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Precision Measurement of Half-Lives and Specific Activities of ^{235}U and ^{238}U†

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New determinations of the half-lives of ^{235}U and ^{238}U have been made. Improved techniques have allowed the half-life values to be measured with greater accuracy than has been heretofore achieved. Samples were prepared by molecular plating and counted in a intermediate-geometry α-proportional counter with an extremely flat pulse-height plateau. The small amount of residual nonplated uranium was counted in a 2π counter. Energy analysis with a silicon-junction detector was used to measure the presence of "foreign" activities. For ^{235}U, the measured specific activity was (4798.1 ± 3.3) (dis/min)/(mg ^{235}U), corresponding to a half-life of $(7.0381 \pm 0.0048) \times 10^8$ yr. For ^{238}U, the specific activity was measured as (746.19 ± 0.41) (dis/min)/(mg ^{238}U), corresponding to a half-life of $(4.4683 \pm 0.0024) \times 10^9$ yr. Errors quoted are statistical (standard error of the mean), based upon the observed scatter of the data. This scatter exceeds that expected from counting statistics alone. We believe that systematic errors, if present, will no more than double the quoted errors.

I. INTRODUCTION

Because they are so long lived, the uranium isotopes have proven to be quite useful for measuring geological ages through evaluation of the amounts of ^{206}Pb and ^{207}Pb formed in the minerals of interest.[1-3] The ^{235}U half-life is shorter (7×10^8 yr) and decay has occurred over several half-lives for minerals of great age. On the other hand, for the ^{238}U isotope, the half-life is so long (4.5×10^9 yr) that decay in a mineral is always less than one half-life. Since the demands on the accuracy of the half-life value increase with the age of the mineral, one might hope for a more accurate value for ^{235}U, for which the need is greater. The facts are, however, that to this point the ^{238}U half-life was known more accurately, presumably with an error of <1%; the accuracy in the ^{235}U value was commonly thought to be about 2%.

We felt it desirable at this time to apply improved techniques to the measurement of the ^{235}U half-life, with the goal of reducing the error to no more than 0.2%. As a preliminary experiment, partly designed to test the procedure, we measured the ^{238}U half-life. To our surprise, we found that our value was lower than hitherto accepted values by amounts exceeding their alleged errors. This measurement is then to be considered not as a calibration of the method, but as a more accurate redetermination of the ^{238}U half-life.

II. EXPERIMENTAL METHOD

In principle, the experiment simply required determination of the α-emission rate from a known weight of isotope. In practice, determining the weight required: (1) chemical analysis of the amount of uranium present, (2) tests for the presence of other interfering elements, (3) mass spectrometric analysis to determine the fraction of uranium present in the form of the desired isotope, and (4) a sample-preparation technique which ensured that each sample contained an accurately known weight. Determining the α-emission rate required that (1) each sample be counted with a precisely known counting efficiency, (2) each sample be counted long enough to make negligible the over-all statistical counting error, and (3) an energy analysis be made to determine the fraction of the total α activity derived from the isotope of interest.

Each of these requirements was met using methods which preserved the level of accuracy we aimed at. For the chemical analysis, we were fortunate that the importance of uranium has led to the development of highly accurate analytical methods. The spectroscopic analysis showed no impurities which would have interfered with a gravimetric procedure (precipitation of uranium and controlled ignition to U_3O_8). Nevertheless, we preferred to use a titration method which has been shown to be extremely accurate and to be insensitive to impurities which would have interfered with a gravimetric approach. Since the samples used were of almost pure isotopes, the mass spectroscopic analyses were used only to make the small mass corrections required by the presence of "foreign" isotopes.

Samples were prepared by using known aliquots of analyzed solutions. The required accuracy was achieved by using weight aliquots rather than volume aliquots. Each sample was molecular-plated onto a disk from a solution in organic solvent. After plating, the solution was evaporated and the small residual activity was counted in a 2π counter. The activity of the molecular-plated sample was measured with a medium-geometry counter of accurately defined aperture, under conditions in which 100% of the particles entering the defined aperture were counted. A thin sample was used for energy analysis, which was achieved with a silicon solid-state detector. α activity due to other isotopes was low enough to allow accurate determination of the foreign activity.

We have devoted considerable effort to reduce the error in each step to a small value, and we believe that the measurements described here have succeeded in yielding quite accurate half-life values. For this reason, we describe the measurements in detail, to permit a close scrutiny of possible sources of error.

III. α COUNTING

A. α Counter

Because the two uranium isotopes have low specific activities, it was necessary to count with as high a counting efficiency as possible. Yet if a uranium sample is deposited on a plate and counted in a 2π counter (efficiency $\simeq 0.5$), corrections are necessary for sample self-absorption and α-particle backscattering.[4] Such corrections, while feasible, are difficult to make to the desired accuracy. Both corrections are unnecessary when the measured particles are restricted to those emitted at fairly large angles to the surface of the sample plate. This is due to the fact that for the uniform deposit in a molecular-plated sample, absorption in the sample occurs only for α particles emitted at small angles to the sample surface, while backscattered α particles, arising as they do from multiple electronic scattering, are also a relatively small-angle phenomenon.

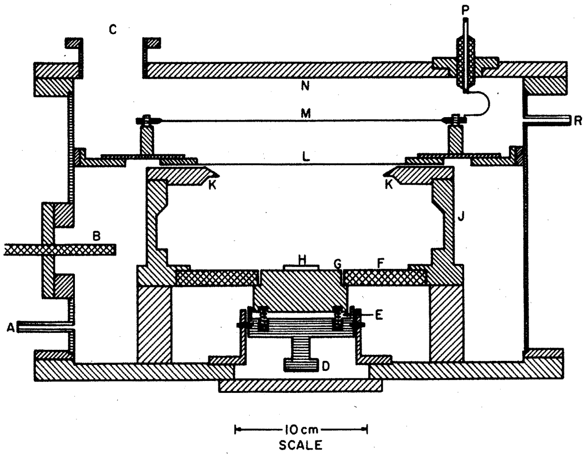

FIG. 1. The intermediate-geometry α counter (IGAC). A and R: gas inlet and outlet for flowing argon (10% methane). C: to vacuum pump. G: sample support, precisely positioned. H: sample mount, centered on sample support. K: accurately machined circular aperture, with 0.001-in.-thick edge and precisely measured diameter. L: thin plastic film with evaporated-gold conducting layer, $\sim$0.6 mg/cm^2. M: proportional-counter wires, spanning the circular area. P: high voltage and signal lead.

Thus, we needed a counter with geometry reduced below 50%, but as high as possible within the limits set by exclusion of backscattering and sample absorption effects. A further requirement was that the counter aperture be large relative to the diameter of the sample deposit. This was necessary, because it was desired to keep the detector geometry reasonably constant over the sample surface. Were the geometry to vary markedly, we would have had to *accurately* determine the distribution of sample mass over the sample surface – a possible, but tedious task. The variation of geometry over the sample is minimized if the detector aperture is made sizably larger than the source diameter. Thus, a counter of fairly large dimensions was required. A large counter also served to make the geometry less sensitive to slight errors in measuring the thickness of the sample mount.

For our specific-activity measurements we used such a counter already well tested and calibrated in other experiments – the intermediate-geometry α counter (IGAC),[5] diagrammed in Fig. 1. The geometry was defined by the diameter of the circular aperture KK and by the vertical distance GK of the sample surface from the plane of this aperture. The precisely machined structure of stabilized stainless steel was designed to be susceptible to precise dimensional measurement. The aperture, accurately circular, was 10.169 cm in diameter. To avoid uncertainties due to penetration of a knife edge, this edge was machined to a thickness of about 0.003 cm, adequate to stop α particles, and yet thin enough to avoid difficulties with edge scattering. The vertical distance to the sample support was 7.237 cm, giving a geometry of 0.090 87 for a centered point source of zero sample mount thickness. The samples were mounted (Sec. IV) on flat polished aluminum disks 4.42 cm in diameter and approximately 0.135 cm thick. The plates were made flat, with parallel sides, and the side receiving the electroplating was given a satin finish. The thickness of each plate was accurately measured with an error of ±0.0003 cm. This value was subtracted from the previously measured vertical distance GK to give the effective vertical distance. The critical dimensions of the system were known with high accuracy; possible errors in these measurements should have resulted in an error in defining the geometry of <0.01%.

Immediately above the aperture was the detector itself, a large multiwire proportional counter bounded on its lower side by a window made of a thin plastic film [Mylar, Dupont, Wilmington, Delaware], with a vacuum-evaporated gold coat which provided a conducting boundary. The window allowed α particles to penetrate with only modest energy loss, but served both to provide an electrical plane to define the detector's sensitive volume and to exclude ions formed outside this volume. The window was about 0.005 mm thick; the film itself had a surface density of 0.56 mg/cm^2 with a gold layer of 0.04 mg/cm^2, for a total of 0.60 mg/cm^2. The entire chamber was filled with a gas mixture of argon with 10% methane at low pressure. The path of the α particle emitted from the source past the aperture edge and into the detecting region was about 10 cm long, aside from the absorption in sample and window. It was thus necessary to reduce the pressure to about 30 to 35 Torr. An α particle had to pass through the sample thickness, the gas absorber, and the window thickness, yet have enough energy left for reasonable ionization in the detector. Of all the α particles, those with a diagonal path suffered the greatest absorption. The pressure was reduced sufficiently to allow these diagonal tracks to yield adequate ionization. The combined equivalent thickness of sample, gas, and window absorption was <2 mg/cm^2 for the longest diagonal track. Further pressure reduction served only to reduce the ionization in all the tracks, most seriously for the vertical ones, which were the shortest.

