Half-life of the 188-keV isomer of ^{184}Re

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We report a new half-life measurement of the 188-keV isomeric state in ^{184}Re. The ^{184m}Re isomer was produced by means of neutron activation of natural rhenium foil in the MARIA nuclear reactor at the National Centre for Nuclear Research (NCBJ). The observation of the ^{184m}Re decay for over two half-life periods has yielded the new value of $T_{1/2} = 177.25 \pm 0.07$ d. Our result is the most precise determination of the half-life of the ^{184m}Re isomer and improves the precision of the previously known value of $T_{1/2} = 169 \pm 8$ d by two orders of magnitude.

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I. INTRODUCTION

The doubly odd ^{184}Re nucleus has an unstable ground state with $I^\pi = 3^{(-)}$ and half-life of 35.4 ± 0.7 d [1], and a β-unstable isomer with $I^\pi = 8^{(+)}$ that was found by Johnson in 1963 [2]. The production of both states in the (γ, n) and $(n, 2n)$ reactions allows for the study of the dependence of the level density on the excitation energy and angular momentum [3,4]. The similarities between ^{184}Re and the adjacent odd-odd isotope ^{186}Re supported the assignment of new levels and transitions above the 148.2 keV ^{186m}Re isomer [5]. While the ground-state half-life in ^{184}Re is known with fairly good precision [1], the half-life of the ^{184m}Re isomer $T_{1/2} = 169 \pm 8$ d [2] is known with a large uncertainty. The experimental value was determined almost 60 years ago and, despite the development of modern methods of nuclear spectrometry, it has not been improved so far. Here in National Centre for Nuclear Research we investigated the production methods of rhenium isotopes, especially in the isomeric state. In our on-site research reactor MARIA [6], rhenium isotopes were produced by means of thermal-neutron activation of a natural rhenium foil. Among them was the ^{184}Re isotope and its ^{184m}Re isomer, produced in ^{185}Re$(n, 2n)$ reaction. We took advantage of having both the unique material and suitable measurement setup for the time long enough to tackle the challenge of improving the available result.

II. EXPERIMENTAL SETUP

Natural rhenium (37.4% ^{185}Re, 62.6% ^{187}Re) foil in the form of a 25 mm × 25 mm × 100 μm, 1.32 g leaf, procured from Goodfellow, was irradiated for 122 hours in the MARIA research reactor [6]. The activated sample was then let to cool down for another 32 months. At this point, after over 28 full half-life periods $T_{1/2} = 35.4 \pm 0.7$ d [7], all produced ^{184}Re ground state was gone apart from the residual amount remaining in a state of equilibrium with the ^{184m}Re isomer, which has a significantly longer half-life $T_{1/2} = 169 \pm 8$ d. This sample was measured for 24 hours, periodically at least once per ten days, from January 2021 to February 2022. The detection setup consisted of two independently configured high-purity germanium (HPGe) Canberra GX3520 and Ortec GLP-25300/13-S detectors. The sample was placed in a fixed geometry of about 3 cm between the detectors' entrance windows. Each acquisition track consisted of a NIM Ortec 672 spectroscopy amplifier and a Tukan multichannel analyzer [8].

III. DATA ANALYSIS

^{184}Re decay is complex; see Fig. 1. Both the ground state and the isomer can undergo the electron-capture process, each with its own Q_{EC} values, and each feeds different levels in ^{184}W. ^{184m}Re may also deexcite to the ^{184}Re ground state via γ or conversion-electron emissions. As the sample is in a state of equilibrium, it may be safely assumed that the activity of any transition should follow the law of radioactive decay with the half-life of ^{184m}Re. In the analysis it is preferable to take into account all of those transitions, at least as long as they can be identified in the accumulated energy spectra; see Fig. 2. Note that the 83.3 and 188 keV IT decays from the isomer (see Fig. 1) are not visible in the γ-ray spectrum because they are highly converted. The 104.7-keV decay, however, is not, thus it is clearly present in Fig. 2. When measuring decay half-lives, an incorrect approximation of the instrumentation dead time is the capital contribution to systematic errors. For this reason, rather than directly using the intensity of a given transition, or more technically its net peak area, it is better to calculate the peak area ratio of that transition and another one from some reference isotope. The peak net area is derived

