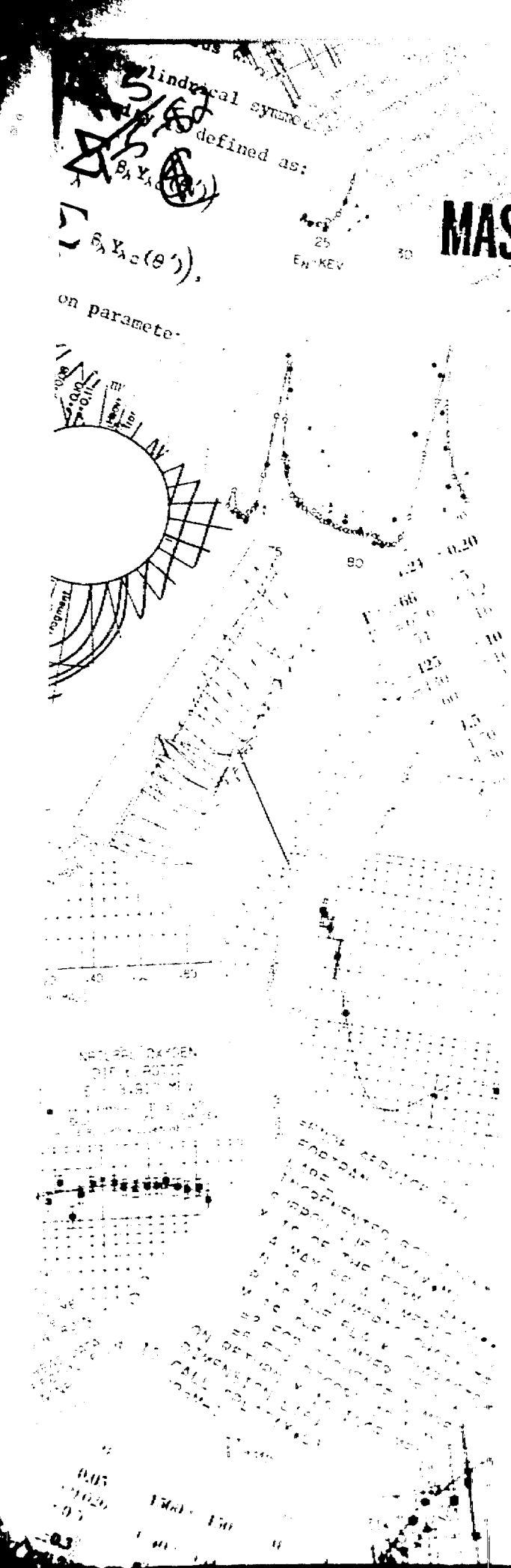

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THE URANIUM HALF-LIVES: A CRITICAL REVIEW

Norman E. Holden

January 1981

INFORMATION ANALYSIS CENTER REPORT

NATIONAL NUCLEAR DATA CENTER
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UPTON, NEW YORK 11973

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UNITED STATES DEPARTMENT OF ENERGY

ABSTRACT

This report evaluates the experimental data and recommends values for the spontaneous fission half-life of ^{238}U, and the total half-lives for ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{236}U, and ^{238}U. The report also discusses the variation of the isotopic abundance of ^{234}U in nature and the error involved in the assumption of secular equilibrium between ^{234}U and ^{238}U in the determination of the specific activity of natural uranium samples.

The recommended half-life values and 95% confidence limits are: ^{238}U spontaneous fission: $8.09 \pm 0.26 \times 10^{15}$ years; ^{232}U total: 69.8 ± 1.0 years; ^{233}U total: $1.592 \pm 0.002 \times 10^{5}$ years; ^{234}U total: $2.454 \pm 0.006 \times 10^{5}$ years; ^{235}total: $7.037 \pm 0.011 \times 10^{8}$ years; ^{236}U total: $2.342 \pm 0.003 \times 10^{7}$ years ^{238}U total: $4.468 \pm 0.005 \times 10^{9}$ years.

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I. INTRODUCTION

The half-lives (partial or total) of the uranium isotopes are of interest for many applications. The total half-lives are of interest for standards purposes and the spontaneous fission half-life of ^{238}U is used in the assay of low-enrichment uranium oxide in bulk containers for safeguards purposes via spontaneous fission neutrons.

The impetus for this study is a re-evaluation of the thermal neutron cross sections and related parameters of the uranium and plutonium fissile nuclides. The half-lives are required to determine the amounts of materials from the measured activities of the nuclides.

A number of measurements of the ^{234}U and the ^{238}U half-lives have been based on the assumption of secular equilibrium; i.e., the much shorter-lived ^{234}U daughter has the same specific activity in a sample as it's longer-lived ^{238}U parent (see Appendix I). However, knowledge of the isotopic abundance variation of ^{234}U in nature[1,2] (see Appendix II) and the $^{234}U/^{238}U$ disequilibrium studies in groundwater since 1955[3] imply that caution should be exercised in utilizing the assumption of secular equilibrium in half-life measurements. Unless a separate determination of the isotopic composition of a sample is made, measurements on natural uranium may not provide an accurate value for the half-life of ^{234}U or ^{238}U.

