Half-life of ^{176}Lu

Y. Nir-El and G. Haquin
Soreq Nuclear Research Centre, Yavne 81800, Israel
(Received 15 September 2003; published 11 December 2003)

The half-life of ^{176}Lu has been measured using gamma-ray spectrometry, and was found to be $(3.677\pm0.075)\times10^{10}$ yr. The measurement geometry eliminated losses of counts due to true coincidence summing. Gamma attenuation in the sample and its container was corrected for. Agreement with previous results is discussed.

DOI: 10.1103/PhysRevC.68.067301 PACS number(s): 21.10.Tg, 27.70.+q

The long-lived naturally occurring radionuclide ^{176}Lu has been used, with its β-decay product ^{176}Hf, as a geochronometer and cosmochronometer to determine the age of rocks. Chronologies of *s*-process (slow radiative capture of neutrons) nucleosynthesis were based on ^{176}Lu. The dating results are influenced by the value taken for the disintegration half-life of ^{176}Lu and its associated uncertainty. Early values scattered broadly between $(7.3\pm2)\times10^{10}$ yr to $(2.1\pm0.2)\times10^{10}$ yr, reported by Libby [1] and Glover and Watt [2], respectively. Even later measurements, e.g., $(4.05\pm0.09)\times10^{10}$ yr obtained by Gehrke *et al.* [3] and $(3.73\pm0.05)\times10^{10}$ yr obtained by Dalmasso *et al.* [4], disagree to within −3.6 or +6.4 standard uncertainties, respectively. This status of widely discrepant values was presented in a recent evaluation of the mass number $A=176$ [5], where the unweighted average $(4.00\pm0.22)\times10^{10}$ yr was given. The uncertainty of this average was expanded to include the most precise value known at that time.

In our previous work [6], we measured the activity of lutetium oxide samples dissolved in hydrochloric acid and found the half-life of ^{176}Lu to be $(3.69\pm0.02)\times10^{10}$ yr.

In most recent works, an improvement in the situation has been partially achieved. Scherer *et al.* [7], measured Lu and Hf isotope ratios in minerals by the multiple collector inductively coupled plasma mass spectrometry (MC-ICP-MS) method, and obtained $(3.717\pm0.030)\times10^{10}$ yr, by calibration against the uranium-lead decay schemes. This value agrees very well with our previous result and with published values which cluster in the rather narrow region $(3.6–3.8)\times10^{10}$ yr. Grinyer *et al.* [8], measured the half-life of ^{176}Lu using a γ-γ technique and found $(4.08\pm0.03)\times10^{10}$ yr. Bizzarro *et al.* [9] reported Lu-Hf isotope measurements of chondrites and basaltic eucrites, using the MC-ICP-MS method, and their half-life was $(3.495\pm0.058)\times10^{10}$ yr.

In the present work, the activity of lutetium oxide in powder form was measured by gamma-ray spectrometry. Of special concern in this work were the detection efficiency calibration, gamma-ray attenuation in the powder and its container, and the quantifying of the half-life uncertainty.

A sample of 0.7834 g of Lu_2O_3 powder (Aldrich, Product No. 20366-1, Milwaukee, WI) was compacted on the bottom of a cylindrical glass container, and formed a layer of 2.54 cm diameter and 0.15 cm thickness. The chemical purity of the Lu_2O_3 was 99.99%. The atomic abundance of ^{176}Lu was analyzed by the ICP-MS method and found to be 2.62 ± 0.04%, in good agreement with the value 2.59 ± 0.02% of natural lutetium [10]. This analysis was performed because lutetium compounds, enriched or depleted in ^{176}Lu, are commercial materials for production of ^{177}Lu in radiopharmaceuticals, or in lutetium oxyorthosilicate (LSO) scintillators, respectively. Hence, the number of ^{176}Lu atoms in the sample was $N=(6.141\pm0.047)\times10^{19}$.

The sample was positioned at a distance of 20 cm from a Ge detector (Ortec, Model No. GEM-60195, Oak Ridge, TN), of 64% relative efficiency and 1.78 keV energy resolution equal to full width at half maximum (FWHM), both specified at the 1332.5 keV gamma ray of ^{60}Co. The 20 cm geometry was chosen to minimize to less than 1%, losses of counts due to true coincidence summing in gamma cascades. A computation by the GESPECOR software [11] yielded corrections of 0.7% and 0.6% for the 201.83 keV and 306.84 keV gamma rays, respectively, of ^{176}Lu. Counting live-time was $T=234447$ s.

The detection efficiency at the 20 cm geometry was calibrated by a set of reliable point source standards, covering the range 59.5–1332.5 keV. The detection efficiency ε (in counts/gamma ray) was given by the polynomial expression, derived by a weighted least-squares fitting,

$$\log(\varepsilon)=\sum_{i=-1}^{i=n} a_i\left(\frac{1}{E}\right)^i, \tag{1}$$

where E (in keV) is the gamma energy and n is the polynomial degree. In the range 276.4–1332.5 keV, a weighted least-squares fitting of a straight line was performed according to

$$\ln(\varepsilon)=a+b\,\ln(E). \tag{2}$$

The detection efficiencies at the 201.83 keV and 306.84 keV306.84keV gamma rays of ^{176}Lu, were calculated by Eqs. (1) and (2), respectively. These efficiencies include a measured correction for the deviation of the circular Lu_2O_3 sample from the calibration geometry of point standards.

