant neutron absorbing impurities. The ThO_2 particles were assumed to have a density of 9.86 g/cm^3 .

The graphite particles were prepared by collecting particles milled from nuclear grade graphite blocks. These particles were then separated into two size portions by screening. Half of the graphite particles used were less than 210 microns (- 65 mesh) and the other half were 350 to 590 microns (- 28 mesh, + 42 mesh).

To load the fuel blocks, the coated PuO_2 microspheres, the ThO_2 and each of the two graphite particle sizes were weighed separately in amounts sufficient to fill one fuel block plus 10%. One end of the fuel block was sealed with half-inch long graphite end plugs screwed in flush with the block surface. The fuel was loaded in the other end through a specially constructed funnel with 25 holes. During the loading the block was set in a vertical position on a vibrator which was used to compact the fuel mixture into the channels. After vibration, the excess fuel blend was

removed and the open end sealed with graphite end caps. The finished blocks thus contain 23 inches of fuel blend in each channel and 0.5 inch of graphite on each end.

Using the above method for loading fuel blocks leads to small variations in the amount of material in each block. These variations mainly occur because of small differences between the blocks (i.e. the exact way in which the holes were drilled) and the differences between the way the fuel material packs into each hole. These variations were kept to a minimum and in particular, the blocks used for the central cells were carefully selected and extra care was taken with the fuel loading. Of the remaining blocks, each was characterized by the C/Th ratio and the total thorium in the block. Blocks within specifications but differing significantly from the mean loading were placed into the reactor at the outer edge of the buffer region. Blocks whose loadings came closest to the mean value were placed in the vicinity of the central cell insofar as possible.

The loading procedure described above was developed in an attempt to achieve a homogeneous fuel distribution throughout each fuel channel. Various graphite size fractions were tried before the final blend was adopted. The central cell blocks were X-rayed after loading. Visual inspection of the X-rays showed a uniform distribution of ThO_2 and PuO_2 particles in the fuel channels. The composition of the central cell is listed in Table II. Without the end cap graphite, the C/Th atom ratio is 251.6 and the C/Pu ratio is 7541. If the end caps are included these ratios are 264.1 and 7916, respectively.

TABLE II. Composition of Central Cell

<u>Region Component</u>	<u>Grams of Element (a)</u>	<u>Atoms/barn-cm (Each Region)</u>
<u>Moderator Region</u> (Excluding End Caps)		
$V_{\text{Mod}} = 14661.72$		
C	23809.	0.08142
N ^(b)	4.87	0.0000143
<u>Fuel Region</u>		
$V_{\text{Fuel}} = 6539.04 \text{ cm}^3$		
C	6223.	0.04775
N ^(b)	4.15	0.0000273
O	329.2	0.001895
^{232}Th	2306.	0.0009167
^{238}Pu	0.05	0.00000002
^{239}Pu	57.69	0.00002225
^{240}Pu	18.22	0.00000700
^{241}Pu	2.64	0.00000101
^{242}Pu	0.75	0.00000028
^{244}Pu	0.0008	0.0000000003
^{241}Am	0.16	0.00000006
<u>End Cap Region</u>		
$V_{\text{End Cap}} = 921.77 \text{ cm}^3$		
C	1491.	0.08092
N ^(b)	0.077	0.0000144

- a. Total weight in the 4 central cell blocks; region volumes are also totals for the 4 central cell blocks.
- b. Amount at 20°C in N₂ atmosphere. The nitrogen density was taken proportional to $1/T^2(^{\circ}\text{K})$ at elevated temperatures. Density changes in other cell components were assumed to be negligible.

B. TEST LATTICE DESCRIPTION

The HTGR test lattice is arranged in an 8 x 8 array, shown in Figure 4, in the center of the HTLTR. The blocks containing $\text{ThO}_2\text{-PuO}_2$ are located in a 6 x 6 array and are surrounded on the radial perimeter by one layer of blocks containing $\text{ThO}_2\text{-}^{235}\text{UC}_2$.⁽¹¹⁾ The central 4 x 4 array which contains the four central cell blocks is three blocks long. The outer two layers of the 8 x 8 HTGR block array are two blocks long.

C. HTLTR CRITICAL LOADING

Surrounding the HTGR blocks are UO_2 shims, 5% enriched UO_2 drivers, gadolinium shims, control rods and fuel-poison column. The HTLTR critical loading is shown schematically in Figure 5. In this loading, the inner ring surrounding the HTGR blocks contained driver fuel. In the next ring, gadolinium shims were located interspersed with natural UO_2 shims. Additional driver fuel and the control rods were located outside these zones. Only the adjacent portion of the 10-foot cubic main graphite stack is shown. Located two rings outside the horizontal control rods are fuel-poison columns⁽¹²⁾ used to add reactivity to make up for the loss with temperature increases. No other changes in loading were made.

FIGURE 4. ThO₂-PuO₂ HTGR Test Lattice (Central Region of HTLTR)

- X** 1 FIVE FOOT DRIVER + 2-10' END DRIVERS
U 4 NATURAL URANIUM SHIMS
G 2 GADOLINIUM TEMPERATURE SHIMS
1-10' END DRIVER AT FRONT + 1 AT REAR
2-10' END DRIVERS AT FRONT + 2 AT REAR

FIGURE 5. HTLTR Fuel Loading Diagram

D. SPECIAL SAMPLES

Special blocks and samples were used to measure various perturbations necessary in obtaining k_{∞} and in correlating worths of absorber materials in the $\text{ThO}_2\text{-PuO}_2$ HTGR lattice with similar measurements in ^{235}U and ^{233}U fueled HTGR lattices. To determine k_{∞} it is necessary to measure the reactivity worths of the standard central cell and copper poison. The copper poison was inserted on a graphite carrier. Two carriers were used, one to hold the copper and an identical one to measure the carrier worth without the copper. Two 3-inch by 24-inch pieces of 0.010-inch copper foil weighing a total of 205.018 grams were placed flat, side by side, on the carrier. Each graphite carrier weighed 2038 grams.

In addition to the standard central cell, a special three-block central cell, illustrated in Figure 6, was used for single block reactivity worth measurements. Single HTGR blocks fueled with $\text{ThO}_2\text{-}^{235}\text{UO}_2$,⁽¹¹⁾ $\text{ThO}_2\text{-}^{233}\text{UO}_2$,⁽¹³⁾ $\text{V}_2\text{O}_3\text{-PuO}_2$, or $\text{ThO}_2\text{-PuO}_2$ (the same as the standard central cell) were inserted to fill the single block void in the three-block central cell. Special poison blocks were also inserted in the three-block central cell to measure their reactivity worths in the $\text{ThO}_2\text{-PuO}_2$ HTGR lattice. These poison blocks are the same as the single fuel blocks except for the replacement of the normal fuel mixture with neutron absorber material (either Eu, Sm, Hf, Rh, or Gd) mixed with graphite. The contents of these blocks are presented in Table III.