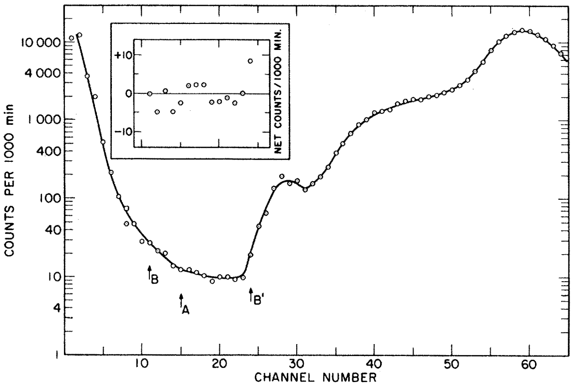

FIG. 2. Pulse-height distribution of pulses from the proportional counter. The ordinate is gross counts per 1000 min. The main spectrum is from one sample of ^{238}U, batch II; the region between B and B′ is an average over the 12 samples. In the insert is shown the net rate per channel in BB′, after background subtraction. The operating point for counting was at point A. The BB′ region showed a total of 2321 counts/1000 min for the 12 samples, or for one sample, an average of 193.4 ± 4.0 counts/1000 min. The background over the same region was 198.0 ± 9.8 counts/1000 min, or a net (over background) of −4.6 ± 10.6 counts/min. This is to be compared with an average over the 12 samples of about 2×10^5 counts/1000 min for each sample. Clearly, the numbers of counts attributable to the sample in the BB′ region is <0.005%.

The multiwire anode was placed equidistant between the thin film and the top of the chamber. The counting volume was approximately 19 cm in diameter by 6.35 cm deep. At 30 Torr, an α particle passing vertically through the chamber would pass through approximately 0.4 mg/cm^2 of gas absorber, less if it happened to hit the wire. Despite the small amount of ionization formed by the particles, the gas gain of the proportional counter could be made large enough so that preamplifier noise was negligible.

Both the original ionization and the multiplication factor were a function of the gas pressure, which was carefully chosen and kept constant. When the operating pressure was chosen, its value was measured with a 0.1% differential gauge and the same pressure was used for all samples of the same type. Counting gas was kept flowing through the system at a rate of 150 ml/min, and a precision manostat was used to keep the pressure constant.

To rapidly identify the proper gas pressure and voltage, the output pulses were examined with a multichannel analyzer. A pulse-height region ranging from 0.15 to 0.4 of the most common pulse size was found to have very few pulses (Fig. 2). A pulse analysis was made for each of the samples counted, as well as for several background measurements. When background was subtracted, the region BB′ (Fig. 2) was found to have zero counts. Pulse analyses for the various samples, when taken at precisely the same operating pressure and high voltage, gave the same pulse-height distribution in the region BB′, and nearly the same at larger pulse heights. Tests showed the spectral shape in region BB′ to be quite insensitive to small changes in gas pressure.

The operating pulse-height-selection level was chosen approximately midway in the region at point A, but the lack of net counts in BB′ would have allowed any point in the region to have been used.

The relatively large number of pulses at the very lowest pulse heights were identified as arising from β-particle ionization. β-active radioactivities with the approximate intensities and approximate energies of ^{234}Th (UX_1) and ^{234}Pa (UX_2) grown from the ^{238}U gave nearly the same pulse-height distribution. With the ^{235}U samples, the ^{231}Th (UY) gave a similar distribution of pulses at the very lowest pulse heights.

Although the existence of a sizable pulse-height range with no pulses represented a counter with a completely flat plateau, it was felt desirable to check the counter stability by examining successive counts on the same sample. The accumulated numbers of counts were recorded every 100 min, and the values were checked for consistency with a χ^2 test. These tests indicated no inconsistencies. To avoid possible timing errors arising from changes in frequency of the power line, timing was derived from a quartz crystal whose frequency was known to 3 in 10^6.

B. Geometry Factor in the α Counter

The geometry factor for a circular aperture and a point source at P' on its axis is given by

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

where a = radius of aperture, z = vertical distance from the plane of the aperture to the source surface, and $D = (a^2 + z^2)^{1/2}$. For a point source at P, off the axis, the geometry is[6]

$$G_P(r) = G_{P'} - \tfrac{3}{8} r^2 \frac{a^2 z}{D^5} + \tfrac{15}{32} r^4 \frac{a^2 z}{D^9}(z^2 - \tfrac{3}{4}a^2) + \cdots , \tag{2}$$

where r is the distance of P from the axis ($r = PP'$). The vertical distance from the top of the sample support to the upper edge of the collimator slit was $z = 7.2375$ cm and $a = 5.0844$ cm. Then $D = 8.8449$ cm and $G_{P'} = 0.090\,867 = 1/11.005$. With a sample plate 0.1364 cm (0.0537 in.) thick, $z = 7.1011$ cm; then $D = 8.7337$ cm and $G_{P'} = 0.093\,463 = 1/10.699$. With $R = 1.65$ cm, the maximum value of r, from Eq. (2),

$$\begin{aligned} G_P(R) &= 0.093\,463 - 0.003\,688 + 0.000\,067 \\ &= 0.093\,463(1 - 0.039\,46 + 0.000\,72) \\ &= 0.961\,26 G_{P'} . \end{aligned}$$

In the light of the intended accuracy, G_P was not constant over the sample area.

Were the deposition uniform, then the average geometry would be given by[6]

$$G_S = G_{P'} - \tfrac{3}{16} R^2 \frac{a^2 z}{D^5} + \tfrac{5}{32} R^4 \frac{a^2 z}{D^9}(z^2 - \tfrac{3}{4}a^2) + \cdots . \tag{3}$$

For the same sample, $G_S = G_{P'}(1 - 0.019\,73 + 0.000\,24) = 0.980\,51 G_{P'}$. However, previous experience in scanning [with the sample activity distribution scanning α counter (SADSAC)][7] samples prepared with the molecular-plating technique had shown that deposition was not uniform.

It is feasible to determine the mass distribution by measuring the α activity at each of many small areas over the sample. However, because of the small amount of activity in a sample, it was not feasible to scan the sample point by point. Instead, we utilized the fact that $G_P(r)$ is constant for a ring-shaped region having constant r.

A series of disks were made of aluminum 1.59 mm (0.0625 in.) thick, and closely spaced holes about 2.0 mm (0.0810 in.) in diameter were drilled at constant radius. Each disk had holes at only one radius. The first disk had one hole at the center, the next one had as many equally spaced holes as could be drilled on a 2.06-mm (0.0810 in.) radius, the next disk was drilled at a 4.11-mm (0.1620 in.) radius, etc. A sample was covered with one of these disks and counted in a 2π counter. Using all of the disks, in turn, on one sample provided an approximate scan of the activity distribution as a function of the radius.

If $A_P(r, \theta)$ is the mean disintegration rate per unit area at point P (coordinates r, θ) of the sample, then the mean counting rate from the entire sample area S is

$$C_S = \int_S G_P(r) A_P(r, \theta) dS , \tag{4}$$

and the average geometry is

$$G_{\text{av}} = \frac{\int_S G_P(r) A_P(r, \theta) dS}{\int_S A_P(r, \theta) dS} . \tag{5}$$

Or, if $A_r(r)$ is the mean disintegration rate per unit length of radius for a constant radius ring with thickness dr, then

$$G_{\text{av}} = \frac{\int_S G_P(r) A_r(r)\, dr}{\int_S A_r(r)\, dr} . \tag{6}$$

Then, with $\mathfrak{A}(r_i)$ = mean counting rate for the disk with holes at the distance r_i from the center,

$$G_{\text{av}} \cong \sum_{i=1}^{n} G_P(r_i)\mathfrak{A}(r_i) \Big/ \sum_{i=1}^{n} \mathfrak{A}(r_i) , \tag{7}$$

where the sums in Eq. (7) approximately replace the integrals in Eq. (6). r_i is the distance from the disk center to the center of the drilled hole.

Equation (7) is only approximate, primarily because the requirement that there be an integral number of holes around a circle at radius r_i made it impossible that $n_i(r_i)$, the number on this circle, be strictly proportional to r_i. However, the deviation from proportionality is small, so unless the distribution if very nonuniform, and G_{av} deviates considerably from $G_{P'}$, the approximation (7) is a

TABLE I. Radial activity distribution over several ^{235}U samples.

Collimating disk number (i)	1	2	3	4	5	6	7	8	9[a]	Sample mount thickness (mm)	$G_{P'}$	$\frac{G_{av}}{G_{P'}}$	$\frac{G_S}{G_{P'}}$
Radius, r_i[b] (mm)	0.000	2.06	4.11	6.17	8.23	10.29	12.34	14.40	16.46				
Number of holes on circle	1	5	11	16	22	28	33	39	44				
$G_P(r_i)/G_{P'}$[c]	1.000 00	0.999 39	0.997 55	0.994 48	0.990 18	0.984 66	0.977 91	0.969 93	0.960 73				
Counting rate (counts/min)													
Sample No. 1	3.4	15.7	34.7	54.9	77.5	100.1	118.4	144.5	67.0	1.364	0.0934 65	0.980 06	0.980 51
Sample No. 2	2.6	14.4	32.1	48.0	68.3	89.0	105.1	128.4	63.1	1.374	0.0934 84	0.979 95	0.980 51
Sample No. 3	2.5	10.4	23.1	35.3	49.7	66.5	82.0	101.3	46.9	1.364	0.0934 65	0.979 73	0.980 51
Sample No. 4	3.0	14.0	30.9	46.1	65.5	82.6	103.1	124.0	60.4	1.382	0.0934 99	0.979 94	0.980 50
Counting rate per hole (counts/min)													
Sample No. 1	3.4	3.13	3.15	3.43	3.52	3.58	3.59	3.71	1.52				
Sample No. 2	2.6	2.88	2.92	3.00	3.10	3.18	3.19	3.29	1.43				
Sample No. 3	2.5	2.08	2.10	2.20	2.26	2.38	2.48	2.60	1.07				
Sample No. 4	3.0	2.81	2.81	2.88	2.98	2.95	3.12	3.18	1.37				
Relative counting rate per hole[d]													
Sample No. 1	0.92	0.843	0.849	0.924	0.949	0.965	0.968	1.000	0.410				
Sample No. 2	0.79	0.875	0.888	0.912	0.942	0.966	0.969	1.000	0.434				
Sample No. 3	0.96	0.800	0.808	0.846	0.869	0.915	0.953	1.000	0.412				
Sample No. 4	0.94	0.883	0.883	0.906	0.940	0.928	0.981	1.000	0.431				

[a] The radius of the sample R (determined by dimensions of the electroplating cell) fell in the middle of the hole, i.e., $R = r_9$.

