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Absolute intensities of γ rays in ^{182}Hf decay

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The absolute intensities of γ rays produced in the decay of ^{182}Hf were determined by measuring its γ-ray spectra with high-resolution Ge spectrometers. Because the sample was chemically purified more than 30 years ago, the daughter ^{182}Ta ($t_{1/2}$=114.43 d) was in secular equilibrium with ^{182}Hf ($t_{1/2}=8.90\times10^6$ yr). The absolute intensities of ^{182}Hf γ rays were determined with respect to the intensities of ^{182}Ta γ lines. In order to minimize summing losses from the peak areas, spectra were measured at low absolute efficiencies. The absolute intensity of the 270.4-keV-γ ray was found to be (79.0 ± 0.6)% per ^{182}Hf β^- decay.

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The nuclide ^{182}Hf was first produced [1–3] by the neutron irradiation of natural Hf and enriched ^{180}Hf targets in the Materials Testing Reactor at Idaho Falls. In each case, the Hf was chemically purified after ^{175}Hf ($t_{1/2}$=70.0 d) and ^{181}Hf ($t_{1/2}$=42.4 d) had substantially decayed and the new isotope ^{182}Hf was identified by mass spectrometry. By determining the number of ^{182}Hf atoms by mass spectrometry and measuring the γ-ray decay rate of ^{182}Hf or the daughter ^{182}Ta with a sodium iodide detector, these authors were able to determine the half-life of ^{182}Hf as 8.5×10^6 yr [1], $(8\pm5)\times10^6$ yr [2], and $(9\pm2)\times10^6$ yr [3].

In order to study the level structure of ^{182}Ta, larger quantities of ^{182}Hf were produced at Idaho Falls by irradiating both enriched ^{180}Hf and natural Hf samples in the Materials Testing Reactor [4]. After the decay of shorter-lived activities, the Hf was chemically purified and its γ-ray spectra were measured. One sample was mass separated and the spectrum of the collected ^{182}Hf sample was measured with a Ge detector. These studies showed that ^{182}Hf decays by β^- particle emission and all of the β^- decays populate a single state at 270.4 keV in ^{182}Ta which deexcites by 270.405, 172.54, and 156.09 keV γ rays. A level scheme constructed in Ref. [4] is displayed in Fig. 1 with γ-ray energies measured in the present study and the ^{182}Hf half-life from Ref. [5]. After two years, the daughter ^{182}Ta($t_{1/2}$=114.43 d) reached secular equilibrium, to 1.5 %, and its γ rays were used to determine the absolute intensities of ^{182}Hf γ rays. A value of (80 ± 5)% per β^- decay was found for the intensity of the 270.4 keVγ ray.

Currently there is interest in the half-life of ^{182}Hf because the composition of ^{182}Hf–^{182}W samples can be used as a chronometer for early solar system evolution [6,7]. One component in the determination of the ^{182}Hf half-life is the branching ratio of the 270.4-keV-γ ray. The large uncertainty in the intensity of the 270.4-keV-γ ray as measured by Helmer *et al.* [4] comes from the uncertainty in the intensity of the ^{182}Ta γ ray which was used as standard. Because the accuracy of the ^{182}Ta γ-ray intensities has improved since the previous measurement and a 30-year old source was available to us, we undertook a new measurement of ^{182}Hf γ-ray intensities.

In the present work we have used a sample that was produced by Helmer *et al.* [4] for the measurement of ^{182}Hf γ-ray intensities. We have measured the γ-ray spectra of the sample with a 25% Ge detector and a high-resolution $2\ \mathrm{cm}^2\times10$ mm low energy photon spectrometer (LEPS) with the sample at different distances from the detector. In the decay of the daughter ^{182}Ta, β^- decay populates excited states above 1 MeV, which first decay by 100–300 keV γ rays and are then followed by ~1 MeV γ rays. Thus, all of the ^{182}Ta γ rays have γ rays in coincidence, in contrast to ^{182}Hf decay where the 270.4-keV-γ ray has no γ ray in coincidence. Summing corrections should therefore be applied

