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Half-lives for Selected Actinides and Long-lived Radionuclides

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Abstract: Long-lived actinide nuclides are of interest for their use in nuclear reactors, for nuclear reactor burnup studies in waste management, and for safeguard applications, *e.g.* α counting is used to determine the amount of material present. Some long-lived radionuclides are of interest for their use in determining geological ages using various dating methods, and in calculating the cosmic-ray exposure ages of meteorites. Recommended values are presented for both the total half-life and for the spontaneous fission half-life of $^{232-238,238}$U, of $^{236,238-242,244}$Pu, of ^{241,242m,243}Am, and of $^{242-248,250}$Cm. Problems with the presentation of uncertainties are discussed. The impact of the revised ^{14}C half-life on the carbon dating technique and various ^{14}C ages is discussed. The possible primordial occurrence of ^{92}Nb is now definitely ruled out. Based on examination of the ^{26}Al half-life, the calculated value for the cosmic-ray exposure age of meteorites remains too high compared to the age calculated using other radionuclide half-life values. ^{204}Pb, which was once thought to be radioactive, is shown to be stable.

(total half-life, spontaneous-fission half-life, U, Pu, Am, Cm, ^{14}C, ^{26}Al, ^{92}Nb, ^{204}Pb)

I. Introduction

The total half-life, which is often synonomous with the half-life for alpha decay, and the half-life for spontaneous fission are evaluated for various long-lived nuclides of uranium, plutonium, americium and curium. For many of these nuclides, there is also a decay mode of heavy fragment radioactivity,[1] which is ignored here although this decay mode may be comparable with the spontaneous fission decay mode,[2] but is usually only a small perturbation on the value of the half-life for the alpha decay mode. Various experiments have been reanalyzed and recommended half-life values are presented for $^{232-238,238}$U, $^{236,238-242,244}$Pu, ^{241,242m,243}Am, and for $^{242-248,250}$Cm. These recommended half-life values supersede the estimates previously presented.[3,4,5] Many of the uncertainties presented exceed, by up to an order of magnitude, those quoted by individual authors in their publication, *e.g.* the total half-lives of ^{240,241}Pu, ^{241}Am, ^{248,246}Cm. The half-lives for ^{14}C, ^{26}Al, ^{92}Nb, and ^{204}Pb are evaluated and recommended values are presented with their uncertainties.

The general procedure has been to review each experiment and to revise published values for the latest estimates of various parameters originally reported by the authors. Where available in each of these experiments, the standard deviation was combined with one third of the systematic error to provide the uncertainty quoted for each experiment. The result of this procedure should be that the limit of error of the half-life would be obtained by multiplying the quoted uncertainty by a factor of three, or three standard deviations, i.e., 3σ. The uncertainty in the recommended value was then calculated from a weighted average of the listed measurements using a variance weighting technique, i.e., the reciprocal square of the author's reported uncertainty. Some exceptions were made using either unweighted averages, selecting a value which was considered superior to the other listed measurements, or an average half-life was calculated for each of the different experimental techniques used. These half-lives were then averaged and the resulting value was recommended. The actinides are presented in Table I and radionuclides in Table II.

II. Uranium Isotopes

For ^{232}U, Barwick[2] measured a heavy fragment radioactive decay comparable to the spontaneous fission mode half-life,[6] but it was not considered here. For ^{233}U, many details were missing from Geidel'man's paper.[7] It could not be evaluated on the same basis as other results, so the reported uncertainty was increased by 50%. For ^{234}U, the DeBievre[8] measurement has been revised by the authors, who used a variety of methods on over 80 sources. Results by Lounsbury[9] have been revised for better estimates of the specific activity of the other uranium nuclides present. Contamination by ^{232}U was not discussed and the uncertainty was increased by 50% to account for this potential error in a source which was only 1% enriched in ^{234}U.

