A New Determination of the Half-Life of ^{194}Hg

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Using experimental data on excitation functions of ^{197}Au(p, x n)$^{198-x}$Hg reactions the half-life of ^{194}Hg has been determined to be 367 ± 55 y.

There are some uncertainties in the literature concerning the half-life of the isotope ^{194}Hg which decays by electron capture to the ground state of ^{194}Au. The Nuclear Data Sheets compilation of 1972 [1] presents a list of experimental values in the remarkably wide range from 0.4 to 1.9 y. Remarkable are also the implications of the values as such: The observation of (L + M + ···) capture only [2] sets the upper limit $Q_\beta < 81$ keV for the decay energy; half-lives and Q_β-limit together imply surprisingly small log ft values for the $0^+ \rightarrow 1^-$ ground state transition. The ad hoc introduction of a low-lying, $J^\pi = (0, 1)^+$ level in ^{194}Au would raise new problems [2]. More recently, Orth et al. [3] produced ^{194}Hg by spallation of Pb with 600 MeV protons. They obtained two estimates for $T_{1/2}(^{194}\mathrm{Hg})$ by two different methods: (i) $T_{1/2} \geqq 15$ y from observation of the time-dependence of the activity, with special precautions against long-time Hg loss, and (ii) $90\ \mathrm{y} \leqq T_{1/2} \leqq 540$ y from comparison of the observed γ-ray activities of ^{194}Au and ^{195}Au with spallation-yield trends. The authors [3] mentioned the possibility that earlier $T_{1/2}$-measurements went awry by volatilization losses of Hg.

The present value of $T_{1/2}(^{194}\mathrm{Hg})$ is an additional result of the investigation [4, 5] of ^{197}Au(particle, ypxn) excitation functions. The usual stacked-foil technique, which was used in these experiments, requires knowledge of spectroscopic data on the residual nuclei, such as half-lives and absolute γ-ray abundances. If for a residual nucleus the γ-ray abundances are only relatively known, it may nevertheless be possible to get an absolute yield curve for the corresponding reaction by taking into account the additional argument [5, 6] of the smoothness of the sum of a class of excitation functions. A similar approach is used in this work where the smooth-sum argument is applied to the ^{197}Au(p, xn) yield curves. This provides us with absolute ^{194}Hg production cross sections which in turn yield $T_{1/2}(^{194}\mathrm{Hg})$.

Excitation functions for ^{197}Au(p, xn)$^{198-x}$Hg and ^{197}Au(p, pxn)$^{197-x}$Au reactions were obtained over the $E_p = 7$–45 MeV range by irradiation of two stacks, one at $E_p = 25$ MeV and the other at $E_p = 45$ MeV, with the proton beam of the Jülich isochronous cyclotron JULIC. Each stack contained Au foils and suitable Al degraders. Gamma-ray spectra of the irradiated Au foils were measured with a Ge(Li) spectrometer shortly after irradiation, for the determination of the yield curves corresponding to short-lived ($T_{1/2} < 7$ d) residual nuclei[1]. Similar γ-ray measurements were performed about six months later when almost all induced activities had decayed. The ^{194}Au nuclei originally produced directly by the ^{197}Au(p, p3n) reaction had decayed by then ($T_{1/2}(^{194}\mathrm{Au}) = 40$ h), so that the ^{194}Hg activity (no γ-rays observed [1, 2]) could now be determined from the γ-ray spectrum of the ^{194}Au decay. The $E_\gamma = 294$ and 329 keV γ-lines with absolute abundances [7] of 11 and 63 photons/100 decays, respectively, were used for this determination. The weaker line which is moreover somewhat disturbed by a 295 keV room-background line from the U-Ra chain was used for consistency checks only. The measurement of the stronger line required the extinction especially of the $E_\gamma = 328$ keV line from ^{196}Au ($T_{1/2} = 6.2$ d) and in general of the background due to other induced activities. Possible contamination was checked carefully; the only observed room-background line (a weak $E_\gamma = 328$ keV line from the ^{232}Th chain) was taken into account. Contributions from the ^{197}Au(n, α)^{194m}Ir(β^-; $T_{1/2}$ = 171 d)^{194}Pt process were ruled out on the basis of line intensities in the present γ-ray spectra; the main argument comes from non-observation of ^{194}Pt γ-rays just below the (p, 4n) threshold. The loss of Hg which is homogeneously distributed in the 20 μm thick Au foils is assumed to be negligible at room tem-

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[1] The cross-section data and their analysis will be published in a separate paper.

perature; in a test experiment substantial escape of Hg could be observed only near or at the melting point of Au. Furthermore, measurements of (particle, $xnyp$) reaction cross sections involving residual Hg nuclei were found to be in agreement with experimental results of other authors and with theory [4, 5].

