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SUBJECT: Determination of $\bar{\eta}$ by Comparison of $\overline{\eta\sigma}_a$ for U^{233} and Pu^{239} with $\overline{\eta\sigma}_a$ for U^{235} in a Flux Trap Critical Assembly

TO: Distribution

FROM: R. Gwin and D. W. Magnuson

Abstract

The values of $\bar{\eta}$ for U^{233} and $\bar{\eta}$ for Pu^{239} have been determined by a reactivity coefficient measurement. An aqueous solution of the isotope was introduced axially into a critical cylindrical annular flux trap reactor, and the resulting reactivity change measured by period determinations. From these data the ratio $\overline{\eta\sigma}_{ax}/\overline{\eta\sigma}_{a(U^{235})}$ was obtained. Using recently measured values of $\sigma_{a(U^{235})}$ and $\bar{\eta}$ for U^{235} in this ratio gives the thermal value of 2.308 ± 0.040 for $\bar{\eta}$ of U^{233} and 1.995 ± 0.053 for $\bar{\eta}$ of Pu^{239}. Correction to a neutron velocity of 2200 m/sec by using the appropriate g-factor gives a value of 2.308 ± 0.040 for η of U^{233} and 2.044 ± 0.054 for η of Pu^{239}.

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INTRODUCTION

The measurement of the value of η, the number of neutrons produced in a fissionable isotope per neutron absorbed, has been a continuing interest with investigators at the ORNL Critical Facility, both because of the primary importance of this value in any program of breeding of fissionable material and because of the proper desire to improve the precision to which the important physical constants are known. The employment of varied and distinct experimental techniques provides useful and independent verifications of accepted values.

The present paper reports a measurement of the ratio of the product $\overline{\eta\sigma_a}$ for U^{233} and Pu^{239} to $\overline{\eta\sigma_a}$ for U^{235} by means of the familiar reactivity coefficient technique. From this ratio, in conjunction with a recently measured value of $\bar{\eta}$ for U^{235} plus the most recent values of the absorption cross sections, values of $\bar{\eta}$ for thermal energies have been derived for both U^{233} and Pu^{239}.

EXPERIMENTAL PROCEDURE

A drawing of the experimental arrangement is shown in Fig. 1. The assembly consisted essentially of a 15-cm-dia water column located coaxially inside a critical cylindrical (38.1-in.-OD) annulus of uranyl fluoride solution enriched to 93.2% in the U^{235} isotope. Samples of aqueous solutions of either fissionable or neutron-absorbing materials, in various dilutions, were inserted axially into the water column after a stable critical system was achieved, and the resulting reactivity changes measured by means of period determinations. Samples were contained within a 136-ml

