Half-life of ^{176}Lu

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The half-life of ^{176}Lu was deduced using Ge γ-ray spectrometry from the mass and activity of a lutetium oxide sample enriched in ^{176}Lu. The half-life was found to be $4.05\pm0.09\times10^{10}$ y. The relative intensities of the γ rays were also measured and are compared to previously reported values.

INTRODUCTION

The half-life of ^{176}Lu has been of considerable interest over the past 50 years. A number of papers have been published in which the half-life has been determined.[1–18] A process for using the ^{176}Lu/^{176}Hf couple for age dating of rocks which contain a sufficient amount of the Lu and not too much of the Hf has been developed, and ^{176}Lu has been used to determine the age of the *s*-process nuclei. Results from such techniques depend on the value used for the half-life. Unfortunately, the reported values have varied from 2.1 to 7.3×10^{10} yrs and even those more recent ones which agree more closely vary from 3.27 to 4.56×10^{10}yrs.

Table I presents the previous work and the half-life values reported. Many of the earlier attempts are probably flawed by the difficulty of measuring samples of low activity (the natural abundance of ^{176}Lu is 2.6% and its half-life is very long) and of obtianing pure samples, as natural Lu is likely to be contaminated with Th and its daughters. Determinations of the half-life by beta counting are particularly likely to be affected by Th contamination. Other measurements reported have used limited or poor experimental methods or have not reported all the detail relevant to the measurement. In Addition, as can be seen from Table I, many of the uncertainties assigned to the half-life values appear to be unrealistically small and may include only statistical uncertainties from the counting process.

Some recent experimenters have used a γ-γ sum-peak coincidence technique originally suggested by McNair[8] to measure the detector efficiency. However, a difficulty with this method has been pointed out by Sutherland and Buchanan;[19] for extended sources, the variation in response of the detector to different parts of the source must considered. Experimenters who have used this method have also assumed that the 202- and 307-keV transitions are converted only negligibly, which is not the case.

Both Norman[15] and Sato *et al.*[17] used γ-ray spectrometry to determine the absolute activity of an Lu sample of known ^{176}Lu mass. Norman applied corrections for summing effects, internal conversion, and attenuation in the sample. It is not clear whether or not Sato *et al.* applied corrections for summing effects to their data, but if not, it could account for their value being different and lower than Norman's.

To clear these difficulties, we decided to measure the half-life of ^{176}Lu measuring the 88-, 202-, and 307-keV γ-ray emission rates with a Ge detector. The γ-ray emission rates were converted to activities by dividing by the respective γ-ray emission probabilities. The activities were averaged and the ^{176}Lu half-life was deduced by dividing the activity by the number of ^{176}Lu atoms in the lutetium oxide sample.

EXPERIMENTAL

From Oak Ridge National Laboratory (ORNL), we obtained a sample of Lu_2O_3 enriched to (44.23 ± 0.23)% in ^{176}Lu. The sample, including impurities as analyzed and reported by ORNL, weighed 34.1 mg. The sample was counted in the glass vial in which it arrived from ORNL. The vial has a cross sectional area 1 cm^2 and the Lu_2O_3 powder could be made to form an evenly distributed layer that just barely covered the bottom of the vial by carefully tapping the side of the vial. This vial was then mounted to the sticky side (facing up) of a piece of tape stretched across a 2.54-cm diameter hole at the center of an aluminum card. In this way the sample was counted on a vertically mounted Ge detector in a reproducible source-to-detector distance. Following three spectral measurements, the Lu_2O_3 powder was removed from the vial and its mass determined. The powder was transferred to a small Al foil boat and the mass was determined to be 33.75 mg, about a 1% difference from the ORNL reported mass. The powder was then returned to the original container. Several months later the mass of Lu_2O_3 was remeasured and found to be 33.8 mg. This value was used in calculating the number of ^{176}Lu atoms present. The mass of the chemical impurities, as listed on the sample identification and analysis sheet from ORNL, was subtracted and the Lu_2O_3 mass was converted to the number of ^{176}Lu atoms presented in the sample. The number of ^{176}Lu atoms in the sample is $(4.42\pm0.05)\times10^{19}$ atoms.