The derivation of $T_{1/2}(^{194}\mathrm{Hg})$ assumes the absence of (i) long lived and/or e-capture active isomers in ^{194}Hg and ^{194}Au, and (ii) α-decay of ^{194}Hg. These assumptions are very safe in view of the present-day spectroscopic data [7] and the small value $Q_\alpha = 2.69$ MeV [8] for ^{194}Hg. In addition we assume, in accordance with Ref. [3], that irradiation, cooling and measuring times (all < 6 months) are much shorter than the half-life of ^{194}Hg. In this case the relation between the production cross section and the half-life is simply $\sigma = A T_{1/2}$ where A contains measurable quantities as number of target atoms, particle flux and γ-ray activity. Taking into account the above considerations, a relative ^{197}Au(p, 4n)^{194}Hg excitation function (essentially $\sigma(E)/T_{1/2}$) was derived from the six months delayed γ-ray measurements.

The excitation functions for two (p, xn) reactions could not be determined experimentally in the region of interest ($25\ \mathrm{MeV} \leqq E_p \leqq 45\ \mathrm{MeV}$): that for ^{197}Au(p, n)^{197g}Hg because of small cross sections and unfavourable spectroscopic conditions, and that for ^{197}Au(p, 2n)^{196}Hg because it leads to a stable residual nucleus. From an extrapolation of the (p, n)^g data at lower energies the total (p, n) excitation function was approximated taking twice the measured ^{197}Au(p, n)^{197m}Hg cross sections; for the (p, 2n) reaction the result of a hybrid-model [9] calculation (cf. footnote [1]) was used which turned out to be in reasonable agreement with experimental data for ^{197}Au(p, yp xn) excitation functions in general as well as for that of ^{197}Au(p, 2n) below $E_p = 15$ MeV [10]. Both approximations, however, do not introduce additional errors because the contribution of these reactions to the total sum of (p, xn) cross sections is small (about 1% and 5% for (p, n) and (p, 2n), respectively) and the excitation functions are flat in the region of interest.

The half-life of ^{194}Hg was obtained employing the following fitting procedure: In the energy range from 25 to 45 MeV ten intervals were chosen which differ in position and width (typically 8—12 MeV wide). For each interval the sum of all (p, xn) cross sections was assumed to be linear in a semilogarithmic plot and a least-squares fit was performed with three free parameters: the two constants of the straight line and the scaling factor of the (p, 4n) excitation function which is closely related to the ^{194}Hg half-life value. The values for the single intervals did not exhibit any systematic behaviour with respect to the choice of the interval; their mean turned out to give $T_{1/2}(^{194}\mathrm{Hg}) = 367$ y. Nine of the ten single values differ from the mean value by less than 6%. The overall error in $T_{1/2}(^{194}\mathrm{Hg})$ has been estimated to be 15% (one standard deviation). It contains contributions — ordered according to their importance — from the ^{194}Au decay data, from the fitting procedure, from the decay data of the neighbouring Hg isotopes and from the statistics of the ^{194}Au decay measurements. Other quantities like proton beam intensity, foil thicknesses and spectrometer efficiency do not affect the results in first order; the statistical errors of the ^{193,195}Hg spectra can be neglected. Figure 1 shows the sum of the (p, xn) excitation functions and illustrates how variations of the $T_{1/2}(^{194}\mathrm{Hg})$ value influence its shape. The new value for the half-life of ^{194}Hg, $T_{1/2} = 367 \pm 55$ y, is in agreement with the much less precise data of Orth et al. [3] given above. Our

