

New determination of the ^{233}U specific activity and half-life

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 (Received 7 January 1974)

A new determination of the ^{233}U specific activity has been made using a method developed for measuring the longer-lived uranium isotopes. Samples prepared by molecular plating were counted in an intermediate-geometry α proportional counter with a flat pulse-height plateau. For each sample, the small amount of residual non-plated uranium was evaluated by counting in a 2π counter. Energy analysis with a silicon-junction detector allowed accurate measurement of the small amount of activity not ascribable to ^{233}U and ^{234}U . The ^{234}U activity was corrected for by calculation from the mass-spectroscopically-measured isotopic concentration. The specific activity was measured as $21\,405 \pm 20$ -dis/min μg ^{233}U , corresponding to a half-life of $(1.5911 \pm 0.0015) \times 10^5$ yr. The quoted error is statistical (standard error of the mean), based upon observed scatter of the data.

$$\left[\text{RADIOACTIVITY } ^{233}\text{U}; \text{ measured } T_{1/2}. \right]$$

I. INTRODUCTION

The value used for the α -decay half-life of ^{233}U has an important effect upon the values calculated for the various fission constants used by reactor physicists.^{1,2} These include the fission cross section, the capture cross section, and the neutron yield per fission of ^{235}U , ^{233}U , and ^{239}Pu . Such extensive interrelatedness arises because many measurements yield ratios of the values from the several nuclides, and because each measurement requires an evaluation of the amount of nuclide present.

Experiments in which fission constants are measured generally require thin and uniform samples. Suitable sample preparation techniques are, on the whole, not quantitative, i.e., they do not allow the deposition of a known weight of material, since they often involve electroplating (which does not deplete the solution) or vacuum sublimation. It is convenient, therefore, to first prepare high-quality samples, and then to assay them by α counting in a counter of known efficiency. This mode of assay has also been used even when a potentially quantitative sample-preparation method has been used, such as evaporation of a solution aliquot.

The α -counting assay approach has been commonly applied to ^{233}U and ^{239}Pu , which have relatively high specific activities (dis/min mg) and has even been used for the low-specific-activity ^{235}U through "spiking" with known amounts of ^{233}U .³ In the latter case, the ^{233}U is in a low concentration (evaluated by mass spectrometry), and ^{235}U dominates the fission activity while ^{233}U dominates

the α activity. Thus, the ^{233}U half-life is an important input datum for the least-squares reconciliation of the many measured values and ratios of the fission constants.^{1,2,4}

Until a few years ago, the half-life value available seemed to be a good one. All the measurements since the first one in 1949 agreed and the true value appeared to be close to 1.620×10^5 yr with an error of perhaps 0.2–0.3%. Then two very low measurements (see Table VI) appeared in 1967 and 1968, both agreeing and giving about 1.54×10^5 , a drop of 5%. Our reexamination of one of the very low values (see Sec. III B) indicated that it needed correction upward, but only by 1.3%, still not enough to reconcile the polarized set. In 1969, a preliminary (as yet unpublished) value appeared which was intermediate between the two groups. Those investigators who had to decide what to use for the input data of a least-squares analysis were in a quandary. In Ref. 1 the problem was resolved by discarding the older measurements, averaging the remainder to 1.593×10^5 yr, and increasing the assigned error to ($\pm 1.5\%$). In Ref. 4 the decision was made to discard all of the earlier (hence, higher) values and to average the two latest (and lower) ones as 1.554×10^5 yr ($\pm 0.2\%$). It will appear from our results that the method of Ref. 1 happened to have been a more fortunate choice.

It is difficult to explain such gross discordance between a number of experiments, each of which seems, by itself, to be carried out with a reliable method. In attempting to resolve the problem, we decided to redetermine the specific activity,⁵ using an experimental technique previously applied by

us to the precision measurement of the ^{235}U and ^{238}U half-lives, quantities of considerable interest in geological dating.⁶

II. EXPERIMENTAL METHOD

This measurement used a technique which was largely the same as that described in Refs. 6 and 7, with some modifications made necessary by the fact that the ^{233}U specific activity is much higher than those of ^{235}U , ^{236}U , or ^{238}U . A brief description is given here under the headings: uranium samples, sample preparation, chemical analysis, α counting, pulse-height analysis, and mass spectroscopic analysis.

A. Uranium samples

The ^{233}U oxide received from the Isotopes Division of Oak Ridge National Laboratory was dissolved and a portion was subjected to a routine anion-exchange purification step. A pulse analysis showed a small amount of ^{238}Pu to be present, an unexpected contingency for which the purification step was not adapted. In later runs, a reducing agent (ammonium iodide) was included to insure that plutonium would not follow the uranium, and the pulse analyses showed this procedure to have been successful.

