



Experimental electron binding energies for thulium in different matrices



A.Kh. Inoyatov^{a,b,*}, A. Kovalík^{a,c}, D.V. Filosofov^a, M. Ryšavý^c, L.L. Perevoshchikov^a,
Yu.V. Yushkevich^a, M. Zbořil^{d,1}

^a Laboratory of Nuclear Problems, JINR, Dubna, Moscow Region, Russian Federation

^b Institute of Applied Physics, National University, Tashkent, Uzbekistan

^c Nuclear Physics Institute of the ASCR, CZ-25068 Řež near Prague, Czech Republic

^d Institut für Kernphysik, Westfälische Wilhelms-Universität Münster, D-48149 Münster, Germany

ARTICLE INFO

Article history:

Received 6 December 2014

Received in revised form 25 February 2015

Accepted 25 February 2015

Available online 6 March 2015

Keywords:

¹⁶⁹Tm

¹⁶⁹Yb

Atomic environment

Electron binding energy

Chemical shift

Natural atomic level width

ABSTRACT

The L₁, L₂, L₃, M₁, M₂, N₁, N₃, O₁, O₂, O₃, and P₁ subshell electron binding energies (related to the Fermi level) in thulium generated by the electron capture decay of radioactive ¹⁶⁹Yb atoms implanted at 30 keV into polycrystalline platinum and aluminum foils and deposited by vacuum evaporation on surfaces of polycrystalline platinum, carbon, and aluminum foils were determined by the internal conversion electron spectroscopy. The greatest differences in the electron binding energies (4.5 ± 0.1 eV in the average without the P₁ shell and 7.0 ± 0.5 eV for the P₁ shell alone) were found between the matrices of the evaporated ytterbium layer on the aluminum foil and the bulk of the high purity polycrystalline platinum. The thulium electron binding energies in the matrices of the evaporated ytterbium layers on both the platinum and carbon foils and in the aluminum bulk were observed to be the same within the experimental uncertainties. The N₁, N₃, and O_{2,3} electron binding energies most frequently presented in data compilations were found to be higher by about 3 eV. Natural widths of most of the K, L₁, L₂, L₃, M₁, M₂, M₃, N₁, N₃, and O₁ subshells in Tm in the investigated matrices were also determined. No significant differences in the natural widths were found among the matrices. The results obtained demonstrate that the physicochemical surrounding of the radioactive atoms should be well defined and understandable for any type of electron calibration source particularly in the case of the super stable calibration ⁸³Rb/^{83m}Kr electron sources for the KATRIN neutrino mass experiment.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

It is well known that the exact value of the binding energy of an electron depends not only on the atomic level, but also on the oxidation state of the atom and the local chemical and physical atomic environment. Induced changes in the electron binding energies (so-called chemical shifts) were both extensively and intensively studied in the past by the X-ray-absorption method and particularly by the X-ray photo-electron spectroscopy (XPS), the precision of which is considerably higher. The discovery of the chemical shifts in photo-electron spectra and their interpretation led to the invention

of the ESCA method (electron spectroscopy for chemical analysis; see, e.g., Ref. [1]) – a non-destructive technique well-suited for investigation of the electronic properties of atoms, molecules and solids and for elemental and chemical analysis of surfaces. The ability to discriminate between different oxidation states and chemical environments is one of the major strengths of the ESCA technique. However, despite the extensive ESCA investigations of chemical shifts in electron binding energies, there is a scarcity of data for deep-lying core electrons. The investigation was mainly concentrated on outer electrons also due to inherent technical limitations in the ESCA method and equipments used.

The “Internal conversion electron spectroscopy” (ICES) [2] can in principle be applied for the same purposes as the ESCA method but exhibits some considerable advantages in this field such as: (i) natural line widths Γ_γ of the “exiting gammas” are extremely small (typically 10⁻⁸–10⁻⁴ eV for low-lying excited nuclear states) thus no monochromators are needed (it should be, however, emphasized that internal conversion electrons result from a direct

* Corresponding author at: Laboratory of Nuclear Problems, JINR, Dubna, Moscow Region, Russian Federation. Tel.: +7 4962163956; fax: +7 4962165805.

