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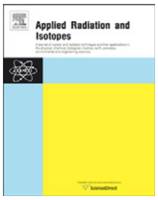

Activity determination and nuclear decay data of ^{113m}Cd

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ABSTRACT

The radionuclide ^{113m}Cd was extracted from a Cd component, which has been irradiated in a research reactor during its routine operation. A validated separation procedure was then used to obtain a ^{113m}Cd solution of high radiochemical purity. The solution was measured by means of liquid scintillation counting for activity determination. To this end, the CIEMAT/NIST efficiency tracing method as well as the triple-to-double coincidence (TDCR) counting technique were used. Measurements in a low-background gamma-ray spectrometer were applied to identify any potential photon-emitting impurities and to measure the 263.7 keV photons from the isomer transition. A gamma emission probability of 0.01839(29)% was determined for this line. The liquid scintillation measurements were also used to evaluate the half-life of ^{113m}Cd. The decay was followed for about 11 months and a half-life $T_{1/2}$=13.97(13) years was obtained.

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1. Introduction

^{113m}Cd is mainly created by neutron activation in control rods and shielding of nuclear reactors. Due to its half-life of about 14 years it must be taken into account when considering nuclear facilities decommissioning and disposal of nuclear waste. The nuclide has also been detected in environmental samples as a consequence of nuclear weapons testing (see, e.g. Dunn and Tisue, 1989 and references therein). However, techniques for accurate activity measurements of ^{113m}Cd were not well established before 2008 and nuclear decay data have only been measured in a limited number of studies.

^{113m}Cd predominantly disintegrates via beta decay, but knowledge of the decay scheme parameters is scarce. The beta transition is of the first-forbidden, non-unique type with an end-point energy of 580(4) keV and a probability of about 99.86% to the ground state of ^{113}Sn (Blachot, 2005). The isomer transition to the ^{113}Cd ground state has an energy of 263.7 keV and a probability of 0.14%. A theoretical total internal conversion coefficient for the E5 transition is $\alpha=5.1$, i.e. in most cases conversion electrons are ejected and the gamma emission probability is only 2.3×10^{-4} per disintegration of ^{113m}Cd. This makes quantitative activity measurements by means of gamma-ray spectrometry difficult.

In this work liquid scintillation (LS) counting is used to determine the activity concentration of a ^{113m}Cd solution. Liquid scintillation counting in combination with the CIEMAT/NIST efficiency tracing technique was already successfully applied in a recent work (Warwick and Croudace, 2009). A deeper investigation of that method is presented here. Moreover, the triple-to-double coincidence ratio (TDCR) method was used. The results of both methods agree well and make the determination of the activity concentration with low uncertainty possible.

Aliquots of the solution were also measured by means of a low-background gamma-ray spectrometer, which was calibrated with activity standards from PTB.

A combination of the results of liquid scintillation counting and gamma-ray spectrometry yield a new accurate value of the gamma emission probability of the 263.7 keV line.

Finally, the LS measurement data were used to determine the half-life of ^{113m}Cd.

2. Methodology

2.1. Preparation of the solution

A purified solution of ^{113m}Cd was prepared from irradiated Cd metal using anion exchange chromatography. Approximately 0.6 g of irradiated Cd was dissolved in HCl and the solution evaporated to dryness. The residue was dissolved in 10 mL of 3 M HCl and the solution transferred to a 5 × 2 cm (i.d.) Eichrom 1 × 8 anion exchange column previously conditioned with 3 M HCl. The column was washed with 2 × 2.5 mL 3 M HCl rinsings followed by an additional 4 × 20 mL 3 M HCl. Cd was eluted with 70 mL 3 M HNO_3. (Warwick and Croudace, 2009). The solution was evaporated

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to dryness and the residue dissolved in 100 μL conc. HCl. The final solution had a hydrochloric acid concentration of approximately 0.1 mol/L and the Cd^{2+} concentration was estimated to be about 28.4 mg per gram of solution. A nominal activity concentration of the solution of about 220 Bq/g (reference date—1 January 2010) was determined by means of gamma-ray spectrometry measurements using a gamma emission probability of 0.023%. About 20 g of the solution were shipped to the PTB for further measurements.

2.2. *Gamma-ray spectrometry*

A weighed aliquot of 10.9378(10) g of the ^{113m}Cd solution was transferred into a 20 mL polyethylene vial. A small amount of hydrochloric acid was added to obtain a filling volume of 12 mL and a total HCl concentration of 1 mol/L. The vial was used for Čerenkov measurements in commercial liquid scintillation counters, but the experimental counting efficiency was found to be lower than 1%, which is too low for reliable application.

The vial was then measured by means of a low-background gamma-ray spectrometer. The spectrometer is normally used for certifications of environmental reference materials. The detector is of the coaxial type with a relative efficiency of 50%. The energy resolution is 1.0 keV at 122 keV and 1.9 keV at 1.33 MeV. The peak to Compton ratio is 60:1. Nitrogen gas evaporating from the Dewar vessel is directed via silicon rubber hoses into the lead shielding in order to reduce radon and radon daughter-nuclide contributions to the background spectrum. The shielding consists of 20 cm thick walls made of low-level lead with a specific activity of less than 2 mBq/g. The inner volume of the lead shield has the dimensions of 20 cm × 20 cm × 25 cm (l × w × h) allowing for measurements of environmental samples contained in an 1 L Marinelli beaker. Around the detector end cap there is copper shielding with a wall thickness of 7 mm in order to reduce the influence of Pb X-ray fluorescence photons and serving as the support for Marinelli beakers. The top of the copper shielding is formed by a circular copper disc of 5 mm thickness.

