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ANALYSIS OF EXISTING DATA AND SPECIFICATION OF AN EXPERIMENT TO DETERMINE THE ^{252}Cf HALF-LIFE TO THE REQUIRED DEGREE OF ACCURACY

I.A. Kharitonov

(Translated from a Russian original published in Yadernye Konstanty 4/1987)

Translation editor: Dr. Alex Lorenz

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ABSTRACT

The methods used and the results obtained in measurements of the ^{252}Cf half-life are analyzed. In calculating the weighted mean value, additional error components as well as the those given by the authors are taken into account. In order to reduce the error in the weighted mean value to less than 0.1%, the need and the requirements for an exact measurement are specified. A half-life of 2.6473±0.0028 years is recommended.

The average number of neutrons per fission, **ν**, and the prompt ^{252}Cf neutron spectrum are recommended in Ref.[1a] as the standards to be used in experiments to measure nuclear physics constants.

Lengthy experiments are performed to calculate the decay of this nuclide since an error in determining its half-life would have a direct effect on the measurement result. In Ref. [2] the optimum value for the error in measuring the number **ν** for ^{252}Cf is estimated to be 0.25%. Since the accumulation of data in experiments on measuring this value may take as long as the half-life itself [3], we can assume that the error should not exceed more than 0.1%.

The aim of this paper is to evaluate the half-life by analyzing experimental results and to establish the experimental requirements to ensure that the half-life is measured with a sufficient degree of accuracy.

ANALYSIS OF RESULTS AND MEASUREMENT METHODS

Table 1 lists values for the half-life T_i obtained experimentally [3-13] and the recommended values in Refs [1b, 14] and also a short description of the measurement methods.

Comparison of the values for T_i shows that:

- the standard deviation of the result is estimated by the authors to be $\sigma_i \leq 0.4\%$;
- the deviation of the results from the ones recommended for example in Ref. [14] is 0.7%;
- the range is 1.5%;
- the results of experiments conducted from 1981-1985 [3, 11-13] are grouped around a mean value which is 0.3% higher than the value recommended in the Ref. [14].

Thus the discrepancy between the measurement results significantly exceeds the errors given by the authors This prompted the authors of ref. [1b] to describe the present situation regarding the measurement of the half-life of ^{252}Cf as highly confused.

The same method of determining the half-life is used in Refs. [3-13]: the decay constant λ is calculated from the results of the relative activity measurements (or neutron flux) of the radionuclide in the source over a given period of time T_o.

The references can be divided into two groups according to the measurement principle used:

- response of the detector proportional to the neutron flux of vial-enclosed source;
- counting rate of pulses recording the radiation from an open source.

In the first case, the sample is fixed in the space restricted by the vial which prevents it from leaking as a result of evaporation. The following slow neutron activation methods are used: manganese nuclei are activated in a manganese bath [3,5,7,9], detectors are placed in a moderator in a fixed geometry, gold foils in distilled water [13], grains of pure manganese in polyethylene [11], boron proportional counters in graphite [8]. The uniformity of the measuring system's efficiency over time and the irradiation conditions are maintained by using the coincidence method for measuring the activity [3,7,13] or by using control neutron sources (e.g., Ra(α,n)Be, Ra(γ,n)Be) whose neutron source strength changes little over time [5,8,9].

In the second case, in which open sources are used, a substrate holding the sample is placed in a pulse fission chamber [4,10,12] or a two-chamber $4\pi\beta$ counter [3]. The measurement consists of recording the counting rate of fission products [4], fragment-fragment [3,12], or fragment-neutron [10] coincidences. Uniformity of the measuring system's efficiency is ensured by using the coincidence method [3,10,12], or by means of instrumentation [4].

In both cases, in addition to the random measurement error, there are systematic errors caused by:

- presence of associated, spontaneously fissile nuclides of ^{250}Cf and ^{254}Cf in the source;
- instability of the measuring conditions over time.

The method of measuring the constant λ which was used in Refs. [3-13] does not make it possible to separate experimentally the effect resulting from decay of the main nuclide from the effects caused by the associated nuclides. In the majority of experiments [3,4,7,9,10,12,13] allowance is made for this effect, introducing corrections calculated on the basis of the composition of the initial sample measured by means of mass spectrometry. In the experiments reported in Refs. [3,10-13], the sample was kept for 1-5 years, for various reasons, before the start of the measurements and the effect of the ^{254}Cf (which has a half-life of 60.5 days) was therefore disregarded. In Refs. [5 and 8], the composition of the initial sample is not given, and no correction was made for the effects of the associated nuclides.