The specific activity measurement was completely repeated three times after the first run. Runs II and III were made from the same ^{233}U batch. Run IV was made on uranium which was gathered together from samples previously used for chemical analysis in runs I-III, and then purified. The anion-exchange column purification was intended to separate the uranium from the impurities introduced in the chemical analysis titration. The procedure was effective for most of these, but it was not realized that the method was ineffective for iron, and that the ^{233}U sample in run IV contained some iron impurity. The $\text{Fe}^{++} \leftrightarrow \text{Fe}^{+++}$ transformations respond to the analytical titration procedure like the $\text{U}^{+4} \leftrightarrow \text{U}^{+6}$ reactions, with a resulting error in the run IV chemical analysis. A precision measurement of the iron present was made, and it was found to correspond to 3.03% of the uranium. With this correction, run IV agreed with the other runs. Some of the uranium batch used for runs II and III was then analyzed and was found to have a small amount of iron, equivalent in titration strength to 0.15% of the uranium present. None of the stock solution used for run I was left so no iron analysis was feasible. We do not know whether the iron in runs II and III was present in the original Oak Ridge sample, or whether it was introduced as a contaminant during the lengthy evaporation-to-dryness step used for

these two runs.

We have not, therefore, made any iron correction for run I. It was felt that, in view of the small effect in runs II and III, the run I results should not be discarded. Nevertheless, because of the uncertainty, we have down weighted the run I result to one half that of the other three runs.

B. Sample preparation

Sample preparation, as in Ref. 6, involved transfer of solution aliquots from a polyethylene bottle which was weighed before and after the transfer. Unlike the ^{235}U and ^{236}U measurements, an added dilution was required before the counting sample weighings, because of the high specific activity of ^{233}U and the requirement we set that aliquot solutions weigh about 100 mg (to insure $>0.03\%$ accuracy in weighing).

Using the molecular plating technique of Ref. 8,

^{233}U Plating Cell

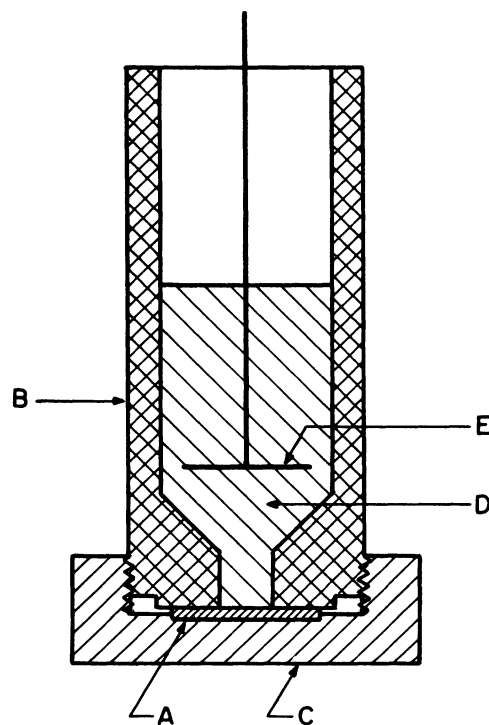


FIG. 1. Molecular plating cell. A, aluminum sample plate and cathode of cell. B, Teflon cell wall chimney with smaller bottom section to reduce area of deposition. C, brass bottom plate. D, solution of ^{233}U and La in isopropyl alcohol. E, anode, of 2.54-cm-diam platinum disk welded to a platinum wire. The cell is fastened together by the pressure of the screw thread. The reverse taper at the Teflon edge which touches the sample plate is described in Ref. 6.

as described in Ref. 6, we deposited the uranium from isopropyl alcohol onto a mechanically polished aluminum disk, covering an area 1.07 cm in diameter (Fig. 1). To achieve >99% uranium deposition for every sample, we found it necessary to add 100 μg of lanthanum to the ^{233}U sample (5 to 50 μg). The surface density of the heaviest sample remained <200 $\mu\text{g}/\text{cm}^2$. The non-plated uranium was recovered from the alcohol solution, concentrated, deposited on a plate, and counted in a 2π counter.

C. Chemical analysis

Samples used in analysis contained 35- to 60-mg uranium. The analytical method was like that used in Refs. 6 and 7, involving prior reduction of uranium with metallic lead, titration with Ce^{4+} , and back titration with Fe^{2+} . Standardization was against National Bureau of Standards standard uranium samples. The accuracy of the method is discussed in Ref. 6. The iron was spectrometrically measured as an o-phenanthroline complex with an accuracy of 2 to 3% in runs II and III and 1% in run IV.

The analytical result for each chemical sample, corrected for the iron content and the dilution factor, yielded the value of ^{233}U weight/mg solu-

tion. Combined with the solution weight for each counting sample, this yielded the uranium weights listed in Table III.

D. α counting

The intermediate geometry α counter (IGAC) is described in Ref. 6 and is shown in Fig. 2. It is an absolute counter,⁹ with geometry calculable from its precisely measured dimensions. Under the correct operating conditions, the lower tail of its α pulse-height distribution has a negligible, almost zero, number of counts. Setting the operating point in the midst of this tail allows 100% of the particles passing within the aperture KK (Fig. 2) to be counted.