A special sample holder was made of acrylic glass in order to reproducibly position the background vial, the calibration vials and the ^{113m}Cd vial. The acrylic glass disc itself was centred by an edge fixing it on the outer diameter of the detector end cap.

The system was calibrated by measuring three samples in the same geometry. The first sample contained weighed portions of standardised ^{57}Co, ^{109}Cd and ^{241}Am solutions. Another sample contained ^{54}Mn, ^{51}Cr and ^{137}Cs. These samples also had a total volume of 12 mL in 20 mL polyethylene vials. The final HCl concentration was 1 mol/L. A third sample was prepared with ^{210}Pb. In this case, the total volume was also 12 mL, but nitric acid was used to achieve high sample stability. The acid concentration was 0.5 mol/L in order to obtain a comparable density with samples in hydrochloric acid. All radioactive solutions are standard solutions from PTB and the corresponding activities can be traced back to primary activity standardization. Corrections for coincidence summing effects were not required for establishing the efficiency curve of the detector. The best fit of the curve was represented by a polynomial of the 5th order. The closest neighbouring lines (the 136.5 keV line of ^{57}Co and the 320.1 keV line of ^{51}Cr) were used as supporting points. For the evaluation of the ^{113m}Cd data, an efficiency value of (8.67 ± 0.09) % at the photon energy of 263.7 keV was used.

Data acquisition was performed by use of analogue electronics consisting of a high voltage supply (Mod. 3106D, Canberra), a spectroscopy amplifier (Mod. 2024, Canberra) and a 6-port multichannel analyser (Mod. Multiport II, Canberra). The spectrometer is connected via the local area network with a personal computer located in a separate operating room. The software package WINNER6 (version 6.00.2067, Canberra) was used for both the spectrometer operation and spectra evaluation.

The ^{113m}Cd sample was measured for 15 days. A dead time correction is negligible due to the low total counting rate of about 2.682 s^{-1}. ^{60}Co and ^{85}Sr were identified as photon-emitting impurities and their activity ratios at the reference date were found to be $A(^{60}\mathrm{Co})/A(^{113m}\mathrm{Cd}) \approx 3 \times 10^{-6}$ and $A(^{85}\mathrm{Sr})/A(^{113m}\mathrm{Cd}) < 10^{-6}$, respectively. A measured spectrum is shown in Fig. 1. It also contains other well-known background peaks originating both from naturally occurring radionuclides and from cosmic ray interaction with detector materials.

The photon emission rate divided by mass of solution n_γ=5.2189(79) $10^{-2}\ s^{-1}\ g^{-1}$ at the reference date (1 January 2010) was determined for the 263.7 keV gamma-rays. The relative combined standard measurement uncertainty of n_γ is 1.51%. The uncertainty budget is listed in Table 1. Dominant contributions are assigned to the detector efficiency determination and to the production tolerance of the set of LS vials used. Potentially differing thicknesses and/or shapes of the bottom walls of the LS vials might cause small differences in the distance between the detector end cap and the bottom of the solution in the vials used for background, calibration or the ^{113m}Cd measurements. As the vials are mass produced, this uncertainty component was estimated as 1%. Also, the precise inner diameters of the individual vials could not be determined. Therefore, a contribution of 0.4% was taken into account in order to cover potential non-uniformities in the shape of the radioactive solution masses.

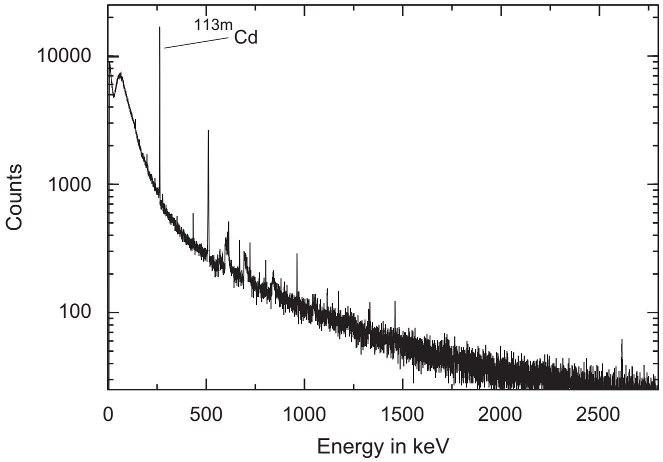

Fig. 1. Gamma-ray spectrum of a ^{113m}Cd source measured in a low-background HPGe spectrometer for about 15 days. In addition to the 263.7 keV peak, the spectrum contains some peaks assigned to the photon-emitting impurities ^{60}Co and ^{85}Sr (not visible, very close to the 511 keV annihilation peak) as well as some known background peaks.

Table 1

Standard uncertainty components for the 263.7 keV photon emission rate divided by mass of solution n_γ of a ^{113m}Cd solution measured by means of a low-background high-purity Ge spectrometer.

Component	$\boldsymbol{u(n_\gamma)/n_\gamma}$ **in %**
Net count number	0.39
Peak fit & background subtraction	0.10
Efficiency	0.98
Uniformity between the ^{113m}Cd source and the efficiency calibration sources	0.40
Dead time	0.03
Active volume, in an individual vial	0.05
Production tolerance between the vials	1.00
Square root of the sum of quadratic components	**1.51**

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