[b] From center of disk to circle on which centers of holes are spaced.

[c] For Sample No. 1, but very close for the other samples.

[d] Counting rate per hole, normalized to the largest value, at the disk with r_8.

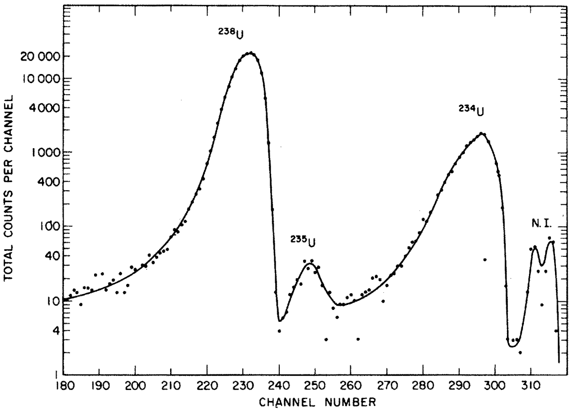

FIG. 3. Energy analysis of α particles from ^{238}U-I with a silicon-junction detector. The peak labeled N.I. was not identified, but contributed only about 0.1% of the activity (Table V).

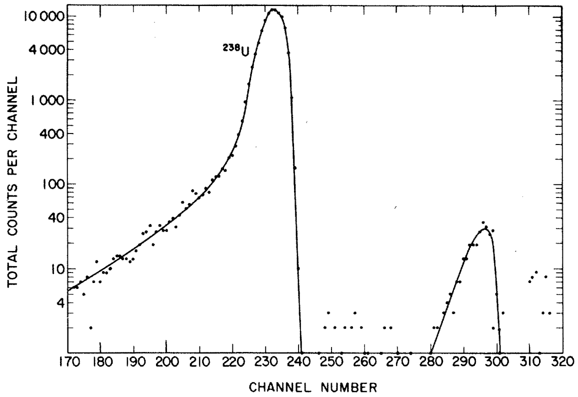

FIG. 4. Energy analysis of α particles from ^{238}U-II with a silicon-junction detector.

very good one. Since the geometry factor for one hole was about 6.8%, the counting rates were quite low. On the other hand, if G_{av} does not differ much from $G_{P'}$, high statistical accuracy[8] is not required of these counts.[5]

Illustrative of these measurements are the data for the first four ^{235}U samples shown in Table I. It is evident that G_{av} differs very slightly from G_S, i.e., the geometry factor is very close to the value calculated as though the sample were uniformly spread.

C. Counting the Supernatant from Molecular Deposition

The yield of the molecular-deposition method used was very high, but was less than 99.9%. It was necessary, therefore, to evaluate the amount of material remaining undeposited. Since the sample was deposited from the organic solvent isopropyl alcohol, it was feasible to evaporate the solution and to count the residual activity. The contents of the solution were evaporated onto a counting plate; this was measured in a 2π-geometry counter, thus giving a factor of 5 increased sensitivity in measuring the residual activity. Since some solid material is always deposited with the uranium activity, the geometry factor could not be the 52% usual for weightless samples, so it was taken as 50%. However, because the residual uranium was very low in amount, an error of 4% in the 2π counting would cause a very small error in the total uranium α count. Thus, even if the undeposited amount were as large as 1%, a 4% counting error in the 2π counter would result in an error of only 0.04% in the uranium assay. In fact, except for a few samples in the first ^{238}U batch, the deposition efficiency exceeded 99%.

D. Pulse-Height Analysis

The distribution of α activity among the various isotopes was measured with samples that were somewhat thinner than the ones used for α counting. These were prepared by the same molecular-plating method. A large-area silicon surface-barrier detector was used for the analyses. For the first ^{238}U sample (^{238}U-I), which had a moderate amount of ^{234}U activity, the activity ratio was also checked with a gridded ionization chamber. Three samples of ^{238}U-I were prepared: A, 24 μg/cm^2; B, 300 μg/cm^2; and C, 110 μg/cm^2. The ^{238}U-II sample was 120 μg/cm^2 and the ^{235}U sample, 80 μg/cm^2. The results are given in Figs. 3–5. To ensure an objective analysis, two operators independently estimated the relative intensities. For the second ^{238}U sample and for ^{235}U, the foreign activity was too low in concentration to cause any difficulties.

E. Checking IGAC Geometry Factor

As a check on the geometry value calculated from the measured dimensions, a special sample counted in IGAC was also counted with a low-geometry counter of accurately defined dimensions. The ^{233}U source, 13 mm in diameter, was scanned with the drilled disks. For the low-geometry counter, the geometry factor was corrected from $G_{P'} = 1/(101.99)$ to $G_{\text{av}} = 1/(102.37)$. The net rate measured was $102\,477 \pm 39$ dis/min. For IGAC, scanning resulted in correcting from $G_{P'} = 1/(10.881)$ to $G_{\text{av}} = 1/(10.921)$, with a net rate of $102\,505 \pm 29$ dis/min. The difference was (28 ± 49) dis/min or (0.03 ± 0.05)%. A similar experiment with a smaller-diameter sample of ^{239}Pu gave $302\,708 \pm 126$ dis/min in IGAC and $302\,731 \pm 150$ dis/min in the low-geometry counter; the difference was (23 ± 196 dis/min) or (0.008 ± 0.06)%.

For the second sample, the counting rate in IGAC was high enough so that dead-time losses were significant. Thus, with a counting rate of about 3×10^4 counts/min, a dead time of 5 μsec gave losses of 0.25%. The time, however, was measured with a live timer. (The quartz-crystal-clock signals were counted only when the counter was open and were blocked for a fixed period after each α particle was counted). Its uncertainty in action was <0.1 μsec. Hence, the uncertainty due to deadtime losses was <0.005%.

F. Backgrounds

The background correction was a small one, though for the ^{238}U samples it was significant, since the measured rates were about 200 counts/min. Prior to this experiment, through extensive use with active samples, the counter background had risen to undesirable levels. Careful cleaning, particularly with an ultrasonic cleaner, reduced the background to less than 1 counts/min. Considering the large exposed area in the counter, this value was felt to be reasonable.

Backgrounds were measured with a dummy plate whose activity was measured to be <0.05 counts/min in a 2π counter. However, it was found that the counter background was the same whether or not the dummy plate was used.

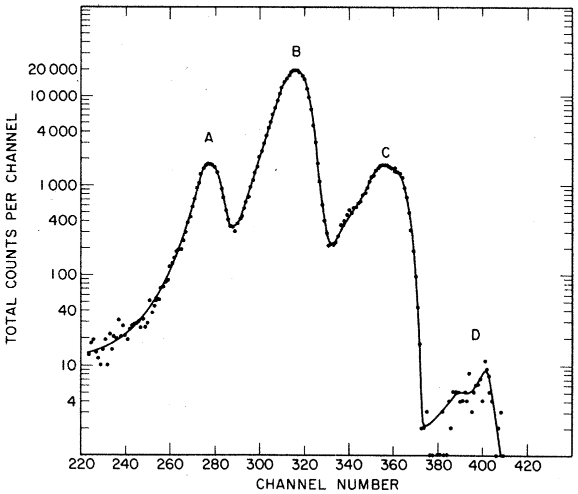

FIG. 5. Energy analysis of ^{235}U sample with silicon-junction detector. The measured relative intensities of the major peaks were approximately A : B : C = 0.089 : 1.00: 0.087. C is obviously a multiple peak. If we use the data of Baranov *et al.* (Ref. 24), we may ascribe B to the mixture of lines at 4.36- and 4.396-MeV, A to the mixture from 4.216- to 4.323-MeV, and C to the mixture of the 4.556- and 4.597-MeV lines. The corresponding relative intensities from Baranov are approximately 0.116 : 1.00 : 0.124. Considering sample thickness and the fact that no attempt was made to carefully analyze the tailing of the peaks, this is a reasonable check. The α particles at D are ascribed to ^{234}U (or ^{233}U).

The exact value of the background depended slightly upon the operating conditions used (gas pressure and chamber voltage). With a specified set of conditions, however, the background counts were consistent over the whole period of counting. For example, one set gave the values: 0.904 ± 0.027, 0.895 ± 0.024, 0.900 ± 0.025, 0.899 ± 0.025. Another set gave the values: 0.819 ± 0.018, 0.816 ± 0.014, 0.806 ± 0.018, 0.880 ± 0.026.

IV. SAMPLE PREPARATION

An aqueous stock solution of the uranyl nitrate salt of each isotope was prepared. The chemical purity of these uranium samples was checked by optical spectrographic analysis and found to be greater than 99.99%. For a single uranium batch, 5-ml polyethylene ampoules[9] were filled from the stock solution and were sealed. Weighed aliquots were subsequently dispensed from these ampoules according to the method described by Merritt and Taylor.[9] About 80 mg of solution per sample was used for α counting and about 2 g of solution per sample was used for mass analysis.

