Branching Ratio and γ-ray Emission Probabilities in the Decay of the $J^{\pi}=13/2^{+}$ Isomer in ^{197}Hg

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Abstract

Gamma rays following the decay of the ^{197g}Hg and ^{197m}Hg radionuclides were measured using chemically-purified sources and a planar HPGe detector. In the case of ^{197m}Hg, precise values of 94.68 (9)% and 5.32 (9)% were determined for the IT and EC branches, respectively, the latter found to differ significantly from previously published data. The half-lives of ^{197g}Hg and ^{197m}Hg were also measured as 64.81 (24) h and 23.82 (4) h.

Keywords: Radioactivity, ^{197g}Hg, ^{197m}Hg, γ-ray emission probabilities, branching ratio, half-life
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1. Introduction

The structure of low-lying levels in ^{197}Hg is well known from various spectroscopy studies (see Ref. [1] and references therein). The ground state, ^{197g}Hg ($J^{\pi}=1/2^{-}$), decays via an electron capture (EC) to low-spin $J^{\pi}=1/2^{+}$ and $3/2^{+}$ levels in the daughter ^{197}Au nucleus. A long-lived isomeric state, ^{197m}Hg ($J^{\pi}=13/2^{+}$, E=298 keV), is known to exhibit both internal transition (IT) and EC decays, the latter directly populating the $J^{\pi}=11/2^{-}$, E=409-keV isomer ($T_{1/2}$=7.73 (6) s [1]) in ^{197}Au, as shown in Figure 1.

There is a continuing interest in the decay properties of ^{197g}Hg and ^{197m}Hg since both radionuclides have potential for theranostic medical applications [3]. However, significant differences were observed in the activity of ^{197m}Hg depending whether γ rays associated with the IT decay branch (134-keV γ ray) or the EC one (279-keVγ ray) were used for its quantification in recent excitation function measurements of proton- and deuteron-induced reactions [4, 5], thus implying inconsistencies in the presently adopted nuclear data for this radionuclide.

The determination of the IT and EC branching ratio of ^{197m}Hg is not straight forward and it requires a detailed knowledge of the decay scheme and a range of complementary nuclear structure information, such as γ-ray emission probabilities, multipolarities, M1+E2 mixing ratios and total electron conversion coefficients. Earlier, branching ratios were recommended in both ENSDF [1, 6, 7] and the Table of Isotopes [8, 9], but the values varied over the years, since they were based on the nuclear data known at the time of publication.

In this paper, we report on new measurements of the emission probabilities for γ rays that follow the decay of ^{197m}Hg. Precise values for the IT and EC branching intensities were determined and the latter was found to differ significantly from the previously published data. The half-lives of ^{197g}Hg and ^{197m}Hg were also measured with a high precision.

2. Experimental methods

2.1. Radionuclide production and source preparation

The radionuclides were produced by deuteron activation of a water-cooled, high-purity (99.99 %, Safina,

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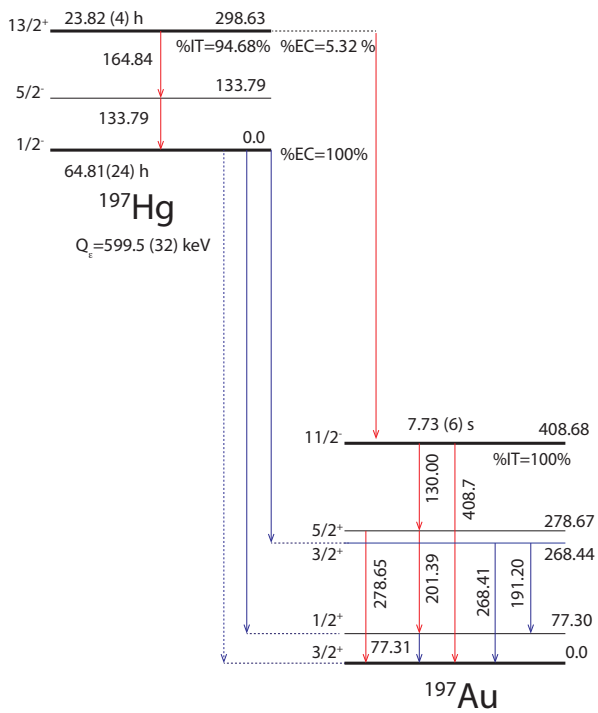