In the following review, the various uranium nuclides are discussed in turn and the significant measurements are described and assessed. An iterative procedure is used whereby the evaluation and recommended values for other isotopes are utilized to update a given half-life, which in turn is used to update the measurements for the other isotopes.

It should be noted that in the following tables, the uncertainty at the 95% confidence level has been listed. It was determined from the author's quoted uncertainty.

The various constants and masses assumed for conversion purposes in this study are discussed in Appendix III. In general, the specific activity has been the measured quantity in the experiments discussed and the values from Appendix III are used to update the half-life value quoted in each experiment.

II. ^{232}U

Sellers[4] used isotope dilution mass spectrometry and alpha proportional counting to determine the activity of a ^{238}U sample containing 0.14% ^{232}U. The specific activity of ^{232}U was found to be 4.65×10^{10} disintegrations per minute per milligram (dpm/mg) (corresponding to a half-life 73.56 ± 1.48 years at the 68% confidence level or 1 standard deviation). The ^{232}U daughter activity was corrected for.

Chilton[5] used both alpha proportional counting and calorimetry to determine the specific activity of a sample of 99.2% enriched ^{232}U. There was a 1.1% difference between Chilton's two results. The weighted mean was $4.772 \pm 0.057 \times 10^{10}$ dpm/mg (corresponding to a half-life 71.67 ± 0.86 years at the 95% confidence limit or 2 standard deviations). ^{232}U daughter activity was corrected for.

Aggarwal[6] determine the specific activity of a ^{238}U sample containing 16.5% ^{232}U using liquid scintillation counting and alpha proportional counting, masses were determined by isotope dilution mass spectrometry. $^{232}U/^{233}U$ relative activity was also used to determine the specific activity of ^{232}U. The specific activity from liquid scintillation counting and alpha counting was $4.96 \pm 0.03 \times 10^{10}$ dpm/mg (corresponding to a half-life of 69.0 ± 0.4 years). The half-life from the $^{232}U/^{233}U$ activity ratio was 68.8 ± 0.4 years. The standard error of the mean and the standard deviations reported have been converted to a 95% confidence limit and the half-life from Aggarwal is 68.90 ± 0.56 years.

The weighted average for the ^{232}U half-life from the three measurements described above is 69.78 ± 0.99 years.

Table 1 ^{232}U Half-Life

Author (year)	Reference	Total Half-Life Value (years)	Method	Comment
Sellers (1954)	4	73.6 ± 3.0	α Proportional counting	
Chilton (1964)	5	71.7 ± 0.9	α Proportional counting and coulometry	50% geometry
Aggarwal (1979)	6	68.90 ± 0.56	α Proportional counting, liquid scintillation counting and $^{232}U/^{233}U$ relative activity	
Holden (1980)		69.8 ± 1.0	Weighted average	Recommended value

III. ^{233}U

Up until 1974, there had been two groups of measurements on the ^{233}U half-life which differed by 5%. It had not been possible to explain the discrepancy between these groups. Durham[7] reported a preliminary measurement on alpha proportional counting with small solid angle, which was intermediate between the two previous groups. However this report has never been published and it, along with all earlier measurements, has been discarded. This evaluation is based on the four subsequent measurement described below. The measurements used highly enriched samples of ^{233}U (98.1% to 99.99% ^{233}U).

Jaffey [8] measured the specific activity of a 99.5% enriched sample of ^{233}U using intermediate geometry alpha proportional counting. ^{238}Pu (later corrected for), ^{232}U and ^{234}U contributed to the activity. Silicon junction detectors were used for the pulse height energy analysis. The specific activity reported was 21405 ± 20 dpm/μgram^{233}U, (corresponding to a half-life of 1.5910 ± 0.0015 x 10^5 years with the uncertainty for the standard error of the mean).

Vaninbroukx[9] determined the specific activity of samples enriched to 99.762% and 99.9986% ^{233}U by alpha counting in low geometry, in a liquid scintillator and by 4π proportional counting. Isotope dilution mass spectrometry was used to determine the ^{233}U content as well as controlled potential coulometry. The resulting half-life is $1.5925 \pm 0.0040 \times 10^5$ years, where the error is the 99.7% confidence limit.

Geidel'man[10] measured the specific activity of a 98.112% enriched ^{233}U sample using 4π alpha-X coincidence. Potentiometric titration was used to determine the uranium concentration in the sample. The result obtained is $1.5937 \pm 0.0022 \times 10^5$ years at the 95% confidence limit.

Aggarwal[65] measured the specific activity of a 99.7% enriched ^{233}U sample by liquid scintillation counting and alpha proportional counting. Isotope dilution mass spectrometry was used to determine the uranium concentration of the sample. The result obtained was $1.5885 \pm 0.0085 \times 10^5$ years at the 95% confidence limit.

The weighted average of these four measurements of the ^{233}U half-life is $1.5922 \pm 0.0017 \times 10^5$ years at the 95% confidence level.

