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Search for the radioactivity of ^{180m}Ta using an underground HPGe sandwich spectrometer

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ABSTRACT

The radioactivity of ^{180m}Ta has never been detected. The present attempt to detect it was carried out using a newly developed HPGe sandwich spectrometer installed 500 m water equivalent underground in the HADES laboratory. The sample consisted of 6 discs of tantalum of natural isotopic composition with a total mass of 1500 g and a total mass for ^{180}Ta of 180 mg. The sample was measured for 68 days and the resulting lower bound for the half-life of ^{180m}Ta was 2.0×10^{16} y, which is a factor of 2.8 higher than the previous highest value.

1. Introduction

Tantalum-180 m is a radionuclide that has generated a lot of interest in recent years for various reasons. The ground state of ^{180}Ta has a short half-life of only 8.1 h but its metastable state is the most long-lived metastable state known to man. Tantalum-180 is the rarest isotope of nature's rarest element and its isotopic abundance was measured recently with high precision by de Laeter and Bukilic (2005) to only 0.01201(8)%. This gives a Solar System abundance of ^{180m}Ta of 2.49×10^{-6} where the reference Si equals 10^6. Recent interest for this nuclide has been due to its potential use in gamma-ray lasers (Carroll, 2007; Coussement et al., 2004) and the debate on its production in the stellar nucleosynthesis (Mohr et al., 2007; Loewe et al., 2003) as well as its wide use in studies of nuclear structures of nuclei with high spin state (Wendel, 2001).

The decay scheme of ^{180m}Ta can be considered to be well understood and was recently (2003) updated in the ENSDF data base (Nuclear Data Sheets, 2008). The part of the decay scheme essential for this investigation is presented in Fig. 1. The decay is composed of an electron capture branch and a β^--decay branch. There is also the possibility for isomeric transition from the 9^+ level to the 2^+ or 1^+ levels. Norman (1981) claims that the decay by isomeric transition should have a half-life greater than 10^{27} y and that the β decays are expected to have higher probabilities.

The radioactivity of ^{180m}Ta is yet to be detected. There are eight attempts to detect it reported in literature (Hult et al., 2006) starting shortly after it was first discovered in White et al. (1955). Presently, the highest value for the lower bound of the half-life is 7.1×10^{15} y. This value was determined in the first underground measurement of the nuclide although the experiment was not optimized for this task (Hult et al., 2006). The experiment reported here was the first ever underground measurement specially designed for searching for the decay of ^{180m}Ta. The measurement was carried out using a newly developed HPGe sandwich spectrometer (Wieslander et al., 2009) installed in the underground laboratory HADES, which is located at a depth of 500 m water equivalent (w.e.).

2. Materials and methods

2.1. Sample

The sample consisted of 6 discs of high purity tantalum of natural isotopic composition. The discs were 10 cm in diameter and together they had a mass of 1500.33 g. Using the natural isotopic abundance of 0.01201% (de Laeter and Bukilic, 2005) gives the total mass for ^{180}Ta of 180 mg. This is to be compared with 73 mg used in the previous underground experiment (Hult et al., 2006) and 8 mg used in the enriched sample by Cumming and Alburger (1985). In order to minimize the background contribution from surface impurities the Ta discs underwent a thorough surface cleaning procedure in a bright dipping solution.

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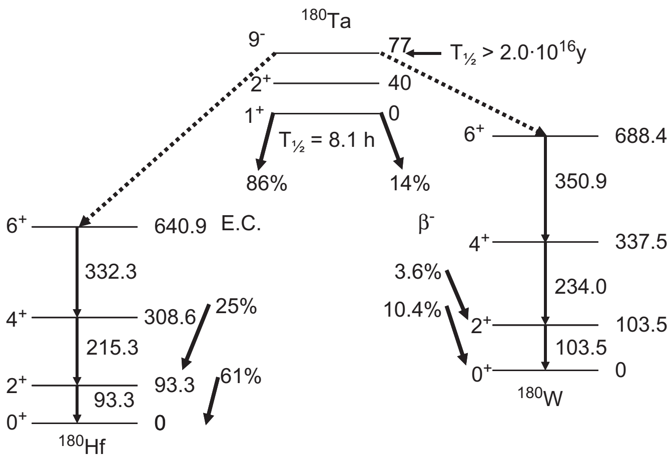

Fig. 1. The decay scheme of ^{180}Ta.