The weighed-aliquot method used in these analyses consisted of the following steps. The uranium solution was drawn up into a polyethylene ampoule, the tip of which had been drawn out into a fine capillary. The tip of the capillary was then heat sealed and the exterior of the ampoule was thoroughly washed and dried. At the time of sample preparation, the tip of the capillary was cut off and appropriate aliquots were squeezed out of the ampoule dropwise. The solution did not wet the capillary tip. The ampoule was weighed before and after each dispension to determine the exact weight of the aliquot. Even though the total time elapsed for the entire ensemble of weighings from each ampoule was short (about 10 min), repeated reweighings of the ampoules over longer periods of time indicated no loss of weight even when an hour or more had elapsed.

Merritt and Taylor showed conclusively that this technique of weighing aliquots by difference from a stock solution is far superior to any of the techniques involving weighing the solution drop itself after it has been dispensed. Two polyethylene ampoules were filled from each stock solution, and six counting samples together with two mass analysis samples were prepared from each ampoule. All weighings were done with a Mettler analytical balance which had been calibrated against National Bureau of Standards (NBS) standard weights in the range of operation used. A typical set of weighings is as follows:

$$\begin{array}{l} 30.611\,65 \text{ g} \\ \underline{30.524\,52 \text{ g}} \\ \;\;0.087\,13 \text{ g aliquot for } \alpha \text{ count} \end{array}$$

$$\begin{array}{l} 30.372\,16 \text{ g} \\ \underline{28.400\,34 \text{ g}} \\ \;\;1.971\,82 \text{ g aliquot for mass analysis} \end{array}$$

The total time lapse for weighing all the samples from each ampoule was about 10 min.

The samples for α counting were prepared in the following manner. Weighed aliquots were dispensed into 50-ml Pyrex centrifuge bottles, and about 10 ml of isopropyl alcohol was added to each container. The bottles were then stored until the time for molecular plating.

The contents of one bottle was then quantitatively transferred to a molecular-plating cell (see Fig. 6) using two 10-ml washes of isopropyl alcohol. The uranium was then molecular-plated onto aluminum plates according to the method described by Parker, Bildstein, and Getoff.[10] Each sample was plated from about 30 ml of isopropyl alcohol at 600 V for at least 4 h. The quantitativeness of the plating technique under the experimental conditions used was checked with ^{233}U tracer and found to be about 99.5%. The residual uranium in the supernate after each plating was corrected for by transferring the supernate solution back into the original centrifuge bottle (using two isopropyl alcohol rinses), evaporating the alcohol in the tube, taking up the residue in nitric acid, and quantitatively transferring this solution to a tantalum disk which was dried and counted in a 2π α counter. This correction varied somewhat from sample to sample but generally amounted to a few 10ths of a percent.

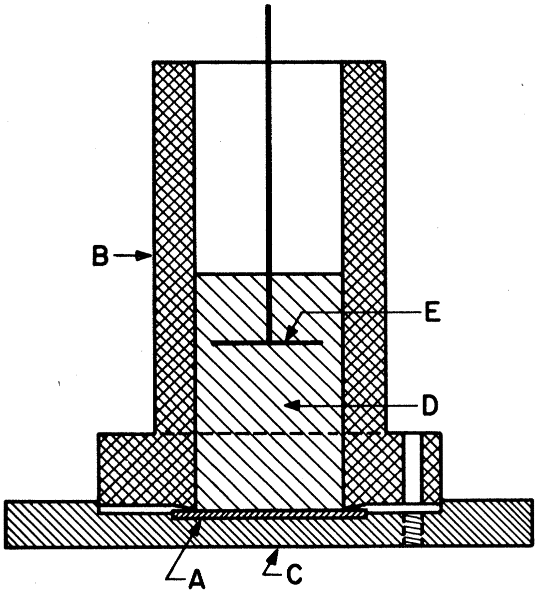

FIG. 6. Molecular-plating cell. A: aluminum sample plate and cathode of cell. B: Teflon cell wall chimney. C: aluminum bottom plate. D: solution. E: anode, of 1-in.-diam platinum disk welded to a platinum wire. The cell was fastened together and sealed by the pressure from three screws passing through the shoulder of the chimney into the bottom plate. To ensure that the sample would not be detached at the edge when the cell wall was removed after plating, the tapered edge of the chimney bottom was given a reverse taper too small to be shown in the drawing. For a distance about 0.25 mm from the inner diameter of the cell, a 30° taper was cut, such that the diameter of the sample plate area exposed to molecular plating was 0.5 mm larger than the chimney inner diameter.

The amount of ^{238}U per sample varied from 2.5 to 4.1 mg, corresponding to a surface density of about 0.3 to 0.5 mg/cm^2. For the ^{235}U samples, 1.8 to 2.7 mg was deposited, yielding 0.2 to 0.3 mg/cm^2 for the surface density.

All 12 of the α-counting samples were weighed out from the two ampoules within a short time. However, a number of days was required for electroplating all 12, since the plating could be completed only at the rate of three per day. The radiochemical purity of each isotope studied was checked by both mass-spectrometric- and α-pulse-analysis techniques.

V. MASS ANALYSIS OF THE URANIUM

The concentration of uranium in the solutions used for preparing α-counting samples was measured by analysis of two larger samples per ampoule. In all, four such samples were analyzed for each batch of uranium. Each sample of weighed uranyl nitrate solution was carefully taken to dryness and treated to convert the salt to the sulfate, completely eliminating the nitrate. The uranium (VI), in a solution of hydrochloric acid and sulfuric acid, was reduced with metallic lead to uranium (IV) and reoxidized with a measured amount of standardized ceric sulfate solution. The excess ceric sulfate was then titrated with a standardized ferrous sulfate solution. The back titration was necessary because the reaction between cerium (IV) and uranium (IV) is too slow at room temperature to permit accurate detection of the end point. Ferroin indicator was used to detect the ferrous ceric end point. All titrations were made with weight burets. The total mass of solution dispensed for each sample ranged from 12 to 15 g, the weighings being made to the nearest mg. The ceric sulfate solution was standardized on a weight basis by titration against both analyzed arsenious acid (50-mg samples) and NBS uranium

TABLE II. Typical standardization of ceric sulfate solution. In units of (mg ^{238}U)/(g Ce^{IV}) solution.

	Using As_2O_3	Using U_3O_8
	5.020_6	5.020_5
	5.029_6	5.020_3
	5.021_7	5.017_9
	5.016_6	5.019_8
	5.014_7	
	5.017_7	
	5.014_2	
Average [a]	$5.019_3 \pm 0.002_0$	$5.019_6 \pm 0.000_6$

[a] The error is the standard deviation of the mean $s/\sqrt{n}$.

oxide (100-mg samples). The two methods generally agreed within 0.02%. A typical comparative analysis is shown in Table II.

The analytical results for the three uranium batches are shown in Table III. The average percentage error per sample is

$$\epsilon_{av} = 0.045\% ,$$

a result with $3(4-1)$ = nine degrees of freedom (d.o.f.). In another experiment evaluating the uranium content of standard NBS uranium oxide, 17 measurements gave an average result deviating by 0.01% from the standard value with an estimated standard deviation of a single measurement (ϵ_{av}) of 0.08%.

These two results, one with nine d.o.f., the other with 16, may be averaged in the usual fashion to give an over-all average $\epsilon_{av} = 0.07\%$, with 25 d.o.f. The χ^2 tables then yield a 95% confidence interval for the percentage error for a single measurement; the interval lies between 0.05 and 0.10%. For the sake of a conservative treatment, we choose the upper limit of this interval, at 0.10%, to use in error analysis. With this value for the error of a single measurement, the error for the average of four measurements, the standard error of the mean, $\bar{\epsilon}_{av}/\sqrt{4}$, is 0.05%. We therefore assign the error in chemical analysis to be 0.05%.

TABLE III. Analytical results for uranium samples used in half-life measurements. In units of (mg/U)/(g solution).

	^{238}U-I	^{238}U-II	^{235}U
Bottle A	42.34	41.14	28.68
	42.34	41.21	28.70
Bottle B	42.34	41.16	28.69
	42.37	41.17	28.69
Average $\bar{x}$	42.35	41.17	28.69
s [a]	0.015	0.029	0.0082
$\epsilon = \frac{s}{\bar{x}} 100$	0.035%	0.070%	0.028%

[a] With $s^2 = \sum (x_i - \bar{x})^2/3$.

The analytical method actually gives the number of moles per gram of solution. This was transformed to mg of U per g solution using the atomic weights derived from the mass spectroscopic analysis. Further details of the analytical method may be found in the work of others.[11-13]

VI. SPECIFIC ACTIVITY AND HALF-LIFE OF ^{238}U

Two different batches of depleted ^{238}U were analyzed. Their isotopic compositions are given in Table IV. The α-energy analyses discussed in Sec. III D are illustrated in Figs. 3 and 4, both taken with a surface-barrier silicon detector; results are summarized in Table V.

The plates of batch ^{238}U-I were counted first, each plate at essentially the same operating point (voltage and gas pressure). Background measurements were interspersed with the plate counts. The counting results are given in Table VI. As described in Sec. IV, samples for plates were taken from two different bottles, labeled A and B, and these are listed separately in the table. The mass of uranium in the sample was derived from the weight of solution in the gravimetric aliquotting (Sec. IV) and the chemical analysis (Sec. V).

