Underground search for the decay of $^{180}Ta^{m}$

Mikael Hult,[1,*] Joël Gasparro,[1] Gerd Marissens,[1] Patric Lindahl,[1] Uwe Wätjen,[1] Peter N. Johnston,[2] Cyriel Wagemans,[3] and Matthias Köhler[4]

[1]*European Commission, Joint Research Centre, Institute for Reference Materials and Measurements, Retieseweg 111, B-2440 Geel, Belgium*

[2]*Applied Physics, Royal Melbourne Institute of Technology, GPO Box 2476V, Melbourne 3001, Australia*

[3]*Gent University, Department of Subatomic and Radiation Physics, Proeftuinstraat 86, B-9000 Gent, Belgium*

[4]*Verein für Kernverfahrenstechnik und Analytik Rossendorf (VKTA), PF 510119, D-01314 Dresden, Germany*

(Received 10 April 2006; published 27 November 2006)

Tantalum-180m is a very rare primordial isotope and is not in its nuclear ground state. The radioactivity of $^{180}Ta^{m}$ has not yet been observed. Previous attempts to measure the half-life of $^{180}Ta^{m}$ have been performed using various detectors located above ground. In this work a 606 g Ta disk of natural isotopic composition was measured for 170 d in the 225 m deep underground laboratory HADES. The new lower bound for the half-life is 1.7×10^{16} y for electron capture decay and 1.2×10^{16} y for β^{-} decay. This gives a total lower bound for the half-life of 7.1×10^{15} y, which is a factor of 6 higher than the previous lower bound.

DOI: 10.1103/PhysRevC.74.054311 PACS number(s): 27.70.+q, 29.30.Kv, 23.40.−s, 29.40.Wk

I. INTRODUCTION

Natural tantalum is composed of ^{181}Ta and $^{180}Ta^{m}$ with relative natural abundances of 99.988(2)% and 0.012(2)% (in atom %), respectively [1]. Tantalum-180 is an odd-odd nucleus, which is only found naturally in an isomeric state (9^{-}). In its ground state, ^{180}Ta, has a half-life of 8.1 h and is thus not present in natural tantalum. The Ta-isotope with mass 180 in natural tantalum was first discovered by White, Collins, and Rourke [2] in 1955. At that time it was not known that it was an isomer. Eberhardt and co-workers [3] tried in the same year to measure its half-life using chemical separations and mass spectrometry. They determined a lower bound of 9.9×10^{11} y for the β^{-} branch towards ^{180}W and in a later paper [4] 4.6×10^{9} y for the electron capture (EC) branch towards ^{180}Hf. Since these early measurements a number of papers have presented improved (higher) lower bounds of the half-life, which are shown in Table I.

Since the discovery of $^{180}Ta^{m}$, there have been efforts to determine its production during nucleosynthesis in stars and various astrophysical scenarios have been presented [10–13]. Tantalum-180 has also been of some interest in recent years for its potential use in γ-ray lasers [14,15]. It has furthermore attracted significant interest in nuclear physics as it provides a case where one can search for possible structure effects induced by the high spin-state of the target. This recent interest has generated several studies on its nuclear properties [16]. In their paper from 1985, Cumming and Alburger [9] gave a good overview of certain nuclear properties of $^{180}Ta^{m}$. In the same paper they published the most recent accepted lower bounds for the half-life of $^{180}Ta^{m}$ (see Table I) of 1.2×10^{15} y. They performed γ-ray spectrometry measurements with a well-type HPGe detector operated in anticoincidence with a large NaI-detector. Cumming and Alburger used a Ta_2O_5 sample containing 151 mg of Ta enriched to 5.47% in $^{180}Ta^{m}$(approximately 8 mg of $^{180}Ta^{m}$). The decay scheme they based their investigation on is presented in Fig. 1. The same decay scheme was used in this investigation.