Older determinations of ^{238}U were based on measurements of the specific activity of natural uranium samples and the assumption of secular equilibrium between ^{234}U and ^{238}U in those samples. This implies that after correction for the small amount of ^{235}U activity, ≈2.2%, the measured specific activity should have been produced equally by ^{234}U and ^{238}U. Holden[3] has shown that this assumption is invalid. There is a disequilibrium in uranium sources found in various parts of the world. The specific activity of natural uranium can vary by up to a factor of two in different sources. As a result, the direct measurement by Jaffey[10] is recommended here. For the spontaneous fission half-life of ^{238}U, results reported using fission track detection in 2π geometry, *e.g.* mica-uranium, or lexan-uranium sandwichs, have a problem with partial fission track fading in the geological materials, pointed out by Storzer and Wagner.[11] Fission track fading would underestimate specific activity and would overestimate the half-life. These measurements do report half-lives which are 10% to 30% larger compared to all of the other techniques. These experiments are not included in the averaging because of this systematic error. The other techniques used in the measurement of ^{238}U have been separately weighted and their results averaged and converted from specific activity into a half-life recommendation.

III. Plutonium Isotopes

If one applies weighted averages to determine the α half-life of ^{240}Pu, three of the five best measurements would fall some ten standard deviations away from the recommended value. A half-life has been recommended on the basis of the unweighted average of the various techniques or methods utilized to measure this half-life. For the case of ^{241}Pu, the total half-life is predominantly β decay rather than either α decay or spontaneous fission decay. If a weighted average was recommended, all of the most recent measurements that were performed at an accuracy level of tenths of one percent would not carry any weight in the evaluation of this half-life. Strohm quotes[12] an absolute accuracy of 486 parts per million, while examining only one sample. This result would eliminate all of the other careful measurements from any consideration. To eliminate this problem, an unweighted average of the various techniques was recommended. The α half-life of ^{239}Pu was measured using a variety of techniques on a single source by a number of laboratories. Highly precise values were reported having a range of results from the various labs which were an order of magnitude larger than the standard deviation quoted in most of these experiments. An unweighted average of the various techniques was again recommended. The α half-life of ^{242}Pu was reported with a range of results from ten to fifteen times larger than the typical uncertainty quoted by any of the authors. An unweighted average of the half-life from the various techniques was once again recommended.

IV. Americium Isotopes

For ^{241}Am, if one uses variance weighting to determine the total half-life, the highly precise measurements reported at the 0.15% to 0.7% level would carry no weight because Ramthun[13] reports an absolute accuracy of 463 parts per million on this half-life. A half-life determined from an unweighted average for various techniques was recommended. For spontaneous fission, the range of reported results was larger than the quoted uncertainties by more than an order of magnitude. An unweighted average of the two most precise values was selected and an uncertainty was chosen which would overlap the range of all of the most recent measurements.

V. Curium Isotopes

For spontaneous fission in ^{242}Cm, Zhang's[14] measurement carries most of the weight. However, the half-life disagrees with the more recent measurements by up to ten standard deviations. For spontaneous fission in ^{244}Cm, the most accurate measurements reported disagree by about sixteen standard deviations. Selected values have been recommended in both of these cases. Similar problems also exist for the reported total half-lives of ^{242,245,248}Cm.

VI. ^{14}C

Early measurements were plagued by very low enrichment or had a problem with the retention of small amounts of high specific activity carbon dioxide during the gas dilution phase.[15] These results were discarded and an unweighted average of the remaining measurements is recommended due to the wide variation in estimating systematic error by authors. In Table III, the measured half-lives used by Libby[16] for his recommended value of 5568 years are displayed along with the most recent half-lives used to derive the recommended value. Values from 4700 years to 10^5 years had been measured at the time that Libby selected the three concordant results to average. The recent measurements are all consistently higher by more than two and one-half percent.