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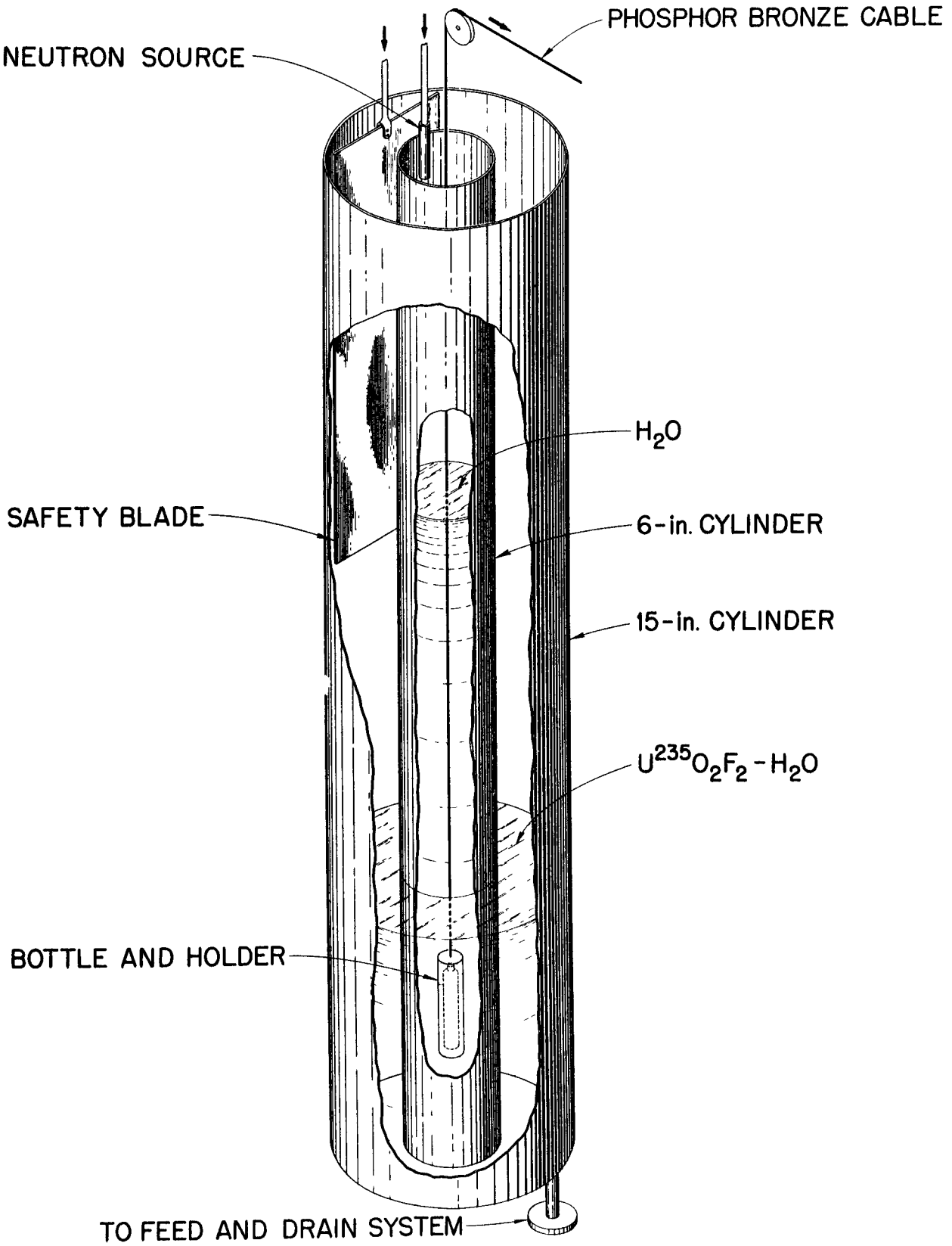


FIGURE 1

polyethylene bottle in a Plexiglas holder. A gravity-actuated cadmium blade served as a safety mechanism. The arrangement possessed two important factors: (1) the neutron flux in the sample region was essentially Maxwellian and was experimentally determined to be constant over the sample region, and (2) the sensitivity of the system was sufficiently high to enable use of samples which were nearly infinitely dilute. (The maximum density of the fissile isotope was 3.2 g/liter in a 136-ml sample.)

A typical series of experiments consisted in first measuring the reactivity of the sample holder filled with a weighed amount of water. (The central water column extended above the level of the critical solution in order to avoid reactivity effects due to changes in the water height upon placing the sample in the system.) A known quantity of the isotope under investigation was then added to the sample bottle and the reactivity again measured. The procedure was repeated for several concentrations of the isotope in order to confirm the expected linearity of response with concentration.

Samples included U^{233}, U^{235}, Pu^{239}, "standard" boron, lithium and indium. The U^{235} measurements were used in a comparison technique to determine the importance of the fission neutrons, while boron served as the primary standard in measuring the reactivity constant of proportionality for absorption. The use of the lithium and indium solutions is discussed later in this report.

The lower practical limit of measurable reactivity change at critical was about 10^{-6}. Smaller increments were experimentally observed, but their determination was made obscure by instabilities possibly due to evaporation, temperature gradients, or bubble formation on the surfaces. The observed

reactivities were shown to be insensitive to changes in sample position of the order of the uncertainty in the determination of sample position. Reproducibility of the reactivity measurements was about ±1% of the average.