Correction factors K_p and K_g for gamma ray attenuation in the Lu_2O_3 powder and in the glass bottom of the sample container were calculated by

TABLE I. Results of the measurement.

E (keV)	a (counts)	K_p	K_g	ε (counts/gamma)	P_γ (gamma/dis.)	$T_{1/2}$ (10^{10} yr)
201.83	22615.84±0.72%	$1.0496\pm7\times10^{-5}$	1.0294±0.0009	3.666×10^{-3}±3.84%	0.780±0.025	3.699±0.189
306.84	21736.32±0.79%	$1.0206\pm3\times10^{-5}$	1.0249±0.0008	2.822×10^{-3}±0.70%	0.936±0.017	3.673±0.082
					Weighted average±standard uncertainty	3.677±0.075

$$K_p = \frac{\mu_p h}{1 - \exp(-\mu_p h)} \tag{3}$$

and

$$K_g = \exp(\mu_g x). \tag{4}$$

Here μ_p and μ_g (in cm^{-1}) are the linear attenuation coefficients for gamma rays at the energy E, in Lu_2O_3 and in the glass, and h and x (in cm) are the thickness of the sample and the glass bottom, respectively. Photon mass attenuation coefficients μ/ρ (in $cm^2\,g^{-1}$) were calculated by the XCOM program [12], at the 2 gamma energies of ^{176}Lu, for Lu_2O_3 and for a soda-lime glass of the following composition: Na_2O:15%; SiO_2:72%; CaO:13%. The density ρ and bottom thickness x of the glass container were measured and found to be 2.339 ± 0.003 g cm^{-3} and 0.0988 ± 0.0008 cm.

Peak areas were analyzed by the Interactive Peak Fit algorithm of the Genie-2000 software (Canberra Industries Inc., Meriden, CT). The half-life of ^{176}Lu was calculated by

$$T_{1/2} = \frac{NT\varepsilon P_\gamma \ln 2}{3.1557\times10^7 a K_p K_g}, \tag{5}$$

where a (in counts) is the net peak area, P_γ (in gamma rays per disintegration) is the gamma emission probability [5] and 3.1557×10^7 s (=365.2422×86400) is 1 tropical year. Results are given in Table I.

The weighted average is dominated by the result of the 306.84 keV gamma ray because of its smaller relative uncertainties associated with the detection efficiency ε and the emission probability P_γ. In the calculation of the correction factors K_p and K_g, it was assumed that the uncertainties of the mass attenuation coefficients μ/ρ are negligible. The main contribution to the uncertainty $\pm0.082\times10^{10}$ yr (±2.24%) of the half-life, determined by the 306.84 keV gamma ray, arrives from the uncertainty (±1.82%) of the emission probability [5]. Therefore, an improvement of its ±0.79% peak area precision, by increasing significantly the counting time, will have a marginal effect on decreasing the ±2.05% uncertainty of the half-life. A similar argument refers to the ±0.70% detection efficiency precision, which can be improved to ±0.40% approximately. The present uncertainty $\pm0.075\times10^{10}$ yr, which is almost four times greater than our previous uncertainty [6], was quantified according to the requirements of the International Organization for Standardization Guide [13], and therefore is more realistic.

The present value $(3.677\pm0.075)\times10^{10}$ yr derived for the half-life of ^{176}Lu agrees very well with our previous result $(3.69\pm0.02)\times10^{10}$ yr [6], with published values which cluster in the range $(3.6\text{ to }3.8)\times10^{10}$ yr, and with the recent value $(3.717\pm0.030)\times10^{10}$ yr, obtained by the measurement of Lu and Hf isotope ratios in minerals by the MC-ICP-MS method [7]. These results do not agree with two much longer half-lives, i.e., $(4.05\pm0.09)\times10^{10}$ yr [3] and $(4.08\pm0.03)\times10^{10}$ yr [8]. An explanation for this discrepancy is unknown.

The authors are thankful to Professor O. Sima for the GESPECOR computation and to Eyal Elish for the ICP-MS analysis.

[1] W. F. Libby, Phys. Rev. **56**, 21 (1939).
[2] R. N. Glover and D. E. Watt, Philos. Mag. **2**, 699 (1957).
[3] R. J. Gehrke *et al.*, Phys. Rev. C **41**, 2878 (1990).
[4] J. Dalmasso *et al.*, Appl. Radiat. Isot. **43**, 69 (1992).
[5] E. Browne and H. Junde, Nucl. Data Sheets **84**, 337 (1998).
[6] Y. Nir-El and N. Lavi, Appl. Radiat. Isot. **49**, 1653 (1998).
[7] E. Scherer *et al.*, Science **293**, 683 (2001).
[8] G. F. Grinyer *et al.*, Phys. Rev. C **67**, 014302 (2003).
[9] M. Bizzarro *et al.*, Nature (London) **421**, 931 (2003).
[10] K. J. R. Rosman and P. D. P. Taylor, J. Phys. Chem. Ref. Data **27**, 1275 (1998).
[11] O. Sima, Appl. Radiat. Isot. **47**, 919 (1996).
[12] M. J. Berger *et al.*, XCOM: Photon cross section database (version 1.2), NIST, Gaithersburg, MD (1999), online available: http://physics.nist.gov/xcom
[13] ISO Guide to the Expression of Uncertainty in Measurement, ISBN 92-67-10188-9 (1993).