Two tests were applied to check data consistency. The first was the Student-Fisher t test for the difference between two means. With $\delta\bar{x} = 823.43 - 822.88 = 0.55$, the criterion for testing whether the data are consistent with the hypothesis that $\delta\bar{x} = 0$ is $t = \delta\bar{x}/s(\delta\bar{x}) = \delta\bar{x}/[\sqrt{2}(s/\sqrt{6})] = \sqrt{3}(0.55/1.38) = 0.63$. This is to be compared with t tables for 11 d.o.f. Since the probability (significance) level exceeds 50%, the test provides no reason for suspecting a difference between the two bottles. Considering another possibility, is there an observable trend? If we consider that x (counts/min mg) may be a linear function of time, with t values uniformly spaced, and if the function fitted is $x_i = \alpha + \beta t_i$, then the least-squares-estimated slope is $\hat{\beta} = S_{tx}/S_{tt}$, with

TABLE IV. Isotopic composition of ^{238}U samples.

	Sample	
Isotope	^{238}U-I (at.%)	^{238}U-II (at.%)
^{238}U	99.9790 ± 0.0003	99.9997 ± 0.0001
^{236}U	$0.000\,04 \pm 0.000\,01$	
^{235}U	0.0204 ± 0.0003	0.0003 ± 0.0001
^{234}U	$0.000\,40 \pm 0.000\,01$	
^{233}U	$0.000\,12 \pm 0.000\,01$	

TABLE V. Pulse analysis of ^{238}U samples.

Sample	Detector	Sample thickness ($\mu g/cm^2$)	^{238}U (%)	^{235}U plus ^{234}U (%) [a]	Higher-energy components (%)
^{238}U-I	Ionization chamber (2π)	24	90.69 ± 0.09	9.19 ± 0.09	0.12 ± 0.02
	Silicon	300	90.69 ± 0.06	9.31 ± 0.06	
	Silicon	110	90.64 ± 0.07	9.36 ± 0.07	
Average			90.67 ± 0.04		
^{238}U-II	Silicon	120	99.73 ± 0.02	0.27 ± 0.02	

[a] For ^{238}U-I, the bulk of the ^{235}U plus ^{234}U activity is ^{234}U (from Table IV, the amount expected for ^{235}U is 0.13%). The pulse analysis does not distinguish between ^{233}U and ^{234}U.

$S_{tx} = \sum(x_i - \bar{x})(t_i - \bar{t})$ and $S_{tt} = \sum(t_i - \bar{t})^2$, and the estimated intercept is $\hat{\alpha} = \bar{x} - \hat{\beta}\bar{t}$. The estimated squared error is $s^2(\hat{\beta}) = s^2/S_{tt}$, where $s^2 = [\sum(x_i - \hat{\alpha} - \hat{\beta}t_i)^2]/(n-2)$, with n = number of points. Then, from the data, $S_{tx} = 15.64$, $S_{tt} = 143.0$, $\hat{\beta} = 0.109$, $\hat{\alpha} = 822.45$, and $s = 1.37$; hence, $s(\hat{\beta}) = 1.37/\sqrt{143.0} = 0.114$. The criterion for testing whether the data are consistent with the hypothesis that $\beta = 0$ is $t = \hat{\beta}/s(\hat{\beta}) = 0.95$. Comparing with t tables for 10 d.o.f., the significance level exceeds 35%, clearly indicating no evidence of a trend.

Since there is no evidence that there was any difference between Bottle A and Bottle B, nor any evidence of a trend, the results of all 12 samples

TABLE VI. Counting data for samples from ^{238}U-I.

Sample number	Net counting rate in IGAC (counts/min [a])	Total number of counts (10^6 counts)	$\frac{1}{G_{av}}$ [b]	Disintegration rate in IGAC (dis/min)	Disintegration rate of the portion not plated [c] (dis/min)	(%)	Total disintegration rate (dis/min)	Uranium mass (mg)	Uranium specific activity (dis/min)/(mg U) [d]
				Bottle A					
1	197.46	0.83	10.921	2156.5	22.4	1.0	2178.9	2.6520	821.6 ± 0.9
2	237.67	0.95	10.919	2595.1	20.4	0.8	2615.5	3.1724	824.4 ± 0.8
3	187.51	1.02	10.910	2045.7	36.8	1.8	2082.5	2.5317	822.6 ± 0.8
4	269.65	1.14	10.911	2942.2	88.2	3.0	3030.4	3.6899	821.3 ± 0.8
5	248.71	1.05	10.911	2713.7	5.6	0.2	2719.3	3.3025	823.4 ± 0.8
6	237.52	1.67	10.907	2590.6	3.9	0.2	2594.5	3.1487	824.0 ± 0.6
								Average	822.88 ± 0.52 [e]
				Bottle B					
7	258.16	3.10	10.911	2816.8	5.7	0.2	2822.5	3.4358	821.5 ± 0.5
8	226.85	1.25	10.906	2474.0	1.8	0.1	2475.8	3.0018	824.8 ± 0.7
9	235.12	1.21	10.906	2564.2	7.0	0.3	2571.2	3.1280	822.0 ± 0.7
10	235.90	0.99	10.903	2572.0	16.6	0.6	2588.6	3.1364	825.3 ± 0.8
11	309.32	1.24	10.909	3374.4	35.9	1.1	3410.3	4.1389	824.0 ± 0.7
12	198.62	1.14	10.912	2167.3	29.2	1.3	2196.5	2.6689	823.0 ± 0.8
								Average	823.43 ± 0.62 [e]
								Grand average	823.16 ± 0.42 [f]

[a] Counting rate in IGAC, corrected for background rate.

[b] G_{av} = geometry of the sample, corrected for actual sample spread.

[c] Activity recovered from the plating solution and counted in a 2π ion chamber.

[d] Errors noted are counting errors only. Weighing, geometry, and other errors are not included.

[e] Error $s/\sqrt{6}$, calculated from $s^2 = \sum_{i=1}^{6}(x_i - \bar{x})^2/5$. For Bottle A, $s = 1.27$; for Bottle B, $s = 1.53$.

[f] Since the two averages $\bar{x}_1$ and $\bar{x}_2$ are consistent under a statistical test, the grand average is $\bar{\bar{x}} = \frac{1}{2}(\bar{x}_1 + \bar{x}_2)$, and the error $s/\sqrt{12}$ is calculated from $s^2 = \sum_{i=1}^{12}(x_i - \bar{\bar{x}})^2/11$ with $s = 1.38$.

are averaged to give the result 832.16 ± 0.42 (dis/min)/(mg uranium).

The error is the standard error of the mean $= s/\sqrt{12}$, where $s = 1.38$ is the estimated standard deviation for a single sample. It is measured solely by the scatter of the results, but does not have folded into it the estimated error in the chemical analysis. A χ^2 test of these results indicates that they are not consistent with the Poisson errors due to counting alone. This is not surprising, since it is generally true, when counting errors are reduced to very low levels by long counting, that other sources of small error become evident.

We may use the relationship $\eta_{\text{tot}}^2 = \eta_{\text{ct}}^2 + \eta_{\text{oth}}^2$, where η_{tot} is the total percentage error corresponding to the scatter of specific-activity results, η_{ct} is the percentage error due to counting error, and η_{oth} is the percentage error due to other factors, such as weighing, plating, etc. Since most of the samples were counted for about 10^6 counts, we may take η_{ct} as about 0.1%. If we estimate η_{tot} as $(s/\bar{x})100$, we find that

$$\eta_{\text{oth}} = \{[(1.38/823.)100]^2 - (0.1)^2\}^{1/2} = 0.13\%$$

For ^{238}U-II, a similar analysis gives

$$\eta_{\text{oth}} = \{[(1.03/748.)100]^2 - (0.1)^2\}^{1/2} = 0.09\% .$$

Such results are, of course, subject to statistical error. However, we may say that other errors are of the order of 0.1% per sample. Thus, with this method, by replication with 12 samples, we may expect to cut the random error in preparing and counting samples to the order of $0.15/\sqrt{12} = 0.044\%$.

To correct the results from Table VI to the specific activity of ^{238}U we refer to the data from Tables IV and V. Thus, for ^{238}U-I,

$$\text{Sp. Act. of } {}^{238}\text{U} = \frac{823.16 \times 0.9067}{0.99979}$$

$$= 746.52 \pm 0.52 \text{ dis/min mg} ,$$

including the pulse-analysis error and the mass spectrometry error, but not the error in chemical analysis. If we include the error of 0.05% for the chemical analysis, then this value becomes 746.52 ± 0.64.

TABLE VII. Counting data for samples from ^{238}U-II.

Sample number	Net counting rate in IGAC (counts/min[a])	Total number of counts (10^6 counts)	$\frac{1}{G_{\text{av}}}$[b]	Disintegration rate in IGAC (dis/min)	Disintegration rate of the portion not plated[c] (dis/min)	(%)	Total disintegration rate (dis/min)	Uranium mass (mg)	Uranium specific activity (dis/min)/(mg U)[d]
				Bottle A					
1	224.64	1.19	10.910	2450.6	16.9	0.7	2467.5	3.3031	747.0 ±0.7
2	180.59	0.98	10.914	1971.0	25.0	1.3	1996.0	2.6736	746.6 ±0.8
3	194.97	1.10	10.915	2128.1	2.3	0.1	2130.4	2.8461	748.5 ±0.7
4	191.89	1.08	10.903	2092.2	17.7	0.8	2109.9	2.8210	747.9 ±0.7
5	185.82	1.59	10.918	2028.8	9.9	0.5	2038.7	2.7267	747.7 ±0.6
6	215.07	1.01	10.916	2347.7	2.4	0.1	2350.1	3.1396	748.5 ±0.7
								Average	747.70 ± 0.29[e]
				Bottle B					
7	251.34	1.03	10.912	2742.6	9.4	0.3	2752.0	3.6884	746.1 ±0.7
8	246.86	1.07	10.912	2693.7	6.1	0.2	2699.8	3.6052	748.9 ±0.7
9	198.13	1.05	10.909	2161.4	1.2	0.1	2162.6	2.8885	748.7 ±0.7
10	203.23	1.12	10.909	2217.0	2.7	0.1	2219.7	2.9655	748.5 ±0.7
11	194.20	1.11	10.915	2119.7	16.2	0.8	2135.9	2.8601	746.8 ±0.7
12	201.52	1.72	10.910	2198.6	2.8	0.1	2201.4	2.9379	749.3 ±0.6
								Average	748.05 ± 0.52[e]
								Grand average	747.88 ± 0.30[f]

[a] Counting rate in IGAC, corrected for background rate.