Uncertainty in the composition of the initial sample and in the nuclear decay data of the associated nuclides is the reason to include systematic errors in the measurement results. In Table 2 this is considered as uncertainty due to corrections for the contribution of ^{250}Cf and ^{254}Cf radiation.

Despite the measures taken to ensure uniform efficiency, instability of the equipment over the time of the experiments cannot be completely excluded. Similarly, when Ra(α,n)Be and Ra(γ,n)Be sources are used for control purposes, the empirical time dependence of the neutron source strength of these sources has to be taken into account when normalizing the results of the individual measurements. Instability of the equipment and error in the determination of the time-dependence of the source strength of the radium sources are causes of systematic error. In Table 2, these are considered as uncertainties due to long-term instability.

In the second group of experiments, there is a specific source of systematic errors which are associated with the uncontrolled losses of californium as a result of the evaporation of the sample. Although measurements were made over 2π steradian angle in all experiments using open samples, it is still not possible to entirely exclude changes in the radiation detection conditions during the transfer of the sample from the support to the walls of the chamber, resulting in systematic errors caused by the evaporation of the sample.

It can be seen from Table 2, that for each of the results analysed, we can assume that there are residual systematic errors which cannot be excluded, and whose combined effect leads to a shift of the experimental result away from the "true value". The total measurement error should include not only the standard deviation of the result of calculating λ using statistical methods, but also a specific systematic component which is the cause for the discrepancy in the measurement results beyond the margin of error given in Table 1.

The measurement results have been analyzed and the half-life evaluation has been published in Ref. [15]; however, in view of the subsequent publications of experimental results given in Refs. [3,13] this evaluation needs to be refined.

VALUE OF $\bar{T}_1$ "WEIGHTED" ONLY BY THE ERRORS GIVEN BY THE AUTHORS

In the σ_i errors that they report in Refs. [3,7,9], the authors include not only the standard deviation of the calculated value for λ, but also an evaluation of the systematic error determined by the authors from an analysis of the measurement procedure. Therefore, although this was not done in the other experiments, the first approximation in the methodology proposed in the present work is the evaluation of $\bar{T}_1$ taking only σ_1 into account.

The weighted mean values for the half-life $\bar{T}$ and their standard deviation S were calculated using the normal formulas

$$\bar{T} = \sum_{i=1}^{L} g_i T_i \ ;$$

$$S^2 = L^{-1}\left[\sum_{i=1}^{L} g_i \sigma_i^2 + (\bar{T})^{-2} \sum_{i=1}^{L} g_i (T_i - \bar{T})^2\right] ,$$

where $g_i = \sigma_i^{-2} \Big/ \sum_{i=1}^{L} (\sigma_i)^{-2}$. The parameters used to calculate the mean weighted evaluated values are given in Table 3. According to the calculation results, $\bar{T}_1 = (2.6457 \pm 0.0026)$ and $S_1 = 0.1\%$.

"WEIGHTED " VALUE OF $\bar{T}_2$, TAKING INTO ACCOUNT THE ADDITIONAL ERRORS ARISING AS A RESULT OF NOT ALLOWING FOR ^{250}Cf AND ^{254}Cf EFFECTS

The work reported in Refs. [5 and 8] does not allow for the effect of the associated nuclides on the measurement results, and Ref. [11] does not allow for the effect of ^{250}Cf. As can be seen from Refs. [3,7,9,13], in which the composition of the initial sample is given in full, the change in the contribution of ^{250}Cf to the neutron source strength over one year may be as much as

0.2 to 0.4%. According to the data in Ref. [4], the contribution of 254Cf was 0.15% two years after the production of the sample, and according to the data in Ref. [9], the contribution was 0.076% after one and a half year.

In Refs. [5, 8 and 11] the composition of the sample is either not given at all, or given in a form which makes it impossible to introduce a correction for the effect of the associated nuclides. In the present work, this effect is taken into account by introducing an additional error θ_i which consists of the following three components:

- for not allowing for the 0.2% from ^{250}Cf,
- for not allowing for the 0.1% from ^{254}Cf, and
- for not allowing for the 0.3% effect of both nuclides.