Table 2 ^{233}U Half-life

Author (year)	Reference	Total Half-life Value (10^5 years)	Method	Comment
Jaffey (1974)	(8)	1.5910 ± 0.0030	α proportional counting	Intermediate geometry
Vaninbroukx (1976)	(9)	1.5925 ± 0.0027	Liquid scintillaton +4πα proportional counting	Low geometry
Geidel'man (1979)	(10)	1.5937 ± 0.0033	4πα - x coincidence	*
Aggarwal (1980)	(65)	1.5885 ± 0.0085	Liquid scintillation +4πα proportional counting	
Holden (1980)		1.592 ± 0.002	Weighted average	Recommended Value

*Geidel'man's value has the error increased by 50% because some details are missing e.g. the measured specific activity, the mass of ^{233}U assumed, the mass spectrometric analysis, etc.

IV ^{234}U

Most of the early measurements on the ^{234}U half-life were based on the assumption of secular equilibrium between the ^{234}U and ^{238}U specific activity in a natural sample of uranium. As noted in Appendices I and II, the variation of ^{234}U in natural uranium ore concentrates invalidates the assumption of secular equilibrium. Smith and Jackson [11] found an 8% variation in the ^{234}U content of natural uranium ore concentrates from sixteen world sources, while Shields[12] noted ^{234}U abundance variations from a 59% deficiency to a 72% excess in uranium found in sandstone deposits in the United States.

One could, in fact, use best estimates of the uranium half-lives, and a measured specific activity of a uranium sample to derive an approximate

estimate of the weight percent of ^{234}U in the sample. Since there was no associated information on the ^{234}U content in the natural samples of these early measurements, they have been uniformly ignored in this evaluation.

Fleming[13] determined the specific activity of a ^{234}U sample enriched to 95.99%. No pulse height analysis was performed since the ^{235}U and ^{238}U activity would be negligible. Alpha counting in medium geometry was used to determine the specific activity to be $1.370 \pm 0.009 \times 10^7$ dpm/mg ^{234}U. At the 95% confidence limit, the half-life becomes $2.475 \pm 0.032 \times 10^5$ years. White[14] measured the specific activity of two enriched ^{235}U samples that had significant amounts of ^{234}U. One sample containing 1.19% ^{234}U was destructively assayed to determine the isotopic composition and provided a half-life of $2.458 \pm 0.031 \times 10^5$ years. The second sample, enriched to 0.116% ^{234}U, used back-to-back fission counting to determine the isotopic composition and was not destructively tested. A half-life of $2.616 \pm 0.042 \times 10^5$ years was obtained. From an analysis of the alpha spectrum, White obtained a specific activity of $1.371 \pm 0.02 \times 10^7$ dpm/mg ^{234}U (corresponding to a half-life of $2.47 \pm 0.03 \times 10^5$ years for ^{234}U).

Meadows[15] measured the half-life of ^{234}U using alpha counting in low geometry, 2π proportional counting and chemical analysis on samples containing 0.856%, 0.662%, and 1.093% ^{234}U. For the low geometry measurements, the half-life was $2.439 \pm 0.048 \times 10^5$ years (95% confidence limit). No details were given.

DeBievre[16] used a variety of methods on 83 sources, which were enriched from 0.17% to 99.87% in ^{234}U to determine the half-life. The resulting value was $2.446 \pm 0.007 \times 10^5$ years at the 99.7% confidence limit. Specific details are missing but were provided by Vaninbroukx[16]. The specific activity of ^{236}U was underestimated by 2.5% in the work. ^{232}U content was determined for the highly enriched samples of ^{234}U by alpha spectrometry but it is not clear that this was the case for the less enriched samples. ^{232}U present to 0.03 parts per million (ppm) or $3. \times 10^{-8}$ grams of ^{232}U per gram of source would cause a source enriched to 1% ^{234}U content to give a half-life which was 1% too low. The highly enriched sources appear to have a slightly larger half-life value than the other sources. In addition, mass determinations by controlled potential coulometry appear to underestimate the mass compared to the isotope

dilution technique especially for highly enriched samples. To account for possible systematic errors of the above nature, the recommended value for this experiment is 2.446 ± 0.011 x 10^5 years (95% confidence limit).

Lounsbury[17] used low geometry alpha counting and mass spectrometric isotope dilution analysis to determine the half-life of ^{234}U. Six sources of 1% ^{234}U were prepared. Correcting the measured specific activity of the samples for the present best estimated half-lives of ^{235}U, ^{236}U and ^{238}U gives an increased activity about 20% larger than Lounsbury used. This reduces the ^{234}U specific activity of the samples by 0.5%. This correction increases the half-life of ^{234}U by 0.5% to 2.458 ± 0.012 x 10^5 years.

The latest half-life measurement for ^{234}U is by Geidel'man[18] who used 4π alpha-x coincidence to determine the specific activity and potentiometric titration to determine the uranium content of a sample enriched to 93.437% in ^{234}U. The value reported for the half-life is 2.459 ± 0.007 x 10^5 years (at the 95% confidence limit).