A typical pulse-height spectrum in IGAC, uncorrected for background, is shown in Fig. 3. Background causes the rise in intensity at low energy and is largely due to β activity in the chamber walls. Although most of the spectrum shown is that of a single sample, in the neighborhood of the operating point (shown as square points) the spectrum is an average over 10 samples. Over six channels in this region, the average count per channel is 0.016 counts per minute (cpm). Then, for the 13 channels below the operating point, the rate should be 0.208 cpm,

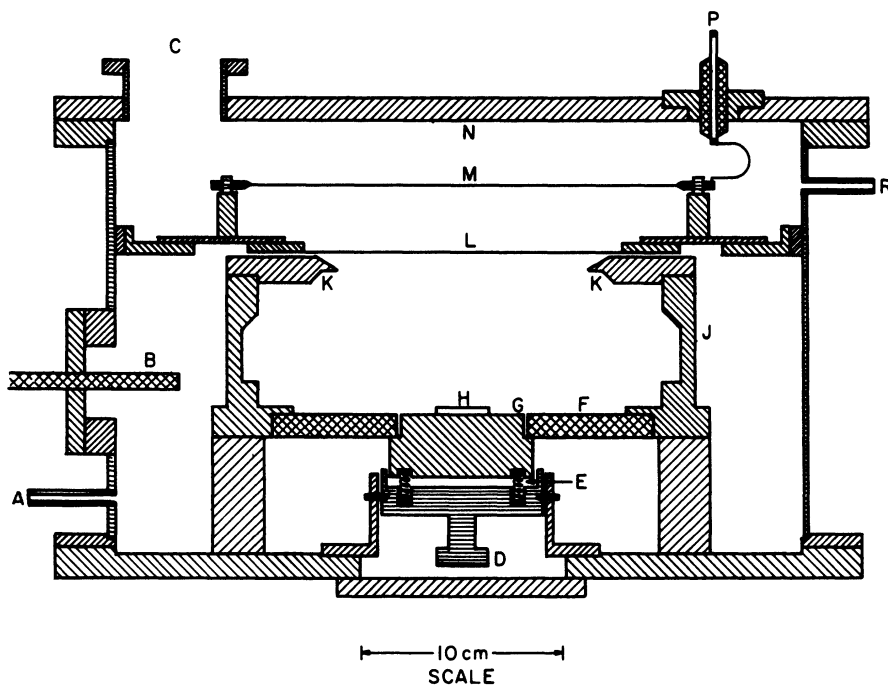


FIG. 2. IGAC, the intermediate geometry α counter. A and R, gas inlet and outlet for flowing argon-10% methane. C, to vacuum pump. G, sample support, precisely positioned. H, sample plate, precisely centered on sample support. KK, precisely machined circular aperture, with 0.003-cm-thick edge and precisely measured diameter. L, thin plastic film with evaporated gold conducting layer, $\sim 0.6 \text{ mg}/\text{cm}^2$ total thickness. M, a set of parallel proportional-counter wires, spanning the circular area. P, high voltage and signal lead.

assuming that the pulse-height distribution remains constant down to zero pulse height rather than tailing off. Comparing this quantity to the total counting rate of 10^4 cpm in IGAC, it is evident that the small pulses which are not counted are $< 0.002\%$ of the total.

Relative to sample rates of 10^4 to 10^5 cpm, the backgrounds of 2–4 cpm were negligible. The dead time was important for the higher counting samples (dead-time losses $\leq 1\%$). The uncertainty in action of the precision live timer⁶ corresponded to $< 5\%$ of the dead time, hence $< 0.05\%$ for the highest count rate sample.

The geometry factor for each sample was calculated from the known counter and sample dimensions and from the measured sample activity distribution.

For a point source P' on the axis, a circular aperture of radius a subtends the geometry

$$G_{P'} = \frac{1}{2} \left(1 - \frac{z}{D} \right) = \frac{1}{2} \left(\frac{a^2}{D(D+z)} \right) \quad (1)$$

with z equal to the vertical distance from aperture plane KK (Fig. 2) to sample surface H, and D equal to the diagonal distance $(a^2+z^2)^{1/2}$. For a point source P off the axis, the corresponding geometry is¹⁰

$$G_P(r) = G_P' - \frac{3}{8} c_2 r^2 + \frac{15}{32} c_4 r^4 + \dots \quad (2)$$

with

$$c_2 = \frac{a^2 z}{D}, \quad c_4 = \frac{a^2 z}{D^3} \left(z^2 - \frac{3}{4} a^2 \right).$$

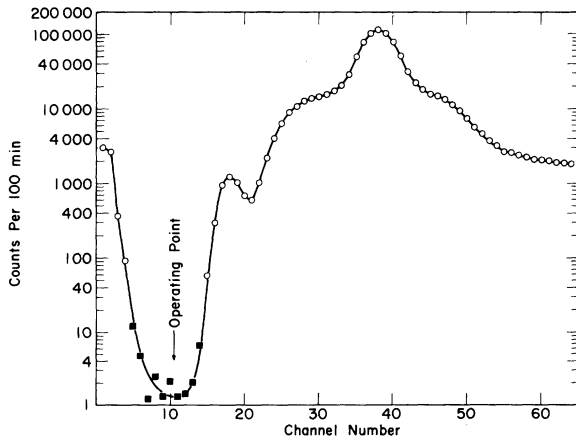


FIG. 3. Pulse-height distribution of pulses from the proportional counter. For clarity, each channel number listed corresponds to eight channels in the original spectrum. The main spectrum (circles) is from one of the samples. In the neighborhood of the operating point [squares], the points are averages over 10 samples. Above the operating point, background has a negligible effect on the spectrum.