II. MATERIALS AND METHODS

A. Sample

The tantalum sample that was used in this study was a disk that was 18 mm thick and with a radius of 25 mm and weighing 606.04(1) g. Assuming natural isotopic abundances gives a $^{180}Ta^{m}$ mass of 73 mg in the sample. The disk was obtained from Goodfellow Cambridge Ltd. and had a purity of 99.9%. The only radioimpurity that it was possible to detect using underground γ-ray spectrometry was ^{182}Ta (half-life 114.43 d), which is formed from neutron activation by environmental thermal neutrons. In fact, the sample that was used in this investigation was initially employed as a highly sensitive neutron detector and for this reason it was initially placed in the underground laboratory Felsenkeller in Dresden (Germany). Felsenkeller has an overburden of 50 m of rock (125 m water equivalent). After one year in Felsenkeller the sample was transported 800 km to the much deeper laboratory HADES (High Activity Disposal Experimental Site), which is located at a depth of 500 m water equivalent at the premises of the Belgian nuclear centre SCK·CEN in Mol and which is operated by EURIDICE (European Underground Research Infrastructure for DIsposal of nuclear waste in Clay Environment). The transport was carried out by car in less than 10 h with the sample covered in 1 mm cadmium and 10 cm paraffin.

B. Measurements

The first measurement of the sample in HADES lasted for 58 d. It was carried out with the aim of obtaining a value for the average thermal neutron fluence rate in the Felsenkeller laboratory based on the ^{182}Ta activity. After being stored in

*Corresponding author. Phone: ++32 14 571 269; FAX: ++32 14 584 273; Email address: mikael.hult@ec.europa.eu

TABLE I. Published values for the lower bound for the half-life of $^{180}Ta^{m}$.

Reference	Technique[a]	Year of publication	Lower limit of half-life (y)		
			Electron capture	β^- decay	Total half-life
Eberhardt *et al.* [3]	Mass spec.	1955		9.9×10^{11}	
Eberhardt *et al.* [4]	Mass spec.	1958	4.6×10^{9}		4.5×10^{9}
Bauminger and Cohen [5]	γ-spec. using NaI	1958	2.3×10^{13}	1.7×10^{13}	9.7×10^{12}
Sakamoto [6]	γ-spec. using NaI	1967	1.5×10^{13} [b]		
Ardisson [7]	γ-spec. using Ge(Li)	1977	2.1×10^{13}	"Impossible"	
Norman [8]	γ-spec. using Ge(Li) and enriched Ta	1981	5.6×10^{13}	5.6×10^{13}	2.8×10^{13}
Cumming and Alburger [9]	γ-spec. using HPGe and enriched Ta	1985	3.0×10^{15}	1.9×10^{15}	1.2×10^{15}

[a]The description of each technique is significantly simplified in the table.
[b]A possible positive detection.

HADES for 46 months a second measurement started since it was realized that this sample could provide a possibility of detecting the radioactivity of $^{180}Ta^{m}$ or obtaining a higher lower bound for the half-life of $^{180}Ta^{m}$. The sample was measured for another 112 d (in total 170 d) on the same ultra low background HPGe-detector with a relative efficiency of 106% and with a crystal radius of 4 cm [17]. The sample was placed in a special Teflon container directly on the endcap of the detector. The deep location of HADES reduces the cosmic-ray induced muon flux more than a factor of 10^3, which significantly reduces the background count rate in γ-ray spectrometry [18,19]. In addition to lowering the number of direct hits on the detector by muons, the background associated with activation of the sample, detector and shield is significantly reduced when measuring underground. It is this very low background that creates the possibility of improving the previous "best" measurement by Cumming and Alburger from 1985.

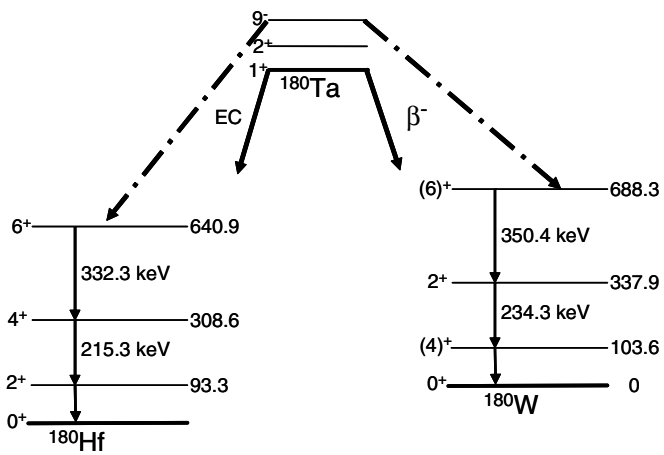

FIG. 1. The decay scheme of $^{180}Ta^{m}$.

