Project: EMPIR 20FUN04 PrimA-LTD

Deliverable no: D4

Due date: 31 May 2024

Actual delivery date: 31 May 2024

Lead partner: PTB

Related activity: A4.3.6

Deliverable description

Summary report of five open access papers, based on experimental results, submitted to peer review journals describing

a) the MMC spectrometer extension for multiple channel measurements >10^8 events per spectrum – obj. 1

b) the validation of MMC activity standardisation of ^{241}Am – obj. 2

c) the MMC source preparation with ion-implantation – obj. 2

d) the high-resolution, high statistics measurement of the ^{55}Fe EC spectrum a lower energy threshold (< 50 eV) – obj. 3

e) the high-resolution, high statistics measurement of the ^{129}I beta spectrum to determine the beta spectrum shape of ^{129}I down to 0 keV – obj. 3

The PrimA-LTD consortium

This project has received funding from the EMPIR programme co-financed by the Participating States and from the European Union's Horizon 2020 research and innovation programme.

The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

Preface

During the project the consortium experienced several delays due to unforeseen problems during MMC production and source preparation. In addition, long lead times hindered the full setup of the multichannel readout even until the end of project. Although a mitigation plan was developed early in 2023 by the consortium, it became clear during the M27 meeting that within the Deliverable 4

> *“The scope of the five publications may not be fully met. Instead, it might become necessary to publish results obtained within the project with a different focus.”*

This deviation from Annex 1 was communicated to MSU during the reporting, correspondingly.

Since the performance of the full spectrometer setups at CEA and PTB could not be proven and since source preparation turned out to be more complex and experimentally demanding instead of a paper describing

> *“a) the MMC spectrometer extension for multiple channel measurements >10^8 events per spectrum -obj. 1”*

a paper on MMC development and an additional paper on gold nanofoams embedded in absorbers were published.

The paper about

> *“b) the validation of MMC activity standardisation of ^{241}Am – obj. 2”*

has been drafted, however some results need further verification and validation. The submission of this paper is foreseen in the next months.

Since sources of ^{55}Fe and ^{129}I became available only in the last months of the project, a publication of

> *“d) the high-resolution, high statistics measurement of the ^{55}Fe EC spectrum a lower energy threshold (< 50 eV) – obj. 3”*

and

> *“e) the high-resolution, high statistics measurement of the ^{129}I beta spectrum to determine the beta spectrum shape of ^{129}I down to 0 keV – obj. 3”*

turned out to be unfeasible. Preliminary results of measurements on ^{55}Fe sources and ^{129}I sources have, however, been obtained and are promising as documented in Deliverable 5. With the analysis procedures for experimental data that were developed early in the project, existing data were analyzed and linked to theory. The results for ^{99}Tc and ^{151}Sm were published and are listed here.

To summarize, six experimental papers have been written by the consortium. Of these papers three are published, two have been submitted and one is drafted.

Publication (submitted)

Authors: Thorben Niemeyer, Daniel Mowitz, Sebastian Berndt, Jörn Beyer, Holger Dorrer, Christoph E. Düllmann, Alexander Göggelmann, Raphael Hasse, Sebastian Kempf, Tom Kieck, Nina Kneip, Karsten Kossert, Andrea T. Loria Basto, Christoph Mokry, Michael Müller, Ole J. Nähle, Dennis Renisch, Jörg Runke, Dominik Studer, Marcell P. Takács, Klaus Wendt

Title: **Ion implantation of the electron-capture nuclide ^{55}Fe for measurements by means of metallic microcalorimeters**