This procedure involved degreasing in perchloroethylene, ultrasound rinsing in soapy solutions as well as in ultrapure water, immersion and stirring in the bright dipping solution, rinsing several times with ultrapure water and finally drying in ethanol. This cleaning resulted in a drastic change in surface colour from grey to silver and a loss of mass of 10%. The discs were kept almost 2 years in HADES prior to starting the measurement in order to reduce the activity of the cosmogenically produced ^{182}Ta (half-life: 114 d). However, during disc cleaning the discs spent 3 weeks above ground 3.5 months prior to commencing the measurements. This generated some ^{182}Ta that contributed to the background of this measurement.

2.2. Detection system

The measurements took place in the HADES underground laboratory (Hult et al., 2006), which is operated by EURIDICE (European Underground Research Infrastructure for Disposal of nuclear waste In Clay Environment) and located at the premises of the Belgian Nuclear Centre SCK●CEN in Mol, Belgium

The tantalum sample was placed between two ultra-low-background HPGe-detectors as depicted in Fig. 2. The shape of the sample enabled a very tight fit and thus minimizing the air volume inside the shield, which consequently limited the radon induced background. The average radon activity concentration in the laboratory was 7 Bq/m^3 during the measurement. Inside the detector-shield, the radon level is expected to be lower than 7 Bq/m^3 because of the flushing with boil-off nitrogen from the Dewars. For this measurement two p-type coaxial crystals were used. The upper detector (Ge-7) has a submicron deadlayer (~0.3 μm) and a relative efficiency of 89.4%. The lower detector (Ge-6) has a deadlayer of 0.9 mm and a relative efficiency of 80.5%. The germanium sandwich spectrometer is described in detail by Wieslander et al. (2009). The background reduction is achieved through (i) placement of the system underground at 500 m w.e., (ii) use of two plastic scintillators as a muon shield and (iii) time stamped list-mode data in order to enable anti-coincidence background reduction off line from e.g. Compton scattering.

Spectra were collected in 24 h intervals and an energy calibration was performed generally every 7 days. All spectra were looked at and checked for inconsistencies and energy calibration. The measurements were interrupted at several occasions in order to measure samples from other projects so the time difference between the first spectrum and the last spectrum was 258 days. The energy calibration was very stable over each measurement period for one or two weeks. But due to change of hardware the energy calibration changed at some occasions of the 258 day period. This was taken care of by adding spectra after converting the *x*-axis to energy in keV.