Figure 1: Decay schemes of ^{197g}Hg and ^{197m}Hg. The level and γ-ray energies, as well as the half-lives and %IT and %EC branching ratios are from the present work. The Q_ε value is from Ref. [2], while the J^π values, the half-life and % IT of ^{197m}Au are from Ref. [1].

Czech Republic) ^{197}Au target that had a diameter of 10 mm and a thickness of 60 μm. The target was irradiated for 1.5 h with a 19 MeV, 6.5 μA deuteron beam provided by the U-120M cyclotron of the Nuclear Physics Institute of the Czech Academy of Science.

A chemical separation of mercury radionuclides from gold ones, including ^{198}Au, that was produced in the ^{197}Au(d,p) reaction, was performed after the irradiation was completed. The target was dissolved in aqua regia prepared from 525 μl of 30% HCl and 175 μl of 65% HNO_3 (purity TraceSelect, Sigma-Aldrich). A 300 μl of 6M HCl was added to the final solution, which was then loaded into a column filled with LN resin (TRISKEM, France). The column was prepared by soaking 3.6 g of LN resin for 15 min in 6M HCl. It was then transferred to a 10 ml Eppendorf pipette tip plugged with glass wool and rinsed slowly with 30 ml of 6M HCl. Altogether 15 fractions of 1 ml volume each were collected and their relative activities were measured using an ionizing chamber (BqMetr4, Empos, Czech Republic). It was found that the ^{197g}Hg and ^{197m}Hg activities were eluted almost entirely (> 95 %) in the fractions 8 and 9. All fractions were also sampled and measured with a γ-ray spectrometer equipped with a HPGe detector (GMX45Plus, Ortec, U.S.A.) at the distance of 600 mm. No traceable activity of ^{198}Au was observed in any of the fractions, indicating a good separation of Hg from the Au radionuclides. Two sources with a diameter of 5 mm each were prepared by evaporating 25 μl from the strongest fraction on a thin filtration paper, which were sealed between two 150 μm thick polyethylene foils. These sources were used in γ-ray spectroscopy measurements aimed at determining the half-lives of ^{197g,m}Hg and the relative γ-ray emission probabilities following their decays.

2.2. Measurements

One of the two sources was measured with a γ-ray spectrometer comprising of a planar HPGe detector (Canberra, GL0515R) at a distance of 200 mm from the detector, which had an energy resolution (full width at half maximum (FWHM)) of 0.5 keV at E_γ=81 keV. The results from these measurements were used to determine the emission probabilities of γ-rays following the decays of ^{197g}Hg and ^{197m}Hg. The other source was used in a set of singles measurements using a planar HPGe detector (Canberra, GL0515R) located at a fixed distance of 120 mm from the source. The activities were followed for approximately 232 h and the results were used to determine the half-lives of ^{197g}Hg and ^{197m}Hg. The HPGe spectrometers were carefully calibrated for energy and efficiency with a set of ^{241}Am (489.3 kBq, combined standard uncertainty 0.4 %), ^{133}Ba (75.77 kBq, combined standard uncertainty 0.5 %) and ^{152}Eu (477.2 kBq, combined standard uncertainty 0.6 %) standards that were provided by the Czech Metrology Institute, Prague. The nuclear data for the standard radionuclides were taken from Ref. [10].

The analysis of the γ-ray spectra was carried out with the $gf3$ program of the RADWARE spectroscopy package [11]. The basic parameters, such as peak shape and FWHM as a function of the γ-ray energy, were determined from the calibration sources spectra, and were fixed during the fitting procedure. A parallel analysis using the

Table 1: Gamma-ray energies and emission probabilities determined in the present work, together with the transition multipolarities, mixing ratios and total conversion electron coefficients used to determine the EC and IT branching ratios of ^{197m}Hg.