The weighted average in Table 3 is 2.455 ± 0.006 x 10^5 years

Table 3 ^{234}U Half-life

Author (year)	Reference	Total Half-Life Value (10^5 years)	Method	Comment
Fleming (1952)	13	2.475 ± .048	α proportional counting	Medium geometry
White (1965)	14	2.47 ± 0.06	α proportional counting	
Meadows (1970)	15	2.439 ± .072	α proportional counting	Low geometry*
DeBievre (1972)	16	2.446 ± .011	Variety of methods	**
Lounsbury (1972)	17	2.458 ± 0.012	α proportional counting	Low geometry
Geidel'man (1980)	18	2.459 ± .011	4α -x coincidence	*
Holden (1980)		2.455 ± 0.006	Weighted average	Recommended

**see text
*Uncertainties increased by 50% because of missing details

V ^{235}U

Nier[19] measured 21 samples of radiogenic lead and from the $^{207}Pb/^{206}Pb$ ratio determined that the actinium series was (4.6 ± 0.1)% (assumed standard deviation) as active as the uranium series. Using a best estimate for the $^{235}U/^{238}U$ atom ratio for natural uranium of 0.00725 (see Appendix II), the half-life of ^{235}U can be calculated to be 7.04 ± 0.31 x 10^8 years (95% confidence limit) by using the expression: atom percent ratio ($^{235}U/^{238}U$) x half-life ratio ($^{238}U/^{235}U$) = 0.046.

Sayag[20] measured the relative alpha activity of uranium due to ^{235}U by energy analysis in an ion chamber and obtained 0.0408 ± 0.0015 (1 standard deviation + systematic error). Correcting for the loss of alpha particles outside the central peak (10% assumed by Sayag) using White's measurement[14]

that 87.4% of the ^{235}U activity is not hidden under the peaks for ^{234}U or ^{238}U gives a value 0.0467 ± 0.0017. Using the above expression, the half-life is 6.94 ± 0.40 x 10^8 years (95% confidence limit).

Fleming[13] measured the specific activity of a 99.94% enriched source of ^{235}U using medium geometry alpha counting. Linear differential pulse height analysis was used to separate the ^{234}U and ^{235}U activity. The specific activity was found to be 4740 ± 100 dpm/mg ^{235}U (1 standard deviation) corresponding to a half-life of 7.12 ± 0.31 x 10^8 years (95% confidence limit).

Knight[21] used the same enriched ^{235}U sample as Fleming, which had a mass spectrometry analysis of ^{235}U = 99.94 ± .05%, ^{234}U < 0.022%, and the remainder assumed to be ^{238}U or 0.038% (all assumed to be in atom percent). Knight used ion chambers in 50% geometry and pulse height analysis to determine the ^{235}U alpha contribution. He reported a specific activity for the sample of 7279.4 ± 23.2 dpm/mg uranium (1 standard deviation). Fleming reported that 65.3% of the alpha activity was due to ^{235}U. Using this value, one obtains a specific activity for Knight's measurement of 4756 ± 200 dpm/mg ^{235}U corresponding to a half-life of 7.10 ± 0.32 x 10^8 years for a 95% confidence limit. This estimate does not include systematic error.

Wurger [22] measured the alpha activity of ^{235}U in natural uranium. He used Ghiorso's value[23] of 35.6% for the percentage of alpha particles from ^{235}U that are not hidden under ^{234}U or ^{238}U alpha peaks, and obtained a value of 0.04776 ± 0.00094 (assumed to be 1 standard deviation) for the relative alpha activity of ^{235}U. Using White's estimate[14] of exposed ^{235}U alpha particles of 87.4% rather than Ghiorso's value, one obtains a ^{235}U relative activity of 0.04678 and a half-life of 6.93 ± 0.27 x 10^8 years (95% confidence limit).

White[14] assayed enriched ^{235}U samples by alpha counting and extracted the ^{235}U fraction by energy analysis. White found that 87.4% of the ^{235}U activity was not hidden under the ^{234}U or ^{238}U alpha peaks. The measured specific activity was 4741 ± 60 dpm/mg ^{235}U (assumed to be 1 standard deviation) (corresponding to a half-life of 7.12 ± 0.189 x 10^8 years at 95% confidence limit).

Banks[24] determined the ^{235}U decay constant from the $^{207}Pb/^{206}Pb$ ratios of zircon and uranothorite. A value of 7.087 $^{+\ 0.073}_{-\ 0.029}$ x 10^8 years (assumed to be 1 standard deviation) was obtained using a ^{238}U half-life of 4.51 x 10^9

years. Correcting to a value of 4.468×10^9 years, one obtains a half-life of $7.02 \begin{smallmatrix} + 0.14 \\ - 0.06 \end{smallmatrix} \times 10^8$ years at 95% confidence limit.

Deruytter[25] determined the ^{235}U alpha activity in natural uranium using energy analysis with a silicon solid state junction detector. He corrected his measured ratio of 0.0406 ± 0.005 (standard deviation of ten results) using Ghiorso's value [23] of 85.6% for ^{235}U alpha particles not hidden under ^{234}U or ^{238}U alpha peaks. If one uses White's value of 87.4% instead, one obtains a ratio of 0.0465. Using 4.468×10^9 years for the ^{233}U half-life, one obtains $6.97 \pm 0.19 \times 10^8$ years for the ^{235}U half-life at 95% confidence.