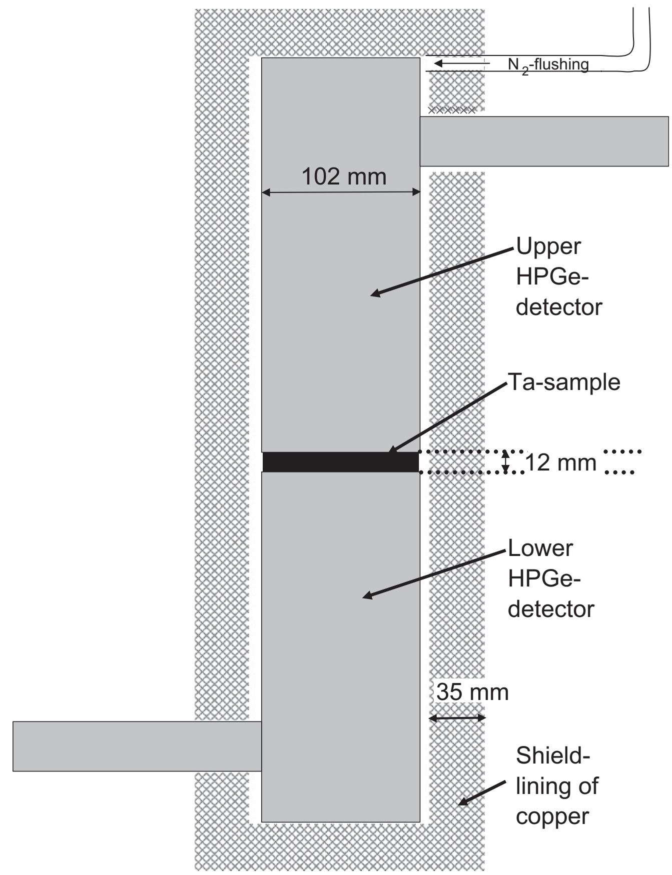

Fig. 2. Sketch of the sample, the detector configuration and innermost shield at a cross section taken where the detector cryostat arms enter the shield. Outside the copper lining there is 17 cm of lead. The drawing is to scale and describes a cylindrical geometry.

2.3. Efficiency calculation

The full energy peak (FEP) efficiencies per decay were calculated using the Monte Carlo code EGS4. The computer model of the sandwich spectrometer that was used in the Monte Carlo code was based on radiographs of the detectors. The thicknesses of the deadlayers on the front, side and back of the HPGe-detectors in the model were adjusted in an iterative procedure in which the FEP efficiency for a number of point sources measured at three different distances were compared with the efficiency calculated using the Monte Carlo code. The iterative procedure was stopped when the relative differences were better than 3%. The computer simulation also contained the complete decay scheme of ^{180m}Ta including internal conversion and X-rays. The expected cascading gamma-rays following the decay of ^{180m}Ta will result in a loss of counts in the FEP peaks due to coincidence summing. For a small sample in a well detector (Cumming and Alburger, 1985) it would be advantageous to detect the sum peaks. In this case, the large sample and sandwich design with independent detection using two detectors favored detection of the single lines. The efficiencies for the two major sum peaks are also stated in Table 1. This approach has been validated by using a

Table 1
Half-life limits obtained from the different energy intervals of interest from the measurements of the Ta-disc.

Energy of expected peak (keV) and decay mode	Full energy peak efficiency per decay (%)	Lower bound of half-life (y)
93.3 (EC)	0.020(2)	3.68×10^{14}
215.3 (EC)	0.96(10)	1.44×10^{16}
332.3 (EC)	2.40(24)	4.45×10^{16}
215.3+332.3 (EC)	0.087(9)	2.39×10^{15}
103.5 (β^-)	0.035(4)	5.27×10^{14}
234.0 (β^-)	1.16(12)	1.79×10^{16}
350.9 (β^-)	2.54(25)	3.65×10^{16}
234.0+350.9 (β^-)	0.099(10)	2.17×10^{15}

The full energy peak efficiency per decay is also shown.

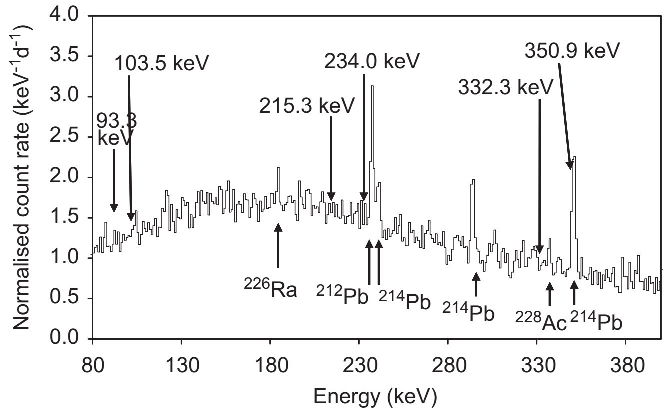

Fig. 3. The part of the γ-ray spectrum collected for 68 days that encompasses the energy regions with the γ-rays expected from the decay of ^{180m}Ta. The spectrum has been re-binned to enhance visibility. The peaks are all from background of the naturally occurring decay chains and the radionuclides being the major contributors to those peaks are indicated.