A BF_3 ionization chamber, in conjunction with a logarithmic amplifier, was the basic device for stable period determination, supported by independent measurements of periods with timed pulse counters.

ANALYSIS OF RESULTS

The reactivity change in these experiments is a function of the number and energies of the neutrons absorbed and produced in the sample. Since the majority of the neutrons are absorbed at thermal energies and those produced are at fission spectrum energies, the relative importances of the neutrons absorbed and produced are different. With the assumption of the neutron distribution over the sample region as a Maxwellian at the system temperature, which was 24 to 25°C, joined discontinuously to a 1/E spectrum at 0.2 ev, an expression for the reactivity may be written as:

$$\rho = C_1 \int_0^\infty \eta(E)\Sigma_{af}(E)\,\phi(E)\,dE - C_2 \int_0^\infty \Sigma_{at}(E)\,\phi(E)\,dE \qquad (1)$$

where

ρ = reactivity (cents),

C_1 = measure of the importance of fission neutrons (cents/n),

C_2 = measure of the importance of thermal neutrons (cents/n),

$\eta(E)$ = the average number of neutrons produced per neutron of energy E absorbed,

$\Sigma_{af}(E)$ = macroscopic absorption cross section of the fissile isotope,

$\Sigma_{at}(E)$ = total macroscopic absorption cross section,

$\phi(E)$ = neutron flux at energy E,

$\phi(E) = \phi_M$ = Maxwellian flux for $0 \leq E \leq 0.2$ ev,

$\phi(E) = \lambda/E + \phi_M$ for $0.2 \text{ ev} \leq E \leq 10^6$ ev,

λ = proportionality constant relating the Maxwellian spectrum to the epithermal 1/E spectrum.

ϕ_M is normalized so that $\int_0^\infty \phi(M)\, dE = 1$.

The constant C_1 is a function of the energy spectrum of the fission neutrons, and in principal could be different for each fissile isotope. However, critical experiments[1] have shown that the effective ages of fission neutrons from U^{233} and U^{235} are essentially the same, and thus the fission spectra of all the fissile isotopes were assumed to be the same.

The constant C_2 is used for the epithermal absorptions as well since most of these absorptions are at energies near thermal. No appreciable error is introduced by this assumption.

The joining of the Maxwellian to the 1/E flux has been investigated by performing numerical integrations over the joining region, assuming the flux dependence on energy measured by Poole.[2] The results show that the total number of absorptions over this joining region (0.15 ev to 0.42 ev) is underestimated when a constant value is used for λ, but that the resulting effect on the thermal value of $\overline{\eta\sigma_a}$ is less than 0.1%. λ was experimentally determined by bare and cadmium-covered gold foil and U^{235} foil measurements, as well as by comparing the reactivity effects of a lithium solution and an indium solution.