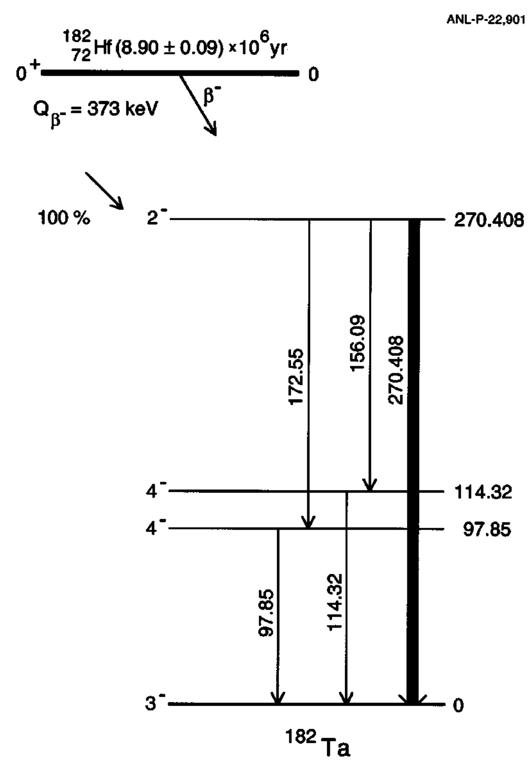

FIG. 1. Decay scheme of ^{182}Hf constructed in Ref. [4]. Energies are from the present measurement and the half-life from Ref. [5].

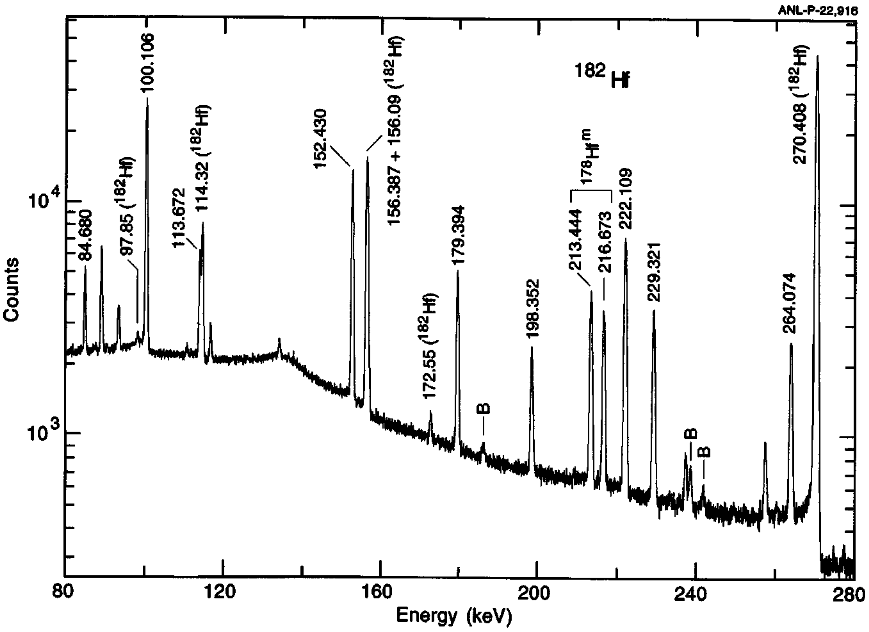

FIG. 2. The γ-ray spectrum of a 400-Bq ^{182}Hf source measured with a $2\ \mathrm{cm}^2 \times 10$ mm LEPS spectrometer. In addition to ^{182}Hf lines, the spectrum contains γ rays from the daughter ^{182}Ta and 31 yr ^{178}Hf^m. B denotes background peaks. The source-to-detector distance was 0.7 cm and the counting time was 12 d.

to the ^{182}Ta γ rays in order to use them as standard for the intensity of the 270.4-keV-γ ray. We used two approaches to measure the absolute intensities of ^{182}Hf γ rays. One is to use a counting setup with low absolute efficiencies and the other is to use a system where one can measure the summing effect. For the former, we used a $2\ \mathrm{cm}^2 \times 10$ mm LEPS detector because most of the high energy γ rays pass through the 10 mm thick Ge crystal requiring a small summing correction. For the second approach, we used a 25% Ge detector and measured the photopeak–photopeak summing with a pure ^{182}Ta source. The bigger Ge crystal has another advantage that the detector efficiency is almost flat in the 200–300 keV range, which causes less uncertainty in the intensity of the ^{182}Hf γ rays from efficiency correction.

The γ-ray spectra were measured by placing the source at 0.7 and 3 cm from the LEPS spectrometer. At the shorter distance, the spectrum was counted for 12 days, and was used to determine the energies and the relative intensities of ^{182}Hf γ rays. This spectrum is displayed in Fig. 2 and the energies measured relative to the ^{182}Ta γ-ray energies [8] are given in Table I. The energy of the 156.0-keV-γ ray could not be determined from this spectrum because it overlaps the 156.3865 keV peak from ^{182}Ta decay. In the table we give the energy of the 156.09-keV-γ ray as measured by Helmer *et al.* [4].