For example, since the vertical distance from G to KK (Fig. 2) is 7.2375 cm, $z = 7.0851$ for a sample plate thickness of 0.1524 cm. With $a = 5.0845$, $D = 8.7207$, whence $G_{P'} = 0.93778 = 1/10.664$. For the outer rim of the sample ($R = 0.535$ cm), $G_P(R) = G_{P'}(1 - 0.00416 + 0.00002) = 0.99586G_{P'} = 1/10.708$.

The average geometry for a uniformly spread sample (radius R) is

$$G_S = G_{P'} - \frac{3}{16} c_2 R^2 + \frac{5}{32} c_4 R^4 + \dots \quad (3)$$

and for a nonuniform distribution, it is

$$G_{av} = \frac{\int_S G_P(r) A_P(r) dr}{\int_S A_P(r) dr} = \frac{C}{V}, \quad (4)$$

where S is the sample area, $A_P(r)$ is the mean sample disintegration rate in an annular ring at radius r , C is the mean sample counting rate, and V is the mean disintegration rate.

The sample distribution was measured by counting the sample in a 2π counter with a series of collimators, each of which measured the relative activity of the sample at a different radius. The i th collimator was a disk with n_i closely spaced holes drilled around a circle with radius r_i . Successive r_i -values gave a nonoverlapping survey of the sample. With $a(r_i)$ the measured counting rate of the sample with the i th collimator, Eq. (4) is well approximated as

$$G_{av} \cong \frac{\sum_{i=1}^n G_P(r_i) a(r_i)}{\sum_{i=1}^n a(r_i)} \quad (5)$$

because $G_P(r_i)$ varies by $< 0.5\%$ over the sample. The values of $1/G_{av}$ used for the samples ranged from 10.550 to 10.740 over the four runs. Further details of the scanning process are given in Ref. 6.

Each sample of run I was counted for a minimum of 3.5×10^6 counts, corresponding to a statistical counting error of $\sim 0.05\%$; for runs II–IV, total counts ranged from 10^7 to 10^8 counts. Statistical tests showed no evidence for drifts in counting efficiency.

For each sample, the α activity from the residual which was not molecular plated was gathered together and counted with a standard 2π proportional counter. Because the unplated fraction was small (generally $< 1\%$), it was unnecessary to evaluate the counting efficiency of the 2π counter accurately. It was taken to be 0.515.

E. Pulse-height analysis

Energy analysis was carried out on a thin sample ($\sim 15 \mu\text{g}/\text{cm}^2$), using a large-area solid-state detector. A typical result is shown in Fig. 4, and

results for all four runs are given in Table I. For run I, correction was made for ^{232}U and ^{238}Pu ; for runs II–IV, only the ^{233}U was present. Although the ^{234}U α activity was significant, the overlap of the ^{233}U and ^{234}U peaks did not allow estimation by energy analysis.

F. Mass spectroscopic analysis

The isotopic composition measured mass spectrometrically before run I agreed with the results measured before run IV; the average is given in Table II. Since pulse analysis did not allow separation of ^{233}U and ^{234}U α peaks, the ^{234}U activity was calculated from the mass ratios (Table II). The correction factor for ^{234}U activity is

$$F_{234} = \frac{1.00000}{1.00119} = 0.99881.$$

III. SPECIFIC ACTIVITY AND HALF-LIFE OF ^{233}U

A. Experimental results

The counting data are shown in Table III. Also given is the percentage of non-plated uranium in each sample; this is measured by 2π counting. The results from the four runs are averaged in Table IV. The final result is: total activity per μg of uranium = 21320 ± 20 dis/min.

Correcting for the ^{234}U activity (F_{234}) and the mass concentration of ^{233}U (Table II), the specific activity of $^{233}\text{U} = 21320 \times (0.99881/0.99484) = 21405 \pm 20$ dis/min μg ^{233}U . The corresponding half-life, using the constants of Table V, is calculated to be

$$T_{1/2}(^{233}\text{U}) = (1.5911 \pm 0.0015) \times 10^5 \text{ yr.}$$

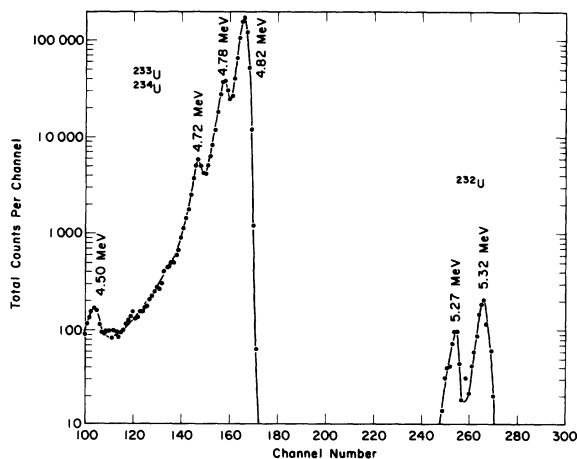


FIG. 4. Energy analysis of α particles from a ^{233}U sample, using a silicon-junction detector. Only ^{232}U and mixed ^{233}U - ^{234}U peaks are evident.