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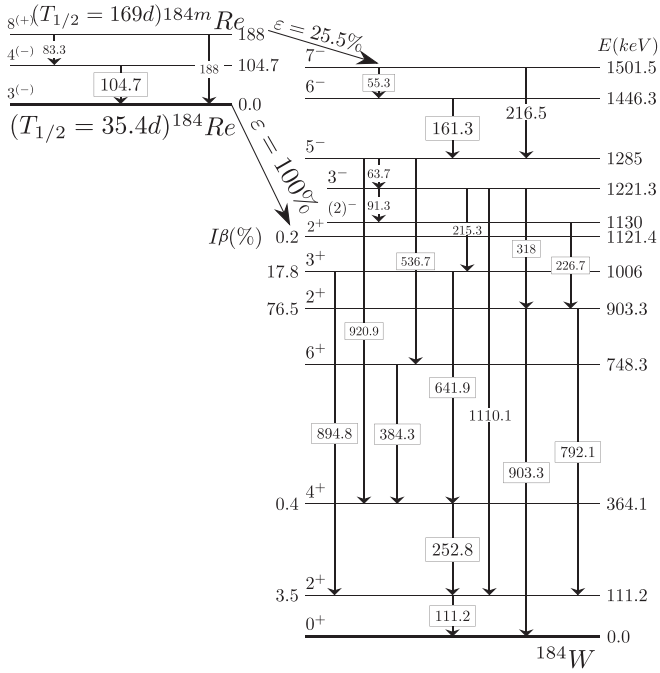

FIG. 1. A schematic view of the ^{184}Re decay and partial level scheme of ^{184}W. The data are taken from Ref. [7]. Note that the $T_{1/2}$ given in the figure is from the literature and not from the present work. The lines marked in the frames were used in the analysis.

from a Gaussian peak fit with a linear background approximation. Only transitions which could be fitted with a single gaussian curve were analyzed. The reference source should always accompany the measurement of the sample. In fact, it should be a part of the measured sample, in order to guarantee the preservation of geometry for the duration of the entire experiment. The reference should have a well known half-life, preferably longer than that of the investigated isotope, and its measured γ lines should have adequate intensity/statistics. This approach is impeccably presented by Pommé [9], and successfully applied by Pibida *et al.* [10].

Evolution of a given ratio can be described by a function

$$A(t) = A_0 \exp(-\lambda t) = \frac{A_{\mathrm{Re}}(t)}{A_{\mathrm{ref}}(t)} \tag{1}$$

with its decay constant λ as a free parameter. The sought half-life is calculated according to the equation

$$T_{1/2} = \frac{\ln(2)}{\lambda_{\mathrm{ref}} + \lambda} \tag{2}$$

where λ_{ref} is the decay constant of the reference isotope. The discrepancy of $T_{1/2}$ is derived according to the law of error propagation. Uniquely for our analysis, activated contaminations of the rhenium sample provided two excellent reference candidates: ^{134}Cs with $T_{1/2} = 2.0652 \pm 0.0004$ yr [11] and ^{60}Co $T_{1/2} = 1925.28 \pm 0.14$ d [12]. Our criteria for a reference were also fulfilled by the 137.2 keV transition assigned to the decay of ^{186m}Re. Only the estimation of the isomer's half-life is known, but its value 2×10^5 yr is long enough to disregard λ_{ref} from Eq. (2) completely. The example of the 161.3 keV line decay, with three different references, is presented in Fig. 3. The derived half-lives for all ratio combinations of rhenium and reference isotope lines, for both detectors, are collated in Table I. Graphical representation of the Table I can be seen in Fig. 4. Fourteen γ transitions times eight γ lines from the ^{60}Co, ^{134}Cs, and ^{186m}Re references, times two detectors (not in all cases though) yielded 208 independent half-life values for the ^{184m}Re isomer. The weighted average of this set, with the weight defined as

