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A Direct Comparison of Some Nuclear Properties of U-233 and U-235

J. T. THOMAS, J. K. FOX, AND DIXON CALLIHAN

Oak Ridge National Laboratory, Oak Ridge, Tennessee

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Nuclear properties of U^{233} and U^{235} are compared using data obtained in a series of critical experiments. Aqueous solutions of uranyl oxyfluoride containing uranium enriched to about 90% in each of the two isotopes have been made critical in water-reflected spherical reactors having diameters of 26.4 and 32.0 cm. Assuming the reported nuclear constants for U^{235} are reliably known and assuming equality of the neutron leakage spectra of U^{233} and U^{235} for the same water-reflected critical sphere, the value of $\eta(U^{233})$ at 0.026 ev was determined to be 2.31 ± 0.03. The critical masses for the two isotopes in these systems have been measured over the temperature range from 20°C to 100°C; corresponding values of the reactivity temperature coefficient are reported. Delayed neutron yields for the two isotopes were compared by noting the periods resulting from the withdrawal of a boron poison from the critical spheres. It is shown that the yield from U^{233} is about one-third that from U^{235}, in agreement with other determinations.

INTRODUCTION

Microgram quantities of the U^{233} isotope were available as early as 1944 when its nuclear properties were investigated at the Argonne and Los Alamos Laboratories (*1–5*). By 1948 measurements of the absorption and fission cross sections were made at Chalk River (*6*, *7*) and an extensive experiment was performed using "swing" measurements (*8*) in the low-power heavy water pile (ZEEP) to obtain a value of η, the number of neutrons produced per thermal neutron absorbed by U^{233}. The results were in agreement with those obtained earlier by Zinn at Argonne using the same method. Since these experiments yielded the product ηp, the result depended on the accuracy of the resonance escape probability for the particular reactor. "Swing" measurements produced, however, the best value of η up to that time. An accurate value of η for U^{233} is required for solution of power and breeding problems of the reactor development program.

The variation of $\eta(U^{233})$ over the limited energy range from 0.026 to 0.032 ev has been determined by a series of comparative critical experiments using U^{235},

the nuclear constants of which are well known, and U^{233}, the nuclear constants of which are uncertain, in identical geometry. Two different diameter spherical reactor vessels, with water reflectors, were used. These were made critical at various temperatures over the range from 20° to 100°C. Criticality was also attained with the larger reactor unreflected at room temperature.

Positive period measurements were obtained in both U^{233} and U^{235} by the displacement of a small diameter boron rod, thus enabling a comparison of their delayed neutron yields.

This work was done at the critical experiments facility of ORNL during the period from August 1953 to February 1954.

EXPERIMENTAL MATERIALS

Solutions

Aqueous solutions of uranium oxyfluoride containing predominately the U^{233} and U^{235} isotopes were used in these experiments. The isotopic concentrations were 98.7% U^{233} and about 90% U^{235}. Spectrographic analysis of the solutions and reflector water were made during the experiments and no significant buildup of impurities was observed.

The volume coefficient of thermal expansion for uranium oxyfluoride solutions in the specific gravity range from 1.15 to 1.57 was measured with a pycnometer. The results are presented in Fig. 1, where the ratio of the volume at temperature, t, to the volume at 23°C is plotted as a function of the temperature.

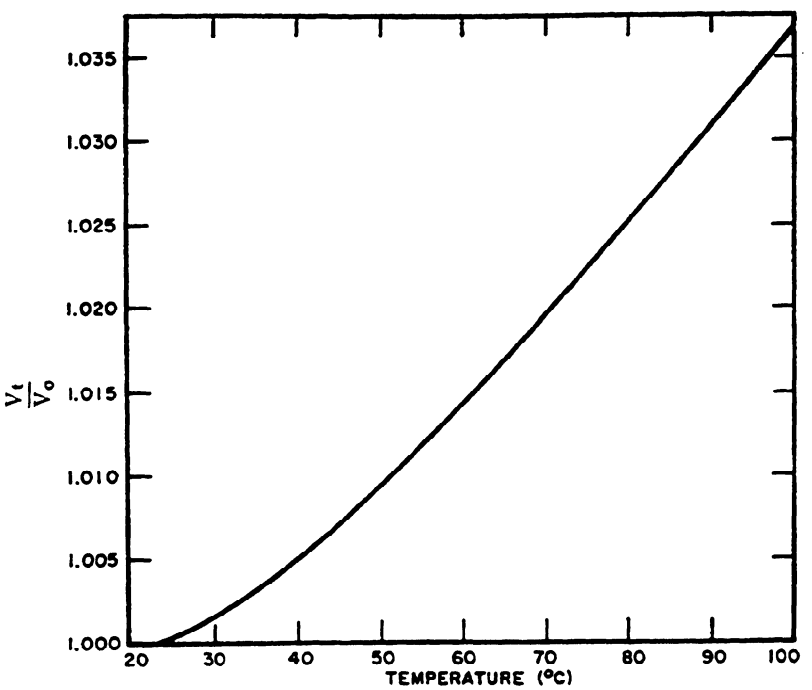

FIG. 1. Volume ratio of UO_2F_2 solutions vs temperature.