A single block containing coated PuO_2 microspheres and graphite, but with V_2O_3 in place of ThO_2 particles, was used to measure the thorium resonance integral. The $1/v$ absorption cross section of vanadium in 272.72 grams of V_2O_3 just compensates for the $1/v$ portion of the absorption cross section of thorium in 657.03 grams of ThO_2 . The V_2O_3 powder was screened to particle sizes less than 210 microns (~ 65 mesh). This block was loaded in the same manner as the standard $\text{ThO}_2\text{-PuO}_2$ blocks.

FIGURE 6. Special Three-Block Central Cell

TABLE III. Composition of Poison Blocks

<u>Poison</u>	<u>Amount of Poison (g of element)</u>	<u>Total Block Weight (g)</u>
Eu	1.96	7888
Sm	1.67	7718
Gd*	1.69*	7722
Hf	82.3	7926
Rh	44.4	7824

* Rare earth mixture containing approximately 50% Gd_2O_3 mixed with other rare earths (mostly Sm_2O_3); weight shown is total rare earth. The cross section at room temperature is at least 80% due to Gd.

V. EXPERIMENTAL MEASUREMENTS

A. FLUX MATCHING

To determine the reactor configuration likely to give a good flux match, calculations were made using the two-dimensional diffusion theory code 2DB⁽¹⁴⁾ and a two-energy-group model of the HTLTR in R - Z geometry. The HTGR test core is arranged in an 8 x 8 array (shown in Figure 4) in the central region of the HTLTR. Calculations were performed for the progressive substitution of the 8 x 8 array of ThO_2 - $^{235}\text{UO}_2$ fueled blocks with ThO_2 - PuO_2 fueled blocks. The epithermal-to-thermal flux ratio in the central cell of a 6 x 6 array of ThO_2 - PuO_2 fueled blocks was about 8% lower than the fundamental mode value. A full 8 x 8 array of ThO_2 - PuO_2 fueled blocks was still too thermal by about 6%. Since a reasonably good neutron spectrum match could be obtained in the 6 x 6 array, containing 1747 grams of plutonium in 88 blocks, this loading was used because it required only 60% of the plutonium required for an 8 x 8 array. The two-group fluxes at 20°C calculated using the 2DB HTLTR model are shown in Figures 7 and 8 as a function of distance from the reactor centerline, radially and axially.

In order to measure the spatial variation of the epithermal-to-thermal flux ratio, radial and axial activation rates were measured using bare and cadmium-covered gold foils. The neutron spectrum index (gold cadmium ratio -1) is directly proportional to the thermal-to-epithermal flux ratio. The calculated thermal-to-epithermal flux ratio was normalized to the measured gold cadmium ratio -1 at the reactor centerline. Comparison of measured and calculated (gold cadmium ratio -1) values in the radial direction, shown in Figure 9, and in the axial direction, shown in Figure 10, indicate excellent agreement. This correlation indicates that the 2DB calculations may be used to provide reasonable estimates of the two-group flux mismatch.

FIGURE 7. Two-Group Fluxes as a Function of Radial Distance from the Reactor Centerline, Calculated Using the 2DB HTLTR Model

FIGURE 8. Two-Group Fluxes as a Function of Axial Distance from the Reactor Centerline, Calculated Using the 2 DB HTLTR Model

FIGURE 9. Comparison of Measured and Calculated (Gold Cadmium Ratio -1) As a Function of Radial Distance from the Reactor Centerline

FIGURE 10. Comparison of Measured and Calculated (Gold Cadmium Ratio -1) As a Function of Axial Distance from the Reactor Centerline

B. MEASUREMENT OF REACTION RATES

The measurement of relative thermal reaction rates in the actual cell and copper absorber is carried out at room temperature using foils. These foils are normalized to foils placed on a rotator at a location near the edge of the HTLTR. The rotator ensures that each type of foil sees an identical flux.

The reaction rate in the copper poison was measured by placing the poison strips in the cavity left when the central cell was removed and irradiating the material. Foils were punched from selected locations in the strips and the copper activity determined, to obtain the integral of absorptions in the poison strips.

The cell reaction rate was measured using 0.005-inch copper foils to evaluate l/v absorptions and fuel foils to evaluate fuel absorption. The copper foils were placed axially and radially between the central cell blocks to allow an integral of the flux over the cell to be calculated. A special fuel block, shown in Figure 11, containing the same $\text{PuO}_2\text{-ThO}_2$ fuel mixture as the central cell blocks was used to hold foils for the reaction rate measurements. This block had the fuel in two channels contained in removable aluminum cylinders, three in each channel, between which foils could be inserted. Special foils were fabricated to fit in the fuel channels. Foils made of copper and foils containing a mixture of coated PuO_2 microspheres, ThO_2 particles, and graphite identical to the fuel were used. These fuel foils were used to evaluate the relative absorption rates in plutonium and thorium.

In order to evaluate relative thermal absorptions, both bare and cadmium-covered (0.040-inch thick cadmium) foils were irradiated. The measured relative reaction rates were then corrected for the perturbations caused by the aluminum, the foils, and cadmium covers.⁽¹⁵⁾ These corrections were calculated using the two-dimensional transport theory code DOTSN.⁽¹⁶⁾ The corrections to the bare activity were small (about 0.5% or less). The corrections for the cadmium cover perturbations were larger, however.

FIGURE 11. Special HTGR Fuel Block Used in Foil Irradiations

These perturbations arise from depression of the epithermal flux in the vicinity due to thermal absorption in the cadmium, epithermal absorption in the cadmium, and due to production of thermal neutrons by scattering inside the cadmium cover. The DOTSN calculation provides the thermal and epithermal flux inside the cadmium cover relative to the unperturbed flux far from the foil. Using the DOTSN results, the thermal and epithermal activation rates in the fuel were evaluated by the relations:

$$A_2 = 1.004 A^{\text{bare}} - 1.036 A^{\text{Cd}} \quad (7a)$$

$$A_1 = 1.034 A^{\text{Cd}} - 0.0006 A^{\text{bare}}. \quad (7b)$$

It was also necessary to correct for epithermal absorption in the monitor foils located near the edge of the HTLTR. Measured copper and gold cadmium ratios at the monitor location were used to evaluate these corrections. These corrections decreased the measured monitor foil activities by about 7% in thorium, 1% in copper, and 0.4% in plutonium. The relative thermal activation rates (in the central cell fuel to the monitor location) for the corrected thorium activation measurements agreed within experimental uncertainties with the corrected copper activation measurements. Since the thermal absorption cross sections of both thorium and copper are $1/v$, this agreement between relative thermal activation rates tends to confirm the corrections made for epithermal absorption in the monitor foils.

Thermal cross sections at the monitor location were evaluated using a spectrum from a THERMOS⁽¹⁷⁾ calculation of the HTLTR. The cross sections of the materials in the PuO_2 microspheres and the ThO_2 particles at the monitor location were calculated using an averaging routine CROSAV⁽¹⁸⁾ which uses a collision probability routine together with the THERMOS fluxes and cross sections from the THERMOS BNW master library to calculate effective cross sections in the particle.