$$w_i = \frac{1}{S_{x_i}^2}, \tag{3}$$

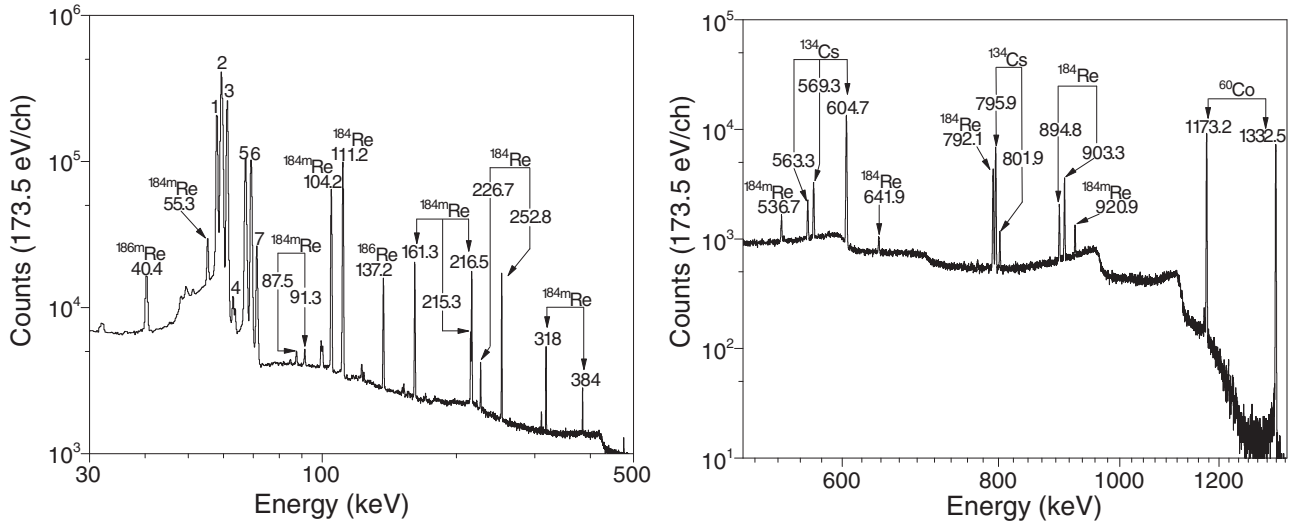

FIG. 2. An example of a gamma-ray spectrum measured by an HPGe Ortec detector (d1) for 24 h. On the left graph, the several γ rays of the ^{184}Re decay are clearly observed. The x-ray area marked with numbers 1–7 comes from Re{**3**: $K_{\alpha 1} = 61.1$ keV, **2**: $K_{\alpha 2} = 59.7$ keV, **6**: $K_\beta = 69.3$ keV}, W{**2**: $K_{\alpha 1} = 59.3$ keV, **1**: $K_{\alpha 2} = 58.0$ keV, **5**: $K_\beta = 67.2$ keV}, Os{**4**: $K_{\alpha 1} = 63.0$ keV, **3**: $K_{\alpha 2} = 61.5$ keV, **7**: $K_\beta = 71.4$ keV}. Another six γ lines from the decay of ^{184}Re are shown on the right graph. Additional reference γ lines for ^{186}Re (137.2 keV), ^{134}Cs, and ^{60}Co used to derive the half-life are also presented.

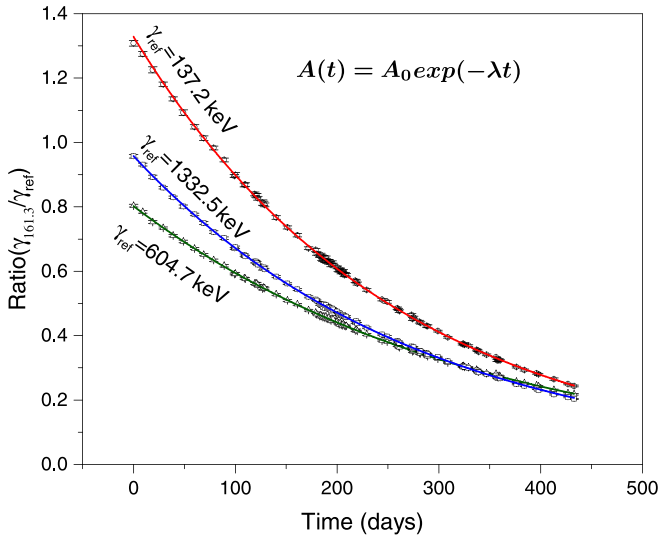

FIG. 3. Time evolution of the 161.3 keV γ line intensity with three different references. Each point on the graph represents the ratio of the γ-ray emission rate of the 161.3 keV line from ^{184}Re to the appropriate reference value lines (137.2 keV from ^{186}Re, 604.7 keV from ^{134}Cs, and 1332.5 keV from ^{60}Co).