Reactors

Two spherical reactors having diameters of 26.4 cm and 32.0 cm were fabricated of 3S aluminum 1.27 mm thick. (These diameters are nominal in the sense that they are determined from the calibrated volumes.) To provide 15 cm of water as an effectively infinite neutron reflector, the spheres were mounted in cylinders of appropriate dimensions. The inside surfaces of the spheres were coated with a plastic to prevent excessive corrosion.

Water was used to measure the capacity of the reactors as a function of liquid depth in the temperature range from 20° to 100°C. At 26°C, the volume of the 26.4-cm sphere was 9.660 ± 0.005 liters and that of the 32.0 cm sphere was 17.020 ± 0.005 liters.

Apparatus

The solutions were stored in a manifold of suitable dimensions which was connected to the reactor by flexible tubing and which could be moved vertically by a signal from a remote control point. In this manner the solution could be added to the reactor in increments corresponding to changes in liquid height of the order of 0.2 mm. The volume of solution present in the sphere was determined by a power driven probe which closed a signal circuit when it made

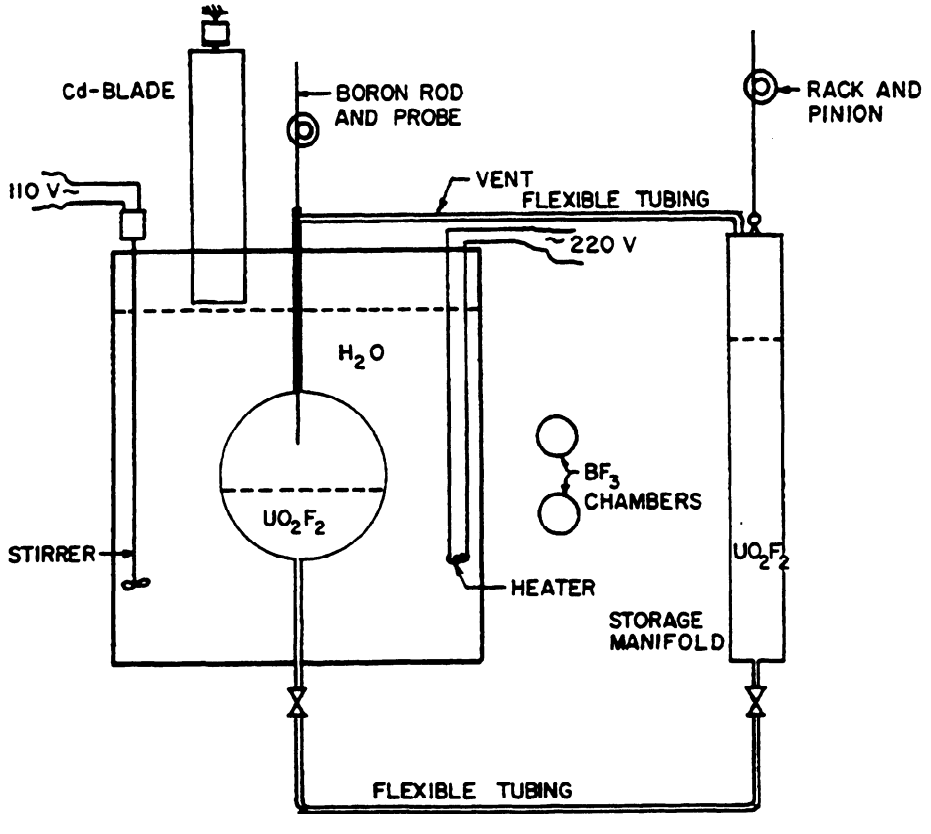

Fig. 2. Schematic diagram of apparatus.

contact with the liquid surface. The positions of the manifold and probe were indicated by selsyns which could be read to the nearest 0.2 mm.

