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The Infinite Neutron Multiplication Constant of Homogeneous Hydrogen-Moderated 2.0 wt%-U^{235}-Enriched Uranium

JOHN T. MIHALCZO

Oak Ridge National Laboratory, Oak Ridge, Tennessee

AND

VICTOR I. NEELEY

*General Electric Company, Hanford Atomic Products Operation, Richland, Washington**

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The infinite medium neutron multiplication factor, k_∞, of a mixture of 92.1 wt% UF_4 and 7.9 wt% paraffin has been measured both in the Physical Constants Testing Reactor at the Hanford Atomic Products Operation and in critical experiments at the Oak Ridge National Laboratory. The density of the mixture is 4.5 gm/cc and the U^{235} enrichment of the uranium is 2.0 wt%, resulting in an $H:U^{235}$ atomic ratio of 195. The values of k_∞ from the two experiments are 1.216 ± 0.013 and 1.197 ± 0.015, respectively.

In the analysis of the critical experiments a two group model was assumed for the nonleakage probability. The neutron age to thermal was determined from buckling perturbation measurements as 43.1 ± 3.4 cm^2. The critical buckling was measured to be (4344 ± 65) × 10^{-6} cm^{-2}, the bare extrapolation distance 2.7 ± 0.3 cm, and the fast fission factor 1.039 ± 0.004.

Within the experimental error, the values of k_∞ from critical experiments at ORNL and from the PCTR at HAPO agree.

INTRODUCTION

The Physical Constants Testing Reactor (PCTR) in the Hanford Laboratories of the General Electric Company has been used to measure the infinite-medium thermal-neutron multiplication factor, k_∞, of various combinations of fissionable material and moderator (*1*). These experimental determinations of k_∞ require the assumption of a particular neutron spectrum and the error associated with this assumption is not always well defined. Since a large number of the experiments in the PCTR utilized low enriched uranium, it was desirable to compare the value of k_∞ thus determined with that determined by independent measurements. The value of k_∞ of 2.0 wt%-U^{235}-enriched uranium, homogeneously moderated by hydrogen, was measured at the Oak Ridge National Laboratory Critical Facility (*2*) from the steady state critical dimensions and the reactivity resulting from small buckling perturbations applied to a critical system. This material was also used in the PCTR in order to compare the values of k_∞ determined by the two methods.

MATERIALS

The material investigated was a homogeneous mixture of 92.1 wt% UF_4 and 7.9 wt% paraffin. The U^{235} enrichment of the uranium was 2.0 wt%, resulting in an $H:U^{235}$ atomic ratio of 195. Its overall density was 4.5 gm/cc and its uranium density was 3.1 gm/cc. The mixture was formed into blocks 0.25, 0.5, 1.0, 2.0, and 4.0 in. high with base dimensions 1, 2, and 4 in. square. The blocks were covered with 0.0008-in.-thick aluminum foil to facilitate handling. The UF_4 particle size was less than 300 μ, the average being about 150 μ. The materials were free of significant amounts of neutron-absorbing impurities.

The thermal neutron absorber used in the PCTR experiments was principally at 4 wt% boron carbide-impregnated polyethylene sheet, 0.005 in. thick, supplemented by some copper foil. Helium was used as the reference standard in the reactivity measurements since its low neutron scattering and absorp-

* *Present Address:* University of Oregon, Eugene, Oregon.

