Measurement of the absolute gamma-ray emission intensities from the decay of ^{103}Pd

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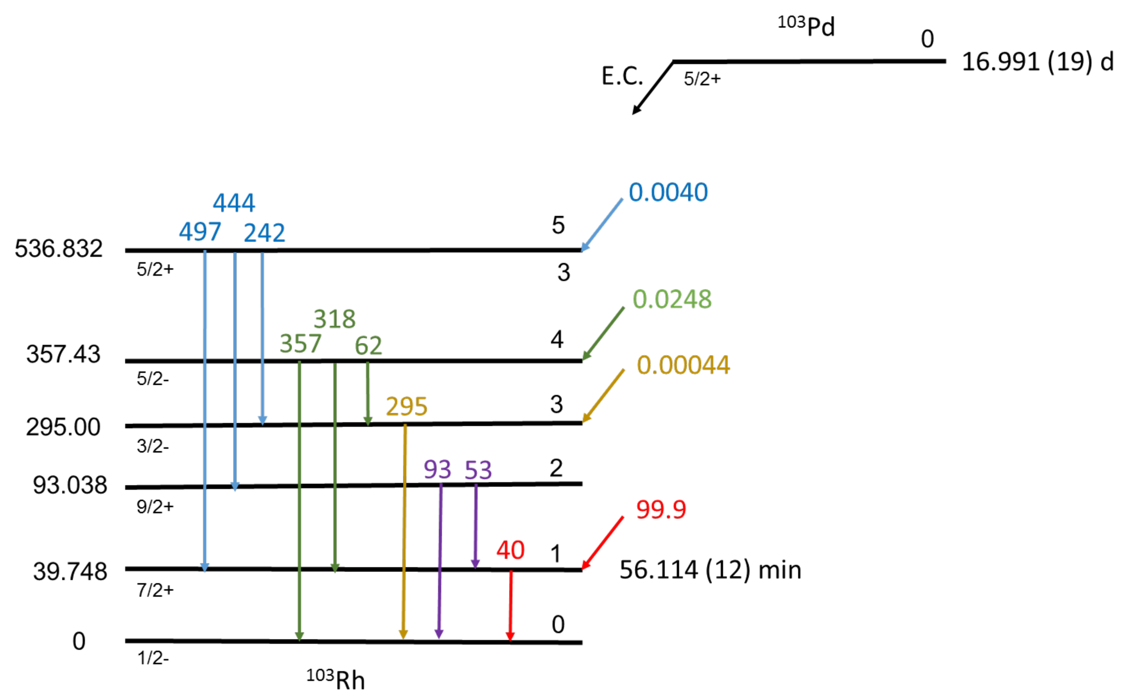
Introduction

Palladium-103 is a radioisotope of interest in medical applications which is used in brachytherapy implants for the treatment of prostate cancer.
It decays through electron capture to excited levels of ^{103}Rh, and especially (99.959%) to the 39.748 keV metastable state.
The radioactive equilibrium between ^{103}Pd (half-life = 16.991 (19) d) and ^{103m}Rh (half-life = 56.114 (12) min) is reached within about 9 hours.



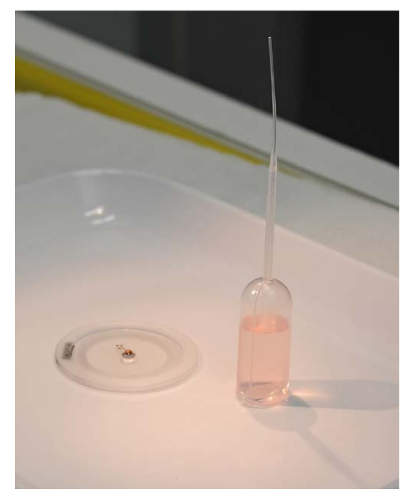
Energies are quoted in keV

Sources preparation

Initial solution : Palladium chloride in ammonium hydroxyde
Mass activity ~ 37 GBq.g^{-1}

- 6 point sources for gamma- and X-ray spectrometry

Dilution to 10 MBq.g^{-1} using 3 mol·L^{-1} HCl with 10 µg.g^{-1} of stable ruthenium to ensure chemical stability of ^{103m}Rh in equilibrium with ^{103}Pd.