The underground laboratory has very stable conditions regarding, e.g., temperature (less than 1° variation) and Rn-concentration (around 7 Bq/m^3). Furthermore the electrical connection was via an uninterruptible power supply that filters the incoming power and delivers a very stable and clean current. The HPGe-detector was connected to a single digital unit (DSA-2000) and the γ-ray spectra were recorded using the Genie-2000 data acquisition system. Each spectrum was recorded for typically 24 h and was inspected visually for any gain shifts or abnormalities. Gain shifts are normally very small and do not require correction. In cases where settings are changed the background peaks at 1460 and 609 keV were used for adjusting the energy calibration. The data analysis was performed by manually inspecting the spectrum.

C. Efficiency calculations

The decay scheme of $^{180}Ta^{m}$ (Fig. 1) indicates a relatively high probability for cascade summing, which means that two or more cascading decay radiations may be detected in coincidence and consequently reducing the full energy peak efficiency of each γ ray. In addition to this, new sum-peaks appear in the spectrum. This leads to fewer counts in the peaks of interest and makes the search for the decay more difficult. In order to quantify these effects Monte Carlo calculations using the EGS4 software [20] were conducted. The complete decay scheme of $^{180}Ta^{m}$ was entered into the Monte Carlo calculations as well as the geometry of the HPGe-detector and the sample. In the Monte Carlo technique a pseudo random number generator determines the origin and direction of each γ ray. Each photon is tracked until it is either fully absorbed or leaves the space that is defined in the model. This space includes the sample, the detector and the shield. It is inherent to the Monte Carlo calculation that the self attenuation in the sample is taken into account. The energy intervals of most

TABLE II. Half-life limits obtained from the different energy intervals of interest from the measurements of the Ta-disk. The full energy peak efficiency per decay is also shown.

Energy of expected peak (keV)	Full energy peak efficiency per decay (%)	Energy interval (keV)	Lower bound of half-life (y)	Decay branch
93.3	0.014	91.5–95.1	1.8×10^{14}	EC
215.3	0.45	213.3–217.3	6.3×10^{15}	EC
332.3	1.07	330.2–334.4	1.7×10^{16}	EC
103.6	0.022	101.8–105.4	2.9×10^{14}	β^-
234.3	0.54	232.0–236.0	7.7×10^{15}	β^-
350.4	1.14	348.8–353.0	1.2×10^{16}	β^-

interest are around the following peaks: 215.3 and 332.3 keV for the electron capture decay and 234.3 and 350.4 keV for the β^- decay. The simulations show for example that some 0.13% of the counts in the 332.3-keV full energy peak will be lost due to summing with x rays and that 11% will be lost due to $\gamma\gamma$ summing. Furthermore, the Monte Carlo simulations served as a means of obtaining the detection efficiency used in the calculation of the half-life. It was assumed that the $^{180}Ta^{m}$ was homogeneously distributed in the sample. The full energy peak efficiencies for the different γ rays are listed in Table II. The number of histories for a simulation was always kept high (10^7) in order to keep the statistical uncertainties low. The main uncertainties arose from the interaction cross sections and the model description of the detector. The Monte Carlo method for determining the detection efficiency of samples with strange shapes or with radionuclides for which there is no reference material, has been successfully used in many laboratories [21,22]. This approach was validated through the use of a reference source composed of ^{176}Lu, which has a long half-life, $37.9(3) \times 10^9$ y, and a decay scheme that greatly resembles that of $^{180}Ta^{m}$.