Reference: Submitted to Applied Radiation and Isotopes by May 31st, 2024

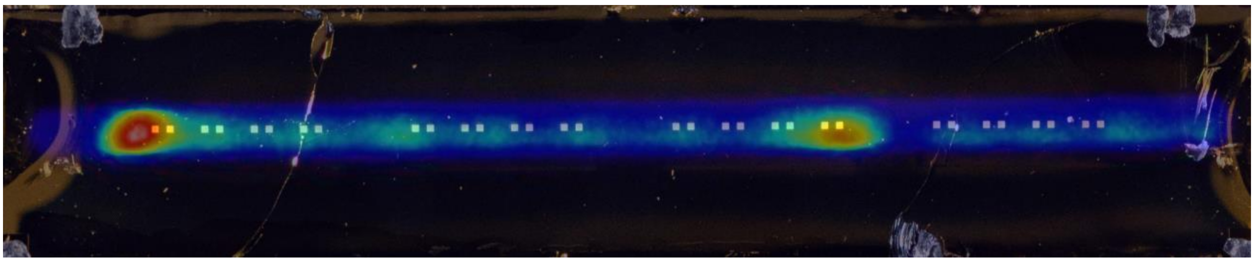
Open Access: Yes

Project partners: JGU, PTB, KIT

Summary

Precise measurements of fundamental decay data such as energies and transition probabilities of radioactive isotopes are important for the development of corresponding nuclear modelling, activity determination and various applications in science and technology. The EMPIR project PrimA-LTD aims to measure the electron-capture decay of ^{55}Fe very precisely using Metallic Microcalorimeters (MMCs) with outstandingly high energy resolution. Using a high-statistics measurement, electron-capture probabilities shall be precisely determined and higher-order effects such as electron shake-up and shake-off shall be examined with unprecedented precision. A key to success for this experiment is sample preparation. This work reports on the implantation of the ^{55}Fe nuclei into the 140 µm × 140 µm gold absorbers of the MMCs as a proof of principle for scalability. Based on preparative laser-spectroscopic studies on stable ^{56}Fe, laser resonance ionization at the RISIKO mass separator was used to produce a monoisotopic ^{55}Fe ion beam with the required high spatial characteristics. Successful implantations of this isotope into 32 test absorbers with about 0.7(2) Bq each and subsequently various on-chip absorbers with an activity close to the requested 5 Bq per absorber are presented. The impact on the quality of spectra is demonstrated on the basis of first MMC test measurements.

The implantation of the radioisotope ^{55}Fe into the MMC chips has successfully been carried out at the RISIKO laser mass spectrometry facility of Mainz University. Four chips with activity levels close to the target value of 5 Bq per detector pixel were prepared. One test chip with 0.7 Bq per pixel was investigated by autoradiography proving the outcome and quality of the implantation process. Preparatory measurements on the resonant laser ionization process have provided deeper insights into the atomic spectrum of iron and delivered necessary information for the work. The atomization process of the sample could be optimized regarding overall efficiency, purity of the ion beam and spatial quality of the beam to permit for this demanding application. The implanted ^{55}Fe sources are undergoing tests and calibration measurements within the labs of the EMPIR PrimA-LTD consortium. First spectra obtained from these sources have already shown significant improvements with regards to spectral distortions compared to source preparation by drop deposition.



Overlay of an optical image of an ^{55}Fe implanted 32-pixel MMC detector chip with the activity distribution visualized by autoradiography.

Publication (published)

Authors: Jawad Hadid, Matias Rodrigues, Abdelmounaim Harouri, Christophe Dupuis, David Bouville, Antoine Martin, Martin Loidl, Laurence Ferlazzo

Title: **Detachable three-layer Au absorber microfabrication for low-temperature detectors**

Reference: Micro and Nano Engineering 20 (2024) 100220
https://doi.org/10.1016/j.mne.2023.100220