Since the error θ_i is a systematic component, the total error σ_{2i} was calculated in Refs. [5,8,11] using the formula $\sigma_{2i} = \sigma_i + \theta_i$, and in the remaining experiments from the formula $\sigma_{2i} = \sigma_i$.

The effect of ^{254}Cf is not taken into account in Ref. [9], but since the composition of the sample is given in full, the value of T = 2.638 years determined by the authors was corrected in the present work and the value of T = 2.6396 years is used in the subsequent calculation. Calculation results yield $\overline{T}_2$ = (2.6468 ± 0.0021) years and S_2 = 0.08%.

PLANNING A MODEL "RATIONAL" EXPERIMENT

If in the equation describing the decrease in the nuclide activity as a consequence of decay, $N(t) = N_0 \exp(-\lambda t)$, the measured value is normalized by the usual method

$$\eta = N(t)/N_0 \tag{1}$$

taking the logarithm yields $\ln \eta = -\lambda t$ (2)
which is the equation for the line y = mx where $y = \ln \eta$, x=t and $m = -\lambda$.

In solving for η using equation

$$y_i = mx_i \tag{3}$$

the standard deviation of the parameter Δm can be calculated by the least squares method using the formula

$$\Delta m = \left\{\left(1/\sum_{i=1}^{n} x_i^2\right)\left[\sum_{i=1}^{n} d_i^2/(n-1)\right]\right\}^{1/2}, \tag{4}$$

where $d_i = y_i - mx_i$.

From equation (4) it is evident that the least squares method does not envisage limitations in the number of measurements, therefore some criterion has to be used in selecting a "rational" value for n. Let us express the time limit of the whole experiment T_o by T:

$$T_0 = pT. \tag{5}$$

Let us assume that n similar measurements $N_i(t)$ with a standard deviation σ_o are planned to be performed over equal time intervals, so that

$$t_i = ipT/n. \tag{6}$$

Let us introduce the following criterion taking the resolving ability of the measurement method into account:

$$y_i - y_{i+1} \geqslant \Delta y_i + \Delta y_{i+1} . \tag{7}$$

Since in accordance with Eqs. (2) and (3), $y_i = -\lambda t_i$, then taking Eq. (6) into account:

$$y_i - y_{i+1} = p\,\ln 2/n. \tag{8}$$

On the other hand, it follows from Eqs. (1) and (3), that $y_i = \ln(N_i/N_o)$ and $\Delta y_i = (\Delta N_i/N_i) + (\Delta N_o/N_o)$. For equivalent measurements

$$\Delta y_i = \Delta y_{i+1} = 2\sigma_0 . \tag{9}$$

Substituting Eqs.(8) and (9) in Eq. (7) we have

$$n \leqslant 0{,}2\, p/\sigma_0 = 0{,}2\, T_0/\sigma_0 T . \tag{10}$$

Introducing Eq. (6), we can express the series summation

$$\sum_{i=1}^{n} x_i^2 = p^2 T^2 \frac{(n+1)(2n+1)}{6n} . \tag{11}$$

For the statistically most probable line, in the absence of systematic errors, the following equation holds

$$\sum_{i=1}^{n} d_i^2/(n-1) = \sum_{i=1}^{n} (2\sigma_0)^2/[n(n-1)] . \tag{12}$$

Substituting Eqs. (11) and (12) in Eq. (4) we find

$$\Delta\lambda/\lambda = 2\sigma_0/p\ln 2\left[6n/(n+1)(2n+1)(n-1)\right]^{1/2} . \tag{13}$$

If we assume that N $\geq$ 5, then Eq. (13) can be simplified

$$\Delta\lambda/\lambda \approx 5\sigma_0/pn . \tag{14}$$

Substituting expression (10) in Eq. (14), and solving the inequality with respect to σ_o, we can determine that

$$\sigma_0 \leq 0{,}2\; T_0/T\,(\Delta\lambda/\lambda)^{1/2} . \tag{15}$$

Let us examine the procedure for planning an experiment to measure λ ^{252}Cf using the selected criterion. In Ref. [16], it is shown that as a function of the signal to background ratio of the measurement system, the duration of the experiment should be selected from the range 0.5T $\leq$ T_o $\leq$ 1.4T, whereby a longer period is permissible for a lower background level. Let us consider the case where T = T_o, i.e. p=1, and assume that the experiment is intended to measure the half-life T with $\Delta\lambda/\lambda$ = 0.1%.