[b] G_{av} = geometry of the sample, corrected for actual sample spread.

[c] Activity recovered from the plating solution and counted in a 2π ion chamber.

[d] Errors noted are counting errors only. Weighing, geometry, and other errors are not included.

[e] Error $s/\sqrt{6}$, calculated from $s^2 = \sum_{i=1}^{6} (x_i - \bar{x})^2/5$. For Bottle A, $s = 0.78$; and for Bottle B, $s = 1.29$.

[f] Since the two averages $\bar{x}_1$ and $\bar{x}_2$ are consistent under a statistical test, the grand average is $\frac{1}{2}(\bar{x}_1 + \bar{x}_2)$, and the error $s/\sqrt{12}$ is calculated from $s^2 = \sum_{i=1}^{12} (x_i - \bar{\bar{x}})^2/11$ with $s = 1.03$.

TABLE VIII. Constants used in calculating half-life.

Atomic weight ^{238}U = 238.05 [a]
Atomic weight ^{235}U = 235.04
Avogadro's number = 6.0226 × 10^{23} per mole
1 yr = 5.2595 × 10^{5} min

[a] C^{12} as standard.

The data for ^{238}U-II are given in Table VII. Applying the same tests as above, $\delta\bar{x} = 0.35$, $s(\delta\bar{x}) = \sqrt{2}s/\sqrt{6} = 1.03/\sqrt{3} = 0.594$; then $t = 0.35/0.594 = 0.59$, corresponding to a significance level >50%. For the trend test, $S_{tx} = 16.70$, $S_{tt} = 143.0$, $\hat{\beta} = 0.117$, $\hat{\alpha} = 747.12$, $s = 0.98$, $s(\hat{\beta}) = 0.98/\sqrt{143.0} = 0.082$. With the test criterion $t = 0.117/0.082 = 1.42$, the significance level is almost 20%. Neither test indicates a lack of consistency, so all 12 results are averaged to give

$$\text{Sp. Act. of } {}^{238}\text{U} = \frac{747.88 \times 0.9973}{1.0000}$$

$$= 745.86 \pm 0.34 \text{ dis/min mg },$$

excluding chemical-analysis error. With a chemical-analysis error of 0.05%, the result becomes 745.86 ± 0.50. Clearly, the results of both experiments are consistent, and so average to

$$\text{Sp. Act. of } {}^{238}\text{U} = 746.19 \pm 0.41 \text{ (dis/min)/(mg } {}^{238}\text{U)} .$$

From the constants in Table VIII, this corresponds to

$$T_{1/2}({}^{238}\text{U}) = (4.4683 \pm 0.0024) \times 10^{9} \text{ yr} .$$

VII. SPECIFIC ACTIVITY AND HALF-LIFE OF ^{235}U

Only one batch of ^{235}U was measured. The isotopic composition is given in Table IX. The pulse analysis, measured with a silicon detector, is summarized in Table X and illustrated in Fig. 5. The counting procedure was the same as for ^{238}U samples and the counting results are given in Table XI.

The tests described above were also applied here. The difference between the means of the two bottles is $\delta\bar{x} = (\bar{x}_2 - \bar{x}_1) = 5.01$, and the estimated standard error of the difference is $s(\delta\bar{x}) = 4.80(\frac{1}{5} + \frac{1}{6})^{1/2} = 2.91$ (footnote f, Table XI). Then the test criterion $t = 5.01/2.91 = 1.72$, when compared with t tables for 10 d.o.f., indicates that the difference is significant at the 12% level. Now we consider the presence of a trend. If we look at the counting rates of the samples prepared from both bottles as a function of their number (corresponding to the order of preparation), there is a definite increasing trend, except for the drop at No. 11. Applying the quantitative test as above, $S_{tx} = 91.90$, $S_{tt} = 110.0$, $\hat{\beta} = 91.9/110.0 = 0.835$, $\hat{\alpha} = 4795.00$, and $s = 4.03$, so $s(\hat{\beta}) = 4.03/\sqrt{110.0} = 0.384$. With the test criterion $t = 0.835/0.384 = 2.18$ the significance level is found to be about 6% from the tables (nine d.o.f.).

The two tests are not independent; they are, in fact, highly correlated.[14] The significance level is such as to indicate that a slight source of drift exists. We have attempted to hypothesize possible causes, but have not arrived at a plausible explanation. The chemical analyses (Table III) indicate no difference between Bottles A and B. Counter drift is not a possible cause, since samples were counted in a different order from the order of sample preparation (hence, number). They were also recounted, in a second round, after all were originally counted. The results checked, with no indication of a drift. Another possible explanation is evaporation of water during the weighing out of samples. This would require that the solution become increasingly concentrated as the sample aliquotting proceeded. Against this is the evidence from the samples taken for chemical analysis. These were withdrawn in the sequence: Bottle A: chemical analysis sample (C.A.) 1, counting sample 1, 2, 3, 4, 5, 6, C.A. 2; Bottle B: C.A. 3, counting sample 7, 8, 9, 10, 11, C.A. 4. The results in Table III indicate no trend in the results from chemical-analysis samples 1 to 4. Since all the aliquots were made on the same morning, but were molecular-plated over a period of days, one might expect that the uranium could have deposited out or entered an unplatable form, and such a reaction might have occurred slowly with time. Against such an hypothesis is the fact that: (1) The isopropyl alcohol was evaporated, leaving behind any nonplated uranium for counting in the 2π cham-

TABLE IX. Isotopic composition of ^{235}U sample.

Isotope	at.%
^{238}U	0.000 87 ± 0.000 02
^{236}U	0.000 238 ± 0.000 007
^{235}U	99.998 86 ± 0.000 02
^{234}U	0.000 0087 ± 0.000 0009
^{233}U	0.000 023 ± 0.000 006

TABLE X. Pulse analysis of ^{235}U sample. Using a Si solid-state detector and an 80-μg/cm^{2} sample.

^{235}U	99.960 ± 0.004
^{234}U ^{233}U	0.040 ± 0.004 [a]

[a] The two isotopes were not resolvable.

ber; (2) the tube was rinsed with nitric acid, to pick up any precipitated uranium. Further, such reactions would at most cause a decrease in count rate for the later samples rather than the observed increase.

In the absence of a plausible explanation, we average $\bar{x}_1$ and $\bar{x}_2$. However, we arbitrarily inflate the observed error by the factor 1.5 to allow for the apparent trend. Were $s(\delta\bar{x})$ and $s(\hat{\beta})$ increased by this factor, the observed difference and trend would not have been significant. Excluding the error due to chemical analysis, the specific activity is

$$\frac{4800.01\times 0.9996}{0.999\,99}=4798.1\pm 2.2\ (\mathrm{dis/min})/(\mathrm{mg}\ ^{235}\mathrm{U}).$$

Taking the chemical-analysis error of 0.05% into account,

$$\text{Spec. Act. of }^{235}\mathrm{U}=4798.1\pm 3.3\ (\mathrm{dis/min})/(\mathrm{mg}\ ^{235}\mathrm{U}).$$

From the constants in Table VIII, this corresponds to

$$T_{1/2}(^{235}\mathrm{U})=(7.0381\pm 0.0048)\times 10^{8}\ \mathrm{yr}\ .$$

VIII. OTHER MEASUREMENTS AND SUMMARY

A. Nuclide ^{238}U

Kovarik and Adams (1932, 1955).[15,16] This measurement was first reported in Ref. 15. The material used was natural uranium, presumably in the form U_3O_8. It had been left by a deceased chemist who had labeled it "very pure U_3O_8." Sample preparation involved slow settling of very fine particles out of chloroform and subsequent weighing of the dried sample. Average sample thicknesses were 0.4- to 1.6-mg/cm² U_3O_8. An ionization chamber was used for counting with a carefully drilled plate of collimator holes over the samples. Each hole, in effect, served as a low-geometry counter whose geometry factor could be calculated. Thus, backscattering and sample self-absorption were eliminated. These samples were later recounted to a total of 3×10^6 counts, and yielded the value 1503 (dis/min)/(mg natural U).

Schiedt (1935).[17] The specific activity of natural uranium was measured. Two samples of surface density 1 mg/cm² were electroplated and were dis-

TABLE XI. Counting data for samples from ^{235}U batch.