Jaffey[26] measured the specific activity of a sample enriched to 99.99886% in ^{235}U using intermediate geometry alpha counting. Jaffey obtained a value of 4798.1 ± 3.3 dpm/mg ^{235}U at the 95% confidence limit for the 11 runs made, the half-life is $7.0373 \pm 0.0106 \times 10^8$ years.

Deruytter[27] measured the branching ratio of the central peak of alpha particles to the total alpha particles emitted by ^{235}U using a sample enriched to 99.999% ^{235}U and used this ratio to recalculate the ^{235}U half-life. Jaffey [28] has pointed out that Deruytter's measurement did not show as good a resolution in the pulse height analysis as the work of White[24], so Deruytter's earlier measurement[25] was used with White's branching ratio above.

The weighted average from Table 4 for the ^{235}U half-life is $7.037 \pm 0.011 \times 10^8$ years.

Table 4 ^{235}U Half-Life

Author (year)	Reference	Total Half-Life Value (10^8 years)	Method	Comment
Nier (1939)	19	7.04 ± 0.31	$^{207}Pb/^{206}Pb$ ratio	
Sayag (1951)	20	6.94 ± 0.40	α Proportional counting	
Fleming (1952)	13	7.12 ± 0.31	α Proportional counting	Medium geometry
Knight (1950)	21	7.10 ± 0.32	α Proportional counting	50% geometry
Wurger (1957)	22	6.93 ± 0.27	α Proportional counting	
White (1965)	14	7.12 ± 0.18	α Proportional counting	
Banks (1966)	24	$7.02^{+0.14}_{-0.06}$	$^{207}Pb/^{206}Pb$ ratio	
Deruytter (1965)	25	6.97 ± 0.19	α Counting	Si Detector
Jaffey (1971)	26	7.037 ± 0.011	α Proportional counting	Intermediate geometry
Holden (1980)		7.037 ± 0.011	Weighted average	Recommended

VI ^{236}U

Jaffey[29] measured the specific activity using samples of ^{235}U enriched to 22% - 38% in ^{236}U. There are no details on the specific activity of the samples or the values assumed for the other isotopes involved. Samples were weighed through fission counting of ^{235}U and ionization chamber energy analysis was used to determine the alpha activity. The half-life obtained was $2.457 \pm .094 \times 10^7$ years (at 95% confidence).

Fleming[13] measured the specific activity of ^{236}U using sources enriched to 96.65%. The alpha activity and the mass spectrometric analysis disagree with the known specific activities of ^{234}U and ^{236}U by more than a factor of two. Details on the actual specific activity of the sample are missing. The resulting half-life obtained was 2.391 ± 0.038 x 10^7 years (at 95% confidence).

Flynn[30] measured the specific activity of a 99.644% enriched ^{236}U sample. From pulse-height analysis, the ^{236}U contributed 98.314% of the measured alpha activity. Data from the isotopic analysis did not agree with the pulse height data. Jaffey[31] has indicated that there were two mass spectrometric analyses performed on the sample, one at Argonne and the other at Oak Ridge. There was a 40% difference in the amount of ^{233}U present in the two analyses and the Oak Ridge values (unpublished) did agree with the pulse height analysis. Using the Oak Ridge value of 99.68% ^{236}U in the sample, the measured specific activity becomes 143552 ± 84 dpm/mg ^{236}U. For the 11 runs, the half-life is 2.3422 ± 0.0031 x 10^7 years (at 95% confidence). The recommended value from Table 5 is 2.342 ± 0.003 x 10^7 years for the ^{236}U half-life.

Table 5 ^{236}U Half-Life

Author (year)	Reference	Total Half-Life Value (10^7 years)	Method	Comment
Jaffey (1951)	29	2.46 ± 0.14	α Proportional counting	*
Fleming (1952)	13	2.391 ± 0.057	α Proportional counting	*
Flynn (1972)	30	2.3422 ± 0.0031	α Proportional counting	Intermediate geometry
Holden (1980)		2.342 ± 0.003	Weighted average	Recommended

*Uncertainties increased by 50% because of missing details.

VII. ^{238}U-Total Half-Life

Except for Steyn[34], all determinations of the specific activity of ^{238}U using natural uranium and assuming secular equilibrium have been discarded (see Appendix I). Kienberger[32] measured both natural uranium and highly depleted uranium which had 0.05 ppm of ^{234}U. The natural uranium results which depended upon secular equlibrium and should carry a large systematic error (see Appendix I), have been ignored. Details are sketchy and the systematic errors such as accurate corrections for backscattering and sample self absorption are difficult to estimate. The quoted specfic activity was 742.7 ± 1.6 dpm/mg ^{238}U corresponding to a half-life of 4.489 ± 0.010 x 10^9 years.

Leachman[33] performed 2Π alpha counting on depleted uranium samples deposited on platinum. The experiment was described in a footnote to an article on neutron cross sections. No details were given except that the ^{238}U mass was determined by weight and calorimetric analysis. The half-life quoted was 4.56 ± 0.03 x 10^9 years. Steyn [34] determined the specific activity of natural uranium using liquid scintillation alpha counting. The measured specific activity is 1520 ± 2 dpm/mg ^{238}U corresponding to a half-life of 4.45 ± 0.02 x 10^9 years. However, the assumption of secular equilibrium carries with it a large systematic error.