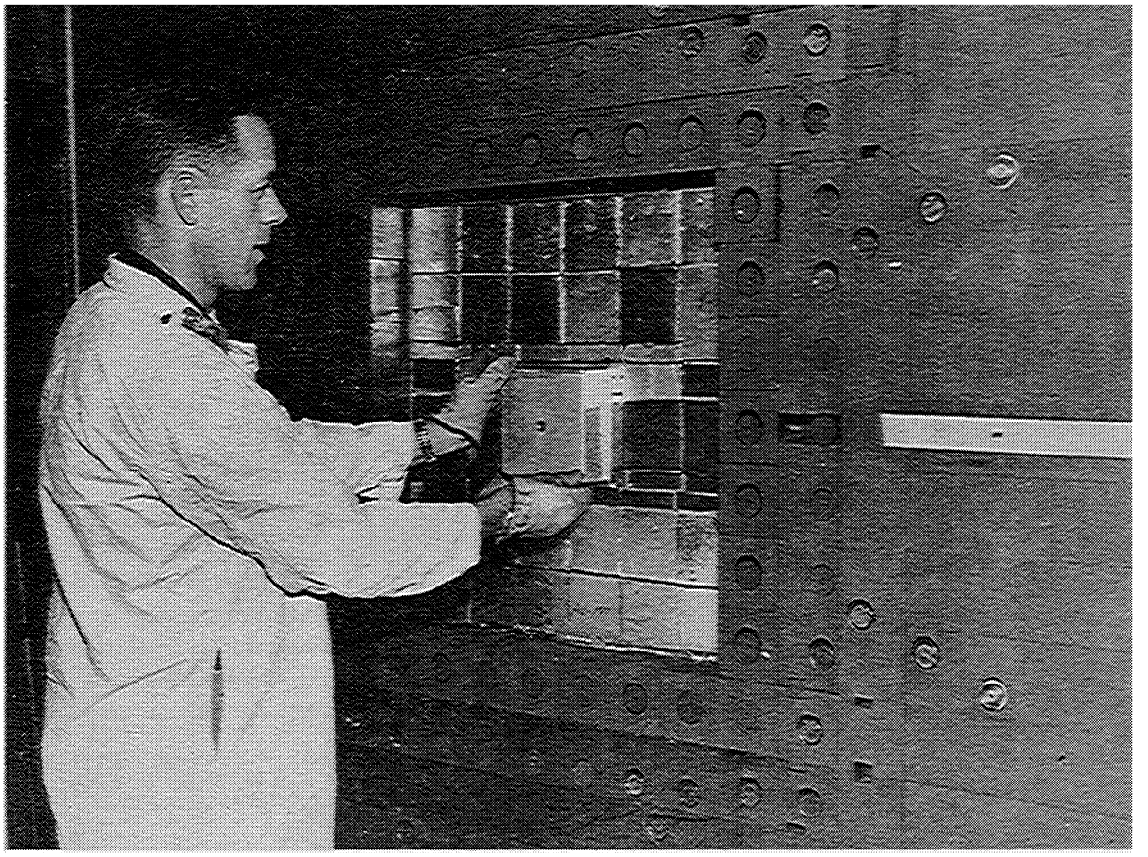

FIG. 1. Test sample and buffer region in the PCTR

tion cross sections make it equivalent to the vacuum specified in the idealized experiment.

EXPERIMENTS IN THE PHYSICAL CONSTANTS TESTING REACTOR

DESCRIPTION OF THE REACTOR

The PCTR has been described by Donahue (*3*) and is a 7-ft cube of graphite with a 2 × 2 × 3 ft central cavity. The reactor is fueled by horizontal rods of U^{235}-enriched uranium metal latticed in the graphite moderator and is controlled by eight rods located in the graphite reflector. A 2-ft long end section of the assembly is mounted on a cart which can be moved a distance of 6 ft, allowing access to the central test region.

In an idealized experiment of the type described in this paper the material under test is modified by the addition of sufficient neutron absorber to allow replacement of a central sample by a vacuum without alteration of the PCTR reactivity. The neutron spectrum at the sample must be that which would exist in an infinite volume of the composite to make the results interpretable. This spectrum is established at the sample by filling the cavity with the poisoned UF_4–paraffin mixture and considering only a small 6 × 6 × 12 in. central volume, contained in a $\frac{1}{8}$-in. thick Type 6061 aluminum can, as the sample. The remainder of the contents of the cavity is called a buffer. Figure 1 is a photograph of the central buffer and sample region.