D. The influence of ^{182}Ta

The decay of ^{182}Ta is followed by x rays and some 41 different γ rays. Six of the γ rays (68, 100, 1121, 1189, 1221, and 1231 keV) have emission probabilities above 10%. Compton scattering from four of those six γ rays generate background in the energy intervals of most interest for this study. This effect was studied using Monte Carlo simulations with the EGS4 code and with the decay scheme of ^{182}Ta introduced in the simulations. The simulations show that e.g. in the energy region around 332 keV, there should be three counts after the first (58-d) measurement from ^{182}Ta. The data show that in the same region there are 133 counts. The background contribution from ^{182}Ta is thus around 2.3% for the first measurement. The relative background contributions of ^{182}Ta to the other energy intervals of interest for this study are of the same order of magnitude. Because of the decay of ^{182}Ta while being stored in HADES, its contribution to the second measurement is negligible.

The $^{180}Ta^{m}$ half-life limit was calculated both with and without the first measurement. It was found that it was better to include the data of the first measurement although the contribution from ^{182}Ta was greater there. There should be no peak from ^{182}Ta in any of the interesting regions. Care was taken not to calculate the continuous background in regions with γ-ray peaks from ^{182}Ta (at 100.1, 222.1, and 229.3 keV). The ^{182}Ta peak at 351.05 keV has such a low emission probability (0.0091%) that it can be neglected.

III. RESULTS

Figure 2 shows the part of the γ-ray spectrum collected for 170 d that encompasses the energy regions with the 350.4, 332.3, 234.3, and 215.3 keV γ rays that are expected from the decay of $^{180}Ta^{m}$. The peaks that are seen are solely coming from common background lines in the natural decay chains. The apparent peak at 181.5 keV (immediately to the left of the 186-keV peak from ^{226}Ra) could not be identified and is most likely due to pulse statistics as it is near to decision threshold. The energy intervals of interest are listed in Table II. Based on the count rates in those intervals the calculation of decision thresholds was carried out following the international standard [23]. All the results were compared to the decision thresholds using $\alpha = 0.05$ (probability of error of the first kind).

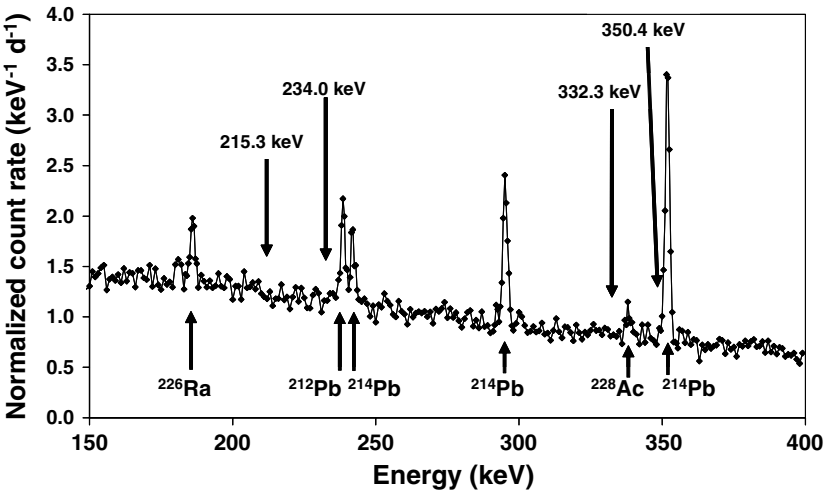

FIG. 2. The part of the γ-ray spectrum collected for 170 d that encompasses the energy regions with the γ rays expected from the decay of $^{180}Ta^{m}$. The spectrum has been re-binned to enhance visibility. The peaks are all from background of the naturally occurring decay chains and the radionuclides being the major contributors to those peaks are indicated.

TABLE III. The new lower bounds of the half-life and corresponding log*ft* values for ^{180}Ta^m arising from this work.