Jaffey[26] measured the specific activity of depleted uranium samples corresponding to 99.9790 atom percent ^{238}U and 99.9997 atom percent ^{238}U using intermediate geometry alpha counting. Jaffey obtained a specific activity of 746.19 ± 0.41 dpm/mg ^{238}U (1 standard deviation). This corresponds to a half-life of 4.468 ± 0.005 x 10^9 years (at 95% confidence).

The recommended value is 4.468 ± 0.005 x 10^9 years, where earlier measurements were discarded because of lack of details prevented an adequate estimate of the systematic error or the known systematic error meant the measurement was not significant.

VIII. ^{238}U Spontaneous Fission Half-Life

There is a wide variation in the measured values of the half-life for spontaneous fission of ^{238}U, although the measurements in the past twenty

years cluster about two values which differ by 20-25%.

The values and the standard deviation for the mica-uranium sandwich or Lexan-uranium sandwich, i.e. fission track technique in 2π geometry, have given a disintegration constant of $7.1 \pm 0.1 \times 10^{-17}$ years^{-1} (see Fleischer[35], Roberts[36], Kleeman[37], Leme[38], Khan[39], Ivanov[40] and Emma[41]). Other techniques lead to larger values for the disintegration constant. Fission track detection in dated uranium glass (Storzer[42], Wagner[43] and Thiel[44]) give a weighted average of $8.59 \pm 0.31 \times 10^{-17}$ years^{-1}. Radiochemical determination of the fission products of ^{238}U by Von Gunten[45] give a value of $8.66 \pm 0.22 \times 10^{-17}$ years^{-1}. The rotating bubble chamber experiments, where centrifugal forces produce negative pressures in a liquid and energy deposition from spontaneous fission can be detected as in a normal bubble chamber, give a weighted average of $8.447 \pm 0.057 \times 10^{-17}$ years^{-1} (Spadavecchia[46] and Galliker[47]). Ionization chamber measurements by Whitehead[48] and Segre[49] give a weighted average of $8.548 \pm 0.253 \times 10^{-17}$ years^{-1}. The decay constant from third order correlations in a modified Rossi alpha experiment is $8.66 \pm 0.43 \times 10^{-17}$ years $^{-1}$ (Thury[50]).

It should be noted that Fleischer and Price[51] found that the ages of Tektites and other natural glasses obtained by the fission track method could be made to agree with ages determined by the K-Ar method where a disintegration constant of 6.9×10^{-17} years^{-1} was used. Storzer and Wagner[52] pointed out the problems of partial fission track fading in geologic materials and detected this in the case of glasses by measuring track lengths or diameters. When a corrections is made for the annealing influence in tektites, the best agreement with K-Ar age determinations results from use of a ^{238}U disintegration constant for spontaneous fission of 8.4×10^{-17} years^{-1} from Gentner[53], Storzer and Wagner[54], and Storzer[55].

Since Storzer [52] pointed out this potential systematic error of fission track fading which when corrected increased the decay constant from 6.9×10^{-17} to 8.4×10^{-17} and the five techniques other than the fission track sandwich give results about 8.5×10^{-17} years^{-1}, elimination of the fission track sandwich results in the overall average is recommended. The weighted average of the remaining methods excluding the rotating bubble chamber is $8.61 \pm 0.14 \times 10^{-17}$ years. A straight average of all 5 remaining methods is $8.58 \pm 0.14 \times 10^{-17}$ years, where the error has been assessed from the other methods since there is no systematic error assessment on the

rotating bubble chamber. The resulting half-life at 95% confidence is $8.08 \pm 0.26 \times 10^{15}$ years.

Table 6

^{238}U Spontaneous Fission Decay Constant, λ_f^{238}, and standard deviation.

Author (year)	Reference	Spontaneous Fission Decay Constant (10^{-17} years^{-1})	Method
Whitehouse (1950)	48	8.38 ± 0.52	Ionization chamber
Segre (1952)	49	8.60 ± 0.29	Ionization chamber
Fleischer (1964)	35	6.85 ± 0.20	Weighted mean of ^{40}K, ^{87}Rb ages of minerals + mica-uranium sandwich
Roberts (1968)	36	7.03 ± 0.11	Mica-uranium sandwich
Spadavecchia (1967)	46	8.42 ± 0.10	Rotating bubble chamber
Von Gunten (1969)	45	8.66 ± 0.22	Fission products from ^{238}U
Galliker (1970)	47	8.46 ± 0.06	Rotating bubble chamber
Storzer (1970)	42	8.49 ± 0.76	Fission tracks in dated uranium glass
Kleeman (1971)	37	6.8 ± 0.6	Lexan-uranium sandwich
Thury (1971)	50	8.66 ± 0.43	Third-order coincidence
Leme (1971)	38	7.30 ± 0.16	Mica-uranium sandwich
Khan (1973)	39	6.82 ± 0.55	Mica-uranium sandwich
Ivanov (1975)	40	7.12 ± 0.32	Mica-uranium sandwich
Emma (1975)	41	7.2 ± 0.2	Mica-uranium sandwich
Wagner (1975)	43	8.7 ± 0.6	Fission tracks in dated uranium glass
Thiel (1976)	44	8.57 ± 0.42	Fission tracks in dated uranium glass

IX Summary

Table 7 summarizes the recommended values for the specific activity and the half-life data for the various uranium isotopes.