Substituting the assumed values in equation (15), we find the value for the error σ_o with which the ratios η_i have to be exchanged to be $\sigma_0 = 0{,}6\%$. (16)

Substituting the Eq. (16) in Eq. (10) we can determine the rational number of measurements to be

$$n_0 = 33. \tag{17}$$

Finally, let us determine, using Eqs. (6) and (17), the interval Δt between consecutive measurements: $\Delta t = T/33 = 29$ days. With the least successful groupings of experimental points with respect to the calculated straight line, in the absence of systematic error, we have

$$d_i = 2\sigma_0 . \tag{18}$$

Substituting Eqs. (11) and (18) in Eq. (14), and performing the calculation, we have

$$\Delta\lambda/\lambda \leq 5\,(\sigma_0/p\sqrt{n}) \tag{19}$$

Inequality (19) can be used to process the results of the experiment as a test of the statistical control of the series of measurements carried out. If the calculated value agrees with Eq. (4) and the value of the standard deviation parameter satisfies inequality (19), the measurement can then be considered to be free from any systematic error that has not been taken into account caused by instability of the equipment or by the fact that the experimental conditions were not maintained.

"WEIGHTED" VALUE OF $\overline{T3}$ TAKING INTO ACCOUNT THE ADDITIONAL ERROR FOR THE "NON-RATIONALITY" OF THE EXPERIMENT

Table 3, which lists the values of p_i and n_i from Refs. [3-13], shows that there is a considerable latitude in the selection of the duration of the experiment (0.5 to 8 years) and in the number of measurements (2 to 50). This makes it difficult to compare the results since both factors affect the accuracy of the calculations of the decay constant using the least squares method. In the absence of any criterion, which makes it possible to give preference to particular values of these parameters, a representation of the "rational" model experiment was used in comparing the results in the present work.

The duration and number of measurements in each actual experiment was compared with the parameters of the "rational" experiment, $p_o = 1$ and $n_o = 33$, directed at achieving the measurement error Δ/λ = 0.1%. The difference between p_i and n_i, and p_o and n_o, was calculated as the factor causing the additional measurement error σ_i^* which was calculated using the formula $\sigma_i^* = \rho_0/\rho_i \times (n_0 \rho_i / n_i \rho_0)^{1/2}\, 0{,}1\%$: Thus all the experiments were "rationalized".

Since σ_i^* was calculated as a random error, it is included in σ_{3i} by the formula $\sigma_{3i} = [(\sigma_{2i})^2 + (\sigma_i^*)^2]^{1/2}$ From the calculational results, T_3 = (2.6473 ± 0.0028) years and S_3 = 0.11%.

Comparison of the $\overline{T1}$, $\overline{T2}$, and $\overline{T3}$ half-lives shows that the introduction of additional errors as a result as a result of not taking into account the associated nuclides and the deviation from the parameters of the "rational" experiments leads to an increase by 0.1% in the value of $\overline{T}$ with the same standard deviation.

The value T = (2.6473 ± 0.0028) years can be recommended on the basis of the results published in Refs. [3-13]. Analysis of the coefficients g_{3i} in Table 3 shows that the contribution of the results of Refs. [3-13] to T_3 is more than 50%.

It would be desirable to reduce the total influence of the results given in Refs. [3-13] on $\overline{T}_3$. In order to do this, it is necessary to carry out at least two independent experiments whose requirements can be formulated as follows:

- Measurement method. Activation detectors (preferably gold foils), enclosed source in fixed geometry in a moderator (water, graphite or polyethylene), foil activity to be measured using a $4\pi\beta-\gamma$ counter and coincidence method; the choice of method excludes sample evaporation and reduces the possibility of instability of the measuring conditions.

- Sample composition. The ^{250}Cf content is not limited, but it should be measured to an accuracy of 3-5%; the ^{254}Cf content in the initial sample should be not more than 0.02% and should be determined with an error of not more than 30%; the time interval between the sealing of the source and the first measurement should not be less than 14 months; a judicious choice of the sample composition minimizes the effect of associated nuclides.

- Measurement procedure. In accordance with the "rational" experiment plan, the length of the experiment should be 2.7 years, the number of measurements should be 33, and the time interval between measurements should be 1 month.

