Half-life of ¹⁸³Hf

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(Received 10 July 2006; published 13 November 2006)

Significant amounts of the neutron-rich isotope ¹⁸³Hf have been produced by (n, γ) reaction on the long-lived ¹⁸²Hf $(t_{1/2} = 8.9 \times 10^6 \text{ y})$ during an irradiation with thermal neutrons. The half-life of ¹⁸³Hf has been remeasured with high precision using the decay curve of its most abundant γ rays. The new half-life value is 1.018 \pm 0.002 h, which is 4.6% shorter and eight times more accurate than the previous recommended value.

DOI: 10.1103/PhysRevC.74.057303

PACS number(s): 21.10.Tg, 23.40.-s, 25.40.Lw, 27.70.+q

The long-lived isotope 182 Hf $(t_{1/2} = 8.9 \times 10^6 \text{ y } [1])$ plays an important role in understanding of heavy-element nucleosynthesis in stars. Anomalies in tungsten isotopic composition in meteorites indicate that the ¹⁸²Hf abundance in the early solar system was high [2,3] (¹⁸²W is the stable decay product of ¹⁸²Hf). Lying on the neutron rich side of the valley of stability, 182 Hf can be produced either by the s or the *r* process. An accurate knowledge of contributions from these two processes is essential to relate the ¹⁸²Hf early solar system abundance to other extinct radionuclides (e.g., ¹²⁹I) and the last nucleosynthesis events [4]. In the case of the *s* process the neutron capture rates for production $[^{181}\text{Hf}(n, \gamma)^{182}\text{Hf}]$ and destruction [182 Hf(n, γ) 183 Hf] have not been measured so far. The latter is subject to a recent study [5] and leads to the short-lived ¹⁸³Hf. An accurate and precise half-life value of this short-lived reaction product is crucial for determination of the neutron capture reaction rate.

During the course of our neutron capture studies on ¹⁸²Hf at stellar and thermal neutron energies a time-dependence of the derived cross section has been observed indicating that the previous half-life value of the reaction product ¹⁸³Hf was inaccurate [5]. Thus a second irradiation with thermal neutrons was performed with a subsequent measurement series focused on a precise half-life determination of ¹⁸³Hf.

Previous half-life measurements were performed in the 1950s and 1960s and are based on the decay of β activity [6–10]. In most of the work ¹⁸³Hf was produced by (n, α) reactions on tungsten targets, except the work of Ref. [9] which used the ¹⁸²Hf (n, γ) reaction. The first half-life measurement was performed by Gatti and Flegenheimer, who reported a value of 1.067 ± 0.050 h [6]. Motavalledi-Nobar *et al.* measured 1.050 ± 0.050 h [8]. The current recommended value 1.067 ± 0.017 h is from Bakhru and Mukherjee [7]. However, their observed 91 d half-life of a proposed isomeric state was later found to be nonexistent [10]. This indicates the difficulty of measuring half-lives accurately using only

 β activities. In contrast, our measurement is based on the decay curve of the γ -ray lines which are unique for a particular isotope.

For our 182 Hf (n, γ) 183 Hf studies two samples were prepared from material used for the half-life measurement of ¹⁸²Hf [1]. The high 182 Hf content of 1.12% of this material was produced about 30 years ago by long-time neutron irradiation of natural Hf for one to two years in the high-flux Materials Testing Reactor in Idaho Falls, USA, at a neutron flux of 5×10^{14} s⁻¹cm⁻². Part of this material was later prepared at the Department of Earth Sciences, ETH-Zurich, Switzerland, and divided into four samples of different ¹⁸²Hf concentration for the half-life measurement of ¹⁸²Hf (for details see Ref. [1]). One of those (sample B3, containing $2.861 \times$ 10¹⁶ ¹⁸²Hf atoms) was further processed at the Atominstitut der Österreichischen Universitäten in Vienna, Austria, which included ¹⁸²Ta removal, the 114 d daughter nuclide of ¹⁸²Hf, by ion-exchange column and drying of the Hf solution in a clean carbon matrix. Two samples were then pressed to form a disk 8 mm in diameter and 0.65 mm in thickness.

The neutron activation was performed at the TRIGA Mark-II reactor of the Atominstitut der Österreichischen Universitäten in Vienna, Austria, using a thermal neutron flux of $4 \times 10^{11} \text{ s}^{-1} \text{ cm}^{-2}$. The high activation of ¹⁸³Hf allows for a direct determination of the half-life of ¹⁸³Hf through the decay of its γ activity. An automated measurement system recorded γ -ray spectra from a 50% HPGe detector at a distance of 8.9 cm integrated for 30 min over a period of 2 d. Typical γ -ray spectra are shown in Fig. 1. Signal processing included a loss-free counting system [11] which ensures accurate γ -ray spectra at total counting rates up to 100 kcps. A separate count rate test was performed by measuring a source in constant geometry and varying the total count rate with a strong ¹³⁷Cs source from 0.5 to 100 kcps. The activities above the Compton background showed deviations of much less than $\pm 1\%$ indicating that the data acquisition works over the entire total count rate range.