Table 7

Recommended Half-life and Specific Activity Values

Isotope	Half-life (years)	Specific Activity or Decay Constant
^{232}U	69.8 ± 1.0	4.90 ± 0.07 x 10^{10} dpm/mg ^{232}U
^{233}U	1.592 ± 0.002 x 10^{5}	2.139 ± 0.003 x 10^{7} dpm/mg ^{233}U
^{234}U	2.455 ± 0.006 x 10^{5}	1.381 ± 0.003 x 10^{7} dpm/mg ^{234}U
^{235}U	7.037 ± 0.011 x 10^{8}	4.7983 ± 0.008 x 10^{3} dpm/mg ^{235}U
^{236}U	2.342 ± 0.0034 x 10^{7}	1.436 ± 0.002 x 10^{5} dpm/mg ^{236}U
^{238}U (total)	4.468 ± 0.005 x 10^{9}	746.2 ± 0.8 dpm/mg ^{238}U
^{238}U (spon. fiss.)	8.08 ± 0.26 x 10^{15}	8.58 ± 0.28 x 10^{-17} $years^{-1}$

Authors's Note:

As this report goes to press, Vaninbroukx (private communication June 1981) has reevaluated the ^{234}U measurement from Geel (DeBievre[16]). Taking into account the items mentioned in this report on that experiment Vaninbroukx recommends $t_{1/2}$ = 2.450±0.008 x 10^{5}years. My recommended weighted average becomes $t_{1/2}(^{234}U)$ = 2.454±0.006 x 10^{5}years.

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Appendix I Specific Activity of Natural Uranium

A number of determinations of the half-life of ^{234}U and ^{238}U are based on measurements of the specific activity of natural uranium samples and the assumption of secular equilibrium in those samples. In a radioactive decay chain, when a daughter nuclide has a much smaller half-life than its parent nuclide, at a time interval very long compared to the daughter's lifetime, the growth and decay of the daughter is controlled by the parent and the two nuclides are in secular equilibrium i.e. their activities, $N\lambda$, are equal, where N is the number of atoms and λ is the decay constant (λ = natural logarithm 2/half-life). Since the half-life of ^{234}U is only 10^5 years and the age of the earth is orders of magnitude greater, 10^9 years, all of the ^{234}U originally formed in natural uranium has long since decayed. The ^{234}U now present in uranium is due to the ^{238}U decay series, i.e., ^{238}U $^{\alpha}$ decay ^{234}Th $^{\beta-}$ decay ^{234}Pa $^{\beta-}$ decay ^{234}U. In a closed system, the ^{238}U and the ^{234}U should be in secular equilibrium. This implies that a measurement of the specific activity of a natural uranium sample when corrected for the small amount of ^{235}U activity (≈2.2%) would provide a determination of the equal amounts of activity of the ^{238}U and ^{234}U present. Given the isotopic composition of ^{238}U and ^{234}U in the natural sample, one could calculate the specific activity and half-life of each nuclide. This procedure has been followed in a number of measurements. However if a closed system is not maintained, the secular equilibrium assumption is not valid. Given the specific activity or half-life for ^{234}U and ^{238}U, one can derive the "theoretical" isotopic abundance of ^{234}U implied by secular equilibrium. From the values in Table 7, the ^{234}U abundance is $54.55 \pm 0.15 \times 10^{-4}$ atom percent (at 95% confidence) or 54.55 ± 0.15 parts per million (ppm) in natural uranium assuming secular equilibrium. The "theoretical" specific activity of natural uranium is calculated from the above abundance to be 1515.7 ± 2.7 dpm/mg.

As shown in Appendix II, Smith[11] has determined a ^{234}U variation from 50.9 ppm to 54.8 ppm in various world-wide sources of uranium ore. Rosholt[56, 57, 58] has found extreme values for ^{234}U of 22.2 ppm to 92.5 ppm in sandstone deposits of natural uranium in the United States. The calculated specific activities are shown in Table 8. With such a large variation (≈2) in the specific activity of natural uranium, no value can be quoted without knowledge of the isotopic composition of the sample.

Table 8 Specific Activity of Natural Uranium

Source	Reference	Specific Activity (dpm/mg)
Secular Equilibrium	This report	1515.7 ± 2.7
Smith	11	1465 to 1519
Rosholt	56, 57, 58	1077 to 2031

Appendix II Natural Variation of the Uranium Isotopic Composition

This discussion should be considered as an update of reference 1 for uranium. Smith[11] has measured the variation of ^{234}U on 16 world sources, while Cowan[59] has measured the variation in ^{235}U for some 90 samples, which included the sixteen sources of Smith. Tables 9 and 10 summarize these two sets of results for U.S. and non U.S. ore sources respectively.

Although the ^{234}U abundance value varies significantly in Tables 9 and 10, it can be seen that the ratio $^{238}U/^{235}U$ is fairly constant at 137.9 ± 0.06.