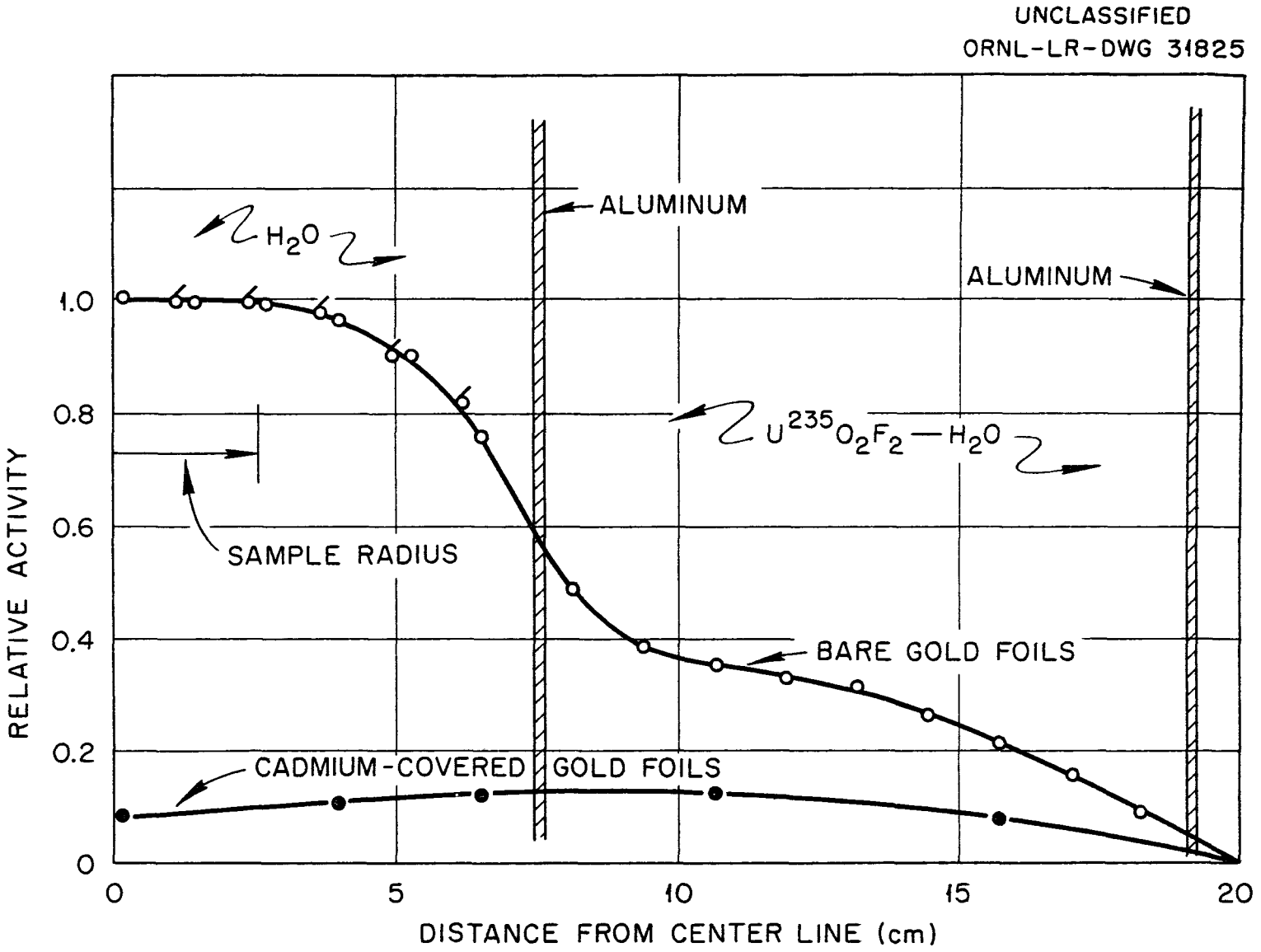
1. R. Gwin and D. W. Magnuson, Critical Experiments for Reactor Physics Studies, CF-60-4-12 (1960).
2. M. J. Poole, A Measurement of the Neutron Spectra in Moderators and Reactor Lattices, J. Nuclear Energy 5, 325 (1957).

RESULTS AND DISCUSSION

The neutron flux within the reactor, measured along a radius with bare and cadmium-covered gold foils, is shown in Fig. 2 and that measured with bare and cadmium-covered U^{235} foils in Fig. 3. All foils were 2 mils thick. The essentially flat flux throughout the sample region is clearly evident in both plots.