$E_\gamma(\Delta E_\gamma)$ [a)] (keV)	$I_\gamma(\Delta I_\gamma)$ [a)] (rel. units)	$I_\gamma(\Delta I_\gamma)$ [b)] (%)	Mult. [1]	$\delta(\Delta\delta)$ [c)]	$\alpha_T(\Delta\alpha_T)$ [13]
^{197m}Hg; %IT=94.68 (9)%					
133.79 (5)	1000 (6)	34.8 (3)	E2		1.705 (24)
164.84 (5)	8.09 (5)	0.2816 (26)	M4		337 (5)
^{197m}Hg; %EC=5.32 (9)%					
77.31 (5)	0.410 (16) [d)]	0.0143 (6)	M1+E2	-0.352 (11)	4.14 (10)
130.00 (5)	4.92 (4)	0.1713 (21)	E3		29.9 (4)
201.39 (5)	1.55 (5)	0.0540 (18)	E2		0.366 (5)
278.65 (5)	108.8 (6)	3.79 (4)	M1+E2	-0.40 (4)	0.368 (9)
408.7 (1)	0.164 (16)	0.0057 (6)	M4		3.87 (5)
^{197g}Hg; %EC=100 %					
77.31 (5)	1000 (10)				
191.20 (5)	28.4 (3)				
268.41 (5)	1.97 (5)				

[a)] Determined in the present work, unless otherwise stated.
[b)] Determined in the present work using the new %EC and %IT branches.
[c)] See Table 2 for details.
[d)] Determined in the present work from I_γ^{tot}(77.31-keV γ ray)=I_γ^{tot}(201.39-keV γ ray).

GENIE2000 software [12] was also carried out and the results were found to be consistent with those obtained using the $gf3$ program.

3. Results and Discussion

The γ-ray spectrum that was used to determine the relative γ-ray emission probabilities in the decays of ^{197g}Hg and ^{197m}Hg is shown in Figure 2. Measured γ-ray energies and emission probabilities are presented in Table 1, together with values for the transition multipolarities, M1+E2 mixing ratios (δ) and total electron conversion coefficients (α_T). Given the assigned γ-ray transition multipolarities, the α_T values were determined using the *BrICC* code [13] and for γ rays of a mixed M1+E2 multipolarity they were calculated as:

$$\alpha_T(M1+E2) = \frac{\alpha_T(M1) + \delta^2\alpha_T(E2)}{1+\delta^2} \quad (1)$$

where α_T(M1) and α_T(E2) are the total conversion coefficient for pure M1 and E2 multipolarities, respectively. The δ values for the 77.31-keV and 278.65-keV γ rays were obtained from a least-squares fit to the available experimental data, presented in Table 2, using the *briccmixing* program [14].

The quoted uncertainties in the γ-ray emission probabilities represent one standard deviation and these were obtained from the quadratic sum of individual uncertainties that arise from the statistics of the full-energy peaks, relative detector efficiency (~0.5 %), dead-time corrections (~0.2%), and peak/background modeling (~0.2%). Using the established decay scheme of ^{197m}Hg, shown in Figure 1, and the data presented in Table 1, the %EC and %IT branching intensities of ^{197m}Hg can be determined from:

$$\%EC/\%IT = \frac{I_\gamma^{tot}(130.00\gamma) + I_\gamma^{tot}(408.7\gamma)}{\frac{1}{2}(I_\gamma^{tot}(133.79\gamma) + I_\gamma^{tot}(164.84\gamma))} \quad (2)$$

$$\%EC + \%IT = 100 \quad (3)$$

where I_γ^{tot}=$I_\gamma\times(1+\alpha_T)$ is the total γ-ray emission probability and I_γ is the relative γ-ray emission probability (column 2 of Table 1). Comparison between the presently determined EC and IT branching ratio for ^{197m}Hg with results from previous evaluations is presented in Table 3. It