Rosholt[56, 57, 58] in series of papers reviewed the fractionation of uranium in various sandstone deposits in the United States. Rosholt[58] found samples with a ^{234}U abundance as low as 22.2 ppm in Powder River basin Wyoming. In Shirley Basin, Wyoming sandstone, Rosholt[56] found samples with a ^{234}U abundance as high as 92.1 ppm. Finally, in Gas Hill, Fremont County, Wyoming, Rosholt[57] found samples enriched in ^{234}U to 92.5 ppm less than a half foot from a sample with a ^{234}U abundance of 45.7 ppm. The extreme ranges for the $^{238}U/^{235}U$ ratio found by Rosholt were 137.3 to 138.7.

Table 9 Uranium Isotopic Composition (U.S. samples)

Source	atom % 234 (ppm)	atom % 235 (ppm)	atom % 238 (%)	Half-life ^{234}U (10^5 years) assuming secular equilibrium
Ford, Washington	54.1	7198	99.275	2.435
Lakeview, Oregon	52.2-52.6	7200-7202	99.275	2.349-2.367
Fremont County Wyoming	51.9-52.8	7197	99.275	2.336-2.376
Edgemont, South Dakota	50.9	7196	99.275	2.291
Moab, Utah	53.0-53.8	7197	99.275	2.385-2.421
Kermac, Ambrosia Lake New Mexico	53.6-54.3	7196	99.275	2.412-2.444
Climax Mountain Colorado	54.3	7196	99.275	2.444
Homestake, Ambrosia Lake New Mexico	54.7	7196	99.275	2.462

Table 10 Uranium Isotopic Composition (non U.S.A. Samples)

Source	atom % 234 (ppm)	atom % 235 (ppm)	atom % 238 (%)	Half-life ^{234}U (10^5 years) (assuming secular equilibrium
Belgian Congo	54.8	7200.	99.275	2.466
Radium Hill, Australia	53.3	7198.	99.275	2.399
Blind River, Ontario (Canada)	53.3	7199.	99.275	2.399
Marion River Northwest Terr. (Canada)	53.9	7198.	99.275	2.426
Rum Jungle, Australia	54.3	7198.	99.275	2.444
South Africa	53.3-54.0	7199.	99.275	2.415
Lake Athabaska, Saskatchewan (Canada)	53.9	7199.	99.275	2.426
Bancroft Ontario, Canada	54.4	7198.	99.275	2.448

For the ninety world-wide samples of Cowan[59], the $^{235}U/^{238}U$ ratio has twin peaks at .0072495 and .0072515, with extreme values of .007241 to .007256, while the four U.S. locations that Rosholt surveyed gave an average value for the ratio of .007242 with extreme values of 0.00721 to 0.00728. A best estimate for the ratio is 0.00725.

In addition to the above studies of the variation in isotope atom ratios in samples, $^{234}U/^{238}U$ disequilibrium studies have continued since the 1955 Cherdyntsev report[60] of large deviations in the activity ratio of ^{234}U to ^{238}U occur in nature. The ^{234}U is used to date geological events. Since the activity ratio should be unity for secular equilibrium, a non-unit value

implies the variations of ^{234}U abundance in the uranium sample. Since $A_{234}/A_{238} = N_{234}\lambda_{234}/N_{238}\lambda_{238} = (N_{234}/N_{238})/(t\ 1/2(234)/t\ 1/2(238))$, then $N_{234} = (A_{234}/A_{238}) \times (t\ 1/2(234)/t\ 1/2(238)) \times N_{238}$, where A_{234}/A_{238} = (dpm of ^{234}U)/(dpm of ^{238}U).

Osmond[3] has reviewed the data and for uranium dissolved in groundwater reports extremes corresponding to ^{234}U abundances of 27.4 ppm to 383.5 ppm in well water from Florida and Texas, respectively.

Similarly, Rosholt[61] studied volcanic tuff in California and Utah and determined activity ratios corresponding to ^{234}U abundances of 90.0 ppm and 27.8 ppm, respectively.

For carbonates in ocean water, Thurber[62] found average activity ratios corresponding to a ^{234}U abundance of 62.7 ppm.

The mechanism involved to account for these enhancements and discrepancies are alpha recoil leaching and alpha recoil adsorption since the ^{234}U is the daughter product of the ^{238}U alpha decay. Rosholt[57] has found samples depleted and enhanced in ^{234}U within 1/2 foot of each other. ^{234}U is relocated by recoil and radiation damage into crystal defects where it is more readily accessible to oxidation.

From the above it should be noted that uranium is an element whose isotopic composition is variable in nature and if the ^{234}U abundance is a critical quantity, it should be directly determined for the particular sample of interest.

Appendix III Parameters of the Evaluation

In the computation of specific activity in dpm/mg and half-life in years, the following values were used for the listed parameters:

Parameter	Value	Source
Atomic Weight of uranium	238.0289	Holden[2]
Atomic Mass of ^{232}U	232.03714	Wapstra[63]
^{233}U	233.039629	"
^{234}U	234.040947	"
^{235}U	235.043925	"
^{236}U	236.045563	"
^{238}U	238.050786	"
One year (solar)	525949 minutes	
Avogadro's number	6.022045×10^{23} mol^{-1}	Cohen[64]

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