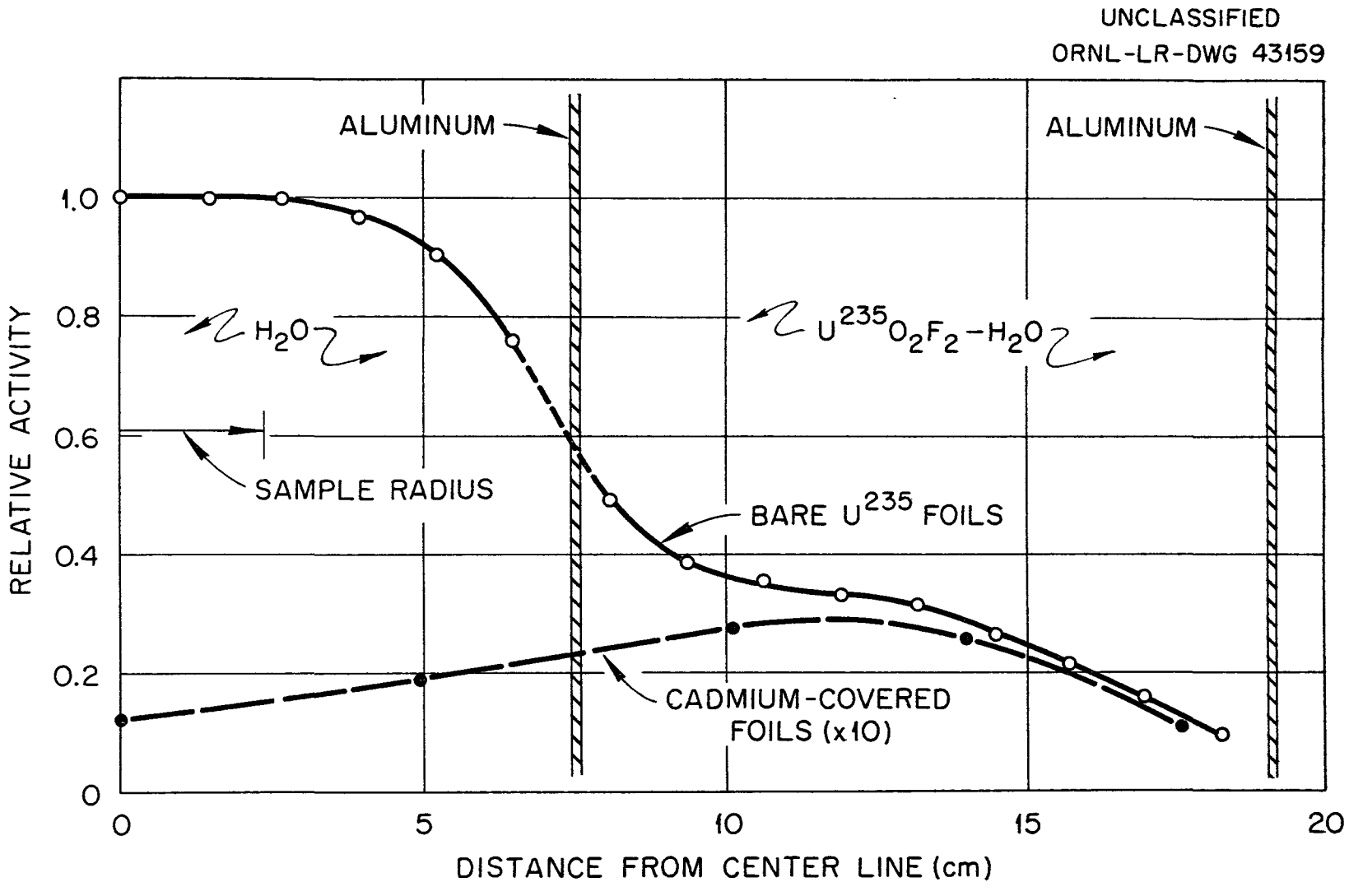
The gold and uranium foil data were also used, as previously noted, to obtain an experimental value of λ, the proportionality constant relating the Maxwellian and epithermal 1/E spectra. In this measurement, however, it was necessary to take into account the decrease in apparent foil activity caused by self-shielding within the foil. In some subsequent experiments[1] made with bare U^{235} foils of varied thicknesses in a system having a U^{235} concentration of $\sim$14.1 g/liter it was observed that the activities induced (primarily by thermal neutrons) in 2-mil-thick foils were decreased $\sim$25% by the effect of their finite thickness. A calculation using the method of Bothe and Tittle[3,4] showed that for a 2-mil-thick foil, the foil activation per unit mass is decreased, because of self-shielding, by 22% in an isotropic distribution of neutrons. Trubey[5] has calculated that the decrease in activation by 1/E resonance neutrons due to self-shielding would amount to $\sim$ 6%. A similar correction was applied to the observed activities of

3. W. Bothe, The Use of Neutron Detectors, CP-G-2964 (1945) from Z. Physik 120, 437 (1943).
4. C. W. Tittle, Nucleonics 9 (1) 60 (1951).
5. D. K. Trubey, private communication, July 30, 1959.