Immersion heaters, placed along the periphery of the water reflector were used to effect the temperature change. The temperature was maintained by a thermoregulator to within ±0.5°C, while a stirrer assured a uniform temperature distribution.

Safety and control were provided by fine adjustment of the elevation of the manifold, by a boron rod which could be immersed in the oxyfluoride solution, and by a sheet of cadmium which could be inserted between the sphere and the reflector water.

Three boron lined proportional counters located under the reactor assembly were used to monitor the neutron density changes during the approach to critical and to provide data for reciprocal multiplication curves when needed. Signals from BF_3 chambers were supplied to a pile power, pile period, and three linear recorders which made a history of the experiment and indicated when criticality was achieved. A signal to a linear recorder from an argon chamber indicated the gamma radiation level present in the reactor room. Scram signals could have originated from any one of the three BF_3 chambers if the neutron level exceeded a preset value. Figure 2 is a schematic diagram showing the relative location of the apparatus.

METHOD AND RESULTS

The experiments consisted, essentially, of determining the chemical concentrations of the solutions of uranium which were required to make the spherical volumes critical. From these results a comparison can be made of the values of η for the two isotopes. The critical masses of U^{233} and U^{235} in the two spheres as a function of temperature can also be obtained from the concentrations. A brief description of the experimental method and a summary and analysis of the data are presented.

Reflected Reactor Experiments

Experimental Procedure and Data

In a typical experiment the concentration of the solution was adjusted to have the sphere critical when filled and at a particular temperature. The concentration was then increased and the temperature raised to make the sphere again critical when filled. Immediately after the reactor had been critical, duplicate samples of the oxyfluoride solutions were taken. In most cases the analyses of these samples differed by no more than 0.5%.

Table I summarizes the data of the reflected sphere experiments. The atomic ratios, H/U^{233} and H/U^{235}, as functions of temperature for both spheres are

TABLE I

CRITICAL CONDITIONS FOR WATER REFLECTED SPHERICAL ALUMINUM VESSELS

Fuel concentration		Temperature (°C)	Volume (L)	Mass of X (gm)
gm X[a]/L (at 25°C)	H/X			
		U^{233} Oxyfluoride Solutions		
26.4-cm diameter sphere				
61.34	418.3	32.0[b]	9.666	591.0
61.89	414.6	39.5	9.675	596.3
64.05	400.5	65.5	9.704	611.0
65.84	389.6	83.2	9.723	623.0
67.80	378.1	96.5	9.737	638.0
32.0-cm diameter sphere[c]				
38.75	663.1	26.3	17.020	659.0
39.97	643.1	56.0	17.042	673.0
42.65	602.8	99.5	17.074	703.0
		U^{235} Oxyfluoride Solutions		
26.4-cm diameter sphere				
95.14	268.8	27.5	9.661	918.3
97.10	263.3	39.5	9.675	935.3
104.09	245.4	74.0	9.713	989.8
106.75	239.3	85.5	9.726	1010.1
32.0-cm diameter sphere[c]				
50.29	515.1	27.0[b]	17.020	855.8
51.50[d]	502.6	43.0	17.032	872.0
51.44[d]	503.4	43.0	17.032	870.8
52.13	496.5	54.0	17.042	879.2
53.07	487.6	64.5	17.049	890.3
54.27[d]	476.8	87.5	17.065	899.6
56.28[d]	459.6	87.5	17.065	932.0

[a] X = U^{233} or U^{235}

[b] In these experiments the vessel was critical with the order of 0.1% of its capacity unfilled and at a temperature 5°C lower than this value. Figure 1 and the capacity of the sphere calibrated over the temperature range were used to obtain this temperature at which the sphere would have been full and critical.

[c] The values of the concentrations and masses measured with this sphere are believed to be about 2% high because of a systematic error.

[d] Duplicate samples.