It can be assumed that the detailed planning of this experiments will make it possible to determine a value for the half-life of ^{252}Cf with an error of less tan 0.1%.

Table 1

HALF-LIFE AND BRIEF DESCRIPTION OF THE METHOD
USED IN ITS DETERMINATION

Year	T_{i}, years	σ_{i}, %	Method	Refs.
1965	2.646±0.004	0.15	N_f, OS, PFS	[4]
1969	2.631±0.006	0.23	Q_n, CS in MB relative to RAN	[5]
1969	2.621±0.006	0.23	Improvement of [5]	[6]
1973	2.659±0.010	0.36	Q_n, CS in MB absolute measurement	[7]
1974	2.628±0.010	0.38	Q_n, CS in GM relative to RAN	[8]
1974	2.638±0.007	0.27	Q_n, CS in MB relative to RGN	[9]
1974	2.637±0.005	0.19	N_f, OS, PFS	[10]
1981	2.640±0.007	0.27	ACT	[11]
1982	2.651±0.004	0.15	N_f, OS, PFS	[12]
1983	2.648±0.002	0.08	Q_n, CS in DW absolute measurement	[13]
1985	2.6503±.0031	0.12	N_f, OS in $4\pi\beta$ detector	[3]
1983	2.64	-	Recommended value	[14]
1985	2.646	-	Recommended value	[1b]

Legend:

N_f - Counting rate measurement: of fission fragments in a pulse fission chamber (PFC), of the fragment-fragment coincidences in a double chamber $4\pi\beta$-counter, or of fragment-neutron coincidence in a system consisting of the PFC and a neutron detector.

Q_n - Measurement of the neutron flux by one of the absolute methods: manganese bath (MB), activation of gold foils in distilled water (DW), in graphite moderator (GM), the graduated assiciated particle method; or relative to reference Ra(α,n)Be or Ra(γ,n)Be neutron sources.

ACT - measurement of manganese detector activity that were irradiated by an open source in polyethylene

OS - Open source

CS - Closed source

PFS - pulse fission chamber

MB - manganese bath

GM - graphite moderator

DW - distilled water

RAN - Ra(α,n)Be source strength

RGN - Ra(γ,n)Be source strength

Table 2

Classification of systematic error components

Source and type of discrepancy	References										
	3	4	5	6	7	8	9	10	11	12	13
Corrrection for the contribution of the ^{250}Cf and ^{254}Cf radiation	+	+	+	+	+	+	+	+	+	+	+
Long term instability due to:											
- instrumentation	+	+	-	-	+	-	-	+	+	+	+
- radium source normalization	-	-	+	+	-	+	+	-	-	-	-
Sample evaporation	+	+	-	-	-	-	-	+	-	+	-

Table 3

Parameters used
in calculating the $\overline{T}_1$, $\overline{T}_2$ and $\overline{T}_3$ quantities

Refs.	T_i, yrs	σ_i, %	g_{1i}, %	θ_i, %	σ_{2i}, %	g_{2i}, %	P_i	n_i	σ^*_i, %	σ_{3i}, %	g_{3i}, %
/3/	2,6503	0,12	17,2	-	0,12	18,6	1,5	39	0,075	0,141	32,5
/4/	2,646	0,15	11,0	-	0,15	11,9	0,82	33	0,135	0,202	15,8
/6/	2,621	0,23	4,7	0,3	0,53	0,9	1,66	45	0,067	0,534	2,2
/7/	2,659	0,36	1,9	-	0,36	2,0	0,38	17	0,263	0,446	3,2
/8/	2,628	0,38	1,7	0,3	0,68	0,6	0,38	4	0,456	0,819	1,0
/9/	2,638	0,27	3,4	-	-	-	-	-	-	-	-
	2,6396	-	-	-	0,27	3,7	1,77	2	0,304	0,355	5,1
/10/	2,637	0,19	6,9	-	0,19	7,4	0,19	6	0,526	0,559	2,1
/11/	2,640	0,27	3,4	0,2	0,47	1,2	1,5	50	0,067	0,475	2,9
/12/	2,651	0,15	11,0	-	0,15	11,9	1,23	3	0,300	0,335	5,7
/13/	2,648	0,08	38,8	-	0,08	41,8	3,07	7	0,125	0,148	29,5

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