^{176}Lu reference source consisting of a liquid solution with 8.95 g of $LuCl_3 \cdot 6(H_2O)$ of natural isotopic abundance (104 mg of ^{176}Lu). It was contained in a cylindrical Teflon container with an O-ring seal. The inside of the container was 80 mm in diameter and 10 mm in height, while the outside measures where 110 mm and 14 mm, respectively. ^{176}Lu has a long half-life (3.6×10^{10} y) and a decay scheme for the β^- decay that is similar to that of ^{180m}Ta. The relative difference of the calculated and measured FEP efficiencies for the peaks at 88, 202 and 307 keV were 12%, −6% and −1%, respectively.

3. Results

The relevant part of the resulting sum spectrum with muon induced counts subtracted is shown in Fig. 3. The resolution of the final spectrum was about 20% worse than in the previous measurement (Hult et al., 2006) due to stability problems with the electronics, which marginally degrades the data. There is no sign of the expected peaks and there were no unidentified peaks in the rest of the spectrum. The only peaks that are not amongst the normal background peaks are from ^{182}Ta, which was produced by neutron activation in the Ta-discs when they were stored above ground. In Fig. 3 (below the spectrum curve) is indicated the location of the visible background peaks. At 122 keV is a peak-like structure that is likely to be attributed to background from ^{57}Co located in copper parts and possibly also in the Ge-crystals as that peak is also seen (but below decision threshold) in the background spectrum. It is worth noting that the count-rate in the region below 300 keV is lower when the Ta-sample is present. This tells us that the sample is very radiopure and improves the shielding. The decision thresholds for the peaks in question were calculated using the formula given in the international standard with the error of first kind (α) set to 0.05 (ISO, 2000). The upper bound of the disintegration constant (λ) was calculated for each decay branch (EC or β^-) by taking the ratio of the decision threshold of the activity and the number of ^{180}Ta atoms in the sample ($n = 6.0 \times 10^{20}$). More details on the calculation are given in the paper describing the previous underground attempt to detect the radioactivity of ^{180m}Ta by Hult et al. (2006). The resulting lower bounds for the half-life calculated for each gamma-line are presented in Table 1. Due to self-absorption in the sample and conversion, the efficiencies for the 93.3 and 103.5 keV peaks, and their respective sum-peaks, are very low and consequently the corresponding half-life limits are also relatively low. The combined lower bound for each decay branch and the total half-life and the corresponding log ft values calculated for third-forbidden non-unique transitions are reported in Table 2. The background count-rate of the background peak at 351.9 keV from ^{214}Pb is 4.4(4) d^{-1}. In this experiment (Fig. 3) the count-rate of that peak was 4.0(4) d^{-1}, which is consistent with the background value. This peak hampers detection of the expected 350.9 keV gamma-ray[1] from ^{180}W. However, the peak count-rate is more or less equal to the count-rate of the continuous background under the peak, so the effect of the background peak is not so severe. Because of (i) the minute quantities of air near the detectors, (ii) the high purity of the thick copper lining, (iii) the low radon concentration in the laboratory air and (iv) the flushing of the sample volume with boil-off liquid nitrogen, the main contribution to this peak is expected to come from ^{226}Ra impurities inside the detectors. This should thus alleviate great variations in the background count-rate, which is also what has been observed so far with this relatively new detection system. The peak count-rates of the 242 and 295 keV peaks (from the decay of ^{214}Pb) were 1.3(4) and 2.0(2) d^{-1}, respectively, which is somewhat higher than in the background spectrum. The same applies to other radon induced background peaks at 609 and 1764 keV. This indicates that the count-rate in the peak at 351.9 keV is due to decay of ^{214}Pb and not ^{180m}Ta.

Table 2
The new lower bounds of the half-life and corresponding log ft values for ^{180m}Ta arising from this work.

	EC decay	β^- decay	Total
Lower bound for half-life (year)	4.45×10^{16}	3.65×10^{16}	2.0×10^{16}
Lower bound for log ft	24.1	22.9	

4. Discussion

The new sandwich spectrometer is a very useful instrument in many projects requiring detection of mBq activities and will be fully occupied for many years to come. Even so it is hoped that there will be time slots available to continue the search for the decay of ^{180m}Ta using this set-up. Another 300 days of data-taking with the present background would result in a detection limit of about 5.2×10^{16} y for the total half-life and 1.15×10^{17} y for the EC

[1] The energy of this gamma transition was until 2003 in the ENSDF evaluation with a value of 350.4 keV.