	EC decay	β^- decay	Total
Lower bound for half life (y)	1.74×10^{16}	1.21×10^{16}	7.14×10^{15}
Lower bound for log *ft*	23.7	22.2	

The disintegration constant, λ, for each branch was calculated using the relation

$$\lambda_{db} = \frac{A_{db}}{n} = A_{db}\frac{M}{m\theta N_A},$$

where n is the number of ^{180}Ta^m atoms in the sample, A is the ^{180}Ta^m activity (in Bq) and the suffix "db" stands for decay branch and is either the EC branch or the β^- branch. The other parameters are the following:

M = atomic mass of ^{180}Ta^m = 179, 947, 464.8(2.4)μu [24]

θ = the natural isotopic abundance of ^{180}Ta^m = 0.012(2)% (atom %) [1]

m = the mass of the sample = 606.04(1) g

N_A = the Avogadro number = 6.0221415(10) $\times$ 10^{23} mol^{-1} [25].

The disintegration constant for the total decay is simply the sum of the two decay constants.

The half-life limits obtained for the different γ rays are listed in Table II. From this it is clear that the 332.3-keV γ ray is best suited to study the EC decay giving a lower bound for the half-life of 1.7×10^{16} y, while for the β^- decay the 350.4-keV γ ray gives the highest lower bound of 1.2×10^{16} y. The γ-ray peak at 350.4 keV would sit on the low energy tail of the background peak at 351.9 keV (^{214}Pb), which hampers the usefulness of that γ ray but it is still preferred to the 234.3-keV and 103.6-keV γ rays that suffer from more self attenuation. The background in the region of interest around the 350.4-keV peak goes up by a factor of 2 due to the ^{214}Pb-peak. The decision threshold for the decay constant is proportional to the square root of the background but inversely proportional to the detection efficiency. The higher detection efficiency (a factor of 2.1) at 350.4 keV compared to at 234.3 keV favours thus the peak at 350.4 keV. Given the data above, the new half-life limits as well as corresponding log*ft* values for these third-forbidden nonunique transitions are presented in Table III.

IV. DISCUSSION

The isomer ^{180}Ta^m has frequently been called "nature's rarest naturally occurring isotope" since it occurs in the solar system with an abundance of only 2.48×10^{-6} atoms per 10^6 Si atoms [26]. To explain this very low abundance, various nucleosynthesis processes (s, r, p) and astrophysical production sites (supernovae, AGB stars, cosmic radiation) have been proposed. A detailed discussion can be found in recent papers by Loewe *et al.* [10] and Arnould and Goriely [12]. Although there is agreement that an r-process origin of ^{180}Ta^m is unlikely, the papers advocate different production processes. Loewe *et al.*, claim that the observed abundance of ^{180}Ta^m can be fully explained by s-process nucleosynthesis, while Arnould and Goriely claim a p-process origin. Considering the arguments it is likely that both nucleosynthesis processes contribute. Detecting the decay of ^{180}Ta^m could help in resolving this issue.

The measurement presented in this paper was not tailor made to search for the radioactivity of ^{180}Ta^m but rather a spin-off from another measurement. Nevertheless, because of the low background in the underground laboratory it was possible to increase the lower bound of the half-life a factor of 6 compared to the previous best value. It should be possible to extend the lower bound at least another factor of 4 using the same technique[1] (underground γ-ray spectrometry) and measurement time but in an experiment that has been designed explicitly for the purpose. The proposed experiment optimizes the sample geometry using Monte Carlo calculations as well as using multiple detectors (including muon veto-shields) for further suppression of background as well as aid in tagging interesting events. In case such a measurement still does not succeed in detecting the radioactivity one could propose to use detectors aimed at measuring neutrinos or double-β decay, e.g., the GERDA detector [27], to gain a factor of 100 or more.

From theory it is difficult to predict what the half-life should be. According to Norman [8], one can say that the decay by internal transition should have a half-life of shorter than 10^{27} y. The β decays should have higher probabilities so the authors believe that it is unlikely that the radioactivity should remain hidden provided the improved measurements techniques as outlined here are used. This study also shows that it is possible to use tantalum with natural isotopic abundance for this type of work. It would, however, be beneficial for the study to decrease the uncertainty of the natural isotopic abundance of ^{180}Ta^m.