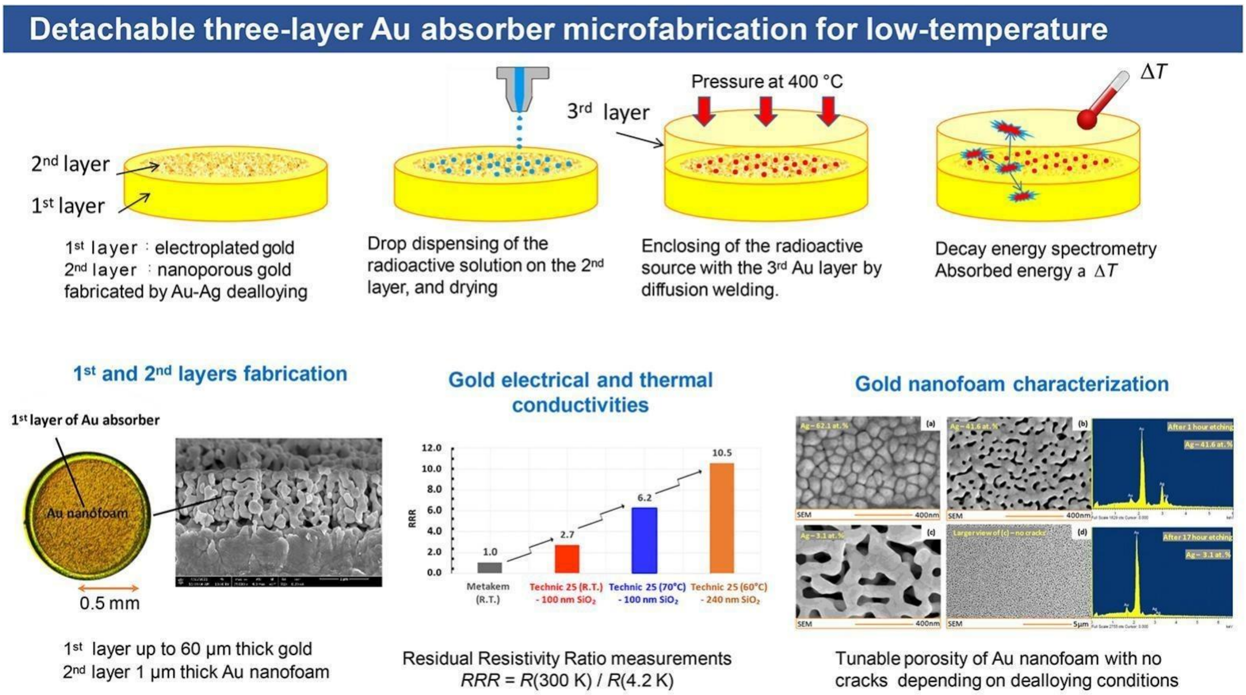
Open Access: Yes

Project partners: CNRS, CEA

Summary

Low temperature detectors (LTDs) used for decay energy spectrometry (DES) can provide accurate and reliable decay data thanks to their high-energy resolution and a near 100% detection efficiency for the radiations of interest. However, it is essential to consider the source quality to mitigate spectral distortion due to the self-absorption of particle energy in the source deposited.

This work aimed to produce a replaceable 4π 3-layer gold absorber for DES in reusable metallic microcalorimeters, a class of LTDs. We present a novel 3-layer microfabrication process for a 1 mm diameter absorber with a total gold thickness ranging from 20 µm to 120 µm depending on the measured radionuclide (^{55}Fe or ^{241}Am). The absorber integrates a gold nanofoam in which the radionuclide is deposited by nanodrop deposition of a few tenths of µL of a radioactive solution. We fabricated a high-quality gold nanofoam layer with controllable porosity through a dealloying process using wet etching and integrating it on a thick electrodeposited gold layer. The fine study of the nanofoam microfabrication is performed using high-resolution scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX).



This work introduces a novel microfabrication process forming three Au layers, which integrates the middle Au nanofoam between the top and bottom absorber layers. This 3-layer Au structure can be easily thermally connected to the sensor, enabling the reusability of MMC. The authors have fabricated the top and bottom layers using electrolytic techniques, thereby reducing the typical imperfections observed when machining noble foils. In addition, they have successfully implemented a gold nanofoam on the bottom Au layer, which exhibits tunable porosity. This feature enables to minimize the thickness of the source layer and therefore reduce the absorption of particles energy in the deposit which is responsible for the spectrum distortion. Structural and chemical characterizations of the microfabricated nanofoam have been performed using high-resolution scanning electron microscopy (SEM) and energy dispersive x-ray spectroscopy (EDX).

Publication (published)

Authors: Michael Müller, Matias Rodrigues, Jörn Beyer, Martin Loidl, Sebastian Kempf

Title: **Magnetic Microcalorimeters for Primary Activity Standardization Within the EMPIR Project PrimA-LTD**