The absorptions in the copper poison and central cell are summarized in Table IV. The errors shown in the relative absorptions (column 5) include uncertainties in the grams of material in the central cell (column 1), uncertainties in the cross section (column 3) (evaluated at the monitor location and normalized to $1/v$ at 2200 m/sec.), and uncertainties in the activity (central cell absorptions/gram normalized to monitor absorptions/gram). Since these errors are to some extent systematic, the error in the total cell absorption has been taken to be the sum of the errors in the absorption in each cell component. Otherwise, it is felt that this error would be unrealistically small. The central cell reaction rate normalized to the thermal reaction rate in 209.9 grams of copper is then found from Table IV to be 39.96 ± 0.60 .

TABLE IV. Evaluation of Relative Reaction Rates

Central Cell Component	Weight (g)	Atomic Weight	$(\sigma_a)_{\phi_o}^{\text{monitor}}$	$\frac{(\sigma_a \phi)_2^{\text{cell}}}{(\sigma_a \phi)_2^{\text{monitor}}}$	$\frac{(\sum_a \phi V)_2^{\text{cell}}}{\phi_o^{\text{monitor}}}$
<u>Moderator Region</u>					
C	23809.	12.0	3.80×10^{-3}	0.659	2.99 ± 0.04
N	4.87	14.0	1.85	0.659	0.26 ± 0.01
<u>Fuel Region</u>					
<u>Fuel Diluent</u>					
C	6223.	12.0	3.80×10^{-3}	0.651	0.77 ± 0.01
N	4.15	14.0	1.85	0.651	0.21 ± 0.01
<u>ThO₂ Particles</u>					
²³² Th	2306.	232.0	7.389	0.651	28.80 ± 0.49
O	318.5	16.0	1.78×10^{-4}	0.651	0.00
<u>PuO₂ Microspheres</u>					
²³⁸ Pu	0.05	238.0	484.	0.713	0.04 ± 0.01
²³⁹ Pu	57.69	239.0	1158.	0.881	148.31 ± 1.29
²⁴⁰ Pu	18.22	240.0	256.1	0.800	9.69 ± 0.08
²⁴¹ Pu	2.64	241.0	1344.	0.767	6.80 ± 0.06
²⁴² Pu	0.75	242.0	17.4	0.730	0.02
²⁴⁴ Pu	0.0008	244.0	1.5	0.713	0.00
²⁴¹ Am	0.16	241.0	548.	0.713	0.16 ± 0.04
O	10.64	16.0	1.50×10^{-4}	0.713	0.00
Total Thermal Absorption Rate in Central Cell =					198.05 ± 2.04
Copper in Central Void	Weight (g)	Atomic Weight	$(\sigma_a)_{\phi_o}^{\text{monitor}}$	$\frac{(\sigma_a \phi)_i^{\text{copper}}}{(\sigma_a \phi)_2^{\text{monitor}}}$	$\frac{(\sum_a \phi V)_i^{\text{copper}}}{\phi_o^{\text{monitor}}}$
Thermal	209.9	63.54	3.79	0.657	4.956 ± 0.054
Epithermal	209.9	63.54	3.79	0.141	1.064 ± 0.012

C. MEASUREMENT OF REACTIVITIES

Reactivity measurements were made by placing a sample in the void and measuring the change in reactor asymptotic period. The change in reactivity was then calculated using the inhour equation.⁽¹⁹⁾ A correction for the reactivity of the nitrogen in the void was calculated from the worth of copper in the void.⁽²⁰⁾ This correction for the 7-1/2 x 7-1/2 x 24 inch void and the corrected reactivity worths for the various central cells, copper, and graphite, are summarized in Table V. The central cell worth, corrected for graphite end caps, is included in Table V. These reactivity worths include the N₂ in the pores of the graphite.

Additional measurements were made at room temperature in air. These measurements included a measurement of the copper worth on the carrier versus the copper worth in the void and the worth of an amount of graphite equivalent to the end cap region on the central cell. The effect of the carrier graphite was found to increase the worth per gram of copper on the carrier, relative to copper placed alone in the void, by $(3.3 \pm 0.8)\%$. This normalization was used to evaluate copper worths in the void at elevated temperatures from the measured copper worths on the carrier. The worth per gram of the dummy end cap graphite was $(5 \pm 2)\%$ larger than the worth per gram of the graphite carrier. Similarly, this normalization was used to evaluate the end cap worths at elevated temperatures from the measured graphite carrier worths.

The ThO₂-PuO₂ HTGR experiment utilized the new sample changer for the HTLTR, shown in Figure 12. This device allows samples to be preheated before insertion in the reactor and thus enables temperature transients during sample changes to be minimized. This results in a time saving during reactivity measurements and an improvement in accuracy. Errors have not been included in Table V, but typical errors might be $\pm 0.02\%$ at room temperature, $\pm 0.04\%$ at 300°C, 500°C, and 750°C, and $\pm 0.07\%$ at 1000°C. Since samples could not be preheated to 1000°C, some temperature transients remained at this temperature and the accuracy is therefore lower. These errors are estimated from the consistency of the data and represent approximately one standard deviation.

TABLE V. Reactivity Worth Measurements

	20°C, ¢	300°C, ¢	500°C, ¢	750°C, ¢	1000°C, ¢
N ₂ Correction	-0.96	-0.49	-0.32	-0.22	-0.16
Standard 4 Block Central Cell (W/End Caps)	+13.57	+11.98	+11.33	+9.91	+7.29
(Without End Caps)	+11.90	+10.10	+9.27	+7.74	+5.05
Copper in Void (209.898 g)	-3.64	-3.40	-3.15	-2.90	-2.75
Graphite Carrier (2038 g)	+2.17	+2.44	+2.67	+2.83	+2.89
Special 3 Block Central Cell (W/End Caps)	+9.78	+8.65	+8.25	+7.24	+5.28
(Without End Caps)	+8.53	+7.24	+6.69	+5.64	+3.60
Single ThO ₂ -PuO ₂ Block (With End Caps)	+3.52	+3.28	+2.93	+2.56	+1.82
(Without End Caps)	+3.10	+2.81	+2.41	+2.03	+1.26
Single V ₂ O ₃ -PuO ₂ Block (With End Caps)	+10.51	+10.12	+10.27	+10.52	+9.69
(Without End Caps)	+10.09	+9.65	+9.75	+9.98	+9.13
Single ThO ₂ - ²³⁵ UC ₂ Block ⁽¹¹⁾ (With End Caps)	+3.75	+3.11	+2.98	+2.84	+2.56
(Without End Caps)	+3.33	+2.64	+2.46	+2.31	+2.00
Single ThO ₂ - ²³³ UO ₂ Block ⁽¹³⁾ (With End Caps)	+3.30	+3.27	+3.45	+3.63	+3.78
(Without End Caps)	+2.88	+2.80	+2.93	+3.09	+3.22
Single Poison Blocks (With End Caps)					
Europium	-4.70	-1.95	-0.77	-0.44	-0.90
Rhodium	-23.93	-22.60	-21.55	-21.10	-21.44
Samarium	-19.66	-18.00	-13.64	-8.98	-5.60
Hafnium	-33.68	-33.30	-32.88	-32.80	-33.41
Gadolinium	-27.44	-19.88	-13.70	-7.93	-4.08

FIGURE 12. HTLTR Remote Sample Changer with Preheating Capabilities

A second advantage of the sample changer is the elimination of the extension effect which created an uncertainty in earlier experiments. (11, 13, 21)

The HTLTR is electrically heated and can be operated for extended periods of time (several weeks) with average temperatures of the 10-foot graphite cube up to 1000°C. Nitrogen gas is used to provide an inert atmosphere to inhibit oxidation of the carbon moderator. A photograph of the HTLTR graphite stack at 1000°C, taken through the viewing part is shown in Figure 13. Preheated samples can be moved along the graphite table, shown in Figure 13, to be inserted and withdrawn from the HTLTR using the sample changer, shown in Figure 12.