The γ-ray spectrometer used in the present experiment is an intrinsic Ge detector of 160 cm^3 nominal active volume. The glass vial containing the sample was counted at a 10-cm distance from the detector housing, and three separate 64-h counts were made. One of the spectra thus obtained is shown in Fig. 1. A fourth spectrum was accumulated under the same condictions after the

TABLE I. Determinations of ^{176}Lu half-life.

Reference	Experiment	Half-life (10^{10} yrs)
1	beta counting	4.
2	beta counting	7.3±2
3	beta counting	2.4
4	γ, NaI	2.15±0.01
5	2*pi* beta proportional	4.56±0.3
6	γ, NaI	2.1±0.2
7	radiogenic	2.17±0.35
8	sum peak	3.6±0.1
9	4 pi beta liq. scint.	2.18±0.06
10	beta-γ coincidence	3.58±0.05
11	sum peak	5.0±0.3
12	liq. scint., coincidence	3.27±+0.05
13	radiogenic	3.3±0.5
14	sum peak	3.79±0.03
15	γ counting, Ge(Li)	4.08±0.24
16	radiogenic	3.53±0.14
17	γ counting	3.78±0.01
18	radiogenic	3.57±0.14

second mass determination. Insufficient statistics were gathered at the 10-cm source-detector distance for the 401 keV line to be observed, but all other expected lines were observed in the spectra.

The efficiency of the detector for the sample in the glass vial geometry at 10 cm was determined using a mixed radionuclide standard solution (traceable to NIST), containing ^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{203}Hg, ^{113m}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y, and ^{60}Co. A ^{182}Ta source was also counted and the resulting relative efficiencies were fitted to the mixed source efficiency curve obtained with the mixed radionuclide source, so that the shape of the efficiency curve could be better defined in the energy region required for the ^{176}Lu γ rays. Attenuation corrections were calculated for the self attenuation in the 33.8 mg Lu_2O_3 sample uniformly distributed over the bottom of the one-centimeter area vial[20] and are given in Table II.

The areas in the peaks at 88, 202, and 307 keV were calculated with a γ-ray spectral analysis package developed at the Idaho National Engineering Laboratory (INEL), called VAXGAP.[21] The uncertainties in the peak areas represent one estimated standard deviation. The

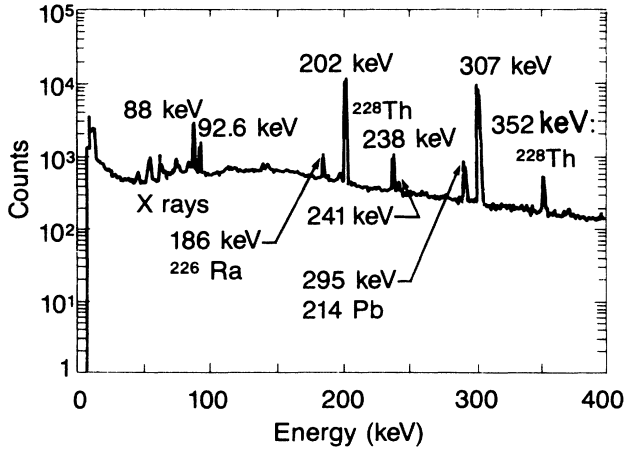

FIG. 1. Spectrum of Lu_2O_3, 44.23% enriched in ^{176}Lu. The Lu_2O_3 was in a glass stable isotope vial at 10 cm from the detector housing and was counted for 64 h.

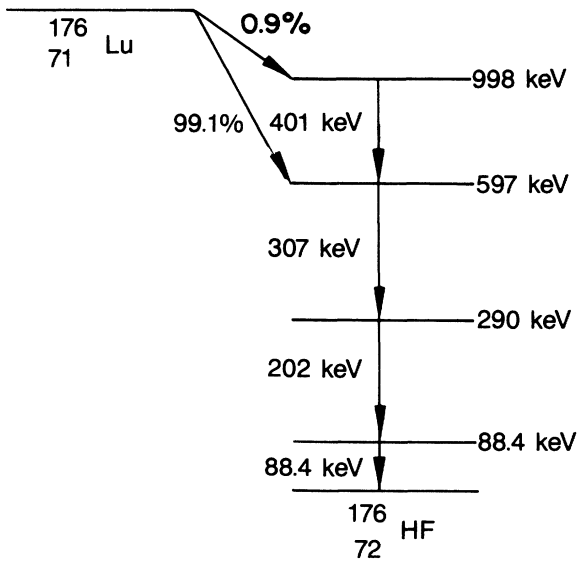

FIG. 2. Decay scheme of ^{176}Lu.