branch solely. In order to reach a total half-life limit of 10^{17} y, it would be necessary to measure during 3.75 y, assuming a constant background. This could be considered the uppermost limit of what this detection system can reach. It is possible to improve the present concept somewhat. Recent developments in producing large HPGe-crystals may continue (Hult, 2007) and the background can be reduced further. Simulations show that by using two detectors with crystals of 9 cm in diameter (instead of 8 cm as in this experiment) combined with using a bigger Ta-disc of 12 cm diameter and assuming a background reduction of a factor of 2 would result in a lower bound of the total half-life of 10^{17} y after a measuring time of 1.7 y. Should it be considered worth the effort to search beyond 10^{17} y, other more highly performing systems can be designed by using enriched samples with ^{180}Ta masses of a few hundreds of mg and/or using big detector systems used for detection of rare events like e.g. the GERDA detector (Schönert and the GERDA Collaboration, 2006).

5. Conclusions

- A new underground experiment designed for measuring the decay of ^{180m}Ta could not detect its radioactivity. It resulted, however, in a new lower bound of its half-life 2.0×10^{16} y, which is a factor of 2.8 higher than the previous lower bound.
- The present system has the possibility of increasing the lower bound of the half-life to 10^{17} y after 3.7 years of data taking.

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References

Carroll, J.J., 2007. Nuclear structure and the search for inducedenergy release from isomers. Nuclear Instruments and Methods in Physics Research B 261, 960–964.
Coussement, R., Shahkmouratov, R., Neyens, G., Odeurs, J., 2004. Quantum optics with gamma radiation. Europhysics News 34/5, 190.
Cumming, J.B., Alburger, D.E., 1985. Search for the deacy of ^{180}Ta^{m}. Physical Review C 31, 1494.
de Laeter, J.R., Bukilic, N., 2005. Isotope abundance of ^{180}Ta^{m} and p-process nucleosynthesis. Physical Review C 72, 025801.
Hult, M., Gasparro, J., Marissens, G., Lindahl, P., Wätjen, U., Johnston, P.N., Wagemans, C., Köhler, M., 2006. Underground search for the decay of ^{180}Ta^{m}. Physical Review C 74 (054311), 1–5.
Hult, M., Preusse, W., Gasparro, J., Köhler, M., 2006. Underground gamma-ray spectrometry. Acta Chimica Slovenica 53, 1–7.
Hult, M., 2007. Low-level gamma-ray spectrometry using Ge-detectors. Metrologia 44 (4), S87–S94 (Erratum in Metrologia 44, No. 5, 425).
ISO, 2000. International Standard 11929-3. Determination of the detection limit and decision threshold for ionizing radiation measurements. International Standards Organization, Geneva, Switzerland.
Loewe, M., Alexa, P., de Boer, J., Würkner, M., 2003. The ^{180}Ta production puzzle and the classical s-process. Nuclear Physics A 719, 275c.
Mohr, P., Käppeler, F., Gallino, R., 2007. Survival of nature's rarest isotope ^{180}Ta under stellar conditions. Physical Review C 75, 012802.
Norman, E.B., 1981. Half-life of ^{180}Ta. Physical Review C 24, 2334.
Nuclear Data Sheets, 〈http://www.nndc.bnl.gov/nds/〉, accessed, September 25, 2008.
Schönert, S., The GERDA Collaboration, 2006. Status of the germanium detector array (GERDA) for the search of neutrinoless ββ decays of 76Ge at LNGS. Progress in Particle and Nuclear Physics 57, 241–250.
Wendel, T., et al., 2001. Low-K two-quasiparticle states in ^{180}Ta. Physical Review C 65, 65.
White, F.A., Collins, T.L., Rourke, F.M., 1955. New naturally occurring isotope of tantalim. Physical Review 97, 566.
Wieslander, J.S.E., Hult, M., Gasparro, J., Marissens, G., Preusse, W., Misiaszek, M., 2009. An ultra low-background HPGe-detector system in a sandwich configuration. Applied Radiation and Isotopes, these proceedings, doi:10.1016/j.apradiso.2009.01.026.