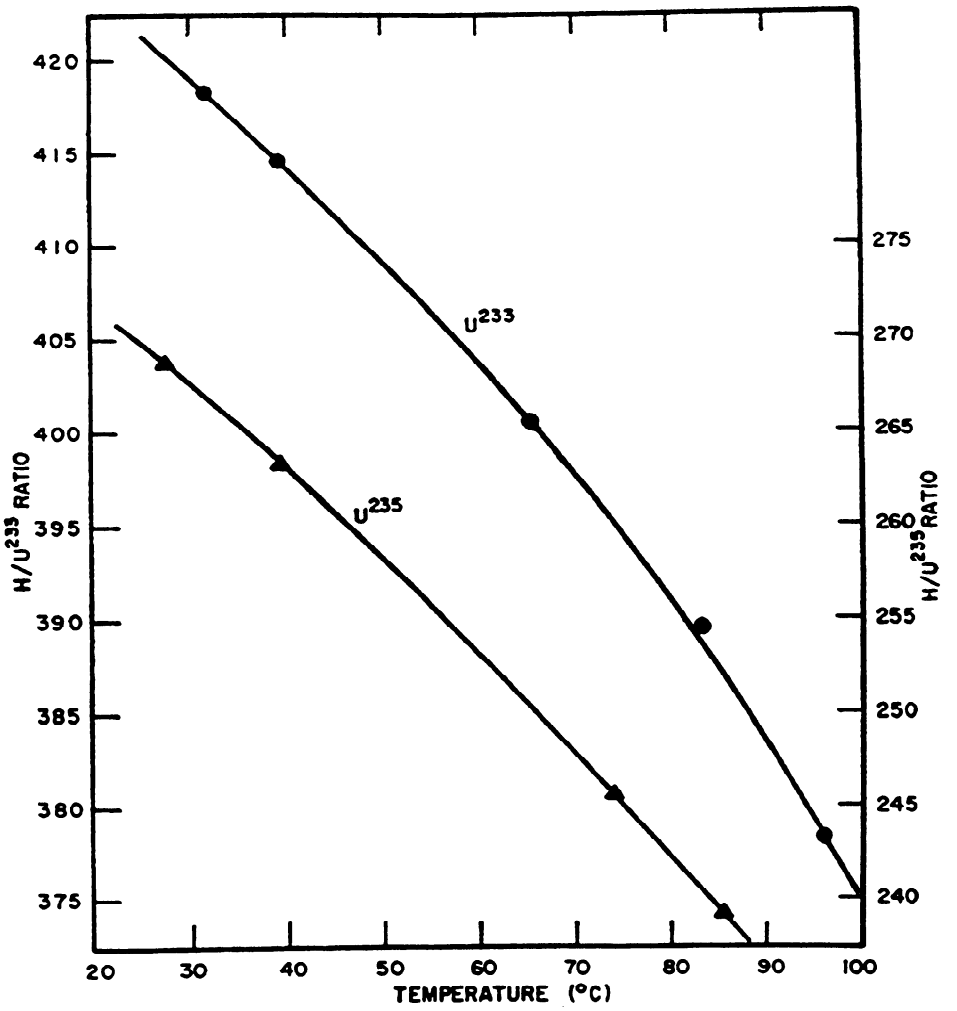

FIG. 3. Ratio of H:U^{233} and H:U^{235} in 26.4 cm diameter water-reflected spherical reactor vs temperature.

presented in Figs. 3 and 4. Figures 5 and 6 illustrate the variation of critical mass with temperature for the two isotopes.

Theory and Nuclear Constants

Accepting the hypotheses that the neutron leakage from a critical spherical reactor is the same for both U^{233} and U^{235} oxyfluoride solutions, and that the two-group critical equation,

$$k_{\text{eff}} = \frac{\eta f}{(1 + \tau B^2)(1 + L^2 B^2)} \tag{1}$$

is applicable, then for a critical reactor at a temperature, t_0, the following condition subsists