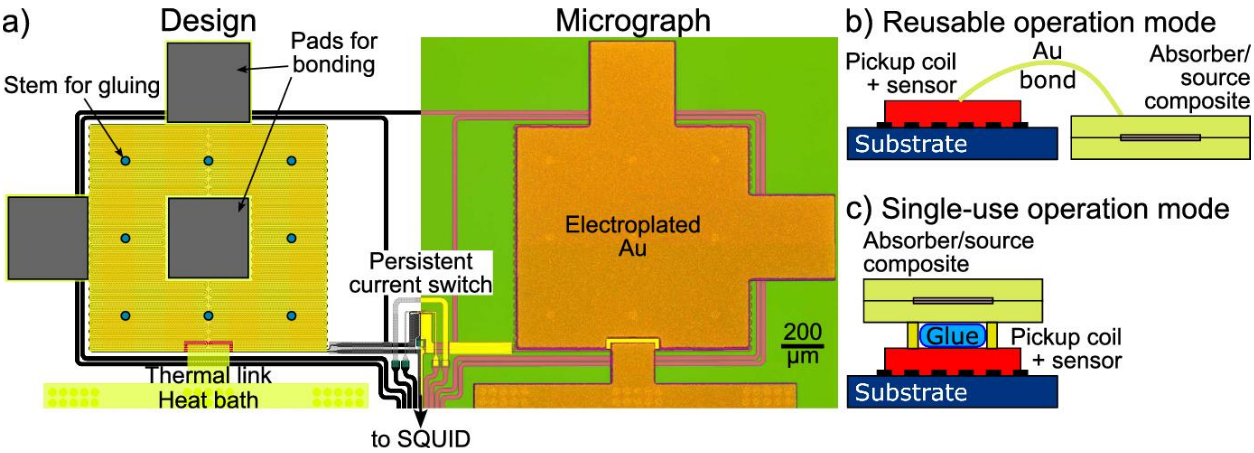
Reference: Journal of Low Temperature Physics 214 (2024) 263-272
https://doi.org/10.1007/s10909-024-03048-7

Open Access: Yes

Project partners: KIT, CEA, PTB

Summary

The precision of existing decay data of radionuclides for activity determination is often a limitation for actual applications in science, society and industry. For this reason, the EMPIR project PrimA-LTD aims to introduce an advanced primary activity standardization technique that is based on magnetic microcalorimeters (MMCs) and that will offer very low energy threshold of few eV and a decay scheme-independent detection efficiency close to 100%. As a proof of concept, we developed two MMC-based detector types in order to standardize an α-decaying, a β-decaying and an electron capture decaying isotope. One detector type aims to introduce a reusable detector setup, while the other aims to provide highly accurate decay spectra by high-resolution measurements with high statistics. The designs, fabrication status and first characterization measurements of both detector types and an outline of the next steps are presented.



Overview of the RoS-L detector showing the design layout of the left pixel and a micrograph of the right pixel. The area covered by sensor material is indicated in yellow in the design layout. Moreover, b and c show the two methods for coupling of an absorber/source composite to the detector.

Magnetic microcalorimeters (MMCs) are an ideal detector technology for determining decay data of radionuclides thanks to their excellent energy resolution and high detection efficiency. For this reason, the EMPIR project PrimA-LTD aims to demonstrate MMC spectrometry as a novel method for primary activity standardization. As a proof of concept, we developed two types of MMC-based detectors that enhance the flexibility and accuracy of decay measurements within radionuclide metrology. The RoS detectors will improve the flexibility and throughput of activity measurements by introducing a reusable detector setup. The implanted Fe detector will improve the accuracy of decay spectra by introducing ion implantation into microfabricated absorbers as a new source preparation technique in the context of radionuclide metrology. Basic functionality of both detector types has been verified by first evaluation measurements using X-rays from external Fe and Kr sources and the 2-step electroplating of the microfabricated absorbers for the implanted Fe detector has been successfully demonstrated. Next steps are the comparison of the RoS detector performances between glued and wire-bonded absorber foils as well as the completion of the implanted Fe detector fabrication after ion implantation.

Publication (drafted)

Authors: Miguel Roteta et al.

Title: **Comparison of activity standardisation of ^{241}Am using metallic microcalorimeters and established techniques**