TABLE I. Pulse analysis of ^{233}U samples.

Isotope	% total α activity	
	Run I	Runs II, III, IV
^{232}U ^a	0.170 ± 0.010	0.170 ± 0.010
^{233}U	99.379 ± 0.013	99.830 ± 0.010
^{234}U		
^{238}Pu	0.430 ± 0.008	
>5.5 MeV	0.021 ± 0.002	

^a By mass, this corresponds to $0.78(10^{-4})$ wt.%, checking with the value in Table II.

The errors in the mass spectrometry value, in F_{234} , and in the pulse analyses, as well as the statistical counting errors, are negligible relative to the error represented in the scatter between runs. The quoted error is the standard error of the mean shown in Table IV.

B. Comparison to previous measurements

Measurements of the ^{233}U half-life have been reported in the following papers.

Hyde (Ref. 11, 1949). Using uranium with 96.4% ^{233}U , the chemical content was analyzed by controlled ignition to U_3O_8 . Small masses were involved, ranging from 1.5 to 8 mg. Dissolved U_3O_8 was deposited on platinum disks through volume aliquotting, and evaporated to dryness with no special spreading techniques. The samples were counted in a 2π ionization chamber operated in air. The dead time was taken as 0.48 msec, and the geometry factor (correcting for backscattering) was taken as 52%. Neither correction was evaluated for the experiment, but was taken as characteristic of the class of instruments used. No correction or allowance for ^{232}U and daughters was made. The result was $T_{1/2} = 1.62(\pm 0.01) \times 10^5$ yr.

Sellers, Bentley, and Studier (Ref. 12, 1953). Details are sparse. The ^{233}U was weighed as U_3O_8 , dissolved, diluted, and weight aliquots were used to prepare counting samples. These were evapo-

TABLE II. Isotopic composition of uranium in the sample.

Isotope	Wt. %	Relative α activity ^a
^{232}U	$0.8(10^{-4})$	0.00174
^{233}U	99.484 ± 0.007	1.00000
^{234}U	0.186 ± 0.003	0.00119
^{235}U	0.064 ± 0.002	0.00000
^{236}U	0.017 ± 0.005	0.00000
^{238}U	0.249 ± 0.003	0.00000

^a Calculated from current half-life values and the mass ratios.

rated onto platinum disks with no special spreading techniques. 2π counting in an argon-CO₂ ionization chamber was used with an assumed 52% geometry factor. No correction or allowance for ²³²U and daughters was made. The result was 1.615(±0.004) × 10⁵ yr.

Bigham et al. (Ref. 13, 1958). Uranium samples used in fission cross-section measurements also yielded α specific activity ratios. The usual chem-

ical mass analysis was made unnecessary through measurement of $R_i = f_i/\alpha_i$, the ratio of fission counting rate to α counting rate, in the same 2π ionization chamber. Samples of natural uranium, ²³³U, ²³⁵U, and mixed ²³³U-²³⁵U of known composition were used. Since the thin aluminum discs onto which the samples were painted were not completely flat, the counting geometry ϵ could not be assumed to be 2π . However, this effect canceled

TABLE III. Counting data for ²³³U samples.

Run no.	Sample no.	Activity ^a (dis/min)	Percent not plated ^a	Uranium mass ^b (μ g)	Specific activity ^c (dis/min μ g)	Run no.	Sample no.	Activity ^a (dis/min)	Percent not plated ^a	Uranium mass ^b (μ g)	Specific activity ^c (dis/min μ g)
Bottle A						Bottle C					
I	1	73 322	0.10	3.4121	21 489	6	1 247 470	1.02	58.483		21 331
	2	97 786	0.18	4.5422	21 528	7	1 170 510	2.11	55.067		21 256
	3	100 677	0.60	4.6871	21 480	8	1 214 020	0.84	57.022		21 290
	4	94 244	0.24	4.3817	21 509					Average	21 292
	5	109 905	0.86	5.1094	21 511						
				Average	21 503						
Bottle B						Bottle D					
	6	90 312	0.32	4.1991	21 507	9	888 560	0.21	41.605		21 357
	7	87 114	0.58	4.0514	21 502	10	787 570	0.78	36.956		21 311
	8	100 284	0.19	4.6618	21 512	11	999 800	1.00	47.011		21 267
	9	103 873	0.68	4.8290	21 510	12	1 097 170	1.21	51.460		21 321
				Average	21 508					Average	21 314
				Run average	21 506					Run average	21 306
Bottle A						Bottle B					
II	1	1 102 100	0.30	51.602	21 358	4	1 659 550	0.34	77.581		21 391
	2	1 241 130	1.43	58.087	21 367	5	1 291 550	0.37	60.382		21 390
	3	1 052 590	0.60	49.283	21 358	6	1 360 040	0.67	63.608		21 382
	4	1 133 590	0.98	53.018	21 381	7	1 491 440	0.57	69.832		21 358
				Average	21 366	8	1 378 150	0.64	64.533		21 356
										Average	21 375
Bottle B						Bottle C					
	5	814 350	0.38	38.073	21 389	9	1 395 310	0.35	65.351		21 351
	6	840 730	0.59	39.328	21 377	10	1 392 620	0.59	65.038		21 412
	7	971 760	1.41	45.495	21 360	11	1 324 100	0.72	61.950		21 374
	8	937 730	1.05	43.822	21 369	12	1 277 230	0.59	59.823		21 350
				Average	21 374					Average	21 372
				Run average	21 370					Run average	21 367
Bottle A						Bottle B					
III	1	1 023 990	0.33	47.959	21 351	3	992 080	1.10	46.562		21 307
	2	1 139 590	1.75	53.533	21 288	4	1 390 910	1.24	65.391		21 271
				Average	21 319	5	1 156 400	0.80	54.246		21 318
										Average	21 298