$$k_{\text{eff}}]_{U^{233}} = k_{\text{eff}}]_{U^{235}} \tag{2}$$

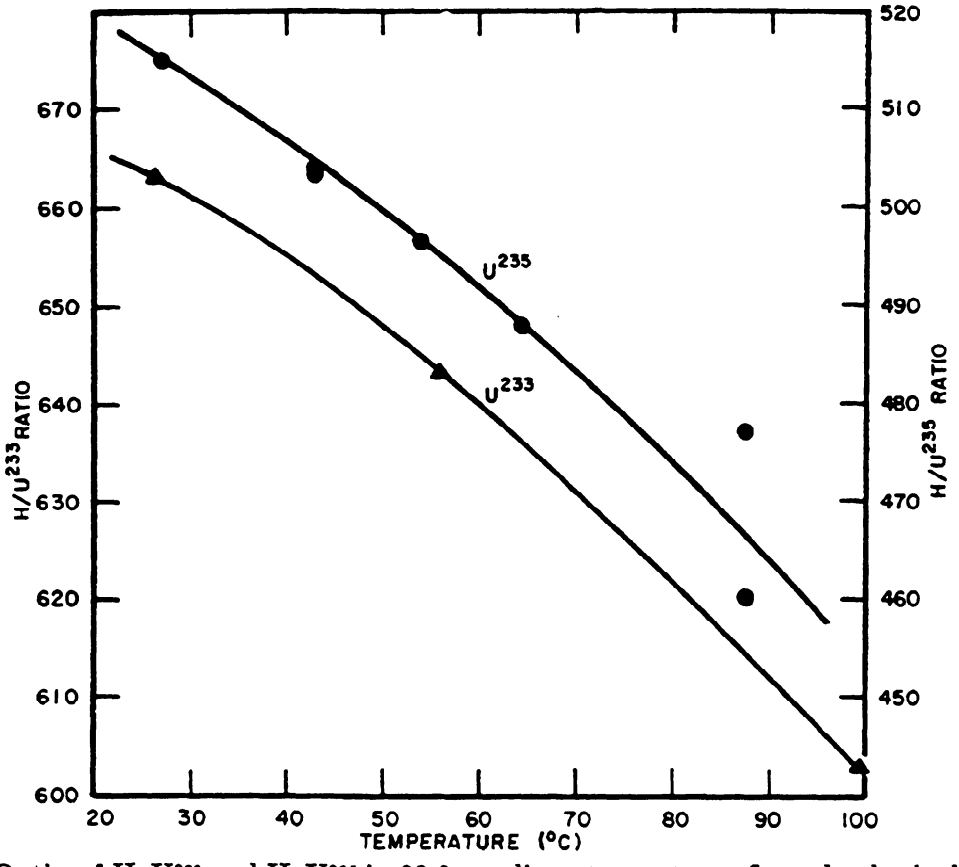

FIG. 4. Ratio of H:U^{233} and H:U^{235} in 32.0 cm diameter water-reflected spherical reactor vs temperature.

where the symbols have their usual meanings. In this paper the subscripts 3 and 5 refer to U^{233} and U^{235}, respectively. Applying the definitions of thermal utilization for both the fuel and the moderator to equations (1) and (2) one obtains

$$\frac{\eta_3}{\eta_5} = \frac{\sigma_a(5)[R_3\,\sigma_a(\mathrm{H})(1 + L_0^2B^2) + \sigma_a(3)]}{\sigma_a(3)[R_5\,\sigma_a(\mathrm{H})(1 + L_0^2B^2) + \sigma_a(5)]} \tag{3}$$

where L_0 is the thermal diffusion length in water; R_3 and R_5 are the solution concentrations expressed as the atomic ratios H/U^{233} and H/U^{235}, respectively; $\sigma_a(H)$, $\sigma_a(3)$, and $\sigma_a(5)$ are the absorption cross sections of hydrogen, U^{233}, and U^{235}. Knowing the cross sections, the value of η_5, and the critical concentrations, equation (3) yields the value of η for U^{233}.

The atomic ratios H/U^{233} and H/U^{235}, corresponding to the critical concentrations at various temperatures were taken from Figs. 3 and 4 and are listed in Table II. The cross section data (*9*) for hydrogen, U^{233}, and U^{235} are presented in Table III. The non-$1/v$ correction has been applied to the U^{235} cross sections for a Maxwell distribution. The scattering cross section has been assumed constant and equal to 10 barns over this energy range for both U^{233} and U^{235}. The value used for η-235 is 2.08.