where S_{x_i} are the uncertainty estimators of x_i, yields the new value of $T_{1/2} = 177.25 \pm 0.07$ d. Please note that the weighted average was calculated using full precision values rather than the rounded ones collated in Table I.

IV. MEASUREMENT ERRORS ESTIMATION

A decay half-life is obviously measured indirectly so its statistical error is reflected in the uncertainty of the free-fit λ parameter of the function from Eq. (1), and then recalculated according to the law of error propagation. The curve fit quality is affected by the constituting individual measurement points: their number, distribution, span, and individual uncertainties. To some extent, the first three factors can compensate for problems with measurement precision, and vice versa. An extremely good measurement precision can compensate for the overall low number of points, their uneven distribution, or the inability to continue an experiment for the duration of two or three full decay periods. The most sensitive part is the measurement precision since it involves a string of mathematical operations: peak fitting, background subtraction, and dividing numbers. The error propagation formula is notorious for being unforgiving for this kind of a process. Ultimately, it is the systematic errors that limit the accuracy of the final result.

There are two main contributions to systematic errors in our experiment. The first one is the acquisition dead time. This issue can be resolved at the cost of statistical uncertainties by means described earlier in the Data Analysis section. The

TABLE I. ^{184m}Re half-life measurements in this work. "d1" denotes Ortec GLP-25300/13-S while "d2" corresponds to Canberra GX3520. γ-ray energies are given in keV while half-life values are in days.