Table 2: Experimental data, including K, L and total electron conversion coefficient, sub-shell electron ratios and mixing ratios from $\gamma\gamma(\theta)$ measurements, used to determine the M1+E2 mixing ratios for the 278.65-keV and 77.31-keV γ rays

278.65-keV γ ray	Reference	77.31-keV γ ray	Reference
α_K=0.29 (3)	[15]	α_T=4.37 (10)	[25]
δ=-0.41 (4)	[16]	α_L=2.46 (26)	[29]
δ=-0.33 (9)	[17]	M1/M2/M3=100/44 (4)/0.41 (4)	[27]
δ=-0.40 (4)	[21]	L1/L2/L3=100/44 (3)/35 (2)	[27]
δ=-0.39 (2)	[22]	L1/L2/L3=100/51 (1)/37 (1)	[26]
α_K=0.32 (4)	[18]	L1/L2/L3=100/44 (4)/32 (2)	[23]
K/L=4.8 (5)	[20]	L1/L2/L3=100/42 (7)/35 (8)	[24]
L1/L2/L3=100/17.9 (27)/4.8 (7)	[19]	δ=-0.352 (5)	[28]

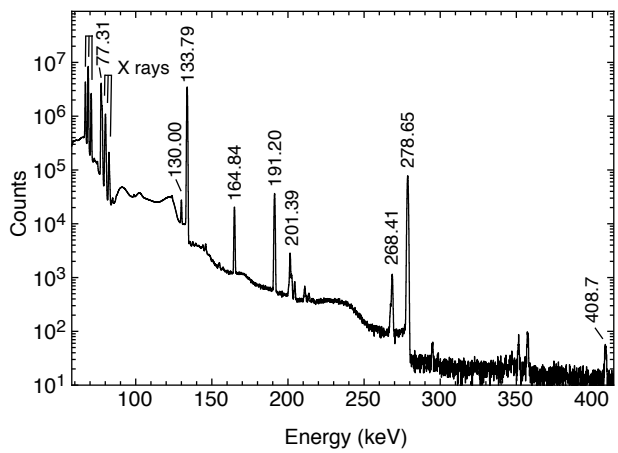

Figure 2: Gamma-ray spectrum of ^{197g}Hg and ^{197m}Hg measured with the planar HPGe detector (Canberra, GL0515R) at a distance of 200 mm.

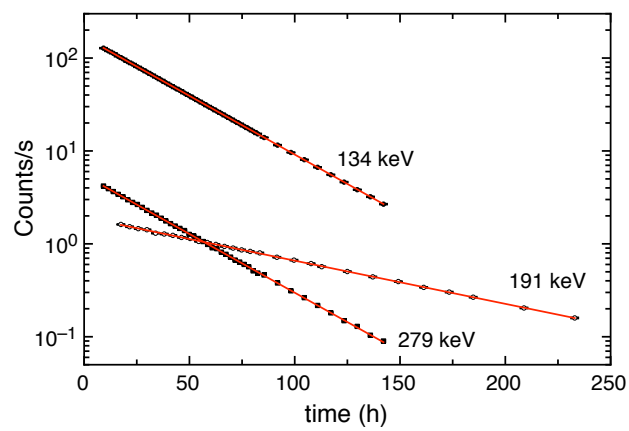

Figure 3: Activities for the 133.79-keV and 278.65-keV γ rays (^{197m}Hg), and 191.20-keV γ ray (^{197g}Hg) as a function of time after irradiation.

should be pointed out that the deduced in the present work EC=5.32 (9)% differs significantly from that of 8.6 (7)%, recommended in the most-recent ENSDF evaluation [1] that is based on the value reported in Ref. [31]. Similarly, the absolute γ-ray emission probability for the 278.65-keV γ rays is approximately a factor of two smaller compared to that recommended in Ref. [1]. Somewhat different branching intensities were reported in the earlier ENSDF evaluation [7], adopted also in the latest edition of the Table of Isotopes [9], which are based only on the γ-ray emission probabilities measured in Ref. [19] and the known at that time values for δ and α_T. The values published in the earlier version of Table of Isotopes [8] were based on the γ-ray emission probabilities reported in Ref. [30].