FIGURE 13. HTLTR Graphite Stack at 1000°C

VI. EVALUATION OF k_{∞}

The value of k_{∞} at each temperature was determined using Equation (1). Quantities used in the evaluation of k_{∞} without end caps are given in Table VI for uniform lattice temperatures of 20°, 300°, 500°, 750°, and 1000°C. The measurements of the reactivities of the central cell and the copper in the void are described in Section V.C. The central cell worth without end caps was obtained by subtracting the end cap worth.

The measurements of relative thermal-neutron absorption rates in the central cell and in the copper in the void at room temperature are described in Section V.B. Relative reaction rates as a function of temperature were deduced from the values measured at room temperature using temperature-dependent thermal-neutron cross section ratios calculated using GRANIT⁽²²⁾. The ratio of thermal neutron fluxes, central cell to copper in the void, calculated using the 2DB⁽¹⁴⁾ HTLTR model, was found to be constant as a function of temperature.

$$\begin{aligned}
 \left[\frac{(\Sigma_a V \phi)_2^{\text{Cu}}}{(\Sigma_a V \phi)_2^{\text{Cell}}} (T) \right] &= \left\{ \left[\frac{(\Sigma_a V \phi)_2^{\text{Cu}}}{(\Sigma_a V \phi)_2^{\text{Cell}}} (20^\circ\text{C}) \right]_{\text{measured}} \right. \\
 &\quad \times \left[\left(\frac{\bar{\sigma}_a(T)}{\bar{\sigma}_a(20^\circ\text{C})} \right)_2^{\text{Cu}} \left(\frac{\Sigma_a(20^\circ\text{C})}{\Sigma_a(T)} \right)_2^{\text{Cell}} \right]_{\text{GRANIT}} \\
 &\quad \times \left[\left(\frac{\bar{\phi}_2^{\text{void}}}{\bar{\phi}_2^{\text{Cell}}} (T) \right) \left(\frac{\bar{\phi}_2^{\text{Cell}}}{\bar{\phi}_2^{\text{void}}} (20^\circ\text{C}) \right) \right]_{\text{2DB}} \left. \right\} \quad (8)
 \end{aligned}$$

TABLE VI. Evaluation of k_{∞} Without End Caps

Quantity	20°C	300°C	500°C	750°C	1000°C
$\Delta\rho_{\text{With End Caps}}^{\text{cell}} (\phi)$	+13.57	+11.98	+11.33	+9.91	+7.29
End Cap Worth (ϕ)	+1.67	+1.88	+2.06	+2.17	+2.24
$\Delta\rho_{\text{Without End Caps}}^{\text{cell}} (\phi)$	+11.90	+10.10	+9.27	+7.74	+5.05
$\Delta\rho_{\text{in Void}}^{\text{Cu}} (\phi)$	-3.64	-3.40	-3.15	-2.90	-2.75
$-\left[\Delta\rho^{\text{cell}}/\Delta\rho^{\text{Cu}}\right]$	3.27 ± 0.03	2.97 ± 0.05	2.94 ± 0.06	2.67 ± 0.06	1.84 ± 0.08
$(\Sigma\phi V)_2^{\text{cell}}/(\Sigma\phi V)_2^{\text{Cu}}$	39.96 ± 0.60	46.33 ± 0.81	52.82 ± 1.09	59.29 ± 1.30	65.70 ± 1.62
$\left[1 + \left(\frac{\phi_1^+}{\phi_2^+}\right) \frac{(\Sigma\phi)_1^{\text{Cu}}}{(\Sigma\phi)_2^{\text{Cu}}}\right]$	1.1557	1.1805	1.2077	1.2344	1.2605
k_{∞} (uncorrected)	1.0945	1.0757	1.0673	1.0556	1.0352

Similarly, the ratio of epithermal-to-thermal reaction rates as a function of temperature for copper in the void was deduced from the measured value at room temperature, thermal and epithermal cross sections calculated using GRANIT and EGGNIT,⁽²³⁾ and the epithermal-to-thermal flux ratios in the void listed in Table VII, calculated using the 2DB HTLTR model.

$$\left[\frac{(\Sigma_a \phi)_1^{Cu}}{(\Sigma_a \phi)_2^{Cu}} (T) \right] = \left\{ \left[\frac{(\Sigma_a \phi)_1^{Cu}}{(\Sigma_a \phi)_2^{Cu}} (20^\circ\text{C}) \right]_{\text{measured}} \right. \\ \times \left[\left(\frac{\bar{\sigma}_a(20^\circ\text{C})}{\bar{\sigma}_a(T)} \right)_2^{Cu} \right]_{\text{GRANIT}} \times \left[\left(\frac{\bar{\sigma}_a(T)}{\bar{\sigma}_a(20^\circ\text{C})} \right)_1^{Cu} \right]_{\text{EGGNIT}} \\ \times \left[\left(\frac{\bar{\phi}_1}{\bar{\phi}_2} (T) \right) \left(\frac{\bar{\phi}_2}{\bar{\phi}_1} (20^\circ\text{C}) \right) \right]_{\text{2DB}}^{\text{void}} \left. \right\} \quad (9)$$

The average thermal flux depression in the copper was 0.8% at room temperature, calculated using DOTSN.⁽¹⁶⁾ For the epithermal range, the resonance self-shielding for the 0.010-inch-thick copper foil was calculated using EGGNIT without including the copper in the unit cell. It was assumed that the epithermal flux depression in the copper, except in the resonances, can be neglected. Using the epithermal-to-thermal flux ratio in the void, calculated at room temperature using the 2DB HTLTR model, and copper cross sections calculated using GRANIT and EGGNIT with the above assumptions, the calculated cadmium ratio for copper agreed with the measured value. The evaluation of the epithermal neutron contribution to the copper worth,

$$\left(\frac{\phi_1^+}{\phi_2^+} \right) \frac{(\Sigma \phi)_1^{Cu}}{(\Sigma \phi)_2^{Cu}}, \text{ used the adjoint fluxes in the void, calculated using the}$$

2DB HTLTR model, with the epithermal-to-thermal reaction rates from Equation (9). The resulting values which account for the epithermal contribution to the copper worth are presented in Table VI.