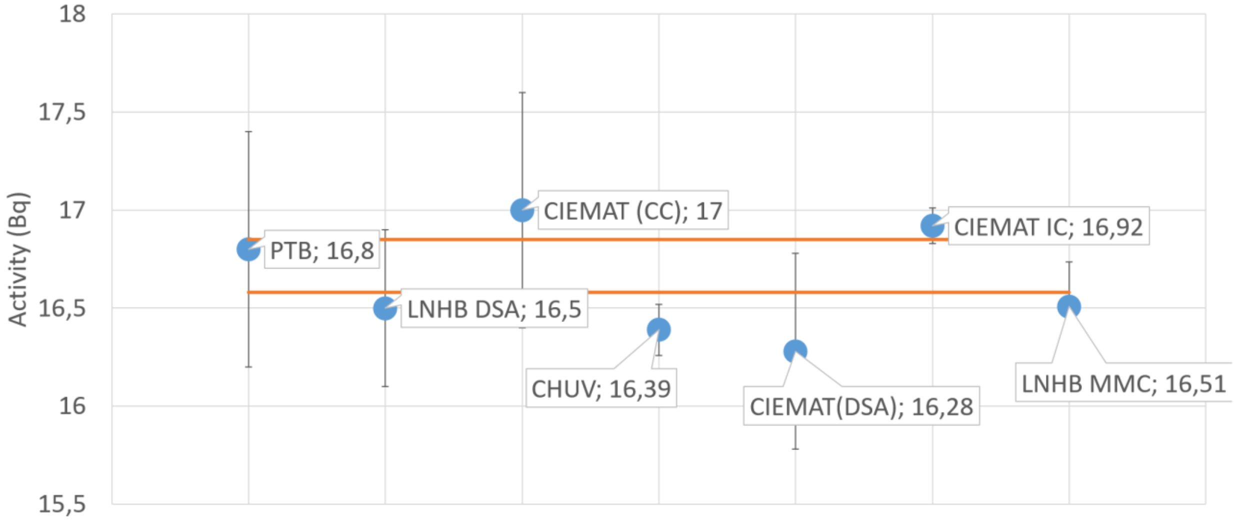
Reference: Drafted. To be submitted to Applied Radiation and Isotopes by Aug 31th, 2024

Open Access: Yes

Project partners: CIEMAT, CEA, CHUV, PTB

Summary

Within the framework of the PrimA-LTD project, radioactive samples have been produced by electroplating on gold foils applying a masking technique. Using this approach, more than 10 radioactive localized sources, compatible in size with magnetic microcalorimeter detectors (MMCs), were prepared on gold foils as carriers and subsequently measured using different established techniques for absolute activity determination. The samples were sent to four NMIs and three different standardization techniques were successfully applied: high pressure ionization chamber with defined solid angle, defined solid angle and alpha-gamma coincidence counting. The individual results are consistent and were used to calculate a reference value for this informal intercomparison. Finally, the gold carrier foils were segmented into individual sources that were enclosed in gold absorbers compatible with MMCs. Residual activity on the carrier foils inbetween the individual sources, however, introduces a significant uncertainty component, Despite this issue, overall agreement between the activity obtained by the established techniques and the MMC measurement is satisfactory.



Preliminary results of the informal intercomparison, taking a correction for the activity loss from source segmentation into account.

Publication (published)

Authors: Karsten Kossert, Martin Loidl, Xavier Mougeot, Michael Paulsen, Philipp Ranitzsch, Matias Rodrigues

Title: **High precision measurement of the ^{151}Sm beta decay by means of a metallic magnetic calorimeter**

Reference: Applied Radiation and Isotopes 185 (2022) 110237
https://doi.org/10.1016/j.apradiso.2022.110237

Open Access: Yes

Project partners: PTB, CEA

Summary

The beta decay of ^{151}Sm was measured by means of a metallic magnetic calorimeter. The source used for the MMC measurement was prepared from a purified ^{151}Sm solution, with a small amount of impurities of ^{154}Eu and ^{155}Eu. Although both are beta emitters, none of the X-ray or gamma-ray lines of either of these nuclides, or any other impurities, were detectable in the MMC spectrum. We thus considered that their small activities did not induce a noticeable distortion of the measured ^{151}Sm spectrum. The initial source was prepared by electrodeposition of ^{151}Sm on a silver foil, which was then repeatedly folded, laminated and annealed to break the samarium oxide/hydroxide layer formed during the electrodeposition process into small particles and incorporate them in the silver volume. The resulting silver foil had a thickness of about 7 μm and was placed between two other silver foils, each 15 μm thick, to ensure absorption of the full beta electron energy. These foils were then bonded together by diffusion welding and the final absorber, with a heat capacity of 18 pJ/K at 10 mK, was then fixed onto an MMC. The MMC signal was read out by a dc SQUID and a Magnicon XXF-1 SQUID electronics; a Stanford Research Systems model SR560 low noise amplifier was used to further amplify the signal and to set an anti-aliasing filter. An external photon-emitting source with about 9.9 kBq of ^{55}Fe and about 260 Bq of ^{109}Cd was used for energy calibration. The MMC was enclosed together with the SQUID and the collimated calibration source in a superconducting lead cylinder for magnetic shielding and mounted in a dilution refrigerator having a base temperature of about 10 mK. The measurement and subsequent analysis yielded a beta spectrum with an outstanding high-energy resolution of about 70 eV (FWHM) at 22 keV and a very low energy threshold well below 400 eV.