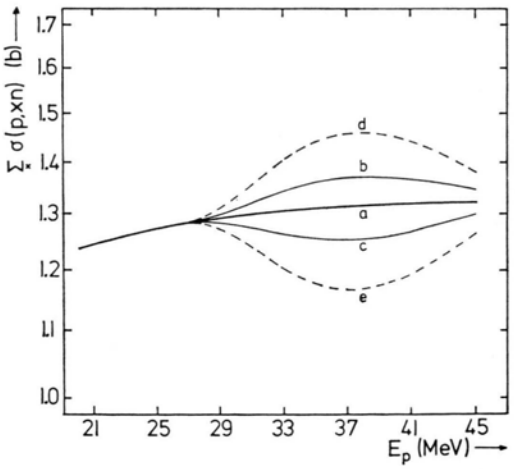

Fig. 1. Sum of the measured ^{197}Au(p, xn) excitation functions, including that of the ^{197}Au(p, 4n)^{194}Hg reaction for different values of $T_{1/2}$ (^{194}Hg); semilogarithmic plot! Curve a) corresponds to $T_{1/2} = 367$ y; curves b), c), d) and e) were obtained by changing the half-life value by $\pm$ 6% and $\pm$ 15%. The 6% curves indicate the uncertainly in the fitting procedure. The 15% curves are shown only to demonstrate a "drastic deviation from smoothness"; they should not be confounded with the 15% total error which contains — besides the fit uncertainty mentioned above — contributions of quite different origins.

value $T_{1/2}(^{194}\mathrm{Hg}) = 260 \pm 40$ y cited in Ref. [7] represents a preliminary result; the present value is based on more experimental data, on a more careful fitting procedure and on a more extensive literature survey concerning the decay data of the nuclei involved. The upper limit for the reduced half-life given in Ref. [2], $\log ft \leqq 5.8$, is now shifted to $\log ft \leqq 8.3$ with the new half-life value. This agrees much better with the expectation for a non-unique first-forbidden transition.

Some comment should be made concerning the ^{194}Au decay γ-ray abundances used in this work. They were derived from intensity-balance considerations within the decay scheme based on γ-ray and e^--spectroscopy [11—13] combined with experimental positron intensities [14] and theoretical electron-capture to positron-decay ratios [15]. Two remarks are made about the latter. First, the ground-state to ground-state β-transition is presumably a $(1^- \rightarrow 0^+)$ non-unique first-forbidden one. The β-decay tables [15] do not cover this type of transition, but the compilers apparently made use of the quasi-allowed (or ξ) approximation which is justified in the case of heavy nuclei and not too high decay energies [16]. Secondly, Firestone et al. [17] reported anomalous electron-capture to positron-decay ratios in the decay of ^{145}Gd which would imply serious consequences for β-decay theory. Recently, however, these discrepancies were shown [18, 19] to be essentially non-existent. So, for the moment, there are no special reasons to distrust the application [7] of the β-decay tables [15] in the present case. In view of a comparison of experiment and theory in Ref. [16] a 20% uncertainty has been attributed to the theory for the error estimate of this work.

As the smooth-sum argument on which the present result is based, was introduced and used before [5, 6] as an empirical means, the following physical picture is sketched to justify the present application: First, the smoothness of the total reaction cross section σ_R is directly related to the optical model and its weakly E_p-dependent parameters. Then, because of the large Coulomb barrier, charged particle emission is practically restricted to the initial stages of the reaction. This implies that the share of the summed (p, xn) cross sections in σ_R is determined in a pre-equilibrium process, which can be well described in terms of smoothly varying parameters by e.g. the hybrid model [9]. For the present case the above is supported by model calculations (cf. footnote [1]). These show that σ_R rises slowly with E_p over the $E_p = 25$—45 MeV range, while the fraction going into the (p, xn) channels decreases smoothly from 89 to 78%; the rest feeds predominantly the (p, pxn) channels. As a consequence, the calculated sum of (p, xn) cross sections is nearly constant over this E_p-range.

Finally, an alternative method for the determination of the ^{194}Hg source strength should be mentioned which could replace the argument with the smooth sum of the (p, xn) cross sections. A ^{194}Hg source with known strength can be obtained in the following way: (i) production of ^{194}Tl, for instance via the reaction ^{197}Au(α, 7n) at $E_\alpha =$ 90 MeV; (ii) removal of Hg, especially of ^{194}Hg from the reaction ^{197}Au(α, p 6n)^{194}Hg, by a suitable chemical separation; (iii) measurement of the decay $^{194}\mathrm{Tl} \rightarrow {}^{194}\mathrm{Hg}$ ($T_{1/2} = 0.5$ h). The decay $^{194}\mathrm{Hg} \rightarrow {}^{194}\mathrm{Au} \rightarrow {}^{194}\mathrm{Pt}$ is measured later as described above. This experiment would only be an independent check for the ^{194}Hg source-strength determination. Since additional uncertainties would enter the evaluation, mainly those concerning the spectroscopic data of the ^{194}Tl decay [7], one cannot expect a significant improvement of the final $T_{1/2}(^{194}\mathrm{Hg})$ value.

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