TABLE VII. Calculated Fluxes and Adjoints

	20°C	300°C	500°C	750°C	1000°C
ϕ_1/ϕ_2 Fundamental Mode (No End Caps)	3.483	3.589	3.780	3.944	4.078
ϕ_1/ϕ_2 Fundamental Mode (With End Caps)	3.336	3.439	3.621	3.777	3.906
ϕ_1/ϕ_2 HTLTR Lattice (Central Cell)	3.076	3.133	3.278	3.414	3.502
ϕ_1/ϕ_2 HTLTR Lattice (Central Void)	2.917	2.941	3.051	3.148	3.225
ϕ_1^+/ϕ_2^+ Fundamental Mode (No End Caps)	0.6904	0.6760	0.6668	0.6594	0.6536
ϕ_1^+/ϕ_2^+ Fundamental Mode (With End Caps)	0.6914	0.6772	0.6678	0.6604	0.6540
ϕ_1^+/ϕ_2^+ HTLTR Lattice (Central Void)	0.7249	0.7065	0.6970	0.6864	0.6773

The temperature-dependence of the corrections in Equation (1) which account for fundamental mode neutron leakage was principally due to changes in the buckling, B^2 , obtained from Equation (6). These corrections to k_∞ listed in Table VI amounted to -0.010 at 20°C, decreasing to -0.005 at 1000°C.

As stated in Section V.A., the spectrum mismatch at the central cell was reasonably small. The corrections to k_∞ due to this mismatch were calculated from Equation (1) using flux and adjoint ratios listed in Table VII. Fundamental mode flux and adjoint ratios were calculated for the lattice with end caps, in addition to the calculations without end caps. Mismatch corrections to k_∞ for the HTLTR lattice, listed in Table VI, were less than -0.001 for all cases.

One of the largest uncertainties in the method of evaluating the experimental k_∞ is due to the necessity of accounting for the end cap effect. As described above, the end cap graphite worth was measured in the void and subtracted from the central cell worth. The value of k_∞ evaluated using this net cell worth can then be compared to the value calculated for a cell without any end caps.

A second method of evaluating k_{∞} is to leave the end caps in the central cell. Parameters for the cell including the end caps must then be calculated. A one-dimensional transport code DTF-IV⁽²⁴⁾ with two energy groups and slab geometry was used to calculate the fundamental mode conditions with end caps. Leakage corrections and mismatch corrections to k_{∞} with end caps calculated using this model are listed in Table VIII. These results are similar to the values in Table VI for the case with end caps subtracted. It is apparent that k_{∞} determined with end caps included is not necessarily much more accurate experimentally. The end cap effect is still present to the same degree in any comparison with calculations. However, a comparison between experimental and calculated $k_{\infty}(T)$ evaluated both with and without end caps provides information on the adequacy of the model used to account for the end caps, and on the effect of the end caps on the temperature dependence of k_{∞} .

TABLE VIII. Evaluation of k_{∞} With Graphite End Caps

Quantity	20°C	300°C	500°C	750°C	1000°C
$\Delta\rho_{\text{With End Caps}}^{\text{cell}} (\%)$	+13.57	+11.98	+11.33	+9.91	+7.29
$\Delta\rho_{\text{in Void}}^{\text{Cu}} (\%)$	-3.64	-3.40	-3.15	-2.90	-2.75
$-\left[\Delta\rho^{\text{cell}}/\Delta\rho^{\text{Cu}}\right]$	3.73 ± 0.03	3.52 ± 0.06	3.60 ± 0.07	3.42 ± 0.07	2.65 ± 0.10
$(\Sigma\phi V)_2^{\text{cell}}/(\Sigma\phi V)_2^{\text{Cu}}$	40.01 ± 0.60	46.38 ± 0.81	52.86 ± 1.09	59.33 ± 1.30	65.74 ± 1.62
$\left[1 + \left(\frac{\phi_1^+}{\phi_2^+}\right) \frac{(\Sigma\phi)_1^{\text{Cu}}}{(\Sigma\phi)_2^{\text{Cu}}}\right]$	1.1557	1.1805	1.2077	1.2344	1.2605
k_{∞} (uncorrected)	1.1077	1.0897	1.0822	1.0711	1.0509
Leakage Correction	-0.0101	-0.0089	-0.0079	-0.0068	-0.0054
Mismatch Correction	-0.0008	-0.0008	-0.0008	-0.0009	-0.0009
k_{∞} (With End Caps)	1.097 ± 0.003	1.080 ± 0.003	1.073 ± 0.003	1.063 ± 0.002	1.045 ± 0.002

VII. EXPERIMENT-THEORY CORRELATION

The values for k_{∞} evaluated from HTLTR measurements using Equation (1) are consistent with the theory used for fundamental mode calculations,

$$k_{\infty} = \frac{\text{Total Neutron Production}}{\text{Total Neutron Absorption}} = \frac{\nu_1 \Sigma_1^f \phi_1 + \nu_2 \Sigma_2^f \phi_2}{\Sigma_1^a \phi_1 + \Sigma_2^a \phi_2} \quad (10)$$

where subscript 2 refers to the thermal range ($E_n < 0.683$ eV) and 1 refers to the epithermal range ($E_n > 0.683$ eV). These quantities are also related by the two-group equations:

$$-D_1 B^2 \phi_1 - \Sigma_1^a \phi_1 - \Sigma_{1 \rightarrow 2} \phi_1 + \nu_1 \Sigma_1^f \phi_1 + \nu_2 \Sigma_2^f \phi_2 = 0 \quad (11a)$$

$$-D_2 B^2 \phi_2 - \Sigma_2^a \phi_2 + \Sigma_{1 \rightarrow 2} \phi_1 = 0 \quad (11b)$$

where $\Sigma_{1 \rightarrow 2}$ is the cross section for net transfer from group 1 to group 2, B^2 is the buckling, and ϕ_1 and ϕ_2 are the fundamental mode fluxes. It is important to point out that the value for k_{∞} calculated using Equation (10) is independent of group structure because the fundamental mode fluxes are characteristic of the medium. Eliminating the fluxes from Equation (10), it is found that:

$$k_{\infty} = \frac{\eta_2 f_2 p + \eta_1 f_1 (1-p) [1 + L^2 B^2]}{1 + (1-p) L^2 B^2} \quad (12)$$

by use of Equations (2) through (5). The values of k_{∞} calculated from this equation, together with the parameters needed in the evaluation are presented in Table IX.

The computer codes GRANIT⁽²²⁾ and EGGNIT⁽²³⁾ with ENDF/B-III cross section data were used to calculate the neutronic parameters listed in Table IX. The GRANIT code is a modification of THERMOS⁽²⁵⁾ that is used to calculate thermal group constants for a lattice cell with a granular composite fuel rod. The granular composite fuel rod consisted of spherical PuO_2 particles and ThO_2 particles in a graphite diluent. Thirty-five energy groups were used in the range 0 to 1.86 eV. The edge of the cylindricized cell was treated as having a white boundary.