For determination of the half-life of ¹⁸³Hf the two main γ -ray lines at 459.1 keV (absolute γ -ray intensity of 27.3% [9])

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FIG. 1. Part of γ -ray spectra recorded for 1800 s 2 h (top line, black) and 8 h (bottom line, grey), respectively, after the irradiation. Peaks used for the analysis are labeled.

and 783.8 keV (65%) were evaluated. The 73.2 keV line (38%) was not used because a 0.55 mm Cd absorber was used to minimize coincidence summing effects with the 783.8 keV line. Less intense γ rays (e.g., 1470 keV) gave consistent decay rates, but could be evaluated only in the first few spectra and thus resulted in larger uncertainties. Figure 2 shows the decay curve of the 784 keV line. For each measurement the activity at the start of the measurement is calculated using

$$A_{\gamma}(t) = \frac{C_{\gamma}(t)\lambda}{I_{\gamma}\,\epsilon_{\gamma}\,k_{\gamma}(1-e^{-\lambda t_{m}})}\tag{1}$$

with C_{γ} the integrated counts of the full energy peak of the relevant γ -ray line, I_{γ} the absolute γ -ray intensity, ϵ_{γ} the γ -ray



FIG. 2. Decay curve of the 784 keV line of ¹⁸³Hf. The solid line represents the fit, whereas the dotted line indicates the expected decay from the previous half-life value.

detection efficiency and k_{γ} the absorption of the γ rays in the sample. The half-life is then calculated from the least-square fit to the measured activities for a time span of several half-lives (Fig. 2). Since the measurement time, t_m , was constant for individual measurements, the correction for the decay during the measurement is a constant factor. Thus the slope of the least-square fit depends only on the time dependence of C_{γ} . Contributions from true coincidence summing depend only on the total γ -ray efficiency and does not affect the slope of the fit. Contributions from random coincidences could affect peak areas depending on the total

TABLE I. Half-lives of measured nuclei. Measured uncertainties (1σ) represent errors from the fit only.

| Nuclide | E_{γ} [keV] | meas. $t_{1/2}$ [h] | recom. $t_{1/2}$ [h] | Reference |
|--------------------|--------------------|---------------------|----------------------|-----------|
| ¹⁸³ Hf | 459.1 | 1.017 ± 0.001 | | |
| | 783.8 | 1.018 ± 0.001 | | |
| | 1470.2 | 1.012 ± 0.005 | | |
| | mean: | 1.018 ± 0.001 | 1.067 ± 0.017 | [7] |
| ^{180m} Hf | 215.3 | 5.547 ± 0.010 | | |
| | 332.3 | 5.541 ± 0.008 | | |
| | 443.1 | 5.534 ± 0.008 | | |
| | mean: | 5.540 ± 0.003 | 5.47 ± 0.04^{a} | [12] |
| ⁵⁶ Mn | 846.6 | 2.576 ± 0.013 | | |
| | 1811 | 2.564 ± 0.055 | | |
| | mean: | 2.575 ± 0.013 | 2.5789 ± 0.0001 | [13] |
| ^{116m} In | 416.9 | 0.9032 ± 0.0026 | | |
| | 1097 | 0.9056 ± 0.0019 | | |
| | 1294 | 0.9017 ± 0.0017 | | |
| | 1507 | 0.9021 ± 0.0044 | | |
| | mean: | 0.9033 ± 0.0011 | 0.9048 ± 0.0028 | [14] |

^aValue at 297 K, at 77 K 5.66 ± 0.04 h was measured [12].

count rate. This was checked by evaluating individually the first section of the decay curve at high count rate and the last section at low count rate. No significant changes were found.

Systematic errors were checked by evaluating the half-lives from 180m Hf, 56 Mn, and 116m In. The last two are contaminations of the sample. No deviations beyond the statistical uncertainty or the error of the recommended values were found (see Table I). However, a systematic error of 0.2% was estimated and added in quadrature to the error of the half-life of 183 Hf from the fit.

The results of the γ -ray lines are shown in Fig. 3 and compared to previous measurements. Our final half-life value for ¹⁸³Hf, calculated as the weighted mean of the γ -ray line results, is $t_{1/2}(^{183}\text{Hf}) = 1.018 \pm 0.002$ h, which is 4.6% shorter and eight times more accurate than the previous recommended value. The improved half-life value allows now an accurate decay correction for the neutron capture cross section measurements [5].

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FIG. 3. Summary of half-life measurements of ¹⁸³Hf. Previous measurements are shown in chronological order and are labeled with the references (Refs. [9] and [10] mention a value without uncertainty). Measurements from this work are labeled with the γ -ray lines.

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