The measurers of radiocarbon dates continue to normalize their dates to a value of 5568 years, which is the average of the three measurements which Libby[16] had recommended. While any reported dates are consistent among themselves, as noted by Libby, these radiocarbon dates would not be agree with dates determined by any other method.[17] To obtain correct radiocarbon dates for any samples, a factor of 1.026 must be applied to the dates as presently reported. All radiocarbon dates are now approximately two and one-half percent too low.

VII. ^{26}Al

The cosmic ray exposure ages of meteorites can be determined from their ^{21}Ne content and the ^{21}Ne production rate,[18] where the production rate is deduced from a variety of cosmic ray produced radionuclides including ^{26}Al. The production rate based on ^{26}Al is larger than that based on other radionuclides[19] and could be explained if the ^{26}Al half-life is between $9 \cdot 10^5$ and $10 \cdot 10^5$ years, or if the cosmic-ray flux was higher in the last million years than it was ten million years ago. The value for the half-life of ^{26}Al, which is recommended in Table II, is based on three recent independent measurements, which agree with the earlier value. In Table IV, these measured values are presented. Different methods have resulted in a consistent value, which agrees with the earlier estimate of $7.2 \cdot 10^5$ years. A variable cosmic-ray flux must now be considered to explain the discrepancy in ^{21}Ne production rates.

VIII. ^{92}Nb

An estimate of approximately $1.7 \cdot 10^8$ years for this half-life[20] indicated its' possible primordial occurrence. Other measurements have been reported, as shown in Table V. When these experiments have been revised for the best set of their auxiliary parameters, the recommended half-life value of $3.7 \cdot 10^7$ rules out any possible primordial ^{92}Nb.

IX. ^{204}Pb

There is no recommended half-life for ^{204}Pb. An earlier experiment[21] recommended a half-life value of $1.4 \cdot 10^{17}$ years based on a photographic emulsion measurement which had detected an alpha particle with an energy of 2.6 Mev. However the available decay energy is only 1.97 Mev, over 30% less.

X. Discussion of Results

It has been noted above that various half-life measurements have uncertainties quoted by authors such that they exclude many other good recent measurements from consideration. Undoubtedly, systematic errors have not been carefully considered in these publications. When experiments are performed at the level of five to ten percent accuracy, counting is an important consideration.

In Poisson statistics, increasing the number of counts can improve the overall accuracy. However, by the time that the overall accuracy reaches the level of one-half percent or better, the estimate of systematic errors controls the total accuracy. Improving the statistical precision, by continuing to collect raw data points, does not improve the total error. If one uses variance weighting indiscriminately, one penalizes the authors who attempt the difficult task of estimating the systematic error, while benefiting the authors who make no such attempt to determine other sources of error.

In the review of nuclear data by the (IAEA), International Atomic Energy Agency[22], their general comment on uncertainties included a statement questioning the validity of any presently stated uncertainties of less than 0.1% for half-lives. The same criteria has been adopted in this paper. No half-life has been recommended with an accuracy of better than 0.1%; see the total half-life of ^{238}Pu as an example. The rationale for this rule is that systematic errors up to ten times smaller than the total quoted uncertainty would have an appreciable effect on that uncertainty, if there were a number of such errors. Recommending values, with an accuracy of hundreds of parts per million, would imply that all potential errors in the experiment at the level of tens of parts per million had been investigated, documented, and their effect on the result taken into account. An experiment in which such a thorough study has been performed and documented, has yet to be reported.