TABLE IX. Calculated Neutronic Parameters

<u>ThO₂PuO₂ Cell</u>		<u>20°C</u>	<u>300°C</u>	<u>500°C</u>	<u>750°C</u>	<u>1000°C</u>
$\nu \sum_1^f$	cm ⁻¹	3.0946 x 10 ⁻⁴	3.0973 x 10 ⁻⁴	3.1092 x 10 ⁻⁴	3.1216 x 10 ⁻⁴	3.1381 x 10 ⁻⁴
$\sum_1^a$	cm ⁻¹	1.1000 x 10 ⁻³	1.1785 x 10 ⁻³	1.2335 x 10 ⁻³	1.2816 x 10 ⁻³	1.3320 x 10 ⁻³
$\sum_1^{tr}$	cm ⁻¹	0.25567	0.25546	0.25536	0.25532	0.25530
$\sum_{1 \rightarrow 2}$	cm ⁻¹	2.3171 x 10 ⁻³	2.2684 x 10 ⁻³	2.2165 x 10 ⁻³	2.1792 x 10 ⁻³	2.1365 x 10 ⁻³
$\nu \sum_2^f$	cm ⁻¹	1.1689 x 10 ⁻²	1.2044 x 10 ⁻²	1.2599 x 10 ⁻²	1.3046 x 10 ⁻²	1.3331 x 10 ⁻²
$\sum_2^a$	cm ⁻¹	7.8782 x 10 ⁻³	7.9976 x 10 ⁻³	8.2777 x 10 ⁻³	8.5069 x 10 ⁻³	8.6472 x 10 ⁻³
$\sum_2^{tr}$	cm ⁻¹	0.34354	0.39609	0.41949	0.43506	0.44568
L^2	cm ²	123.16	105.23	96.06	90.21	86.49
τ	cm ²	375.90	373.66	373.61	372.27	370.90
p		0.67810	0.65810	0.64247	0.62971	0.61597
$\eta_1 f_1$ ($E_n > 0.683$ eV)		0.28134	0.26282	0.25206	0.24357	0.23559
$\eta_2 f_2$ ($E_n < 0.683$ eV)		1.4838	1.5060	1.5220	1.5336	1.5417
B^2 Without		1.982 x 10 ⁻⁴	1.722 x 10 ⁻⁴	1.471 x 10 ⁻⁴	1.223 x 10 ⁻⁴	0.889 x 10 ⁻⁴
k_∞ End Caps		1.092	1.076	1.068	1.056	1.039
<u>Graphite End Cap</u>						
$\sum_1^a$	cm ⁻¹	6.0648 x 10 ⁻⁶	5.8610 x 10 ⁻⁶	5.9003 x 10 ⁻⁶	5.9336 x 10 ⁻⁶	6.0256 x 10 ⁻⁶
$\sum_1^{tr}$	cm ⁻¹	0.28937	0.28970	0.28984	0.28997	0.29023
$\sum_{1 \rightarrow 2}$	cm ⁻¹	2.6821 x 10 ⁻³	2.5817 x 10 ⁻³	2.5227 x 10 ⁻³	2.4804 x 10 ⁻³	2.4310 x 10 ⁻³
$\sum_2^a$	cm ⁻¹	1.9850 x 10 ⁻⁴	1.6367 x 10 ⁻⁴	1.4575 x 10 ⁻⁴	1.3251 x 10 ⁻⁴	1.2161 x 10 ⁻⁴
$\sum_2^{tr}$	cm ⁻¹	0.36246	0.44575	0.47242	0.49014	0.50223
B^2 With		2.093 x 10 ⁻⁴	1.891 x 10 ⁻⁴	1.628 x 10 ⁻⁴	1.386 x 10 ⁻⁴	0.962 x 10 ⁻⁴
k_∞ End Caps		1.101	1.087	1.080	1.068	1.052

EGGNIT is a modification of GAM⁽²⁶⁾ that uses the Nordheim numerical integration method⁽²⁷⁾ extended to multiregion and granular composite geometries, which permits treating both absorber and admixed scatterers exactly, and includes Doppler broadening and spatial self-shielding effects. The EGGNIT code was used to calculate epithermal group constants for neutron energies above 1.86 eV. The fundamental mode buckling was used in the neutron spectrum calculation to account for the effect of leakage on the cross sections obtained for one epithermal broad group.

A comparison of measured and calculated values of k_{∞} as a function of temperature for the ThO₂-PuO₂ HTGR lattice without end caps is presented in Figure 14. The calculated values for k_{∞} are about 0.8% higher than the measured values. This difference is larger than the 0.3% uncertainty in the measured values for k_{∞} , that was obtained by propagating the error associated with the measured and calculated quantities used in Equation (1). The quoted uncertainties are at the 67% confidence level.

A potential source of error in the calculation of k_{∞} is the thorium resonance capture rate. The calculated epithermal-to-thermal capture rate in thorium is about 2% lower than the measured value. Since the calculated ratio of thermal absorptions in thorium to copper is in agreement with the experimentally determined value at room temperature, the epithermal thorium capture rate calculated using EGGNIT may be about 2% low. As discussed in Section IV.A., the ThO₂ particles consist of irregular shapes of various sizes. For purposes of calculation in EGGNIT, the ThO₂ particles were assumed to be spherical. Since the mean chord length of an irregular shaped particle just passing through a given screen opening is less than the mean chord length of a spherical particle just passing through the same screen opening, the assumption of spherical ThO₂ particles could result in an underestimate of the thorium resonance capture rate. The same assumption would have negligible effect on the thermal capture rate in thorium, calculated using GRANIT. It is estimated that a 2% increase in the calculated epithermal capture rate in thorium would reduce the calculated values for k_{∞} by about 0.5%.

FIGURE 14. Comparison of Measured and Calculated Values for K_{∞} Without End Caps

Another source of error in the calculations is the uncertainty in the exact loading of the fuel blocks. Although the weight of pyrocarbon coated PuO_2 microspheres loaded in each block is known with an uncertainty less than 0.1%, the amount of plutonium contained in these microspheres is uncertain by about 1% due to difficulties in determining the average weight fraction of the pyrocarbon coating. A 1% uncertainty in the amount of plutonium loaded in the fuel leads to an uncertainty in the calculated values for k_∞ of about 0.2%. A much smaller uncertainty exists for the amount of thorium loaded in the fuel since the ThO_2 particles were not coated with pyrocarbon. The calculated temperature-dependent changes in k_∞ are not affected by these small normalizations of reaction rates. The changes of k_∞ with temperature, calculated using GRANIT and EGGNIT without end caps, are in agreement with the measured changes of k_∞ with temperature, as shown in Figure 14.

A similar comparison of measured and calculated values of k_∞ for the ThO_2 - PuO_2 HTGR lattice with end caps, as a function of temperature, is presented in Figure 15. The calculated values for k_∞ are about 0.5% higher than the measured values. It is seen that the graphite end caps have little effect on the temperature-dependent changes in k_∞ .

Comparison of the experiment-theory correlation with the end caps included to the experiment-theory correlation for the end caps subtracted indicates qualitatively little difference. The calculated k_∞ with end caps is in slightly better agreement with experiment, but the differences between correlations are within the experimental uncertainties. Some of this difference between correlations may be due to the necessity for measuring the end cap worth in the void. However, it has been shown⁽²⁸⁾ that smearing the end caps into the cell moderator by enlarging the outer cell radius and leaving the Dancoff correction unchanged leads to a slightly larger calculated k_∞ with end caps than the two-group slab geometry end cap model described in Section VI. Thus, the experiment-theory correlations both with and without end caps are consistent, and show that the model used to account for end caps is in reasonable agreement with experiment.