THEORY

The evaluation of k_∞ of the unpoisoned material from the quantity of absorber necessary to make k_∞ of the composite equal to unity is based on the principle that no perturbation of an infinite critical homogeneous medium will result when any volume of the medium is replaced by a vacuum. This principle results from the fact that the multiplication factors of both the infinite critical medium and the vacuum are unity. The PCTR utilizes this principle for k_∞ measurements in a finite sized reactor.

The infinite medium multiplication factor for the unpoisoned test material is given by

$$k_\infty(P_0) = \epsilon p \eta(P_0) f(P_0) \qquad (1)$$

where P_0 refers to the unpoisoned material and the other terms have their usual meaning (*4*).

TABLE I

CADMIUM RATIO FOR GOLD[a] IN A UF_4–PARAFFIN MIXTURE

System	Cadmium ratio
Center of critical assembly	2.39 ± 0.02
Unpoisoned test sample in PCTR	2.37 ± 0.01
Poisoned test sample in PCTR	2.33 ± 0.01

[a] 0.005-in.thick gold foils, 0.040-in. thick cadmium covers; activity determined by scintillator to count the gamma decay of Au^{198}.

The corresponding expression for the poisoned system is

$$k_\infty(P) = \epsilon p \eta(P) f(P) = 1 \qquad (2)$$

where P refers to the unpoisoned material combined with the associated absorber required for k_∞ to be equal to unity. It is assumed that the addition of thermal neutron absorber does not affect the fast fission factor and the resonance escape probability. Eta is changed slightly by the shift in the thermal energy distribution of the neutrons caused by the introduction of the neutron poison.

By combining Eqs. (1) and (2)

$$k_\infty(P_0) = \frac{\eta(P_0) f(P_0)}{\eta(P) f(P)} \qquad (3)$$

$$= \frac{\Sigma_f(P_0)}{\Sigma_f'(P)} \frac{\Sigma_a'(P)}{\Sigma_a(P_0)} \qquad (4)$$

where Σ_f and Σ_a are the Wigner-Wilkins averaged thermal macroscopic fission and absorption cross sections. The prime designates cross sections averaged over the spectrum of the poisoned material.

The requirement that there be no change in the reactivity of the reactor when the poisoned test sample is inserted into the test region leads to an expression for the error in k_∞ which results from spectral mismatching in the test section (*3*). This error will be zero when the cadmium ratio at the sample–buffer interface does not change upon insertion of the poisoned test sample.

MEASUREMENTS

In an actual experiment, the amount of borated polyethylene sheet between the UF_4–paraffin blocks was adjusted until substitution of the sample for the standard in the test hole produced a very small change in the reactivity of the system and no observable change in the neutron spectrum measured with bare and cadmium covered gold foils within the buffer and sample regions. The exact amount of poison required to match the test sample with the helium standard was then determined from three reactivity measurements. One of these was made with the standard inserted and the other two with the sample containing slightly different amounts of poison. A linear extrapolation from these observations yields the amount of poison necessary to exactly match the sample and standard.

The neutron absorption cross sections of the UF_4–paraffin material used in the analysis were obtained from the microscopic thermal cross sections given in BNL-325 (*5*). The neutron absorption cross section of the borated polyethylene sheet was measured with respect to copper by the danger coefficient method in the PCTR.

ERRORS

An appreciable error can be introduced in the value of k_∞ if the neutron spectrum in the PCTR test region is not the same as that which would exist in an infinite medium of the material under study (*3*). In an attempt to evaluate this error the cadmium ratio in the poisoned test sample was varied from 2.31 to 2.35 by changing the quantity and distribution of the poison in the buffer region. The resulting change in $(k_\infty - 1)$ over this range is less than 0.8%. The cadmium ratios for gold for the poisoned and unpoisoned test sample in the PCTR are compared in Table I with the corresponding quantity measured in the center of a critical assembly.

