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THE HALF-LIFE OF ^{194}Hg DETERMINED BY MEANS OF
QUANTITATIVE ON-LINE MASS SEPARATION

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ABSTRACT

The half-life of ^{194}Hg , $t_{1/2} = 520 \pm 20$ y, was measured by means of the specific-activity method by using sources prepared in the ISOLDE on-line electromagnetic isotope separator. The mass of the source was calculated from the collected charge of Hg^+ ions; the disintegration rate was determined γ -spectroscopically via the 328.5 keV γ -ray. The ratio of total β^+ -intensity to the 328.5 keV γ -intensity in the decay of ^{194}Au was redetermined to be 0.022 ± 0.001 . From the intensity balance of the known decay scheme it follows that the absolute intensity of the 328.5 keV γ -ray should be revised to 67 photons per 100 decays.

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1. INTRODUCTION

Previous measurements of the half-life of ^{194}Hg have yielded extremely conflicting results. The published literature for 1955 to 1967 contains six half-life values, all obtained by measuring the counting rate of ^{194}Hg sources over long periods. On that basis, Nuclear Data sheets¹⁾ adopted the value $t_{1/2} = 1.3 \pm 0.3$ y. However, later experiments indicated that the ^{194}Hg half-life could be several hundred times larger. Thus, Orth et al.²⁾ observed no decrease of the activity of a ^{194}Hg source during one year; they quoted the limit $t_{1/2} > 15$ y. The same authors²⁾ estimated the half-life by an indirect application of the specific-activity method. After bombardment of a lead target with 600 MeV protons they measured the activity ratio of the spallation products ^{194}Hg and 183 d ^{195}Au . The ratio of the number of nuclei of the two species was estimated from empirical spallation-yield ratios. The resulting half-life was placed in the bracket $90 \text{ y} < t_{1/2} < 540 \text{ y}$. By utilizing a similar principle, Probst et al.³⁾ calculated the absolute yield of ^{194}Hg in the reaction $^{197}\text{Au} (p, 4n)$ by means of a curve-fitting procedure involving the measured excitation functions for the products ^{193}Hg , ^{194}Hg , and ^{195}Hg . From the interpolated absolute yield and the measured disintegration rate they obtained $t_{1/2} = 260 \pm 40$ y.

The crucial point in the application of the specific-activity method is the determination of the number of radioactive nuclei which most often is done by mass-spectrometric abundance measurements⁴⁾. In the present work the half-life of ^{194}Hg was determined by applying this method directly to mass-separated samples prepared by quantitative ion collection⁵⁾.

2. EXPERIMENTAL METHOD AND RESULTS

The ^{194}Hg sources were prepared in the ISOLDE facility at CERN. A 600 MeV proton beam incident on a molten Pb target produced, among other spallation products, Hg isotopes which evaporated into the ion source of an isotope separator. The separated beam of 60 keV $^{194}\text{Hg}^+$ ions was directed onto an Al collector foil. The ion-implantation process should ensure stability against volatilization of Hg from the source. Two types of sources were prepared:

- i) a strong source of 1.6 μCi ^{194}Hg collected during several days' running time;
- ii) three weak sources of accurately known mass to be used for the half-life experiment.

In the latter case the collector foil was placed in the Faraday cup in order to measure the integrated beam current of Hg^+ ions. The collected charge had to be corrected for the background beam current which was present in the isotope separator when the proton beam was switched off but with the target and ion source still hot; the correction was 0.3%. From the net collected charge, typically 2×10^{-6} Coulomb per source, the number of ^{194}Hg atoms was calculated.

In order to test the over-all efficiency of the ion collection and the reliability of the charge measurement a control experiment was performed in which $^{195}\text{Hg}^+$ ions were collected under conditions equivalent to those of $^{194}\text{Hg}^+$. An implanted source of ^{195}Hg was allowed to decay to ^{195}Au . The disintegration rate of ^{195}Au was determined by absolute measurement, with a 1 cm^3 high-resolution $\text{Ge}(\text{Li})$ detector, of the Pt K X-rays and the 99 keV γ -ray for which the intensities are accurately known⁶⁾. From the number of ^{195}Au nuclei, as measured via the collected charge and corrected for growth and decay, one calculates the half-life of ^{195}Au to be 187 ± 9 d, in good agreement with the adopted⁶⁾ value 183 ± 2 d. On this basis, we shall assume that the Hg^+ ion collection is quantitative.

The disintegration rate of ^{194}Hg was determined for the precisely "weighed" sources by means of γ -spectroscopy with a 15% coaxial $\text{Ge}(\text{Li})$ detector. The γ -spectrum contained the lines^{1,7)} from 39.5 h ^{194}Au in radioactive equilibrium with ^{194}Hg . In addition, the γ -rays from 183 d ^{195}Au were present; this contamination amounted to 0.06-0.3 atom%. In order to suppress coincidence sum effects the γ -ray spectra were always measured with the source 10 cm from the detector. The strong 328.5 keV γ -ray from ^{194}Au was used to determine the disintegration rate. This photopeak is situated on a flat Compton background in a region with no interfering peaks, so its area is easily determined (fig. 1).