The measured spectrum was compared to predictions from an advanced theoretical modeling that includes the atomic exchange effect, precise radiative corrections as well as the realistic nuclear structure that usually plays an important role in first forbidden non-unique transitions. The two first forbidden non-unique beta transitions in ^{151}Sm were studied theoretically. The NushellX code was employed to determine the one-body transition densities (OBTD) involved, selecting the *jj56pn* valence space and the recommended *khhe* interaction. Above the doubly-magic ^{132}Sn core, 12 (13) protons (π) can be distributed in the valence space spanning $1g_{7/2}$ to $1h_{11/2}$ and 7 (6) neutrons (ν) can be distributed in the valence space spanning $1h_{9/2}$ to $1i_{13/2}$ for ^{151}Sm and ^{151}Eu, respectively. The number of possible configurations is so high that the valence space must be constrained in order to limit the computational burden. This was done by filling the lowest orbitals and blocking the highest as: full π $1g_{7/2}$; minimum 2 in π $2d_{5/2}$; minimum 4 in ν $1h_{9/2}$; empty ν $3p_{1/2}$ and ν $1i_{13/2}$. The energy of the first excited states in ^{151}Eu was found to be 98.1 keV, compared to the experimental value of 21.541 keV. As the beta transition from this state is small, this nuclear description is considered sufficiently accurate. Theoretical spectrum shapes were calculated with various hypotheses. As the available energy for the transitions is very low, it has been assumed for decades that this is a case study of the ξ-approximation, which tells that these transitions can be safely calculated as allowed. In this work, we quantified this approximation, demonstrating that the best theoretical description leads to a correction of 2% for the dominant transition and 4% for the second one, which definitely rules out the ξ-approximation in metrological studies.

The measured spectrum was then carefully analyzed to determine the maximum beta energy, which was found to be Q = 76.430(68) keV, much more precise than the recommended value. The dominant beta decay of ^{151}Sm populates the ground state of ^{151}Eu, and a weak beta branch populates the first excited state of ^{151}Eu. From our measurements, the probabilities of these two branches were determined to be 99.31(11)% and 0.69(11)%, respectively. The spectrum exhibited unexpectedly elevated beta emission probabilities at very low energy, below 4 keV, which we were unable to reproduce in our theoretical study.

The data analysis was thus scrutinized and an independent analysis of the same data set carried out. All new approaches have confirmed the previously found shape of the beta spectrum, and this discrepancy still remains unclear.

The analysis presented in this paper showed that MMC measurements can be used to determine maximum beta energies with very high precision, competitive with Penning trap measurements. In addition, beta decay probabilities of two different branches could be determined, though in the presented case the corresponding uncertainties are higher than those obtained when using gamma-ray spectrometry in combination with an internal conversion coefficient. To conclude, this article clearly demonstrated that the experimental data and the theoretical calculations do agree over a wide energy range. However, significant discrepancies are still observed at very low energy, even if this did not affect the determination of the Q-value and the branching ratios.

Publication (submitted)

Authors: M. Paulsen, P. C.-O. Ranitzsch, M. Loidl, M. Rodrigues, K. Kossert, X. Mougeot, A. Singh, S. Leblond, J. Beyer, L. Bockhorn, C. Enss, M. Wegner, S. Kempf, and O. Nähle

Title: **High precision measurement of the ^{99}Tc β spectrum**

Reference: Submitted to Physical Review C

Open Access: https://arxiv.org/abs/2309.14014

Project partners: PTB, CEA, UHEI

Summary

This publication report on three independent measurements of the ^{99}Tc beta spectrum, which were next analysed to correct for small, but relevant distortions due to the detection systems. Two spectra measured with MMC at CEA and PTB were found to be extremely consistent, greatly improving energy resolution (0.1 keV at 383 keV) and energy thresholds (0.65 keV) compared to previous measurements. At energies above 25 keV, the spectrum shapes were further confirmed with a state-of-the-art silicon detector measurement, also performed at CEA.