^a Most of the activity measured in IGAC, at a known geometry of about 1/11. The nonplated portion (of order of 1% or less) evaporated onto a plate and counted in 2π geometry.

^b Total uranium mass in sample (i.e., uncorrected for ²³³U content).

^c Total activity per μ g U, uncorrected for non-²³³U activities.

on the assumption that the ϵ value in each sample was the same for both α and fission counting. Similar cancellation of the α backscattering factor ϕ occurred in the ratio for two isotopes, $r_{ij} = R_i/R_j$, on the assumption that the variation of ϕ over the α -particle energy range 4.2 to 4.8 MeV (^{238}U to ^{233}U) was negligible. The uranium was chemically purified from ^{228}Th and daughters. The ^{232}U present was corrected for by energy (pulse) analysis, which showed no unexpected peaks. Using the atom abundance ratio in natural uranium of $^{235}\text{U}/^{238}\text{U} = 0.007257(\pm 0.1\%)$ from Ref. 14, the ratio of the specific α activity (dis/min mole) of natural uranium to that of ^{233}U was determined as $(7.235 \pm 0.012) \times 10^{-5}$. With the natural uranium value of $1502(\pm 0.5\%)$ dis/min mg U (or 3.576×10^8 dis/min mole) from Refs. 15 and 16, the specific activity of ^{233}U was given as 4.943×10^{12} dis/min mole ($\pm 0.53\%$), corresponding to $T_{1/2}(^{233}\text{U}) = 1.606 \times 10^5$ yr. A recent redetermination of the ^{238}U half-life⁶ with improved precision gave the value 3.6082×10^8 dis/min mole U_{nat} ($\pm 0.07\%$). With a more accurate value¹⁷ of the $^{235}\text{U}/^{238}\text{U}$ atomic abundance ratio [$0.007249(\pm 0.04\%)$] and the new uranium specific activity value, the corrected result is $T_{1/2}(^{233}\text{U}) = 1.590 \times 10^5$ ($\pm 0.18\%$).

Dokuchaev and Osipov (Ref. 18, 1959). Details of the measurements are sparse. Several samples of about 94% ^{233}U were used. Sample aliquots were deposited and counted in a low geometry counter (geometry factor ~ 3000). There is no reference to the chemical assay procedure, to the dead-time correction, or to the correction for the ^{232}U daughters, although ^{232}U constituted about 6% of the ^{233}U activity. Results were given as 20950 ± 100 dis/min μg ^{233}U , corresponding to $T_{1/2} = 1.626(\pm 0.008) \times 10^5$ yr.

TABLE IV. Averaging the four runs.

Run no.	Total specific activity ^a (dis/min μg)	Pulse analysis correction factor ^c	Corrected specific activity ^b (dis/min μg)
I	21 506	0.993 79	21 372
II	21 370	0.998 30	21 334
III	21 306	0.998 30	21 270
IV	21 367	0.998 30	21 331
Average ^d			21 320 \pm 20

^a From Table III. Total activity per μg of total uranium.

^b ^{233}U - ^{234}U activity per μg of total uranium.

^c Correction for α activity not derived from ^{233}U or ^{234}U , using pulse analysis data (Table I).

^d As described in the text, run I is given half-weight. The error is the standard error of the mean, $s(\bar{x}) = s/\sqrt{W}$, where $W = 3.5 = \text{sum of weights}$ and $s^2 = \Sigma w_i \Delta_i^2 / 3$, with $w_1 = 0.5$ and $w_2 = w_3 = w_4 = 1$ ($s = 38$), and $\Delta_i = |x_i - 21320|$.