ACKNOWLEDGMENTS

The authors are grateful to the staff of EURIDICE at the HADES underground laboratory for their technical support. Christoffer Ellmark (Lund University) is acknowledged for his work with the decay scheme of ^{182}Ta. Ron Fleming (University of Michigan) is thanked for his input at the start of this project.

[1] It is worth noting that since this measurement was finished the background characteristics of the HPGe-detector were improved after some changes to the endcap that reduced the amount of ^{40}K and ^{226}Ra significantly. The count rate in, e.g., the 351.9-keV peak from the decay of ^{214}Pb is now a factor of 2.7 lower than what appears in Fig. 2 (i.e., 2.0 ± 0.2 counts per day instead of 5.4 ± 0.3).

[1] N. E. Holden, R. L. Martin, and I. L. Barnes, Pure Appl. Chem. **56**, 675 (1984).
[2] F. A White, T. L. Collins, and F. M. Rourke, Phys. Rev. **97**, 566 (1955).
[3] P. Eberhardt, J. Geiss, and C. Lang, Z. Naturforschg. **10A**, 796 (1955).
[4] P. Eberhardt and P. Signer, Z. Naturforschg. **13A**, 1004 (1958).
[5] E. R. Bauminger and S. G. Cohen, Phys. Rev. **110**, 953 (1958).
[6] K. Sakamoto, Nucl. Phys. **A103**, 134 (1967).
[7] G. Ardisson, Radiochem. Radioanal. Lett. **29**, 7 (1977).
[8] E. B. Norman, Phys. Rev. C **24**, 2334 (1981).
[9] J. B. Cumming and D. E. Alburger, Phys. Rev. C **31**, 1494 (1985).
[10] M. Loewe, P. Alexa, J. de Boer, and M. Würkner, Nucl. Phys. **A719**, 275c (2003).
[11] F. Käppeler, H. Beer, and K. Wisshak, Rep. Prog. Phys. **52**, 945 (1989).
[12] M. Arnould and S. Goriely, Phys. Rep. **384**, 1 (2003).
[13] D. Belic *et al*., Phys. Rev. Lett. **83**, 5242 (1999).
[14] R. Coussement, R. Shahkmouratov, G. Neyens, and J. Odeurs, Europhysics News **34/5**, 190 (2003).
[15] S. A. Karamian, C. B. Collins, J. J. Carroll, and J. Adam, Phys. Rev. C **57**, 1812 (1998).
[16] T. Wendel *et al*., Phys. Rev. C **65**, 014309 (2001).
[17] M. Hult, J. Gasparro, L. Johansson, P. N. Johnston, and R. Vasselli, *Environmental Radiochemical Analysis II, Special publication No. 291*, edited by Peter Warwick (The Royal Society of Chemistry, Cambridge, UK, 2003), p. 373.
[18] M. Laubenstein *et al*., Appl. Radiation Isot. **61**, 167 (2004).
[19] M. Hult, W. Preuße, J. Gasparro, and M. Köhler, Acta Chim. Slov. **53**, 1 (2006).
[20] W. R. Nelson, H. Hirayama, and D. W. O. Rogers, Stanford Linear Accelerator Center Report 265, 1985.
[21] J. C. Hardy *et al*., Appl. Rad. Isot. **56**, 65 (2002).
[22] P. N. Johnston, M. Hult, and J. Gasparro, Appl. Rad. Isot. **64**, 1323 (2006).
[23] ISO, Determination of the detection limit and decision threshold for ionizing radiation measurements – ISO 11929 Part 3, 1st ed. (International Standards Organization, Geneva, Switzerland, 2000).
[24] G. Audi, A. H. Wapstra, and C. Thibault, Nucl. Phys. **A729**, 337 (2003).
[25] http://physics.nist.gov/cgi-bin/cuu/Value?na, accessed April 30, 2006.
[26] E. Anders and N. Grevesse, Geochim. Cosmologi. Acta **53**, 197 (1989).
[27] S. Schönert and the GERDA Collaboration, Nucl. Phys. B, Proc. Suppl. **145**, 242 (2005).