The general purpose databases, such as ENDF [33] and JEFF [34], use the most-recent data recommended in ENSDF [1], although there is a small discrepancy in the absolute γ-ray emission probability for the 133.79-keV γ ray that is quoted in Ref. [33] as 33.9 (3)%. Controversially, while the data for the EC decay of ^{197m}Hg are included in the ENSDF database [1], they are missing in NUDAT [32], which is frequently used by the applications community and considered as a "friendly face"of ENSDF.

Table 3: Comparison between the measured in the present work EC and IT branching ratios and the absolute γ-ray emission probabilities for the 133.79-keV and 278.65-keV γ rays with previously published values.

EC (%)	IT (%)	I_γ(133.79-keV γ ray) (%)	I_γ(278.65-keV γ ray) (%)	Reference
5.32 (9)	94.68 (9)	34.8 (3)	3.79 (4)	present work
8.6 (7)	91.4 (7)	33.5 (3)	6.1 (5)	NDS2005 [1, 31]
8.6 (7)	91.4 (7)	33.5 (3)	6.1 (5)	NDS1995 [6, 31]
7.0 (7)	93.0 (7)	34 (4)	5.0 (7)	NDS1991 [7, 19]
7.0 (7)	93.0 (7)	34 (4)	5.0 (7)	TOI1999 [9, 19]
6.5 (10)	93.5 (10)	34.3 (5)	5.4 (4)	TOI1978 [8, 30]
8.6 (7)	91.4 (7)	33.5		NUDAT [32]
8.6 (7)	91.4 (7)	33.9 (3)	6.1 (5)	ENDF/BVIII.0 [33]
8.6 (7)	91.4 (7)	33.5 (3)	6.1 (5)	JEFF3.3 [34]

In addition, the value for the absolute emission probability of the 133.79-keV γ ray is given without an uncertainty in NUDAT [32], which is misleading.

The half-lives of ^{197g}Hg and ^{197m}Hg were determined by measuring the net-peak areas as a function of time for the strongest γ rays that follow their decay. The corresponding time spectra are shown in Figure 3. The statistical uncertainties of the net-peak areas ranged between 0.11 and 2.5 %, while the systematic uncertainties were estimated to be ~1% and included contributions from geometrical efficiency corrections (~0.6%), the peak/background modeling (~0.2%) and dead-time corrections (~0.2%). The half-life of ^{197m}Hg was determined as 23.82 (4) h and 23.81 (6) h by fitting the net-peak areas of the 133.79-keV and 278.65-keV γ rays, respectively, as a function of time with a single exponential decay curve. The presently determined value is consistent with that of 23.8 (1) h recommended in the most-recent ENSDF evaluation [1], but it is more precise. The half-life of ^{197g}Hg was determined from a two exponential fit to the net-peak area of the 191.20-keV γ ray as a function of time and by fixing the half-life of ^{197m}Hg to 23.82 (4) h. The obtained value of 64.81 (24) h in the present work is somewhat larger compared to that of 64.14 (5) h that is recommended in the most-recent ENSDF evaluation [1], which is based on the yet unpublished data reported in Ref. [35].

In summary, the decays of ^{197g}Hg and ^{197m}Hg were studied by means of γ-ray spectroscopy counting of chemically-purified radioactive sources with a planar HPGe detector. Accurate emission probabilities were measured for γ rays that followed the decays of these radionuclides. The IT and EC branching intensities of ^{197m}Hg were determined and the latter were found to differ significantly from previously published values. The half-lives of ^{197g}Hg and ^{197m}Hg were also measured with a high precision.

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