Sample number	Net counting rate in IGAC (counts/min [a])	Total number of counts (10^6 counts)	$\frac{1}{G_{av}}$ [b]	Disintegration rate in IGAC (dis/min)	Disintegration rate of the portion not plated [c] (dis/min)	(%)	Total disintegration rate (dis/min)	Uranium mass (mg)	Uranium specific activity (dis/min)/(mg U) [d]
				Bottle A					
1	1173.33	6.69	10.917	12809.2	30.6	0.2	12839.8	2.6796	4791.7 ±1.9
2	1042.36	2.92	10.916	11378.4	44.8	0.4	11423.2	2.3818	4796.0 ±2.8
3	782.08	4.31	10.921	8541.1	4.0	0.05	8545.0	1.7808	4798.4 ±2.3
4	1000.90	2.80	10.914	10923.8	8.9	0.1	10932.7	2.2797	4795.7 ±2.9
5	851.53	2.39	10.918	9297.0	17.8	0.2	9314.8	1.9389	4804.2 ±3.1
6	1069.33	3.00	10.921	11678.2	6.2	0.05	11684.4	2.4341	4800.3 ±2.8
								Average	4797.73 ±1.76 [e]
				Bottle B					
7	835.41	2.51	10.920	9122.7	2.8	0.03	9125.5	1.8993	4804.6 ±3.1
8	990.87	6.94	10.922	10822.3	7.7	0.1	10830.0	2.2550	4802.6 ±1.8
9	853.64	4.79	10.917	9319.2	13.7	0.1	9332.9	1.9435	4802.1 ±2.2
10	873.58	9.36	10.916	9536.0	28.9	0.3	9564.9	1.9894	4807.9 ±1.6
11	878.31	4.75	10.916	9587.6	21.7	0.2	9609.3	2.0034	4796.5 ±2.2
								Average	4802.74 ±1.86 [e]
								Grand average	4800.01 ±1.45 [f]

[a] Counting rate in IGAC, corrected for background rate.

[b] G_{av} = geometry of the sample, corrected for actual sample spread.

[c] Activity recovered from the plating solution and counted in a 2π ion chamber.

[d] Errors noted are counting errors only. Weighing, geometry, and other errors are not included.

[e] Error $s/\sqrt{6}$ and $s/\sqrt{5}$ calculated from $s^2=\sum_{i=1}^{r}(x_i-\bar{x})^2/(r-1)$, with $r=5$ or 6. For Bottle A, $s=4.30$; and for Bottle B, $s=4.17$.

[f] A statistical test of the difference between the two averages $\bar{x}_1$ and $\bar{x}_2$ shows significance at the 12% significance level. However, since the chemical analyses of the two bottles shows no difference, we take a grand average $\bar{\bar{x}}=\sum x_i/11$ and the error $s/\sqrt{11}$ is calculated from $s^2=\sum_{i=1}^{11}(x_i-\bar{\bar{x}})^2/10$, with $s=4.80$.

solved and chemically analyzed after counting. Counting was done in an intermediate-geometry α counter with a geometry factor of about 8.3%. The sample spread was assumed to be uniform and a relation analogous to Eq. (3) was used to determine the disintegration rate. The ratio $G_S/G_{P'}$ was 0.988. The measured specific activity was 1517 ± 15 dis/min mg; the author reported a value $T_{1/2}(^{238}\mathrm{U}) = (4.42 \pm 0.03)\,10^9$ yr. (See Table XII.)

Curtiss, Stockman, and Brown (1941).[18] In attempting to develop a method for isotopic analysis, the authors measured the specific activity of natural uranium. A uranium compound, sprayed onto a platinum disk, was ignited to glowing in a bunsen burner flame, presumably to U_3O_8, and weighed. Samples were prepared with average surface densities D_S ranging from 0.96 to 1.84 mg/cm^2 of U_3O_8. The α activity was measured in a 2π counter and S, the counting rate per mg of U_3O_8 was plotted as a function of D_S. Straight-line extrapolation to zero D_S yielded the value $S_0 = 10.608$ counts/sec. Assuming the chemical composition to be U_3O_8 and the counting efficiency 0.500, the specific activity was quoted as 1501 dis/min mg.

There are a number of questions that might be raised concerning this measurement. As an example, can such thick samples be treated as smooth flat deposits? Indeed in a later comment[19]: "Microscopically the films are found to consist of a mass of black particles with the appearance of a very smooth pile of coal." The extrapolation to S_0 is over a large range of D_S. The effect of backscattering (discovered after the time of this experiment) is, of course, not considered at all.

Kienberger (1949).[20] Measurements were made on highly depleted uranium and on natural uranium. The depleted uranium contained $(5 \pm 3) \times 10^{-8}$ parts of ^{234}U; hence required a very small correction for its activity. Samples were prepared by electroplating known weights of uranium and subsequently analyzing for residual uranium in the residual solution. A 2π ionization chamber counter was used, with an estimated backscattering factor of 1.26% on nickel. The specific activity as a function of sample surface density was extrapolated to zero sample weight. The depleted uranium gave a ^{238}U specific activity of 742.7 ± 1.6 (dis/min)/(mg ^{238}U). For natural uranium, the specific activity was 1502 ± 1.5 (dis/min)/(mg matural U).

Leachman and Schmitt (1957).[21] The measurement of the ^{238}U half-life was incidental to the main purpose of the experiment (neutron cross section of ^{238}U), so details are sparse. Depleted ^{238}U was used with a ^{238}U/^{235}U ratio of 10^5, a decrease in the ^{235}U content by a factor of more than 700. Since the ^{234}U content was depleted even more, its α activity (as well as that of the ^{235}U) was negligible. Uranium nitrate samples, painted onto platinum plates, were ignited to U_3O_8 and weighed; the average sample surface density was about 0.2 mg/cm^2. A 2π ionization chamber counter was used, and "the usual corrections for finite thickness of the source and scattering from the

TABLE XII. Measurements of ^{238}U half-life.

Author	Reference	Counter	Material	Measured specific activity (dis/min mg)	Specific activity of ^{238}U [a]	^{238}U half-life (units of 10^9 yr)
Kovarik and Adams (1932, 1955)	15, 16	Grid collimator ion chamber	Natural U	1503 ± 6	739.7 ± 3.0	4.508 ± 0.018
Schiedt (1935)	17	Intermediate-geometry ion chamber	Natural U	1517 ± 15	747 ± 8	4.46 ± 0.05
Curtiss *et al.* (1941)	18	2π ion chamber	Natural U	1501 ± 3	738.7 ± 1.5	4.514 ± 0.009
Kienberger (1949)	20	2π ion chamber	Natural U	1502 ± 1.5	739.2 ± 0.7	4.511 ± 0.005
Kienberger (1949)	20	2π ion chamber	^{238}U	742.7 ± 1.6	742.7 ± 1.6	4.489 ± 0.010
Leachman and Schmitt (1957)	21	2π ion chamber	^{238}U	728.9 ± 4.8 [b]	728.9 ± 4.8	4.57 ± 0.03
Steyn and Strelow (1959)	22	Liquid scintillator	Natural U	1519.7 ± 2.4	748.1 ± 1.2	4.457 ± 0.007
Present work (1971)		Intermediate-geometry proportional counter	^{238}U	746.19 ± 0.41	746.19 ± 0.41	4.4683 ± 0.0024

[a] Measured for depleted uranium; recalculated from Eq. (8), for natural uranium.

[b] No details are given in Ref. 21. The specific activity was calculated from the quoted half-life using the constants of Table VIII.

backing were made." Weighing of U_3O_8 checked within 1% with coulometric assay. The half-life result was given as $(4.56 \pm 0.03)\ 10^9$ yr.

Steyn and Strelow (1959).[22] The specific activity of natural uranium was determined by counting samples dissolved in a liquid scintillator. Because an organic scintillator does not allow discrimination between α and β particles, the daughter β activities (^{234}Th, or UX_1, and ^{234}Pa, or UX_2) were chemically separated. Analysis was carried out by precipitation of the uranium in a measured aliquot, followed by ignition to U_3O_8 and weighing. A small correction was made for growth of daughter activity during the sample counting. Results were 1519.7 ± 2.4 (dis/min)/(mg natural U); the half-life was calculated as $(4.45 \pm 0.01)\ 10^9$ yr.

Summary. The various results are summarized in Table XII. The experiments which yielded the specific activity of natural uranium (S_{nat}) have been recalculated using the relationship

$$S_{238} = \frac{0.5(S_{\text{nat}} - \theta_{235} S_{235})}{\theta_{238}}, \tag{8}$$

where S_i = specific activity (dis/min mg) of the ith component and θ_i is its weight fraction = (weight %)/100. The measured atomic percentages are listed in the second column of Table XIII and the corresponding calculated weight percentages in the third column. The value of S_{235} used is that measured by us (Sec. VII), 4798 (dis/min)/(mg ^{235}U). With the constants in Table VIII, the corresponding half-life values are given in the last column of Table XII.

It is clear that in a number of cases the discrepancies exceed the error limits set by the authors. This is not surprising for those experiments using 2π counting, since corrections for backscattering and sample self-absorption are difficult to make with accuracy. The measurement of Refs. 15, 16 is subject to other potential sources of systematic error. The very close checks of the first, third, and fourth measurements[23,24] are reminiscent of the similar close checks of the pre-World-War-II measurements of c, the velocity of light. (These subsequently turned out to be many standard deviations away from later more accurate c measurements.) Only the measurement of Ref. 22 is reasonably free of possible systematic errors. However, since the measurement was carried out for natural uranium, calculation of the ^{238}U half-life depends upon the constants of Table VIII and on the measured ^{235}U half-life.

TABLE XIII. Composition of natural uranium.

Isotope	(at.%) [a]	Weight (%)
^{238}U	99.2747	99.2838
^{235}U	0.7196	0.7106
^{234}U	0.0057	0.0056

[a] See Ref. 24; and B. R. Grundy and A. N. Hamer [J. Inorg. Nucl. Chem. 23, 148 (1961)], M. Lounsbury [Can. J. Chem. 34, 259 (1956)], and F. A. White, T. L. Collins, and F. M. Rourke [Phys. Rev. 101, 1786 (1956)].

B. Nuclide ^{235}U

Nier (1939).[25] Using uranium ores whose ages had been determined by measuring ^{207}Pb/^{235}U and ^{206}Pb/^{238}U ratios, Nier used the measured ^{206}Pb/^{207}Pb ratios as a function of age to evaluate the present-day ratio R = (activity of ^{235}U)/(activity of ^{238}U), finding it to have the value 0.046 ± 0.001, from which he calculated the half-life of ^{235}U to be $(7.13 \pm 0.15)\ 10^8$ yr.