Popplewell (Ref. 19, 1961). A sample of 83.6% ^{233}U was purified and analyzed by ignition (750 C) to U_3O_8 . A concentration of 0.08% ^{232}U by α activity was measured in a gridded ion chamber. Weight aliquots were used in sample preparation; these were counted in a low geometry counter with geometry factor $709(\pm 0.53\%)$. The measurement yielded $T_{1/2}(^{233}\text{U}) = 1.613 \times 10^5$ yr ($\pm 0.63\%$).

Ihle, Langenscheidt, and Murrenhoff (Ref. 20, 1967). A sample of 87.8% ^{233}U was used, with 2.52% of the α activity due to ^{232}U measured by a surface-barrier detector. Counting was carried on in a liquid scintillation counter, in which a 0.3% extrapolation of counting rate versus pulse height to zero pulse height was required. Interference from ^{228}Th and daughters was avoided by measuring within eight days of chemical extraction, this separation also serving to remove fission products whose β -ray emissions the counter did not discriminate from α radiation. Chemical analysis involved controlled ignition of U_3O_8 and sample aliquots were weighed. The measurements yielded a specific activity value of 2.101×10^7 dis/min mg ^{233}U ($\pm 0.20\%$) or 1.621×10^5 yr ($\pm 0.20\%$).

Oetting (Ref. 21, 1967). This was potentially a very important measurement, since it involved a different technique than other experiments, one inherently capable of high precision. Unfortunately, these virtues were somewhat vitiated, since an important correction was not made and this can now be included only with some uncertainty. The power (W/sec) released from a 17.6-g ^{233}U metal sample of known chemical and isotopic composition was measured in a microcalorimeter. From the measured isotopic composition and known decay energies of the "impurities" ^{232}U and ^{234}U , 0.95% of the measured power was ascribed to ^{232}U decay and 0.60% to ^{234}U decay. With the α -particle energy for the ground-state transition of ^{233}U taken as 4.816 MeV, the total decay energy Q_α was 4.900 MeV. The result, after correction for ^{232}U and ^{234}U , was 2.894×10^{-4} W/g ^{233}U ($\pm 0.17\%$), corresponding to $T_{1/2}(^{233}\text{U}) = 1.540 \times 10^5$ yr ($\pm 0.17\%$). An unconsidered source of decay heat in the sample was that due to the daughters of ^{232}U decay. The first daughter is ^{228}Th (1.90 yr) which grew, in the "seven month" period between chemical separation and calorimetric measurement, to 0.192 of its equilibrium activity. The seven succeeding daugh-

TABLE V. Constants used in calculating half-life.

At. wt. ^{233}U	= 233.039 ^a
Avogadro's number	= 6.02252×10^{23}
One year	= 5.2595×10^5 min.

^a ^{12}C as standard.

ters have such short half-lives as to be essentially in equilibrium with the ^{226}Th . From the decay energies given in Refs. 22 and 23, and assuming the escape of energetic γ rays from the calorimeter, calculation shows that 33.1 MeV are absorbed per ^{228}Th decay, leading to an added correction of 1.1% in the half-life value. This result, plus a more recent value of $Q_{\alpha}(^{233}\text{U}) = 4.909 \text{ MeV}$,²² yielded the corrected value $2.863 \times 10^{-4} \text{ W/g } ^{233}\text{U}$, hence $T_{1/2}(^{233}\text{U}) = 1.560 \times 10^5 \text{ yr } (\pm 0.25\%)$.

Keith (Ref. 24, 1968). Two samples were used: (a) of 99.5% ^{233}U and (b) of 99.99% ^{233}U . (a) contained 1.2% ^{232}U and daughters by α activity and (b) 0.2%, both measured with a gridded ion chamber. The (a) solution was standardized by isotope

dilution, "spiking" with a known amount of natural uranium, followed by mass spectrometric measurement. Samples were prepared by freeze drying and counted in the same low geometry counter used by Popplewell. A linear plot of specific activity versus sample weight gave an extrapolated value (2% higher than the last point) corresponding to $T_{1/2}(^{233}\text{U}) = 1.559 \times 10^5 \text{ yr } (\pm 0.93\%)$. Samples from the (b) solution were electroplated and counted in the same way. Each sample was then dissolved and the amount of uranium measured by isotope dilution, using a known amount of ^{235}U as the "spike." Five results ranging from 1.522 to 1.581 averaged to $1.546 \times 10^5 \text{ yr } (\pm 0.89\%)$. The over-all average was $T_{1/2}(^{233}\text{U}) = 1.553 \times 10^5 \text{ yr } (\pm 0.65\%)$.

TABLE VI. Various measurements of ^{233}U half-life.