- 6 volume sources for liquid scintillation (LS)

New dilution by a factor of 20.
Each source contained about 5 kBq of ^{103}Pd in Ultima Gold scintillator, and about 100 µL of water was added for stabilization purposes.

The impurity checking on the initial solution did not reveal any impurity and the detection limit was less than 2·10^{-4} Bq·Bq^{-1}. A complementary measurement was performed one month after the activity measurement, to benefit from the ^{103}Pd decay: Rhodium-101 (half-life = 3.30 (10) a) was detected with an activity relative to that of ^{103}Pd equal to 10^{-5} Bq·Bq^{-1}.

Activity measurement

The palladium chloride solution was standardized by liquid scintillation, using the Triple-to-Double Coincidence Ratio (TDCR) method.

The LS sources were measured using the RCTD1 counter of LNHB.
Each source was measured 10 times for 1 minute, allowing relative counting uncertainty of 6·10^{-4}.

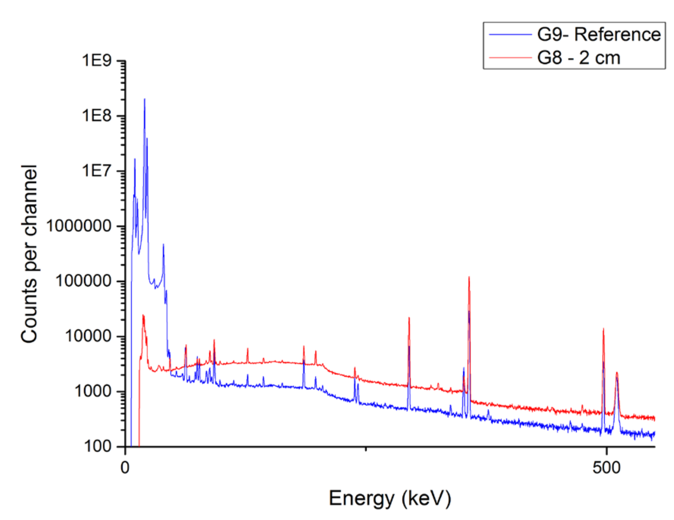
The detection efficiency was calculated taking into account the asymmetry of the photomultiplier tubes, by considering the three individual TDCR values, *i.e.* T/AB, T/BC and T/AC. The absorption of the photons was calculated by Monte Carlo simulation with PENELOPE

The relative standard uncertainty due to the source dispersion was 0.13% and the global uncertainty calculation was achieved by a global Monte Carlo simulation, following the recommendations of the supplement 1 of the Guide to the expression of uncertainty in measurement.

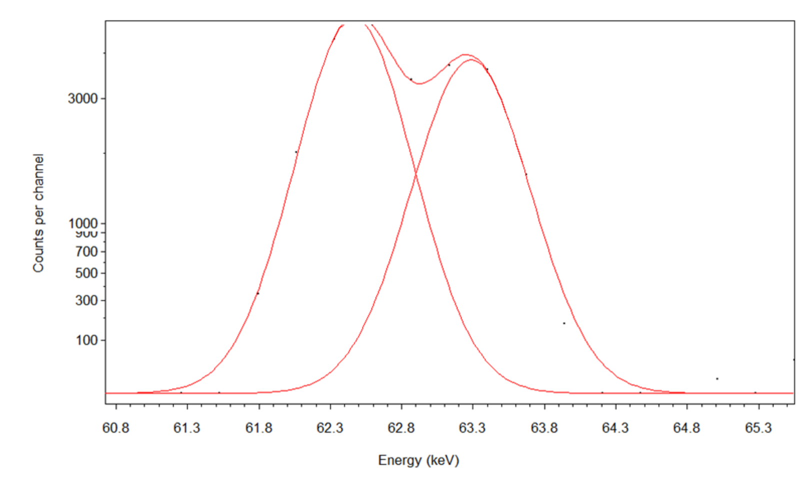
Reference activity: 9 472 (24) kBq.g^{-1}
(Reference date: 19/12/2017, 12h00 UTC)

Photon emission intensities

The absolute photon emission intensities were derived from gamma- and X-ray spectrometry using HPGe calibrated detectors, with different measuring conditions to cross-reference the results.