TABLE II. Conversion coefficients and γ-ray emission probabilities for ^{176}Lu and self attenuation corrections and coincidence summing corrections for the ^{176}Lu sample and counting geometry used in the present experiment.

γ-ray energy (keV)	$\alpha_t(i)$[a]	$P_\gamma(i)$[b]	Attenuation correction[c]	Coincidence summing correction[d]
88.37	5.86±0.18	14.6±0.4	0.9301±0.007	1.029±0.003
201.87	0.281±0.008	78.1±0.5	0.9938±0.0006	1.0175±0.0017
306.91	0.0746±0.0022	93.06±0.19	0.9975±0.0003	1.0157±0.0016
401.13	0.0347±0.0010	0.84		

[a]The conversion coefficients $\alpha_t(i)$ for the 88-, 202-, and 307-keV transitions are taken to have a pure $E2$ multipolarity; the uncertainties in the total conversion coefficients from Ref. 23 were assumed to be ±3% (one estimated standard deviation).
[b]The γ-ray emission probability, $P_\gamma(i)$, expressed as γ rays per 100 decays, was deduced using Eq. (1), assuming that 100% of the decays pass through each transition.
[c]The self attenuation correction is based on the assumption that the lutetium oxide powder was distributed with uniform thickness over the 1-cm^2 area of the bottom of the vial.
[d]Coincidence summing corrections for a Ge detector with the sample counted at a 10-cm distance from the detector housing.

relative intensities of the γrays were measured and are compared in Table III with the relative intensities reported previously. Although the half-life of ^{176}Lu has been measured frequently, the relative γ-ray intensities have not been often reported. The absolute activity was calculated from the peak areas as described below.

As shown in Fig. 2, ^{176}Lu decays by beta decay to the 597- and 998-keV levels of ^{176}Hf. There is no beta feeding to the ^{176}Hf ground state or to the 88- and 290-keV levels. Therefore, all of the decays pass through the 88-, 202-, and 307-keV transitions so that activity can be determined from the γ-ray emission rates and the total conversion coefficients. Table II gives the conversion coefficients (α_t) and estimated standard deviations for each transition.[23] The γ-ray emission probability of each γ-ray, i, is then

$$P_\gamma(i)=100\%\times 1/[1+\alpha_t(i)] \ . \quad (1)$$

These are also given in Table II along with their propagated uncertainties.

Coincidence summing corrections for each γ ray were calculated using the KORSUM program developed by Debertin and Schotzig.[24] The calculated correction factors and their associated uncertainties are give in Table II.

RESULTS

The absolute activity, A, for ^{176}Lu is

$$A = R_\gamma(i)C_s(i)(100/t)C_e(i)C_a(i)P_\gamma(i) \ , \quad (2)$$

where $R_\gamma(i)$ is the peak area in counting rate, $C_e(i)$ is the efficiency, $C_a(i)$ is the attenuation correction, $C_s(i)$ is the cascade summing correction, and $p_\gamma(i)$ is the emission probability (γ rays per 100 decays) of γ ray, i. The γ-ray emission rates for each of the γ-ray energies were converted to ^{176}Lu activities and these data were reduced in a series of three steps.

(i) First, the uncertainties in the net peak areas were combined in quadrature with a ±1% estimated standard deviation in the source-to-detector distance reproducibility to yield the values given in Table IV for each individual measurement. The weighted average of these four values is given in Table IV directly below the individual measurements.