Gold Foil Activations in Annular Critical Experiment

FIGURE 2



U^{235} Foil Activations in Annular Critical Experiment

FIGURE 3

gold foils due to thermal neutrons. The effective epithermal resonance integral of the gold foils was calculated by the narrow resonance relation quoted by Dresner.[6]

From the ratio of the foil activations, written as

$$\frac{A_{Cd}}{A_{bare}} = \frac{\lambda \int_{0.4\ ev}^{10^6} \frac{\Sigma_{act}(E)}{E}\, dE}{\int_{0}^{\infty} \phi_M \Sigma_{act}(E)\, dE + \lambda \int_{0.2\ ev}^{10^6} \frac{\Sigma_{act}(E)}{E}} \qquad (2)$$

and the absorption cross sections and resonance integrals shown in Table 1, the value of λ, using U^{235} foil measurements, was determined as 0.015 ± 0.002; using gold foil measurements, 0.014 ± 0.002. The uncertainty resulted from estimates of the flux depression and the resonance integrals. Both values are in excellent agreement with those measured in water by Poole,[2] who gives $\lambda = 0.016 \pm 0.001$.

An independent series of experiments permitted an evaluation of λ from the reactivity effect as a function of concentration of solutions of indium and lithium introduced in the sample position of the critical assembly. Since the solutions were of sufficient concentration to cause significant self shielding, the relationships are not linear, and it was necessary to select for use in Eq. 1 concentrations of the two elements which produced equal reactivity changes, in order to correctly evaluate λ. The resonance integral for the sample in the geometry of the experiment was calculated by Dresner. The value obtained for λ by this method was 0.018.

6. L. Dresner, Resonance Absorption for Neutrons in Nuclear Reactors, ORNL-2659 (1959).

The value of λ obtained from the U^{235} foil activations was chosen for the analysis of the experimental data, since, if the correction for finite foil thickness is properly made, the source term for epithermal U^{235} fissions is reproduced even if the resonance integral is incorrect. A separate experiment,[1] however, demonstrated that this value of λ, together with the appropriate resonance integral, was equally applicable to the interpretation of the reactivity changes due to either the U^{235} or U^{233} samples. Thin samples of the oxides of both U^{233} and U^{235} were exposed in the same spectrum. The value of λ determined by using the resonance integrals given in Table 1 was found to be the same for either isotope.

A somewhat arbitrary error of $\pm 10\%$, based on foil data, has been placed on the epithermal portion of the source term. Since the ratio of the fission cross section to the absorption cross section is not independent of energy, the products of λ and the absorption integrals were assigned an uncertainty of 25%.

Table 1 gives a list of the experimental and calculated values used in the analysis, while Table 2 summarizes the results. The values of $\bar{\eta}$ given in Table 2 are for the Maxwellian average thermal spectrum. Since $(1 + \alpha)$ for U^{233} and U^{235} are essentially constant over the Maxwellian, η values for 2200 m/sec neutrons are the same as the thermal values. Application of the Hughes[7] g-factor for Pu^{239}, $g_\eta = 0.976$, gives a value of 2.044 ± 0.054 for Pu^{239} and 2200 m/sec neutrons.

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7. D. J. Hughes, Progress in Nuclear Energy, Ser. I. 3, 1 (1959).