Spectrum of ^{103}Pd in the high energy range (8 keV - 550 keV)



Fit of two Gaussian functions to the experimental data of the 62-63 keV doublet

For each energy E_i, the photon emission intensity, I_i, is derived from the count rate in the relevant peak, n_i:

$$I_i = \frac{n_i \cdot \prod C_{ij}}{\varepsilon_i \cdot A}$$

ε_i: FEP efficiency for energy E_i,
A: source activity (Bq),
$C_{i,j}$ represents different correction factors.

	LARA (2019)		Present study	
Energy (keV)	**Photon emission intensity (%)**	**Relative uncertainty (%)**	**Photon emission intensity (%)**	**Relative uncertainty (%)**
XL	8.73 (23)	2.6	8.61 (43)	5
20.07 (Kα 2)	22.05 (22)	1.0	19.59 (21)	1.1
20.22 (Kα 1)	41.70 (40)	1.0	39.48 (42)	1.1
K alpha total	63.75 (46)	0.7	59.07 (47)	0.8
22.81 (Kβ1)	11.34 (15)	1.3	10.27 (11)	1.1
23.20 (Kβ2)	1.88 (7)	3.7	1.781 (19)	1.1
K beta total	13.22 (17)	1.3	12.05 (11)	0.9
K total	76.97 (49)	0.6	71.1 (6)	0.9
39.756	0.0698 (35)	5.0	0.0647 (7)	1.1
53.285	7.2 (20) E-6	28	< 8 E-6	-
62.41	7.8 (23) E-4	29	0.001128 (16)	1.4
241.88	4.9 (5) E-7	10	-	-
294.98	0.00297 (24)	8	0.00315 (7)	2.3
317.72	1.50 (19) E-5	13	1.37 (17) E-5	13
357.43	0.0246 (6)	2.4	0.02486 (17)	0.68
443.8	1.60 (12) E-5	7.5	2.1 (8) E-5	33
497.08	0.00411 (11)	2.7	0.00439 (7)	1.6

Absolute photon emission intensities in the decay of ^{103}Pd

Inconsistencies in the photon emission intensities
Question on the decay scheme equilibrium

Half-life

The half-life of ^{103}Pd was measured by gamma-ray spectrometry on a high-purity germanium (HPGe) detector, using an aliquot of the diluted solution, for 50 days. The measurement was carried out by following the ^{103}Pd main line (357.43 keV).

Authors	Year	Half-life (d)	Absolute uncertainty (d)	Relative uncertainty (%)	Number of half-lives
Rietjens et al.	1954	17.5	0.5	2.86	
Panontin et a.	1968	16.9	0.1	0.59	
Grunditz et al.	1969	18.4	0.5	2.72	4
Czock et al.	1975	16.961	0.016	0.09	2
Vaninbroukx et al.	1981	16.991	0.019	0.11	2-3
Popov et al.	2004	16.8	0.6	3.57	5.9
Present study	**2017**	**17.106**	**0.058**	**0.34**	**2.9**

Conclusions and perspectives

The 357.43-keV photon emission intensity is 0.02464 (16) per 100 disintegrations, in agreement with the tabulated data and obtained with a lower uncertainty.
In addition, the emission intensities of four other gamma rays are also derived with a significant reduction of the uncertainties.
Three gamma emissions (62.43 keV, 295.00 keV and 497.08 keV) from the third, fourth and fifth excited levels have significantly higher intensities than the tabulated data. Conversely, for the emission from the metastable level (39.75 keV), the present result is 7% lower than the tabulated value. Also, the L- and K-X-ray emission intensities are weaker (around 10%) than the tabulated data.
This would suggest that the intensity of the electron capture towards the first excited level should be weaker, while the electron capture branches to levels 3, 4 and 5 should be more intense. Another possibility would be an electron capture branch towards the stable level.
The present results provide new information and should give some useful clues to be exploited in a future evaluation of the ^{103}Pd decay scheme.

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