TABLE I. ^{182}Hf γ rays.

Energy (keV)	Intensity (%)	Transitions Initial–Final
97.85 ± 0.04	0.11 ± 0.01	97.85–0.0
114.32 ± 0.01	3.0 ± 0.1	114.32–0
156.09 ± 0.02[a]	7.0 ± 0.2	270.408–114.32
172.55 ± 0.04	0.20 ± 0.02	270.408–97.85
270.408 ± 0.010	79.0 ± 0.6	270.408–0

[a]This energy is taken from Ref. 4.

In order to obtain a precise intensity of the 270.4-keV-γ ray, the spectrum of the ^{182}Hf sample was measured by placing it 3 cm from the LEPS detector and counting it for 15 days. At this geometry the absolute photopeak efficiency was 0.63% at 100 keV and 0.093% at 270 keV. We did not observe any photopeak–photopeak sum peak in any of the spectra measured with the LEPS detector because of its small peak efficiencies. The contribution of the summing between a peak of interest and the continuous distribution from another γ ray was calculated using the total efficiency of the detector and the ^{182}Ta decay scheme. The intensity of the ^{182}Hf 270.4-keV-γ ray was determined relative to that of the 222.1 keV line of ^{182}Ta.

The intensity of the 270.4-keV-γ ray was also determined from a ^{182}Hf spectrum measured with the 25% Ge detector with the source placed at 10.5 cm. For determining the summing correction and the relative efficiencies at different energies, the spectrum of a pure ^{182}Ta source was measured. In this spectrum, summing of ^{182}Ta γ rays in the 200–300 keV range is with γ rays with energies of 1000–1500 keV. The photopeak efficiency, measured with a calibrated ^{60}Co source at 10.5 cm, was 0.13% at 1.17 MeV and the corresponding total efficiency was 1.1%. We observed several γ-γ sum peaks whose intensities agreed with the values calculated from the measured photopeak efficiencies. The loss of the counts from the 222.1-keV-photopeak was calculated with these total detector efficiencies to be 1.1% of the area. This loss was added to the 222.1-keV-photopeak area and the corrected counts were used to determine the absolute intensity of the 270.4-keV-γ ray.

The absolute intensity of the 222.1-keV-γ ray has previously been measured [9] as (7.48 ± 0.03)% per ^{182}Ta β^- decay. This is in good agreement with the value of (7.49 ± 0.03)% per ^{182}Ta decay deduced by Helmer and Tuli [10] from decay scheme balance of infeed and outfeed transitions. The uncertainties from counting statistics in the peak areas, detector relative efficiencies, and the summing correction are each less than 0.5%. Combining all uncertainties in quadrature, we determine the uncertainty in the 270.4-keV-γ ray intensity as 1.0%. The absolute intensity of the 270.4-keV-γ ray as determined from the LEPS data and the 25% Ge detector data are $(79.1\pm0.8)\%$ and $(79.0\pm0.8)\%$ per ^{182}Hf decay, respectively. The weighted mean of these two numbers gives the intensity of the 270.4-keV-γ ray as $(79.0\pm0.6)\%$ per ^{182}Hf β^- decay.

The absolute intensity of the 270.4-keV-γ ray can also be deduced by normalizing the intensities of γ rays and conversion electron transitions deexciting the 270.4 keV level to 100% as was done in Ref. [4]. The intensities of γ rays in Table I were used as relative intensities for this normalization. We have used internal-conversion coefficients [11] for pure E2 multipolarities for the 270.408, 172.52, and 156.09 keV transitions as was done in Ref. [4]. The absolute intensity thus determined is $(78.8\pm0.6)\%$, in good agreement with the value obtained by direct measurement.

In summary, the absolute intensity of the 270.4-keV-γ ray associated with the decay of ^{182}Hf has been measured relative to the intensity of the 222.1-keV-γ ray of ^{182}Ta. Using the 222.1 keV intensity as $(7.48\pm0.03)\%$ per ^{182}Ta β^- decay, we obtain the intensity of the 270.4-keV-γ ray as $(79.0\pm0.6)\%$ per ^{182}Hf decay. This small uncertainty in the ^{182}Hf γ-ray branching ratio is important to reduce the overall uncertainty in the new half-life of ^{182}Hf [5].

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