(ii) Second, for each γ-ray energy the uncertainties in the respective γ-ray emission probability, coincidence summing correction, and self-absorption correction were combined in quadrature with the uncertainty in the weighted average from step (i) and with the uncorrelated portion of the uncertainty in the full-energy-peak efficiency. [The uncertainty in the full-energy-peak efficiency was divided into two parts: an uncorrelated part (±3% for the 88-keV γ ray and ±1.5% for the 202- and 307-keV γ rays) that can be reduced through the averaging process and a correlated part (±1.5%) that cannot be averaged away.] The results from step (ii) are shown in Table IV to the right of the weighted averages from step (i). A weighted average of these values was then calculated and appears below the three weighted average values.

(iii) Third, the uncertainty in the correlated portion of the full-energy-peak efficiency was combined quadrature

TABLE III. Relative intensities of ^{176}Lu γ lines.

γ line	88.4 keV	201.9 keV	306.9 keV
This experiment	15.5±0.6	83.3±2.2	100
Calculated[a]	15.7	83.9	100
Reference 22	14.09	90.32	100
Reference 4	16.8	90.2	100

[a]Calculation based on 99.1% feeding to the 597-keV level with the 88-, 201-, and 306-keV γ-ray transitions in cascade and the α values of Table II. The 306-keV γ-ray intensity was normalized to 100.

with the uncertainty in the averaged value from step (ii) to yield the ^{176}Lu activity with its uncertainty. The activity with all uncertainties propagated is given at the bottom of Table IV. From this ^{176}Lu activity and the number of ^{176}Lu atoms in the sample, the half-life is computed from the following equation:

$$T_{1/2}[(\ln 2)N_0]/A = (4.05 \pm 0.09) \times 10^{10} \text{ yrs},$$

where A is the activity and N_0 is the number of ^{176}Lu atoms in the Lu_2O_3 sample. This value agrees very well with that obtained by Norman, but is in disagreement with most of the other values in Table I.

CONCLUSIONS

If all measurements of the half-life of ^{176}Lu using beta counting on natural Lu_2O_3 are eliminated, because of the difficulties of accurately counting low-activity samples and because of possible errors due to Th contamination, and if the radiogenic measurements are also eliminated, because of difficulties inherent in the technique used, then we are left with five reported measurements of which three use some variation of the sum-peak method.[10,11,14] Problems with the sum-peak method were discussed in the Introduction. The remaining method[15,17] uses γ-ray spectrometry to measure the absolute activity, from which the half-life is calculated. This method appears to be the best approach to use, provided all necessary corrections are made, and was used by Norman[15] and Sato *et al.*[17] and in the present work. Of these three reported values, two Norman's and the present one, agree quite well, while the value reported by Sato *et al.* is significantly lower. We recommend that the present value of the ^{176}Lu half-life be used even though the reported uncertainty is larger than several previously reported values.

The relative intensities measured by us in the present

TABLE IV. ^{176}Lu activity measurements in units of becquerel (Bq) and propagation of uncertainties.

88-keV data		
Count no.	Activity (Bq)[a]	
1	23.86±1.17	
2	23.63±1.00	
3	25.01±1.20	
4	23.03±0.79	
Wt. average:	23.67±0.50	(23.67±1.08) Bq[b]
202-keV data		
Count no.	Activity (Bq)[a]	
1	24.00±0.28	
2	24.01±0.31	
3	23.87±0.31	
4	23.69±0.32	
Wt. average:	23.90±0.15	(23.90±0.41) Bq[b]
307-keV data		
Count no.	Activity (Bq)[a]	
1	24.02±0.29	
2	24.13±0.31	
3	24.02±0.29	
4	24.04±0.28	
Wt. average:	24.05±0.15	(24.05±0.39) Bq[b]
Activity of ^{176}Lu with all uncertainties propagated: $(23.9_6 \pm 0.4_5)$ Bq[c]		

[a]Uncertainty includes that in the net peak area and in the source-to-detector distance reproducibility.
[b]Uncertainty includes those from the weighted average of the individual counts, the uncorrelated portion of the full-energy-peak efficiency, emission probability, coincidence summing correction, and self-absorption
[c]Uncertainty includes those from the weighted average of the data from the three γ-ray energies, and that in the correlated portion of the full-energy-peak efficiency.

experiment do not appear to agree well with previous results; however, no uncertainties were given with the previously reported values. The present results agree quite well, however, with the absolute intensities calculated by the method described above as shown in Table III. Therefore, we recommend that the relative intensities measured by us should be used.

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