E-mail address: inoyatov@jinr.ru (A.Kh. Inoyatov).

¹ Present address: Physikalisch-Technische Bundesanstalt, Bundesallee 100, D-38116 Braunschweig, Germany.

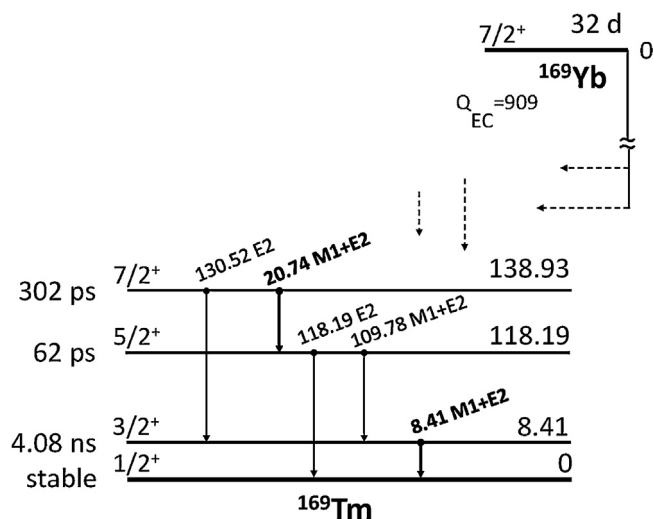


Fig. 1. The incomplete decay scheme of the ^{169}Yb radioisotope [7].

electro-magnetic interaction of an excited nucleus with its atomic clouds (see, e.g., Ref. [3]), i.e., there is no “internal photo effect” of de-exciting gamma-rays with atomic electrons); (ii) choosing a nuclear transition of suitable energy and multipolarity, outermost or deep-lying core electrons with a preferable orbital angular momentum can be investigated; (iii) one can investigate physico-chemical atomic environment (and also a dynamics of its change) at the surface or in the bulk by incorporation there suitable radioactive atoms (radioisotopic labeling, “marker”); (iv) very high sensitivity (relative – in the ppm range, absolute – in the nanogram range) as the investigated radioactive atoms emit electrons due to their intrinsic nuclear-atomic process, i.e. without any external excitation; (v) no sample degradation occurs by heat effects caused by Bremsstrahlung X-rays and/or an anode of the X-ray tube; (vi) a high signal-to-noise ratio; (vii) a wide electron spectrometer energy range (from “0” to 50000 eV in the case of our combined electrostatic electron spectrometer [4]) enabling one to study a large set of atomic levels at the same time; (viii) electrons are emitted only by atoms of a chosen radioactive isotope, etc. The ICES method has, naturally, also its own disadvantages and limitations. They are, first of all, limited choice of suitable radioactive isotopes, complex decay schemes of some of them and the necessity to work with unsealed radioactive materials.

Recently, the ICES method was successfully applied in Ref. [5] to the investigation of atomic environmental effects on electron binding energies, the absolute KLL Auger transition energies, and natural atomic level widths in krypton generated in two different solid hosts and in Ref. [6] on the KLL Auger spectrum of rubidium (energies, relative intensities, and natural widths of all the nine well-resolved basic spectrum components) following the EC decay of the ^{83}Sr and ^{85}Sr isotopes incorporated in different matrices by vacuum evaporation and ion implantation at 30 keV.

In this work we present results of our experimental investigation of the influence of physicochemical surrounding on both the electron binding energies and natural atomic level widths in thulium generated by the electron capture (EC) decay of ^{169}Yb ($T_{1/2} = 32.0$ days, Fig. 1) incorporated into five different matrices. The ^{169}Yb sources used were prepared by vacuum evaporation on three different backings, namely polycrystalline carbon, high purity polycrystalline platinum, and common polycrystalline aluminum and by implantation of ^{169}Yb ions at 30 keV into the mentioned platinum and aluminum materials. Our investigations were performed in the frame of the development of a new technique for the preparation of super stable calibration $^{83}\text{Rb}/^{83\text{m}}\text{Kr}$ electron sources

[8,9] for the KATRIN neutrino mass experiment [10]. The aim was to study influence of atomic environment on energies of the selected conversion electrons emitted in the ^{169}Yb decay and the electron energy stability in time.