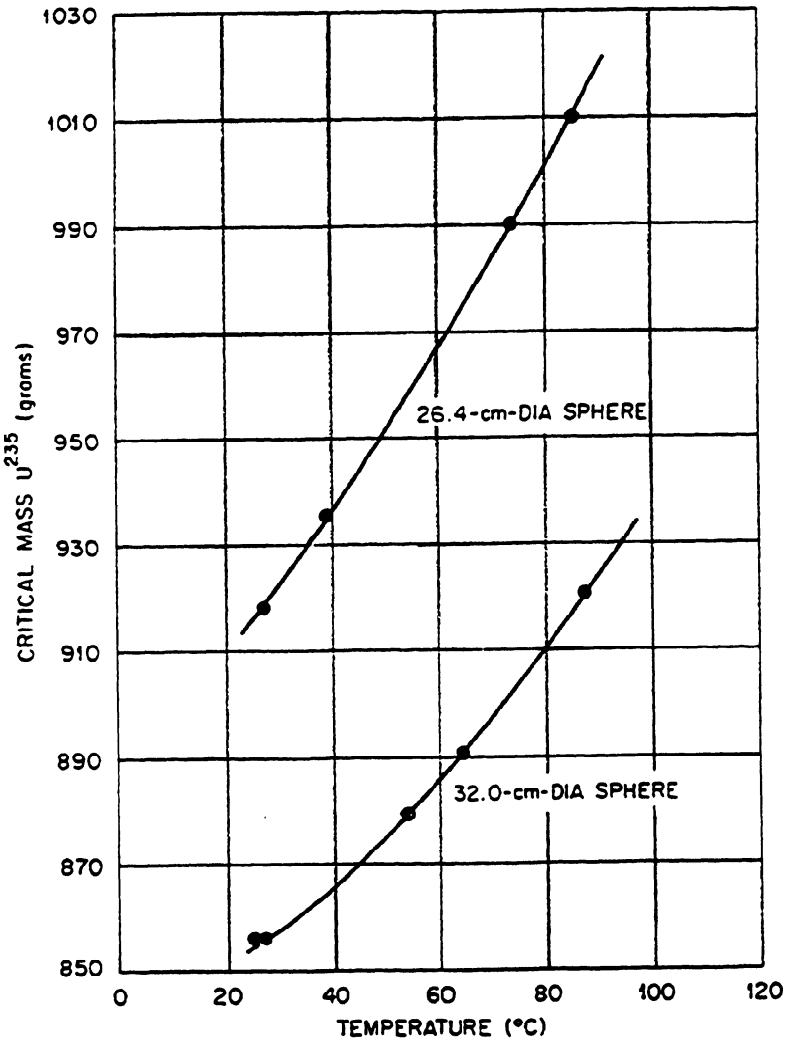

FIG. 5. Critical mass of U^{235} in water-reflected spherical reactors as a function of temperature.

Results and Discussion

The buckling, B^2, was determined from the material rather than geometry as computation of the geometric buckling would involve the use of questionable extrapolation distances for the various temperatures. Using equation (1), the U^{235} buckling[1] was calculated. The results are presented in Table II, together with values of L_0^2 corrected for density changes only. The square of the fast diffusion length, τ, is taken as 27 cm^2 which has been found to give fair agreement between experiment and two-group theory calculations (*11*).

Applying these data to equation (3), η_3 as a function of energy was determined. The results appear in Table IV.

It is shown in equation (3) that $\eta(U^{233})$ depends upon the values of the U^{233}

[1] $(1 + \alpha)$ for U^{235} can be assumed constant from 0.01 ev to 0.08 ev. For more detailed results see (10).

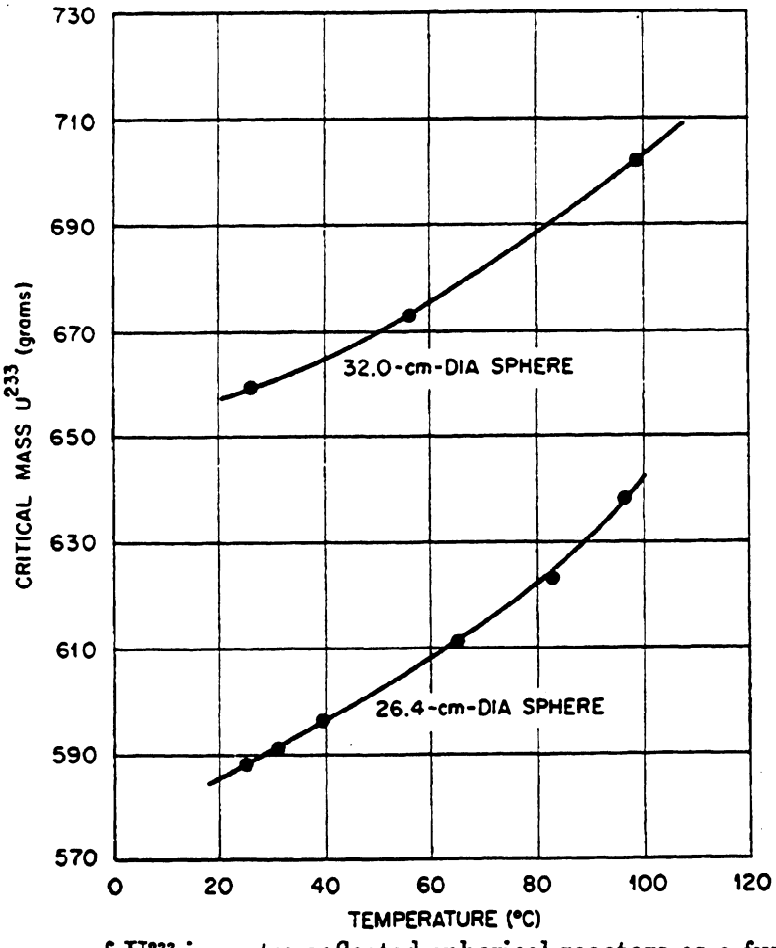

FIG. 6. Critical mass of U^{233} in water-reflected spherical reactors as a function of temperature.