Table 1. Data Used in the Analyses

Sample	Mass (g)	Reactivity[a] (cents/atom)	σ_a^{2200}[b] (barns)	g_a[c]	Σ_{at}[d] (cm^{-1})	Resonance Integrals[e] $\int_{0.2\,ev}^{10^6} \sigma_f(E)\frac{dE}{E}$ (barns)	$\int_{0.2\,ev}^{10^6} \sigma_a(E)\frac{dE}{E}$ (barns)	$\int_{0.4\,ev}^{10^6} \sigma_f(E)\frac{dE}{E}$ (barns)
		($\times 10^{-20}$)						
U^{235}	0.3726	1.0239	682 ± 3	0.9749	0.00466	428	636	330
U^{233}	0.4275	1.0291	577 ± 4	1.0000	0.00475	930	1224	802
Pu^{239}	0.2075	1.6762	1030 ± 10	1.0734	0.00427	1650	2455	
Boron	0.120	- 0.3189	755 ± 2	1.0	0.00649			
Boron	0.201	- 0.3190			0.01088			
Boron	0.300	- 0.3039			0.01620			
Boron	0.307	- 0.2912			0.01658			
Lithium	0.1031	- 0.0305[f]	71 ± 1	1.0	0.00470			
Lithium	0.1342	- 0.02995[f]			0.00610			
Lithium	0.2684	- 0.02834[f]			0.01220			
Lithium	0.4026	- 0.02683[f]			0.01830			
Lithium	0.8050	- 0.02322[f]			0.03670			
Indium	0.509	- 0.11290	196 ± 5	1.017	0.00406		2828	
Indium	1.018	- 0.10461			0.00814		2438	
Indium	1.527	- 0.09911			0.01220		2208	
Indium	2.545	- 0.09343			0.02033			
Gold			98.8 ± 0.3	1.005			481[d]	

a. The reactivities were calculated using the delayed neutron data of Keepin, Wimett and Zeigler, Phys. Rev. 107, 1044 (1957).
b. Cross section values from BNL-325, 2d Ed., and BNL-325, 2d Ed., Supplement 1, Jan. 1, 1960.
c. The g-factors are from C. H. Westcott and D. A. Roy, Supplement to Effective Cross Section Values for Well-Moderated Thermal Reactor Spectra, CRRP-862, Aug. 20, 1959, except for U^{233}. This g-factor must be unity, because the new cross-section data (footnote b) are clearly 1/v over the energy range of the Maxwellian.
d. The macroscopic cross section, Σ_{at}, includes impurities in the sample.
e. The resonance integrals were obtained from numerical integration of BNL-325, 2d Ed., cross sections, calculated by the ORACLE. (Personal communication, W. E. Kinney). L. Dresner calculated the geometric and self-shielding effects in the indium and gold samples.
f. The best-fit equation to all the data on lithium was used to calculate these values. This relation was: (cents - 4.76) = $-27.43\,g + 9.44\,g^2$.

The value of η for U^{233} is in good agreement with the recent results of Macklin and deSaussure,[8] who give η for U^{233} = 2.296 ± 0.010, while the value of η for Pu^{239} is in fair agreement with Hughes,[7] who gives η = 2.10 ± 0.03.

Table 2. Summary of Results and Thermal Cross Sections

Isotope	$\bar{\sigma}_{ax}$(barns)	$\frac{\bar{\eta}_x \bar{\sigma}_{ax}}{\bar{\eta}\bar{\sigma}_{U^{235}}}$	$\frac{\bar{\eta}_x}{\bar{\eta}_{U^{235}}}$	$\bar{\eta}_x$
U^{235}	589.2 ± 3			(2.074 ± 0.015)[a]
U^{233}	511.4 ± 4	0.966 ± 0.014	1.113 ± 0.018	2.308 ± 0.040
Pu^{239}	979.8 ± 10	1.60 ± 0.023	0.962 ± 0.025	1.995 ± 0.053

a. R. W. Gwin and D. W. Magnuson, Critical Experiments for Reactor Physics Studies, CF-60-4-12 (1960).

8. R. L. Macklin, G. deSaussure, J. D. Kington and W. S. Lyon, Manganese Bath Measurements of η of U^{233} and U^{235}, ORNL-CF-60-2-84 (1960).

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