Although the introduction of the hydrogen and carbon of the borated polyethylene sheet into the test region increased the reactivity of the system, an empirical correction was made by observing the effect of the addition of a small amount of unpoisoned polyethylene sheet. This correction was a 2% decrease in the value of $(k_\infty - 1)$.

An additional uncertainty arises in the method of averaging the thermal neutron cross sections. Averages over a Maxwell-Boltzmann spectrum and over a Wigner-Wilkins (*6*) spectrum yielded values of $(k_\infty - 1)$ differing by 3.8%. The result reported is based on the latter method, with the difference included in the error.

The errors due to experimental technique, errors in masses of materials and errors in 2200 meters/sec cross section values amounted to 0.005 in $(k_\infty - 1)$. The total error in the infinite medium multiplication factor from all sources is ±0.013.

RESULT

The value of k_∞ obtained from the PCTR measurements is 1.216 ± 0.013.

CRITICAL EXPERIMENTS AT ORNL

THEORY

The infinite medium multiplication factor can be determined from changes in the neutron-nonleakage probability of a critical system. The interpretation of the experiments is based on the following relations:

the critical relation

$$k_{\text{eff}} = 1 \tag{5}$$

the static relation

$$k_{\text{eff}} = (1 - \beta)k_\infty K_p(B) + \frac{k_\infty}{\epsilon}\sum_i \beta_i K_{d_i}(B), \tag{6}$$

and the kinetic relation

$$\rho = \frac{l}{T} + \frac{k_\infty}{\epsilon}\sum_i \frac{\beta_i}{1 + \lambda_i T} K_{d_i}(B) \tag{7}$$

where

$K_p(B)$ = nonleakage probability for prompt neutrons from fission,

$K_{d_i}(B)$ = nonleakage probability for the ith group of delayed neutrons.

The other terms in Eqs. (5), (6), and (7) have their usual meaning (*4*).

For $T > 100$ sec the l/T term in Eq. (7) can be ignored. The fast fission factor, ϵ, appears in Eqs. (6) and (7) because the delayed neutrons are emitted at energies below the threshold for U^{238} fission. The assumption has been made that the resonance escape probabilities for prompt and delayed neutrons are the same.

The average delayed neutron nonleakage probabilities for the static and kinetic relations respectively may be defined as

$$K_d^{\,s} = \sum \frac{\beta_i}{\beta} K_{d_i}(B) \tag{8}$$

$$K_d^{\,k} = \sum \frac{\beta_i K_{d_i}(B)}{1 + \lambda_i T} \Big/ \sum \frac{\beta_i}{1 + \lambda_i T} \tag{9}$$

The neutron age for the different delayed neutron groups was calculated assuming that the delayed neutrons of a particular group are monoenergetic. For the stable periods used in the experiments the average delayed neutron nonleakage probabilities determined by use of either Eq. (8) or (9) were less than 0.5% different. The error introduced by the method of averaging is included in the error associated with the delayed neutron age.

Equations (6) and (7) can now be written as

$$k_{\text{eff}} = (1 - \beta)k_\infty K_p(B) + \beta \frac{k_\infty}{\epsilon} K_d(B) \tag{10}$$

$$\rho = \frac{k_\infty}{\epsilon} K_d(B) \sum \frac{\beta_i}{1 + \lambda_i T} = \frac{k_\infty}{\epsilon} K_d(B)\rho'. \tag{11}$$

Expanding Eq. (10) in a Taylor's series to first order in ΔB^2 and noting that $\rho = \Delta k$ for the buckling perturbation experiments results in

$$\frac{\rho'}{\Delta B^2} = \frac{\epsilon(1 - \beta)}{K_d(B)} \frac{\partial K_p(B)}{\partial B^2} + \frac{\beta}{K_d(B)} \frac{\partial K_d(B)}{\partial B^2} \tag{12}$$