and U^{235} cross sections. Since, in the concentration range examined in these experiments, $\eta(U^{233})$ is a nuclear constant independent of the geometry in which it is measured, it is possible to show that the values of the cross sections are internally consistent. Equating the expression for $\eta(U^{233})$, equation (3), for the two spheres and letting M represent the quantity $\sigma_a(H)[1 + L_0^2B^2]$ one obtains

$$\sigma_a(3) = \frac{\sigma_a(5)(R_3M - R_3'M') - (R_3'M'R_5M - R_3MR_5'M')}{R_5M - R_5'M'} \tag{4}$$

where the primed and unprimed symbols refer to each of the two spheres, respectively. The values of the cross sections which satisfy this equation at the various temperatures agree with published values (*9*) within their limits of accuracy.

The errors associated with the values of η for U^{233} are attributable to the accuracy with which the other nuclear constants are known. The error has been determined to be less than ± 0.03 from equation (3) and the expression,

$$\Delta\eta_3 = \eta_5 \sum_i \left[\frac{\partial\eta_3}{\partial\alpha_i}\right]_0 \Delta\alpha_i + \left[\frac{\eta_3}{\eta_5}\right]_0 \Delta\eta_5, \tag{5}$$

TABLE II

ATOMIC RATIOS AND MATERIAL BUCKLINGS REQUIRED FOR CRITICALITY

Temperature (°C)	L_0^2 (cm²)	26.4-cm Sphere			32.0-cm Sphere		
		H/U^{233}	H/U^{235}	$B^2 \times 10^3$ (cm^{-2})	H/U^{233}	H/U^{235}	$B^2 \times 10^3$ (cm^{-2})
25	8.1225	421.2	269.8	29.1	663.8	516.0	22.2
40	8.1998	414.5	263.0	29.2	655.2	506.8	22.4
60	8.3482	403.7	253.0	29.6	639.8	491.4	22.6
80	8.5337	390.7	242.3	29.7	620.7	473.0	22.9
100	8.7347	375.6	229.9	30.1	599.7	454.0	23.3

TABLE III

CROSS SECTIONS

Temperature (°C)	Energy (ev)	σ_a(H) (barns)
25	0.0257	0.328
40	0.0270	0.322
60	0.0287	0.312
80	0.0304	0.304
100	0.0321	0.297

TABLE IV

η FOR U^{233}

Temperature (°C)	26.4-cm sphere	32.0-cm sph
25	2.311	2.315
40	2.311	2.315
60	2.312	2.315
80	2.315	2.316
100	2.312	2.313

where the alphas represent the cross sections, buckling, diffusion lengths, and atomic ratios. An accuracy of ±0.5% is assigned to the value of the critical masses measured in the smaller sphere. The precision of the measurements in the larger sphere is somewhat less although the analytical differences noted in Table I are exceptions. The accuracy of these results is, however, affected by a systematic error, making the masses and concentrations in the large sphere about 2% high.

The temperature coefficient of reactivity was obtained by writing equation (1) in the form

$$k_{\text{eff}} = \frac{\eta\sigma_a(\text{U})}{[\sigma_a(\text{U}) + R\sigma_a(\text{H})(1 + L_0^2B^2)](1 + \tau B^2)} . \tag{6}$$

From the definition of the reactivity, $\rho = (k_{\text{eff}} - 1)/k_{\text{eff}}$, and equation (6) one obtains

$$\rho = 1 - \frac{[\sigma_a(\text{U}) + R\sigma_a(\text{H})(1 + L_0^2B^2)](1 + \tau B^2)}{\eta\sigma_a(\text{U})}. \tag{7}$$

The change in reactivity with temperature is then given by

$$\Delta\rho/\Delta T = \sum_i \left(\frac{\partial\rho}{\partial\alpha_i}\right)_0 \frac{\Delta\alpha_i}{\Delta T}, \tag{8}$$

where α_i becomes, successively, $\sigma_a(\text{U})$, $\sigma_a(\text{H})$, R, L_0^2, τ, and B^2. The resulting coefficients for the two isotopes in both spheres are

26.4-cm sphere: -1.4×10^{-4}/°C for U^{233}
-1.3×10^{-4}/°C for U^{235}
32.0-cm sphere: -6.7×10^{-5}/°C for U^{233}
-5.5×10^{-5}/°C for U^{235}.