For the MMC measurement at PTB, an aqueous solution of ammonium pertechnetate ($NH_4\,{}^{99}TcO_4$) in 0.1 mol L^{-1} ammonia (NH_3) with an activity of about 5 Bq was micro-dispensed onto a solid gold absorber substrate with a thickness of 90 µm. The source was then enclosed by diffusion welding a second 90 µm gold layer to the first one. The absorber was attached to one pixel of the MMC detector and a second pixel was equipped with a blank absorber prepared for background monitoring. Energy calibration was performed tanks to an external ^{57}Co source. The setup was completed with a PTB SQUID (model C6X114W) for MMC read-out and Magnicon XXF-1 SQUID electronics, cooled down at 14.5 mK, a full data stream at 200 kS s^{-1} was acquired to hard disc for a measurement's duration of about 20 days. For the MMC measurement at CEA, ^{99}Tc was electrodeposited onto a 10 µm thick gold foil, leading to an activity of about 5 Bq. The foil was then sandwiched between two gold foils (0.9mm×0.9mm×74 µm each) and this stack was diffusion welded. The final absorber had a heat capacity of about 350 pJ/K at 20mK. Energy calibration was performed thanks to an external ^{133}Ba source. The MMC signal was read out by a Supracon VC1A SQUID linked to a Magnicon XBXF-1 electronics. The whole setup was shielded against stray magnetic fields by means of a lead cylinder and cooled down to 12 mK. Data was acquired as a continuous stream over 13.7 days at 100 kS/s. For both measurements, data of each laboratory were treated blindly by the two laboratories with their own analysis codes. This procedure ensured the quality of the reconstructed spectra. Additional, small corrections were applied, in particular to remove the distortion due to the bremsstrahlung photon escape.

The second forbidden non-unique beta transition in ^{99}Tc was studied theoretically. The NushellX code [Bro14] was employed to determine the OBTD. We first considered the effective interaction from Gloeckner [Glo75] with the GL valence space spanning the proton orbitals $2p_{1/2}$ and $1g_{9/2}$, and the neutron orbitals $3s_{1/2}$ and $2d_{5/2}$. With such a description, the ^{99}Tc decay is driven by a single nucleon-nucleon transition, from a neutron in $2d_{5/2}$ to a proton in $1g_{9/2}$. Next, the effective interaction from Mach [Mac90] with the wider GLEKPN valence space was considered. This valence space spans the proton orbitals $1f_{7/2}$, $1f_{5/2}$, $2p_{3/2}$, $2p_{1/2}$ and $1g_{9/2}$, and the neutron orbitals $1g_{9/2}$, $1g_{7/2}$, $2d_{5/2}$, $2d_{3/2}$ and $3s_{1/2}$. To limit the computational burden, the $1f_{7/2}$ proton and the $1g_{9/2}$ neutron orbitals were constrained to be full, and 4 protons were blocked in the $1f_{5/2}$ orbital. As a result, the ^{99}Tc decay is still dominantly driven by the same single-particle transition but a small admixture of a transition from a neutron in $1g_{7/2}$ to a proton in $1g_{9/2}$ appears. Various tests were performed: nuclear structure from GL and GLEKPN valence spaces; without CVC hypothesis; with CVC hypothesis and different methods for estimating the Coulomb displacement energy; various effective values of the axial-vector coupling constant g_A of the weak interaction. As in [Kos17], we found that the spectrum shape is highly sensitive to the effective value of g_A. Next, the best effective value of g_A was obtained by adjusting the theoretical spectrum to the measurement. We also determined the Q-value, the log *ft* value and the average spectrum energy with the highest accuracy ever for ^{99}Tc decay, which are nuclear data quantities of importance for different scientific communities.

No detail comparison was done with previous determinations of the shape of the beta spectrum available in the literature, but we pointed out that the reference parameterization from [Rei74] must now be considered obsolete. It should be noted that all previous measurements of the ^{99}Tc beta spectrum had significantly higher energy thresholds, so that a significant part of the spectrum at low energies had to be considered as being unknown. Combining our measurements with the detailed theoretical calculations, we

extracted new decay data of interest: Q_β = 295.82(16) keV, E_β = 98.45(20) keV, log *f* = −0.47660(22) and log *ft* = 12.3478(23). The spectrum shape was found to be very sensitive to the effective value of the axial-vector coupling constant, with g_A^{eff} = 1.530(83) giving the best agreement with our measurement.
This work demonstrated once again that MMC measurements are excellently suited for determining both the shape and the maximum energy of beta spectra. The measurements are characterized not only by the high linearity and high energy resolution but also by the fact that very low detection thresholds can be reached. The good agreement of two almost independent measurements also in the cross-analysis of the data increases confidence in the obtained beta spectra. Our Q-value is five times more precise than the recommended one [Wan21], an uncertainty definitely competitive with Penning trap measurements.

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