The absolute photopeak efficiency of the detector was measured by means of a calibrated ^{226}Ra source. The ^{226}Ra γ -ray intensities were taken from the work of Zobel et al.⁸⁾, but normalized to 44.9% for the 609 keV γ -ray⁹⁾.

The 328.5 keV γ -ray proceeds from the 2^+ first excited state to the ground state of ^{194}Pt . Its intensity relative to the other γ -rays is well established^{1,7)}, but the absolute intensity per decay of ^{194}Au is uncertain because the exact amount of electron capture (EC) and β^+ feeding to the ground state of ^{194}Pt is not known. In order to clarify this problem, we have measured the total β^+ feeding by comparing the 511 keV annihilation peak with the 328.5 keV peak in the γ -spectrum recorded from the strong ^{194}Hg source sandwiched between 5 mm perspex, 10 cm from the large detector. This experiment yielded the intensity ratio $I(\beta^+)/I(\gamma_{328.5}) = 0.022 \pm 0.001$. By using this number and by exploiting the intensity balance of the ^{194}Pt level scheme^{1,7)} to deduce the EC + β^+ feeding to the 328.5 keV state, it is possible to calculate the EC + β^+ feeding to the ground state. The underlying assumption is that the EC/ β^+ ratios have the theoretical values for allowed beta transitions. However, this is expected to hold, since the first forbidden log ft values are fairly low. Moreover, at least the ground state β^+ -spectrum has been demonstrated to have the allowed shape¹⁰⁾. The resulting β feedings and log ft values are given in table 1.

The intensity ratio of the two β^+ groups in table 1 agrees well with that reported by Thieme and Bleuler¹¹⁾. On the other hand, our total β^+ -to- $\gamma_{328.5}$ intensity ratio is 25% lower than Thieme and Bleuler's value, which was based on an extrapolation of the β^+ -spectrum to zero energy. We feel that our value should be preferred as being the most accurate one, because it was obtained from a simple comparison of two photopeaks, whereas Thieme and Bleuler's extrapolation could be distorted by systematic errors which affect both β^+ groups in the same proportion. This would explain the fact that their ratio of β^+ components agrees with ours.

Finally, from the data of table 1 and the γ -ray data summarized in refs. 1 and 7 it follows that the intensity of the 328.5 keV γ -ray is 67%. This differs from earlier adopted numbers: 61% (ref. 1) and 63% (ref. 7). By using the new

328.5 keV intensity the disintegration rates of the ^{194}Hg sources were determined. From the known number of nuclei in each source the half-life was calculated to be

$$t_{\frac{1}{2}}(^{194}\text{Hg}) = 520 \pm 20 \text{ y} ,$$

as an average over six different experiments. The error is the standard deviation. To this random error one should add a ± 25 y uncertainty coming from possible systematic errors associated with the decay schemes and the data for the calibration source.

3. DISCUSSION

The half-life 520 y, as measured by means of the specific-activity method, is longer than previously measured values, although it agrees with the rather wide limits set by Orth et al.²⁾. The very short half-life values¹⁾ observed in the "decay" experiments with ^{194}Hg are most probably caused by loss of Hg from insufficiently sealed sources, since most mercury compounds are volatile. Our value disagrees with the most recent estimate due to Probst et al.³⁾ by a factor of two. The reason for this is not clear, but one should note that the indirect method used by Probst et al. could be sensitive to certain assumptions. For example, the criterion for obtaining a suitable fit of the unknown absolute yield of ^{194}Hg could be inadequate.

The nucleus ^{194}Hg is known^{1,7)} to decay by electron capture from the L-shell and higher shells. This fact puts limits on the disintegration energy: $14 \text{ keV} < Q_{\text{EC}} < 83 \text{ keV}$. The new half-life value of 520 y leads to an estimate of the Q_{EC} value, if one assumes that the $\log ft$ value for the decay $^{194}\text{Hg}(0^+) \rightarrow ^{194}\text{Au}(1^-)$ is identical to that of the decay $^{194}\text{Au}(1^-) \rightarrow ^{194}\text{Pt}(0^+)$, see table 1, apart from a statistical factor of three from the inverted spin sequence. Thus, with $\log ft = 7.6$ one obtains $Q_{\text{EC}} = 35 \text{ keV}$.

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Table 1

Beta transitions $^{194}\text{Au} \rightarrow ^{194}\text{Pt}$

$I_i \rightarrow I_f$	E_{β^+} (keV)	EC/ β^+ theory	EC + β^+ (%)	β^+ (%)	log <u>ft</u>
$1^- \rightarrow 2^+$	1158	47	32.5	0.68	7.71
$1^- \rightarrow 0^+$	1487	20.4	17.5	0.82	8.12

Figure caption

Fig. 1 : Section of γ -spectrum of mass-separated ^{194}Hg recorded in an 824 min run with the source 10 cm from a 15% Ge(Li) detector. Background lines (B) are from natural U and Th. An ^{228}Ac background line at 328.3 keV constitutes 0.7% of the 328.5 keV ^{194}Au line used here for absolute activity measurement.

Fig. 1