FIGURE 15. Comparison of Measured and Calculated Values for K_{∞} With Graphite End Caps

VIII. CONCLUSIONS

This HTLTR experiment with a $\text{ThO}_2\text{-PuO}_2$ HTGR lattice provides a determination of k_∞ and its change over the range of temperatures from 20° to 1000°C . These data are useful for testing and evaluating the computational methods and cross sections which are being used in the design of plutonium fueled HTGRs.

The GRANIT and EGGNIT codes were used to calculate the temperature-dependent k_∞ for the as-built $\text{ThO}_2\text{-PuO}_2$ HTGR lattice. A comparison of these theoretical values for k_∞ with experimental values shows that the calculated values are about 0.8% higher than the measured values for the $\text{ThO}_2\text{-PuO}_2$ lattice without end caps. Similarly, calculated values are about 0.5% higher than the measured values for the $\text{ThO}_2\text{-PuO}_2$ lattice with graphite end caps. The principal difference between the calculated and measured values for k_∞ may be due to a 2% calculational underestimate of the thorium resonance capture rate. This underestimate could result from the assumption in the calculations that the irregular shaped ThO_2 particles are spherical. A 2% increase in the calculated thorium resonance capture rate would bring the calculated values for k_∞ into agreement with experiment.

An adequate knowledge of the variation in k_∞ with reactor temperature is important for both safety and economic reasons. The reactivity loss due to heatup is a major consideration in determining the amount of control strength that is needed to maintain adequate shutdown margins. This reactivity loss also has a bearing on the initial fuel enrichment needed to achieve burnup goal lifetimes. When plutonium is used as the initial fissile fuel, the temperature coefficient is changed favorably from that of an HTGR lattice fueled initially with ^{235}U .⁽¹¹⁾ The reactivity loss due to elevating the lattice to operating temperatures is about 30% less with plutonium fuel than with ^{235}U fuel. Above the operating temperature range, plutonium tends to make the temperature coefficient more negative due to increased neutron capture in the 1.06 eV resonance of ^{240}Pu . This effect will improve the self-limiting characteristics of the HTGR during a power excursion.

The $\text{ThO}_2\text{-PuO}_2$ HTGR lattice experiment in the HTLTR provides the only available neutronics data base for the development of an HTGR initially fueled with plutonium. The reactivity perturbations of several poison blocks, measured in the center of this $\text{ThO}_2\text{-PuO}_2$ lattice in the HTLTR, provide information on the temperature-dependent effectiveness of europium, samarium, hafnium, rhodium, and gadolinium as control rod materials for this lattice. A plutonium fueled test element has been inserted in Peach Bottom HTGR to evaluate its irradiation performance.⁽²⁹⁾ This test element is surrounded by uranium-bearing fuel elements so it has little effect on the neutronic characteristics of the Peach Bottom HTGR.

At present there is no neutronics data base for testing and evaluating computational methods and cross sections for both plutonium and ^{233}U present in an HTGR lattice. Such a lattice would more nearly represent an actual HTGR situation where plutonium is used as fissile makeup with recycled ^{233}U , or for an HTGR lattice initially fueled with plutonium near end-of-life when substantial ^{233}U has built-in. The reactivity perturbations of a single $\text{ThO}_2\text{-}^{233}\text{UO}_2$ block⁽¹³⁾ as a function of temperature were measured in the center of the $\text{ThO}_2\text{-PuO}_2$ lattice. The reactivity worth of the ^{233}U fueled block increased with temperature when measured in the $\text{ThO}_2\text{-PuO}_2$ lattice, in contrast to the observed reactivity worth decrease of the same block when measured in a lattice of identical $\text{ThO}_2\text{-}^{233}\text{UO}_2$ blocks.⁽¹³⁾ Since ^{233}U has a major low lying resonance at 1.78 eV the observed reactivity increase may be due to increased shielding of the 1.06 eV capture resonance of ^{240}Pu in the plutonium fueled lattice. This additional complexity leads to larger uncertainties in the neutronic characteristics calculated for HTGR lattices containing both plutonium and ^{233}U than exist for lattices containing either plutonium or ^{233}U as the fissile fuel.

Many improvements in the experimental equipment and methods utilized in the HTLTR have led to increased precision in the measured values for k_∞ . The first HTGR experiment in the HTLTR⁽¹¹⁾ had quoted uncertainties of

about 1.3% on measured values for k_{∞} . The temperature control of the HTLTR graphite stack has been improved and the central cell extension was eliminated for the $\text{ThO}_2\text{-PuO}_2$ HTGR experiment. In addition the new sample changer preheated the blocks before insertion in the HTLTR. This enabled temperature transients during sample changes to be minimized. The fuel-poison columns,⁽¹²⁾ used to add reactivity to make up for loss with temperature increases, eliminated the need to make driver fuel loading changes. As a result of these improvements, the quoted uncertainties on the measured values for k_{∞} were reduced to about 0.3% for the $\text{ThO}_2\text{-PuO}_2$ HTGR lattice.

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APPENDIX A

APPENDIX AEVALUATION OF k_{∞} FOR V_2O_3 -PuO₂

As mentioned in Section IV.D., a single block containing coated PuO₂ microspheres and graphite, but with V₂O₃ in place of ThO₂ particles, was used to measure the temperature dependent k_{∞} of the lattice with the thorium resonance capture removed. The $1/v$ absorption cross section of vanadium in 272.72 grams of V₂O₃ just compensates for the $1/v$ portion of the absorption cross section of thorium in 657.03 grams of ThO₂ that it replaces in a single block. The reactivity worth of this block was measured when the special three-block central cell, illustrated in Figure 6, was in the HTLTR central void.

Since the calculated cross sections for the V₂O₃-PuO₂ cell were identical to those for the ThO₂-PuO₂ cell, except for epithermal absorption, the probability of an epithermal neutron escaping to the thermal range was increased in the HTLTR when the V₂O₃-PuO₂ cell filled the central void. This increased the neutron multiplication of the remainder of the reactor outside the V₂O₃-PuO₂ cell due to the presence of this cell. However, the multiplication of excess neutrons leaking from the V₂O₃-PuO₂ cell was reduced from that which would occur if the cell were driven critical by an array of identical V₂O₃-PuO₂ cells. In order to obtain k_{∞} for the V₂O₃-PuO₂ cell in the fundamental mode spectrum of ThO₂-PuO₂ cells, corrections for these flux gradients and the neutron spectrum mismatch were calculated using the 2DB HTLTR model.⁽¹⁴⁾ Calculations were performed as a function of temperature (a) with the V₂O₃-PuO₂ cell installed and (b) with the ThO₂-PuO₂ cell installed. Eigenvalue convergence of 5×10^{-6} was specified in the 2DB calculations.