^{184}Re	Det.	^{186}Re 137.2	^{134}Cs 563.3	^{134}Cs 569.3	^{134}Cs 604.7	^{134}Cs 795.9	^{134}Cs 801.9	^{60}Co 1173.2	^{60}Co 1332.5
55.3	d1	177.9 ± 0.6	176.7 ± 1.1	176.6 ± 0.8	177.2 ± 0.6	177.4 ± 0.7	178.9 ± 1.3	177.9 ± 0.7	177.5 ± 0.7
104.7	d1	177.72 ± 0.26	176.4 ± 0.8	176.4 ± 0.5	177.6 ± 0.3	177.4 ± 0.3	179.2 ± 1.0	178.11 ± 0.28	177.96 ± 0.30
	d2	176.91 ± 0.24	176.6 ± 0.4	176.3 ± 0.4	176.6 ± 0.3	177.9 ± 0.4	176.6 ± 1.0	176.7 ± 0.4	176.2 ± 0.5
111.2	d1	177.73 ± 0.21	176.5 ± 0.8	176.6 ± 0.5	177.81 ± 0.21	177.56 ± 0.28	179.3 ± 1.0	178.24 ± 0.23	178.07 ± 0.24
	d2	176.73 ± 0.22	175.3 ± 0.3	176.69 ± 0.27	176.34 ± 0.26	177.2 ± 0.4	176.8 ± 0.9	176.4 ± 0.3	176.0 ± 0.4
161.3	d1	177.3 ± 0.3	176.0 ± 0.8	175.9 ± 0.4	177.17 ± 0.28	177.1 ± 0.3	178.1 ± 1.0	177.67 ± 0.28	177.47 ± 0.29
	d2	177.67 ± 0.17	176.9 ± 0.4	177.18 ± 0.25	177.50 ± 0.24	178.6 ± 0.3	177.5 ± 1.0	177.7 ± 0.3	177.1 ± 0.4
226.7	d1	178.3 ± 1.3	176.6 ± 1.4	176.3 ± 1.3	175.5 ± 1.2	177.2 ± 1.3	178.5 ± 1.6	178.1 ± 1.2	177.8 ± 1.2
	d2	178.9 ± 1.1	177.9 ± 1.1	178.8 ± 1.1	178.9 ± 1.1	180.5 ± 1.2	179.0 ± 1.4	179.3 ± 1.2	178.7 ± 1.2
252.8	d1	177.4 ± 0.4	176.2 ± 0.8	176.0 ± 0.6	177.1 ± 0.4	177.1 ± 0.4	178.6 ± 1.0	177.7 ± 0.4	177.5 ± 0.4
	d2	177.19 ± 0.21	176.18 ± 0.30	176.56 ± 0.24	176.89 ± 0.19	177.89 ± 0.28	177.1 ± 0.9	177.03 ± 0.27	176.5 ± 0.3
318.0	d1	178.6 ± 0.7	176.9 ± 0.9	176.5 ± 0.9	177.6 ± 0.6	177.7 ± 0.7	179.3 ± 1.2	178.50 ± 0.6	178.2 ± 0.6
	d2	177.4 ± 0.4	176.0 ± 0.4	176.9 ± 0.5	177.3 ± 0.5	178.4 ± 0.6	177.0 ± 1.0	177.5 ± 0.5	176.9 ± 0.6
384.3	d1	177.2 ± 1.3	176.0 ± 1.4	175.3 ± 1.4	176.3 ± 1.3	176.4 ± 1.3	178.1 ± 1.8	176.9 ± 1.3	176.6 ± 1.3
	d2	177.3 ± 0.8	177.1 ± 0.8	177.3 ± 0.8	177.5 ± 0.7	179.2 ± 0.8	176.6 ± 1.3	177.9 ± 0.8	177.3 ± 0.8
536.7	d1	177.6 ± 2.0	176.1 ± 2.2	175.7 ± 2.1	176.8 ± 2.0	177.0 ± 2.1	178.6 ± 2.3	177.5 ± 2.0	177.2 ± 2.0
	d2	180.4 ± 1.2	179.9 ± 1.2	180.0 ± 1.1	180.3 ± 1.1	181.7 ± 1.1	179.8 ± 1.3	180.5 ± 1.2	179.9 ± 1.2
641.9	d1	185 ± 4	183 ± 3	183 ± 4	184 ± 3	184 ± 4	185 ± 4	185 ± 4	184 ± 4
	d2	174.5 ± 1.3	173.8 ± 1.3	174.4 ± 1.2	174.5 ± 1.3	176.1 ± 1.3	175.1 ± 1.8	174.9 ± 1.3	174.3 ± 1.3
792.1	d1	178.0 ± 0.6	176.2 ± 1.0	176.1 ± 0.8	177.1 ± 0.6	177.3 ± 0.6	178.9 ± 1.3	177.8 ± 0.6	177.4 ± 0.6
	d2	177.7 ± 0.6	177.6 ± 0.8	177.3 ± 0.5	177.7 ± 0.5	178.9 ± 0.5	176.2 ± 0.8	177.9 ± 0.5	177.3 ± 0.4
894.8	d1	178.0 ± 1.0	177.1 ± 1.3	176.3 ± 1.1	177.3 ± 0.9	177.6 ± 0.9	179.6 ± 1.4	178.0 ± 0.9	177.6 ± 0.9
	d2	177.2 ± 0.4	176.5 ± 0.4	176.9 ± 0.3	177.12 ± 0.28	178.5 ± 0.3	177.7 ± 0.9	177.36 ± 0.29	176.8 ± 0.3
903.3	d1	178.2 ± 0.6	176.6 ± 0.9	176.6 ± 0.8	177.3 ± 0.6	177.4 ± 0.6	179.1 ± 1.2	177.9 ± 0.6	177.6 ± 0.6
	d2	177.3 ± 0.3	176.5 ± 0.3	176.82 ± 0.22	177.05 ± 0.14	178.22 ± 0.21	177.5 ± 0.9	177.24 ± 0.15	176.67 ± 0.18
920.9	d1	182.4 ± 2.1	180.5 ± 2.2	179.8 ± 2.1	181.3 ± 2.1	181.4 ± 2.1	182.5 ± 2.5	182.1 ± 2.1	181.8 ± 2.1
	d2	176.2 ± 0.7	176.2 ± 0.6	176.2 ± 0.6	176.3 ± 0.6	177.9 ± 0.6	177.9 ± 1.1	176.7 ± 0.6	176.1 ± 0.6