Sayag (1951).[26] By energy analysis in an ion chamber, the author measured the α activity in natural uranium due to ^{235}U. Measurements of the ^{235}U peak gave the activity ratio $R = A_{235}/A_{238} = 0.0408 \pm 0.0015$. Using the data of Perlman, Ghiorso, and Seaborg on the energy distribution of ^{235}U α activity, Sayag corrected for the ^{235}U α-particle peak lost under the ^{234}U peak, giving $R'_{\text{corr}} = A_{235}/A_{238} = 0.0455 \pm 0.0021$. From this was calculated $T_{1/2}(^{235}\text{U}) = (7.07 \pm 0.33)\ 10^8$ yr. The work of Wurger, Meyer, and Huber[27] notes that the correction for lost peaks was not adequately made and that a better value was $R_{\text{corr}} = 0.0479 \pm 0.0021$, yielding $T_{1/2}(^{235}\text{U}) = (6.82 \pm 0.29)\ 10^8$ yr.

Fleming, Ghiorso, and Cunningham (1952).[28] This was a carefully carried out measurement of the ^{235}U specific activity. Aliquots from a solution prepared from weighed U_3O_8 were electroplated and counted in an intermediate-geometry counter of measured dimensions. The disintegration rate was calculated assuming the surface density to be uniform. Comparison of this counter with a low-geometry counter indicated a discrepancy of 0.5%. This would have been of concern except for the fact that another error dominated. Energy analysis in an ionization chamber was used to determine the activity content of the ^{234}U present; this was found to be 34.7%. The authors assigned an error of 2% in the half-life arising from the error in the energy analysis. The results were $(4.74 \pm 0.10)\ 10^3$ (dis/min)/(mg ^{235}U) for the specific activity, and $(7.13 \pm 0.16)\ 10^8$ yr for the half-life.

Knight (1950).[29] Using the same material as in Ref. 28, Knight measured the specific activity as in Ref. 28, but carried out a poorer energy analysis for the ^{234}U α-activity content. The specific activities of the samples measured in Refs. 28 and 29 checked each other. Reference 28 corrected

TABLE XIV. Measurements of ^{235}U half-life.

Author	Reference	Method	Material	Measured activity ratio [a] $\frac{A_{235}}{A_{238}} = R$	Specific activity of ^{235}U [b]	^{235}U Half-life (units of 10^8 yr)
Nier (1939)	25	R measured from Pb/U ratios	Natural uranium	0.046 ±0.001	4800 ±100	7.04 ±0.15
Sayag (1951)	26	R measured by ion chamber energy analysis	Natural uranium	0.0467 ±0.0017	4870 ±180	6.94 ±0.25
Fleming, Ghiorso, and Cunningham (1952)	28	Specific activity	Enriched ^{235}U		4740 ±100	7.12 ±0.16
Knight (1950)	29	Specific activity	Enriched ^{235}U		4753 ±100 [c]	7.10 ±0.16
Würger, Meyer, and Huber (1957)	27	R measured by ion chamber energy analysis	Natural uranium	0.046 77 ± 0.000 73	4880 ±80	6.92 ±0.11
Deruytter, Schroder, and Moore (1965)	30	R measured with a Si detector	Natural uranium	0.046 45 ± 0.000 60	4845 ±50	6.97 ±0.07
White, Wall, and Pontet (1965)	31	Specific activity	Enriched ^{235}U		4741 ±60	7.12 ±0.09
Banks and Silver (1966)	32	R measured from Pb/U ratios	Natural uranium		4809^{-20}_{+50}	$7.022^{-0.029}_{+0.073}$ [d]
Present work (1971)		Specific activity	Enriched ^{235}U		4798.1 ± 3.3	7.0381 ± 0.0048

[a] All activity ratios measured by energy-pulse analyses (i.e., except Ref. 25) have been corrected to the value 87.4% for the fraction of the ^{235}U α activity not hidden by ^{238}U or ^{234}U peaks.

[b] Measured for enriched uranium; calculated from Eq. (9) for experiments with R measurement.

[c] Corrected, as in Ref. 28.

[d] Corrected, using our value of $T_{1/2}(^{238}\text{U})$.

Knight's result using the more accurate energy analysis of Ref. 28. The results were (4753 ± 100) (dis/min)/(mg ^{235}U) for the specific activity and $(7.10 \pm 0.16)\ 10^8$ yr for the half-life.

Wurger, Meyer, and Huber (1957).[27] Using energy analysis in an ion chamber, the authors measured the ^{235}U α activity in natural uranium. The authors checked Ghiorso's proposed decay scheme with α-γ coincidence measurements and used this to correct for the ^{235}U α peaks lost under the ^{234}U and ^{238}U peaks. Taking the "exposed" ^{235}U to be 85.6% of the total, then $R_{\text{corr}} = A_{235}/A_{238} = 0.047\,75 \pm 0.000\,75$, yielding $T_{1/2}(^{235}\text{U}) = (6.84 \pm 0.11)\ 10^8$ yr.

Deruytter, Schroder, and Moore (1965).[30] Using energy analysis with a Si solid-state-junction detector, the authors measured the ^{235}U α activity in natural uranium. $R_{\text{meas}} = A_{235}/A_{238} = 0.0406 \pm 0.0005$ was measured for the exposed peaks. Using Ghiorso's decay scheme and assuming this to have 0.2% error, correction for the ^{235}U peaks lost under the ^{234}U and ^{238}U peaks amounted to 14.4% of the total ^{235}U activity. This resulted in $R_{\text{corr}} = 0.0474 \pm 0.0006$ and a half-life value $(6.92 \pm 0.09)\ 10^8$ yr.

White, Wall, and Pontet (1965).[31] This measurement involved α-counting assayed samples of enriched ^{235}U, followed by energy analysis to extract the ^{235}U fraction. Assay was through weighing the sample plates, followed by spectrophotometry or coulometry of the individual samples after α counting; back-to-back fission counting was also used. Errors in uranium mass assay were about ±0.8%. A low-geometry counter was used for α counting. A solid-state detector was used to determine the ^{235}U spectrum. For natural uranium, the fraction of the ^{235}U activity not hidden under the ^{234}U and ^{238}U peaks was found to be 87.4%, agreeing with Baranov's magnetic-spectrograph work (87%). The specific activity was measured as 4741 ± 60 (dis/min)/(mg ^{235}U), or $T_{1/2}(^{235}\text{U}) = (7.13 \pm 0.09)\ 10^8$ yr. It is noted here that the result of Ref. 30 would be changed to $(7.03)\ 10^8$ yr if the exposed ^{235}U activity value, 87.4%, were used rather than Ghiorso's 85.6%.

Banks and Silver (1966).[32] As in the case of Nier's measurement,[25] lead and uranium isotopes in uranium-containing minerals were measured. Relatively young minerals ($\sim 10^8$-yr age) were used in order to minimize the effect of partial leaching out of the lead component. As above, the experiment included measurement of the isotopic ratio $^{207}\text{Pb}/^{206}\text{Pb}$, which is equivalent to evaluation of R, the activity ratio of ^{235}U and ^{238}U. The authors used the value $T_{1/2}(^{238}\text{U}) = 4.509 \times 10^9$ yr and the concentration of ^{235}U in natural uranium given in Table XIII. Using estimated limits on the possible lead loss and also including other errors, the authors quote their result as: $7.086^{-0.029}_{+0.073} \times 10^8$ yr.

Summary. The various results are summarized in Table XIV. The experiments which yielded a value of $R = A_{235}/A_{238}$ have been recalculated using the relation

$$T_{1/2}(^{235}\text{U}) = T_{1/2}(^{238}\text{U})\,\frac{1}{R}\,\frac{N_{235}}{N_{238}}, \tag{9}$$

where $T_{1/2}(^{238}\text{U})$ is taken as 4.468×10^9 yr and N_{235}/N_{238} is given in Table XIII. Specific activities are calculated using the constants of Table VIII. Values of R have been recalculated using the value 87.4% (from Ref. 31) for the value of the fraction of ^{235}U peaks which are exposed.

IX. ACKNOWLEDGMENTS

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Parity-Violating Asymmetry of the 501-keV Gamma Ray Emitted in the Decay of ^{180m}Hf[†]

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The parity-violating forward-backward asymmetry of the 501-keV γ radiation emitted by ^{180m}Hf polarized at low temperatures is measured to be $-(1.66 \pm 0.18)\%$ at a polarization of 72%. From this asymmetry the magnitude of the irregular $\tilde{E}2/M2$ mixing ratio is deduced to be 0.038 ± 0.004, in good agreement with results of circular polarization measurements. The regular $E3/M2$ multipole mixing ratio of the 501-keV γ ray is deduced to be 5.3 ± 0.3, in agreement with results of directional-correlation measurements. The hyperfine splitting energy of ^{180m}Hf introduced as an impurity in $ZrFe_2$ is deduced to be $-(7.9 \pm 0.5)$ mK.

I. INTRODUCTION

The current-current theory of the weak interaction[1, 2] indicates that nuclear levels may not be states of pure parity, and that there exist small admixtures of wave functions of parity opposite to that of the strong interaction. The relative amplitude of this admixture is estimated[3] to be the order of 10^{-7} the parity-conserving strong interaction, and thus the parity-violating component is expected to produce very small experimental effects. Comprehensive reviews of experiments demonstrating parity-violating effects are given by Hamilton[4] and Henley.[5]

Considerations of nuclear-structure effects may lead to enhancement of the size of the parity-violating experimental effect. The close presence of nuclear states of opposite parity and identical spin may enhance the parity admixture in the states; the retardation of the regular parity-conserving transitions by nuclear-structure effects can often result in a larger laboratory effect. Thus in a