Applied Radiation and Isotopes 67 (2009) 918–921

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

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

Mikael Hult [a,*], J.S. Elisabeth Wieslander [a,c], Gerd Marissens [a], Joël Gasparro [a], Uwe Wätjen [a], Marcin Misiaszek [b]

[a] European Commission, Joint Research Centre, Institute for Reference Materials and Measurements, Retieseweg 111, B-2440 Geel, Belgium
[b] M. Smoluchowski Institute of Physics, Jagiellonian University, ul. Reymona 4, 30-059 Krakow, Poland
[c] Department of Physics, P.O. Box 35 (YFL), FIN-40014 University of Jyväskylä, Finland

ARTICLE INFO

Keywords:
Low-level
Underground
Tantalum
180 m
Half-life
Gamma-ray spectrometry
Muons

ABSTRACT

The radioactivity of $^{180\mathrm{m}}$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 $^{180\mathrm{m}}$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 $^{180\mathrm{m}}$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 $^{180\mathrm{m}}$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 $^{180\mathrm{m}}$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 $^{180\mathrm{m}}$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.

---

[*] Corresponding author. Tel.: +32 14 571269; fax: +32 14 584273.
E-mail address: mikael.hult@ec.europa.eu (M. Hult).

0969-8043/$ - see front matter © 2009 Elsevier Ltd. All rights reserved.
doi:10.1016/j.apradiso.2009.01.057

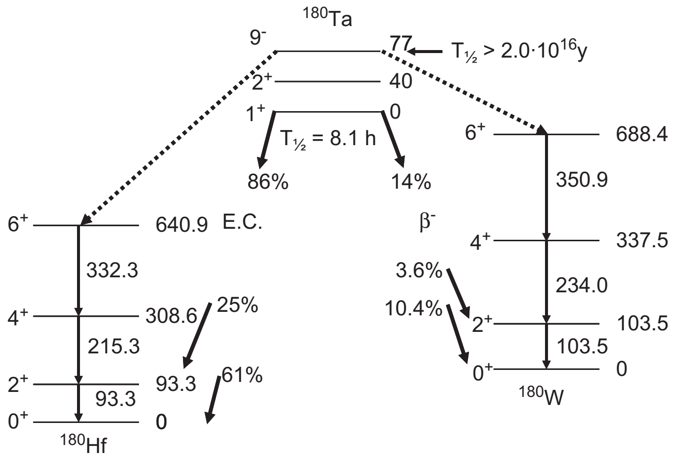

Fig. 1. The decay scheme of ^{180}Ta.

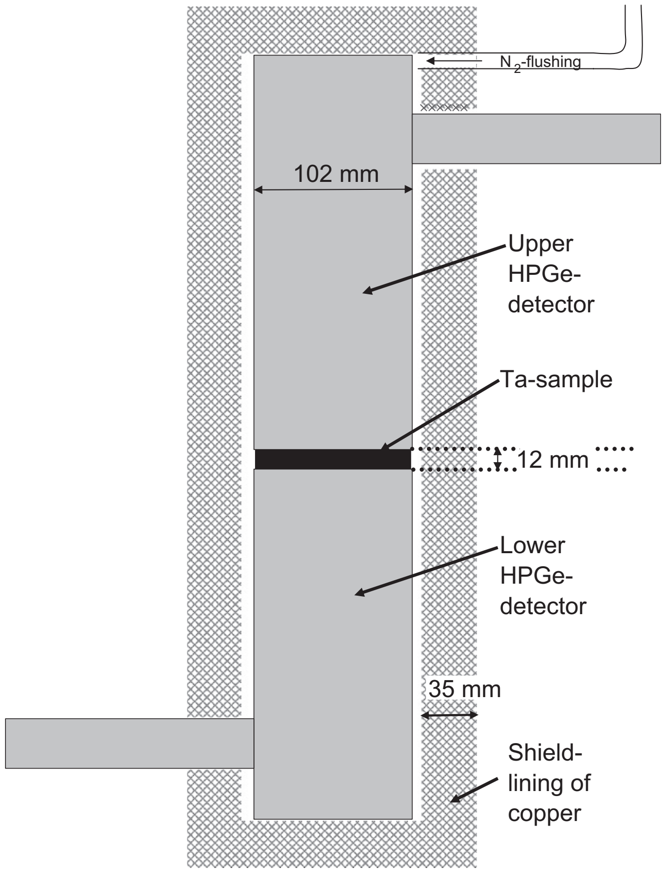

Fig. 2. Sketch of the sample, the detector configuration and innermost shield at a cross section taken where the detector cryostat arms enter the shield. Outside the copper lining there is 17 cm of lead. The drawing is to scale and describes a cylindrical geometry.

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 ($\sim$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.

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