2. Experimental

2.1. Source preparation

The ^{169}Yb isotope was obtained by the spallation reaction of metallic tantalum bombarded by 660 MeV protons from the inner beam of the phasotron particle accelerator at the JINR, Dubna, Russia. After “cooling” for 30 days, the irradiated target (4.5 g weight) was dissolved in the concentrated fluoric acid with the addition of nitric acid. The fraction of ytterbium together with other radioactive lanthanides was separated from the rest target material by the co-precipitation with lanthanum fluoride (2 mg). The subsequent separation of the lanthanides was carried out on a cation-exchange chromatography column (length – 100 mm, 2 mm diameter, A5 resin in the NH_4 form). A preparation obtained was put on the column with a solution of the chloric acid saturated by the boric acid. Then the column was washed with 0.5 M NH_4Cl solution and water. The separation was performed by elution using a water solution of ammonium alpha-hydroxyisobutyrate (alpha-HIB) with consequent increasing of its concentration. The ytterbium fraction gained (consisting of several drops of 0.07 M alpha-HIB) was then used for the two aforementioned methods of electron source preparation, namely mass separation and thermal vacuum evaporation deposition.

2.1.1. Thermal vacuum evaporation deposition

As stated above, three different source backings were used for vacuum evaporation deposition: (i) a polycrystalline carbon foil (thickness – 150 μm , purity – 99.9%); (ii) a high purity polycrystalline platinum foil (thickness – 50 μm , purity – 99.95%); (iii) a common polycrystalline aluminum foil of 200 μm thickness with naturally oxidized surface. Prior to use, surfaces of the foils were mechanically cleansed with application of high purity alcohol.

In all three cases, the evaporation procedure started by transfer of several drops of ytterbium fraction to a Ta evaporation boat annealed at about 1300 $^\circ\text{C}$ and their drying up. To remove possible volatile organic compounds, the Ta evaporation boat with deposited activity was first preheated at 700 $^\circ\text{C}$ for about 60 s. The source backing was shielded throughout this procedure. The source evaporation through an 8 mm diameter circular opening of a mask took place at 1200 $^\circ\text{C}$ for several seconds. During the evaporation, the source backing with the mask rotated around their common axis at a speed of 3000 turns/min at a distance of about 6 mm from the Ta evaporation boat to improve homogeneity of the evaporated layer. No visible effects were observed on the surfaces of the source backings after evaporation. Activity of the ^{169}Yb isotope upon preparation in the sources on the carbon, platinum, and aluminum foils were 5.2, 1.7, and 0.9 MBq, respectively. In all three sources, admixtures of ^{171}Lu ($T_{1/2} = 8.24$ days) with activity of 0.8 (C), 1.0 (Pt), and 0.4 (Al) kBq were detected.

The exact chemical state of the deposited trace amounts of Yb on the surfaces of the source backings in vacuum was unknown. It was most likely Yb_2O_3 or chemical compositions such as $\text{Yb}(\text{HCO}_3)_3$, $\text{Yb}_2(\text{CO}_3)_3$, and $\text{Yb}(\text{OH})_3$. However, the sources were transferred to the electron spectrometer in air after the preparation and thus the evaporated layers were exposed to it. Consequently, ions of Yb were bound with atoms of oxygen in all possible forms (oxides, hydroxides, carbonates, hydrocarbonates, etc. of different proportions) and had the oxidation number +3. This statement is based on: (i) specific chemical properties of ytterbium, (ii) its known

Download English Version:

<https://daneshyari.com/en/article/5395696>

Download Persian Version:

<https://daneshyari.com/article/5395696>

[Daneshyari.com](https://daneshyari.com)