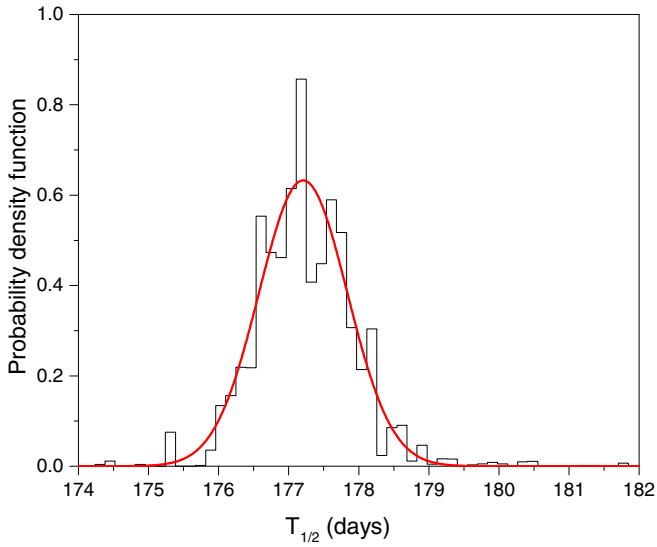

FIG. 4. Graphical representation of Table I content. Contributions of the values to the histogram are weighted by their uncertainties. The symmetric bell shape of the presented distribution testifies that the individual half-life values are truly independent.

second, and the harder one to tackle, is the correct subtraction of background under energy peaks, the reference peaks in particular, in a measured energy spectrum.

In order to estimate the overall impact of our systematic errors, we applied the presented analysis approach to calculate the half-life of ^{134}Cs in our sample. ^{60}Co 1173 and 1332 keV peaks served again as the reference activity. The weighted average yielded a value of 2.064 ± 0.006 yr. It is close enough to the published value of $T_{1/2} = 2.0652 \pm 0.0004$ yr [11], considering the fact our experiment spanned only one-half of the ^{134}Cs decay period.

The weighted average does not eliminate systematic errors but is a tool to appraise their scale. The important assumption is that the weighted results are not correlated and so neither are their discrepancies. We verified this assumption by observing the probability density distribution of over 200 results from Table I; see Fig. 4. The histogram is symmetric and fits well the normal distribution.

Statistics offers two estimators for the weighted average uncertainty:

$$S_{\text{int}}^2 = \sum_{i=1}^{n} \frac{1}{w_i^2} \tag{4}$$

and

$$S_{\text{ext}}^2 = \frac{S_{\text{int}}^2}{n-1} \sum_{i=1}^{n} w_i (x_i - \bar{x})^2. \tag{5}$$

The uncertainty of the weighted average $\bar{x}$ is thus $\text{Max}(S_{\text{int}}, S_{\text{ext}})$. In the ideal situation both estimators are expected to converge. In a real experiment if the S_{ext} estimator is significantly bigger than the S_{int} it indicates underestimated uncertainties S_{x_i} of individual x_i values. For the experiment described in this article S_{ext} was revealed to be bigger than S_{int} by the factor of 2. This suggests that contributions from statistical and systematic errors to the uncertainties of partial results, see Table I, are of the same order, and it would be very hard to point out one single, capital contributor to uncertainty of the final result. Further improvement in this field would require equal improvements in all aspects of the performed experiment.

V. SUMMARY

In this work we report the new value for the half-life of the ^{184m}Re isomer to be $T_{1/2} = 177.25 \pm 0.07$ d. Modern spectrometry resources as well as the accurate analysis approach enabled an increase in the precision by two orders of magnitude. The determined new value of half-life is somewhat longer than the previously reported 169 ± 8 d [2], but well within its discrepancy bars. This result is consistent with Pommé [9] who remarks that incorrect consideration of systematic errors in the analysis leads, in general, to undervaluation of measured decay lifetimes.

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