Assuming a two group model for the nonleakage probability,

$$K_p(B) = \frac{1}{(1 + L^2B^2)(1 + \tau_p B^2)} \tag{13}$$

There is a similar expression for the delayed neutron nonleakage probability. The reliability of this model depends on the validity of the first and second fundamental theorems of reactor theory (*7*) for the particular assemblies of fissionable material studied here. According to the first fundamental theorem the neutron flux in these unreflected systems is separable in space and energy and the neutron flux has the same spatial distribution for all energies, vanishing on the extrapolated boundary. Although some dependence of the extrapolation distance on energy was observed it is not as important for these assemblies as it would be for smaller ones. By the second theorem the nonleakage probability during moderation in these homogeneous bare reactors is the Fourier transform of the slowing-down kernel. If B^2 be sufficiently small, only the second moment will be important in the neutron nonleakage probability.

When Eq. (10) is written for a critical system

$$k_\infty = \frac{1}{(1 + \beta)K_p(B) + (B/\epsilon)K_d(B)}. \tag{14}$$

Substitution of Eq. (13) in (12) yields an expression for τ_p which can be solved if L^2, τ_d, β, $\rho'/\Delta B^2$, ϵ, and B^2 are known. The thermal diffusion area, L^2, computed from thermal cross sections (*5*), is 1.8 cm^2. The average value of the neutron age to thermal for delayed neutrons, determined from Monte Carlo-moments method calculations on the ORACLE (*8*), is 25.6 ± 2.5 cm^2. The value of β is known from the measurements of Keepin et al. (*9*). The remaining quantities, $\rho'/\Delta B^2$, ϵ, and B^2 are measured.

The neutron age is then used in Eq. (13) to determine the nonleakage probability which is used, in turn, in Eq. (14) to determine the infinite medium multiplication factor.

TABLE II

PARAMETERS OF 2.0 wt%-U^{235}-ENRICHED UF_4 IN PARAFFIN H:U^{235} ATOMIC RATIO OF 195

Assembly base dimensions (cm)	71.5 × 71.5	76.7 × 76.7	81.7 × 86.0
Critical height (cm)	94.1	78.1	66.7
B^2 (cm^{-2})	0.004332	0.004344	0.004355
ϵ	1.0380	1.0388	1.0390
τ_p (cm^2)	43.0 ± 3.4	42.0 ± 3.4	43.7 ± 3.5
k_∞	1.196 ± 0.015	1.194 ± 0.015	1.200 ± 0.017

TABLE III

ERROR IN k_∞ MEASUREMENTS WITH A 71.5 × 71.5 × 94.1 cm ASSEMBLY OF 2.0 wt%-U^{235}-ENRICHED UF_4-PARAFFIN MIXTURE

Parameters	Error (%)	Resulting error in	
		τ_p(cm^2)	k_∞
ρ	±1.1	±0.75	±0.0032
ϵ	±0.4	±0.28	±0.0012
τ_d (cm^2)	±10	±0.71	±0.0029
β	±4.7	±3.26	±0.0141
B^2 (cm^{-2})	±1.5	±0.22	±0.0038
ΔB^2 (cm^{-2})	±0.2	±0.13	±0.0006
L^2 (cm^2)	±5	±0.11	±0.0001
		σ = ±3.45	±0.0153

MEASUREMENTS

The buckling, B^2, is obtained from the critical size of the assemblies and the measured extrapolation distance. The critical size of the assemblies was corrected for the neutron reflection effects of the low-mass support structure by experimentally determining the change in critical height caused by the addition of a mock-up of the supporting table on the top of the assembly.

The extrapolation distance was determined from relative flux measurements through the center of the critical assemblies, using a U^{238} fission chamber 1 in. long and $\frac{1}{4}$ in. in diameter. Relative fission rates, as a function of position, were fitted by the method of least squares to the appropriate eigenfunction. To define a distribution free from the higher modes near the edge of the assembly, data from the counters located within 4 in. of the edge of the system were discarded. The extrapolation distance thus determined was 2.7 ± 0.3 cm.