Unreflected Reactor Experiments

Experimental Data

The following are the critical conditions observed for U^{233} and U^{235} uranyl oxyfluoride solutions in the 32.0-cm sphere (17.020 liters volume) at 27.0°C[2] without a water reflector:

U^{233}	gm U^{233}/L (at 25°C):	67.37
	H/U^{233} atomic ratio:	381.0
	Critical mass U^{233}:	1146.0 gm
U^{235}	gm U^{235}/L (at 25°C):	125.18
	H/U^{235} atomic ratio:	203.5
	Critical mass U^{235}:	2128.8 gm

Results

If the assumption of equal leakage from the two uranium solutions in the same unreflected critical sphere is made, equation (3) yields a value of $\eta(U^{233}) = 2.350$ using the following parameters:

$H/U^{233} = 381.0$	$\sigma_a(5) = 662$ barns
$H/U^{235} = 203.5$	$\sigma_a(3) = 575$ barns
$B^2 = 31.35 \times 10^{-3}$ cm^{-2}	$\eta_5 = 2.08$

The difference between this value of η and the one calculated from the results with the reflected spheres indicates that the assumption of equal leakage from the two unreflected critical solutions may be incorrect. Also this may imply

[2] Experiment performed at 21°C and results corrected in manner described in Table I.

TABLE V

POSITIVE PERIOD MEASUREMENTS OF U^{233} AND U^{235} OXYFLUORIDE SOLUTIONS IN A REFLECTED SPHERICAL REACTOR

Rod displacement (cm)	Positive period		Reactivity, ρ		$\frac{\rho\ (U^{233})}{\rho\ (U^{235})}$
	U^{233} (sec)	U^{235} (sec)	U^{233}	U^{235}	
2.54	77.0	230.9	0.144	0.048	3.00
3.81	45.6	139.4	0.205	0.072	2.85
5.08	26.0	106.7	0.280	0.091	3.08

that the age of neutrons in U^{233} oxyfluoride solutions is greater than that in the U^{235} oxyfluoride solutions.

PERIOD MEASUREMENTS

In a series of experiments comparing the fraction of delayed neutrons produced in the fission of U^{233} and U^{235}, the reflected 32.0-cm sphere was made critical successively with the two isotopes, with a boron rod, 0.48 cm in diameter, inserted along the vertical axis to within 5 cm of the center. The rod was withdrawn from each solution in equal increments and the resulting positive reactor periods measured. The results are reported in Table V.

The reactivity responsible for each of the periods was obtained from the inhour equation

$$\rho = \frac{k_{ex}}{k_{eff}} \cong \frac{1}{\sum_i \beta_i} \sum_i \frac{\beta_i}{1 + \lambda_i T}$$

where ρ is the reactivity expressed in "dollars," β_i and λ_i are the abundance and decay constant of the ith group of delayed neutrons, and T is the reactor period resulting from the addition ρ. The values of ρ and their ratios are also given in Table V. The ratio of the reactivities effected by equal rod displacements in the two solutions is inversely proportional to the ratio of the delayed neutron yields from the two isotopes.

SUMMARY

A comparison has been made of the value of η, the number of neutrons emitted per thermal neutron absorbed by U^{233} or U^{235}, by determining the concentration of the aqueous solutions of the two isotopes which could be made critical in the same geometry. A value of 2.31 $\pm$ 0.03 for U^{233} has been obtained from the better known $\eta(U^{235})$, 2.08, and the cross sections. It has been shown that $\eta(U^{233})$ is insensitive to energy changes over the limited range of the experiments. The critical experiments were performed in water reflected spherical vessels of two

sizes giving a satisfactory independent check of the internal consistency of the values of the cross sections of U^{233} and U^{235}. Measurements in an unreflected sphere yielded a value of $\eta(U^{233})$ somewhat different from the one reported above. The difference is probably attributable to the inapplicability of the analytical method to the unreflected experiment.

The temperature coefficients of reactivity of the U^{233} and U^{235} solutions have been measured between 25 and 100°C and found to be approximately equal. The critical masses of U^{233} and U^{235} were measured in two water reflected spheres and in one sphere without reflector.

The delayed fraction of fission neutrons from U^{233} has been shown to be about one-third the number from U^{235}, in agreement with previously reported values. In this experiment the ratio was obtained from measurements of the reactor periods resulting in the two solutions from equal reactivity changes.

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