Author	Ref.	Method	Counter	Measured specific activity dis/min mg $^{233}\text{U} (\times 10^{-7})$	Reported ^{233}U half-life ($\times 10^{-5}$ yr)	Recalc. ^{233}U half-life ^a ($\times 10^{-5}$ yr)
Hyde (1949)	11	Specific activity	2 π air ion chamber	2.10($\pm 0.6\%$)	1.62($\pm 0.6\%$)	
Sellers <i>et al.</i> (1953)	12	Specific activity	2 π ion chamber	2.109($\pm 0.25\%$)	1.615($\pm 0.25\%$)	
Bigham <i>et al.</i> (1958)	13	Ratio of specific activities	2 π ion chamber	2.121($\pm 0.53\%$)	1.606($\pm 0.53\%$)	1.590($\pm 0.18\%$)
Dokuchaev and Osipov (1959)	18	Specific activity	Low geometry counter	2.095($\pm 0.5\%$)	1.626($\pm 0.5\%$)	
Popplewell (1961)	19	Specific activity	Low geometry counter	2.109($\pm 0.56\%$)	1.615($\pm 0.56\%$)	1.613($\pm 0.63\%$)
Ihle <i>et al.</i> (1967)	20	Specific activity	Liquid scintillation counter	2.101($\pm 0.20\%$)	1.621($\pm 0.20\%$)	
Oetting (1967)	21	Specific power	Microcalorimeter		1.540($\pm 0.17\%$)	1.560($\pm 0.25\%$)
Keith (1968)	24	Specific activity	Low geometry counter ^c	2.207($\pm 0.65\%$) ^b	1.553($\pm 0.65\%$)	
Durham (1969) (preliminary)	1, 25	Specific activity	Low geometry counter	2.152($\pm 0.44\%$) ^b	1.583($\pm 0.44\%$)	
Euratom, Geel (preliminary) (1973)	26	Specific activity			1.586($\pm 0.3\%$)	
Euratom, Geel (preliminary) (1973)	27	Specific activity	Liquid scintillation counter		1.585($\pm 0.3\%$)	
Jaffey <i>et al.</i>	Present work	Specific activity	Intermediate geometry counter	2.1405($\pm 0.095\%$)	1.5911($\pm 0.095\%$)	

^a If recalculation was warranted. Large changes are described in the text.

^b Calculated from the reported half-life value.

^c According to Ref. 1, the same counter as in Ref. 19.

Durham (Refs. 1 and 25, 1969). Used a sample of 99.8% ^{233}U with about 0.15% α activity from ^{232}U and daughters (the latter measured with a silicon detector). A low geometry counter was used to count samples prepared by drying weighed aliquots. The master solution was standardized by controlled potential coulometry and by isotope dilution. A preliminary (and as yet unpublished) result was $T_{1/2}(^{233}\text{U}) = 1.583 \times 10^5 \text{ yr}$ ($\pm 0.44\%$).

Central Bureau for Nuclear Measurements Laboratory, Geel (Refs. 26 and 27). No details are given in this progress report for one measurement which yielded the preliminary half-life value $1.586(\pm 0.3\%) \times 10^5 \text{ yr}$. The other measurement was made by liquid scintillation counting of a sample containing 99.9986% ^{233}U . The α and β contribution from daughter products and other uranium α emitters was 0.16% and was corrected for. The uranium mass was analyzed by isotope dilution and controlled potential coulometry. The measurement yielded the preliminary half-life value $1.585(\pm 0.3\%) \times 10^5 \text{ yr}$.

The various results are summarized in Table VI and compared to the result of this work in Table VII. It is evident that most of the results, with the exception of those of Refs. 13 and 25–27, differ from the result reported here by large amounts, either positively or negatively. The Ref. 13 result was one of those discarded in the summary of Ref. 1 because of its use of 2π counting, notoriously inferior for absolute counting. It seems, however, that the use of relative, rather than absolute, counting rates resulted in an accurate ratio of (specific activity of ^{233}U) to (specific activity of natural uranium). This, with a more

TABLE VII. Deviation of various measurements from present result.

Author	Ref.	Half-life ($\times 10^{-5} \text{ yr}$)	Difference from reported value Δ	Normalized difference Δ/error^a
Hyde	11	1.620 ± 0.010	+0.029	+2.9
Sellers <i>et al.</i>	12	1.615 ± 0.004	+0.024	+6.0
Bigham <i>et al.</i>	13	1.590 ± 0.003	-0.001	-0.3
Dokuchaev and Osipov	18	1.626 ± 0.008	+0.035	+4.4
Popplewell	19	1.613 ± 0.010	+0.022	+2.2
Ihle <i>et al.</i>	20	1.621 ± 0.003	+0.030	+10.0
Oetting	21	1.560 ± 0.004	-0.031	-7.8
Keith	24	1.553 ± 0.010	-0.038	-3.8
Durham	1, 25	1.583 ± 0.007	-0.008	-1.1
Euratom, Geel	26	1.586 ± 0.005	-0.005	-1.0
Euratom, Geel	27	1.585 ± 0.005	-0.006	-1.2

^a Δ in units of the quoted error in the third column.

accurate value of the natural uranium value,⁶ resulted in an improved ^{233}U half-life.

ACKNOWLEDGMENTS

We wish to thank Donald Rokop of the mass spectrography group for isotopic analyses of the uranium samples and Frank Telford of the chemical analysis group for analyzing the iron content of these samples. We are indebted to the Isotopes Division, Oak Ridge National Laboratory, for providing us with the ^{233}U sample.

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