Determination of $\rho'/\Delta B^2$ was made from the stable periods resulting from buckling perturbations of the assemblies. The periods measured were less than −150 sec and greater than +125 sec. Instrumentation employed during these measurements included two BF_3 proportional counters, a U^{233} fission counter, and a BF_3 chamber whose output was fed to a logarithmic amplifier and chart recorder. The counter data were machine-fitted to an exponential function by the method of least squares. Analysis showed that both the positive and negative periods obtained were the stable periods.

In determining reactivity from the inhour equation the fast fissions in U^{238} were taken into account in the following equation

$$\sum_{i=1}^{6} \frac{\beta_i}{1+\lambda_i T} = \frac{1}{\epsilon'} \sum_{i=1}^{6} \frac{\beta_i^{25}}{1+\lambda_i^{25} T} + \frac{\epsilon'-1}{\epsilon'} \sum_{i=1}^{6} \frac{\beta_i^{28}}{1+\lambda_i^{28} T} \tag{17}$$

where ϵ' is the ratio of the total number of neutrons from both U^{238} and U^{235} fission to the number of neutrons from U^{235} fissions. The ratio of the number of U^{238} fissions to the number of U^{235} fissions in the assembly is measured by placing a U^{238} fission counter and a U^{235} fission counter in flux-symmetric positions about the center of the system. If the fissions in U^{235} caused by neutrons in the million electron volt energy range can be neglected, this ratio, S, is related to the fast fission factor, ϵ, by

$$\epsilon = 1 + \frac{\nu^{28} - (1+\alpha)}{\nu^{25}} S \tag{18}$$

and to the quantity ϵ' of Eq. (17) by

$$\epsilon' = 1 + \frac{\nu^{28}}{\nu^{25}} S. \tag{19}$$

In these equations α is the average neutron capture to fission ratio in the energy region where U^{238} fission occurs and ν^{28} and ν^{25} are the average number of neutrons per fission. Since the counters were alike except for the uranium isotope, it was assumed that their efficiencies were the same. If C is defined as the ratio of the count rate for the U^{238} fission counter to the count rate for the U^{235} fission counter, then

$$S = \frac{N_R^{28}}{N_R^{25}} \frac{N_E^{25}}{N_E^{28}} \left[\frac{C - (N_D^{25}/N_E^{25})}{(N_D^{28}/N_E^{28}) - C} \right] \tag{20}$$

where N is the atomic density of the fissionable isotope and the subscripts R, E, and D refer to the assembly, the U^{235} fission counter, and the U^{238} counter, respectively.

RESULTS

Three critical assemblies were constructed and the results of the analysis of these experiments are

shown in Table II. The value of k_∞ determined in critical experiments using a two group model for the nonleakage probability is 1.197 ± 0.015 and is to be compared to 1.216 ± 0.013 determined from the PCTR measurements. The effective neutron age to thermal determined from buckling perturbation measurements is 43.1 ± 3.4 cm^2. The buckling was measured to be $(4344 \pm 65) \times 10^{-6}$ cm^{-2}, the extrapolation distance, 2.7 ± 0.3 cm, and the fast fission factor, 1.039 ± 0.004.

Errors

The error analysis is based on the model assumed for the nonleakage probability. The largest source of error in these experiments arises from the uncertainty in the delayed neutron fraction, 0.0064 ± 0.0003. This uncertainty introduces an error of $\pm 4.7\%$ in the absolute reactivity associated with a given perturbation on the system and results in an error in k_∞ of ± 0.014. The standard deviation in the measured reactivity associated with a perturbation in height was between 1.0 and 1.9% depending on the experiment.

The contributions to the final error from different sources are shown in Table III for the assembly with 71.5×71.5 cm base dimensions.

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