The neutron multiplication of the HTLTR with the V₂O₃-PuO₂ cell installed can be expressed in terms of neutron production, P, and absorption, A

$$K_R = \frac{P_R}{A_R} \quad , \quad (A-1)$$

and in the volume of the single V_2O_3 -PuO₂ cell

$$K_C = \frac{P_C}{A_C} \quad . \quad (A-2)$$

The neutron multiplication in the HTLTR, outside the V_2O_3 -PuO₂ cell, is

$$K_O = \frac{P_R - P_C}{A_R - A_C} = \frac{P_O}{A_O} \quad , \quad (A-3)$$

and the net neutron leakage from the V_2O_3 -PuO₂ cell is

$$L_C = P_C - A_C \quad . \quad (A-4)$$

Similarly, the neutron multiplication factor of the HTLTR with the ThO₂-PuO₂ cell installed is

$$K'_R = \frac{P'_R}{A'_R} \quad , \quad (A-5)$$

and in the volume of the single ThO₂-PuO₂ cell

$$K'_C = \frac{P'_C}{A'_C} \quad (A-6)$$

The neutron multiplication in the HTLTR, outside the ThO₂-PuO₂ cell is

$$K'_O = \frac{P'_R - P'_C}{A'_R - A'_C} = \frac{P'_O}{A'_O} \quad , \quad (A-7)$$

and the net neutron leakage from the ThO₂-PuO₂ cell is

$$L'_C = P'_C - A'_C \quad (A-8)$$

The excess neutrons produced in the HTLTR, outside the single central cell due to the presence of the V_2O_3 -PuO₂ cell is

$$\delta P_O = P_O - P'_O \quad (A-9)$$

The excess neutron leakage from the V_2O_3 -PuO₂ cell is absorbed in the HTLTR outside the single central cell

$$\delta A_O = (P_C - A_C) - (P'_C - A'_C) \quad (A-10)$$

Thus, the apparent neutron multiplication of the V_2O_3 -PuO₂ cell in the HTLTR with ThO₂-PuO₂ buffer cells can be obtained from Equations (A-2), (A-9), and (A-10).

$$k_c^* = \frac{P_c + \delta P_o}{A_c + \delta A_o} \quad (A-11)$$

Recall that the neutron multiplication factor of the V_2O_3 -PuO₂ cell in the fundamental mode spectrum of the ThO₂-PuO₂ lattice is

$$k_{\infty}^{V_2O_3} = \left(\frac{\nu \Sigma_1^f (\phi_1/\phi_2) + \nu \Sigma_2^f}{\Sigma_1^a (\phi_1/\phi_2) + \Sigma_2^a} \right) V_2O_3, \quad (A-12)$$

where ϕ_1/ϕ_2 is the epithermal-to-thermal fundamental mode flux ratio from Table VII. Then, the calculated correction to the measured value of k_{∞} for the V_2O_3 -PuO₂ cell is

$$\delta k_{\infty}^{V_2O_3} = k_{\infty}^{V_2O_3} - k_c^* \quad (A-13)$$

This correction, shown as a function of temperature in Figure 16, was added to the measured values of k_{∞} for the V_2O_3 -PuO₂ cell.

The corrected experimental values of k_{∞} for the V_2O_3 -PuO₂ cell as a function of temperature are shown in Figure 17, and compared to values calculated with the GRANIT and EGGNIT codes. The large value of $k_{\infty}-1$ and the large calculated correction, from Figure 16, combine to make the uncertainty in the experimental values for k_{∞} about 1.5%. As shown in Figure 17, the values of k_{∞} for the V_2O_3 -PuO₂ cell, deduced from experiment, fall below the calculated values by an amount of the same size as the uncertainty in the experimental values. This difference may be due to a bias in the calculated two-group flux gradient and spectrum mismatch corrections that were added to the measured values of k_{∞} for the V_2O_3 -PuO₂ cell. If these corrections were recalculated using at least 4 groups, some indication of the bias due to the two-group approximation could be obtained.

FIGURE 16. Temperature-Dependent Correction to K_{∞} for the V_2O_3 - PuO_2 Cell
Due to Flux Gradients and Spectrum Mismatch

FIGURE 17. Comparison of Calculated and Measured Values of K_{∞} for V_2O_3 -PuO₂ and ThO₂-PuO₂

APPENDIX B

APPENDIX BEVALUATION OF THE THORIUM DOPPLER COEFFICIENT

A simple and direct method for measuring the Doppler coefficients, $\frac{1}{I} \frac{dI}{dT}$, for fertile absorbers in an HTLTR test lattice has been developed. (30) This method of analysis was applied to the experimental data for the ThO_2 - PuO_2 HTGR lattice to evaluate the thorium Doppler coefficient. The temperature-dependent k_∞ values for both the ThO_2 - PuO_2 lattice and the V_2O_3 - PuO_2 cell, presented in Figure 17, are required in the analysis.

The temperature coefficient for the ThO_2 - PuO_2 lattice can be expressed by

$$\frac{1}{k_\infty} \frac{dk_\infty}{dT} = \frac{1}{p^{232}} \frac{dp^{232}}{dT} + \frac{1}{f} \frac{df}{dT} + \frac{1}{\eta} \frac{d\eta}{dT} \quad (\text{B-1})$$

A corresponding temperature coefficient for the V_2O_3 - PuO_2 cell can be expressed by

$$\frac{1}{k'_\infty} \frac{dk'_\infty}{dT} = \frac{1}{f} \frac{df}{dT} + \frac{1}{\eta} \frac{d\eta}{dT} \quad (\text{B-2})$$

Substitution of Equation (B-2) into Equation (B-1) isolates the temperature coefficient due to thorium resonance capture.

$$\frac{1}{p^{232}} \frac{dp^{232}}{dT} = \frac{1}{k_\infty} \frac{dk_\infty}{dT} - \frac{1}{k'_\infty} \frac{dk'_\infty}{dT} \quad (\text{B-3})$$

The coefficients on the right hand side of Equation (B-3) were obtained from the data, presented in Figure 17, that was evaluated from HTLTR measurements.

The thorium resonance escape probability, p^{232} , is related to the non- $1/\nu$ capture in thorium by the usual expression (31)

$$p^{232} = e^{-\left[\frac{N}{\xi \Sigma_s} I \right]} \quad (\text{B-4})$$

Using this equation, the Doppler coefficient can be expressed in terms of the effective resonance integral for thorium, I , from Equation (B-3)

$$\frac{1}{I} \frac{dI}{dT} = \frac{1}{\ln p^{232}} \left[\frac{1}{k_{\infty}} \frac{dk_{\infty}}{dT} - \frac{1}{k'_{\infty}} \frac{dk'_{\infty}}{dT} \right] \quad (B-5)$$

The Doppler coefficient was evaluated for the temperature range between 300°C and 750°C using Equation (B-5). The thorium resonance escape probability, p^{232} , was evaluated at 300°C from cross sections calculated using the EGGNIT code⁽²³⁾

$$p^{232} = \frac{\Sigma_{1 \rightarrow 2}}{\left[\Sigma_a^{Th0} - \Sigma_a^{V20} \right]_1 + \Sigma_{1 \rightarrow 2}} = 0.764 \quad (B-6)$$

The resulting Doppler coefficient evaluated from the measured temperature coefficients between 300°C and 750°C is

$$\frac{1}{I} \frac{dI}{dT} = 1.7 \pm 0.5 \times 10^{-4}/^{\circ}\text{C} \quad .$$

If the Doppler coefficient is evaluated from the calculated temperature coefficients between 300°C and 750°C,

$$\frac{1}{I} \frac{dI}{dT} = 1.5 \times 10^{-4}/^{\circ}\text{C} \quad ,$$

the result is within the uncertainty in the value deduced from experimental data.

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