Table I. Recommended Actinide Half-lives

Reference Nuclide	$T_{½}$ (total) (Years)	$T_{½}$ (spont. fiss.) (Years)
^{232}U	70.0 ± 1.5	(8. ± 6.)·10^{13}
^{233}U	(1.592 ± 0.002)·10^{5}	> 2.7·10^{17}
^{234}U	(2.455 ± 0.005)·10^{5}	(1.5 ± 0.3)·10^{16}
^{235}U	(7.037 ± 0.011)·10^{8}	(9.8 ± 2.8)·10^{16}
^{236}U	(2.342 ± 0.003)·10^{7}	(2.49 ± 0.11)·10^{16}
^{238}U	(4.47 ± 0.02)·10^{9}	(8.2 ± 0.1)·10^{15}
^{238}Pu	2.87 ± 0.01	(3.4 ± 1.2)·10^{9}
^{239}Pu	87.7 ± 0.1	(4.70 ± 0.08)·10^{10}
^{239}Pu	(2.410 ± 0.003)·10^{4}	(7.8 ± 1.8)·10^{15}
^{240}Pu	6564. ± 10.	(1.16 ± 0.02)·10^{11}
^{241}Pu	14.4 ± 0.1	< 6.·10^{16}
^{242}Pu	(3.74 ± 0.02)·10^{5}	(6.78 ± 0.04)·10^{10}
^{244}Pu	(8.00 ± 0.09)·10^{7}	(6.6 ± 0.2)·10^{10}
^{241}Am	432. ± 2.	(1.0 ± 0.4)·10^{14}
^{242m}Am	141. ± 1.	> 3.·10^{12}
^{243}Am	7370. ± 22.	(2.0 ± 0.5)·10^{14}
^{242}Cm	163. ± 1. Days	(7.0 ± 0.2)·10^{6}
^{243}Cm	29.2 ± 0.1	(5.5 ± 0.9)·10^{11}
^{244}Cm	18.1 ± 0.1	(1.32 ± 0.02)·10^{7}
^{245}Cm	8410. ± 80.	(1.4 ± 0.2)·10^{13}
^{246}Cm	4760. ± 50.	(1.81 ± 0.01)·10^{7}
^{247}Cm	(1.56 ± 0.05)·10^{7}	--------
^{248}Cm	(3.48 ± 0.05)·10^{5}	(4.15 ± 0.03)·10^{8}
^{250}Cm	≈ 9.7·10^{3}	(1.13 ± 0.05)·10^{4}

If one had a choice in designing the ideal experiment to determine the half-life, one would choose to measure many samples, using a number of duplicate instruments and utilizing a variety of different methods or techniques. If this were possible, the necessary information to correctly estimate the systematic error might be obtained.

Table II. Recommended Half-life of Long-lived Radionuclides

Nuclide	$T_{½}$ (Years)	Comment
^{14}C	5715. ± 45.	Five Measurements
^{26}Al	(7.1 ± 0.2)·10^{5}	Four Measurements
^{92}Nb	(3.7 ± 0.3)·10^{7}	Two Revised Measurements
^{204}Pb	stable	E_{α} > Available Energy

Table III. Comparison of ^{14}C Half-life Measurements

Author	Half-life	Comment
Engelkemeir[23]	5580. ± 45.	Used in Libby's Average[18]
Jones[24]	5589. ± 75.	Used in Libby's Average[6]
Miller[25]	5513. ± 185.	Used in Libby's Average[6]
Watt[26]	5780. ± 65.	*Used in This Work*
Olsson[27]	5680. ± 40.	*Used*
Hughes[28]	5730. ± 50.	Mann[15] Revised; *Used*
Bella[29]	5660. ± 30.	*Used*
Emery[30]	5736. ± 84.	Uncertainty±1.5; *Used*

Table IV. Comparison of ^{26}Al Half-life Measurements

Author	Half-life	Comment
Rightmire[31]	(7.14 ± 0.32)·10^{5}	Revised by Samworth[32]
Norris[33]	(7.05 ± 0.24)·10^{5}	
Middleton[34]	(7.02 ± 0.56)·10^{5}	
Thomas[35]	(7.8 ± 0.5)·10^{5}	Confirms Other Values

Table V. Comparison of ^{92}Nb Half-life Measurements

Author	Half-life	Comment
Apt[20]	≈ 1.7·10^{6}	Primordial Nuclide?
Makino[36]	(3.5 ± 0.4)·10^{7}	Revised
Nethaway[37]	(3.9 ± 0.5)·10^{7}	Revised

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