Publishable Summary for 20FUN04 PrimA-LTD
Towards new primary activity standardisation methods based on low-temperature detectors

Overview

Radionuclide metrology, and more specifically, activity standardisation, is based on well-established measurement techniques that have been used and improved for decades. However, some nuclides such as the α-decaying ^{241}Am, show better achievable uncertainty compared to e.g., ^{55}Fe, that decays by low-energy electron capture (EC). New techniques for activity standardisation using low-temperature calorimeters, namely magnetic microcalorimeter (MMC) were developed defining the starting point of future radionuclide metrology closing the uncertainty gap. The combination of high-resolution decay energy spectrometry (DES) for radioactive decays with sophisticated novel theoretical calculations of the spectrum shape increased our knowledge of fundamental decay data and processes.

In this project, new MMC detectors and associated source preparation techniques were developed and their capability for ultra-high-resolution spectrometry was experimentally demonstrated. In addition, theoretical calculation methods for beta decay and complex electron-capture processes were significantly improved. The experimental results as well as the new calculation methods will help researchers in many disciplines such as radionuclide metrology and nuclear medicine. The use of MMCs for primary activity standardization proved to be very difficult and requires further research and developments.

Need

The composition of radioactive sources used in industry, or for nuclear medicine is challenging to determine. NMIs and DIs follow the ever-changing and increasing demands of emerging radionuclides since each new entrant requires a specifically adapted standardisation procedure. This is important for low-energetic decays since their strong model dependence makes standardisation difficult, resulting in high uncertainties. A solution to this problem is to employ Low Temperature Detectors (LTDs) due to extremely low energy thresholds (down to a few 10 eV) and applicability to all types of nuclear decays. Hence, LTDs offer the possibility to be used as a universal tool for activity standardisation.

The second pillar of radionuclide metrology is the determination of nuclear decay data. The most effective way to accurately determine decay data is to use recently developed theoretical models. Although the newer models include more detailed effects of the underlying nuclear and atomic physics, they still do not cover all types of decays, for example the second forbidden non-unique decay of ^{129}I. Additionally, the theory needs accurate experimental data for validation. LTDs can deliver validation data obtained from high-resolution, high statistics measurements in 4π geometry.

Objectives

The overall objective of the project was to improve the capabilities in radionuclide metrology, by developing a new primary activity standardisation method based on low temperature detectors and improving on fundamental nuclear decay data.

The specific objectives of the project were:

1. To develop a new primary method for decay scheme independent activity determination using low temperature detector-based spectrometers with a quantum efficiency of 100 %, high energy resolution and with the capability of processing measurement statistics which exceed 10^8 events / spectrum.
2. To combine new source preparation techniques (e.g., ion-implantation), and modern detectors (e.g., metallic magnetic calorimeters, etc.), in order to standardise one α emitter (^{241}Am), one β emitter (^{129}I) and one electron-capture nuclide (^{55}Fe). This aimed to considerably reduce the uncertainty compared to existing methods.

3. To develop a method for the measurement of ^{55}Fe energy spectra with a better energy resolution and a lower energy threshold (< 50 eV) than existing techniques, to be used to determine fractional electron-capture probabilities. This included determining L-subshell probabilities and a precise study of shake-up and shake-off effects. In addition, this approach was used to determine the beta spectrum shape of ^{129}I down to 0 keV.
4. To compute beta spectrum shapes and electron-capture decay using new calculation techniques, which consider all relevant effects from atomic and nuclear structure.
5. To facilitate the take up of the technology and measurement infrastructure developed in the project by the measurement supply chain (NMIs, DIs, research laboratories) and users (authorities with responsibilities in radiation protection and environmental monitoring, researchers in allied fields).

Progress beyond the state of the art

The instruments of established techniques for activity standardisation are often used for decades, to keep the results reliable and traceable. Nevertheless, steady re-evaluations and improvements are necessary to keep up with the demand for standardisation of new radionuclides or more precise measurements. In recent years, new spectroscopic measurements performed with LTDs, such as MMCs, in conjunction with improved theoretical calculations have been successfully applied to reduce the uncertainties of relevant nuclear data, leading to a reduced uncertainties in primary and secondary activity standardisation measurements.

Usually, a single MMC detector is prepared for a single spectroscopic measurement and is discarded after the measurement. In this project, re-usable MMC detectors were developed and operated. In addition, the measurement infrastructure was upgraded for multi-detector setups. (Objective 1)

The limiting factor in DES is often the source preparation, as highlighted in various previous experiments. Unfortunately, the most convenient techniques often yield sources with lowest quality. Therefore, other source preparation techniques for activity standardisation and DES, namely ion-implantation, were investigated and used. (Objective 2)

DES has been a regular application for MMCs in radionuclide metrology. An improved resolution and high statistics allowed the observation and characterisation of higher-order processes, such as shake processes in the EC of ^{55}Fe as well as the second forbidden decay of ^{129}I. For the latter, the internal gamma-transition was also measured, enabling the observation of the beta-spectrum at extremely low energies, without being impacted by technical limitations, such as detector threshold. (Objective 3)

The theoretical description of decay spectra improved much in previous EMPIR projects. In this project, the formalisms describing the atomic processes involved in the decays, such as shaking or exchange, were improved, as well as the nuclear physics needed to describe forbidden non-unique decays. By using different and independent models, theoretical calculations were validated against each other and to experimental data obtained during this project. (Objective 4)

Results

Objective 1: Development for the application of advanced MMC detectors for radionuclide spectrometry and activity standardisation

The accuracy of MMC measurements can be improved by increasing statistical power with larger event counts per measurement and by further improving the already good energy resolution. The former can only be achieved in a reasonable time by using a multiple MMC-pixels setup, while the latter rests on a further optimized MMC design (type 1). A novel generation of re-useable MMCs (type 2), where the radionuclide sources are non-permanently attached to the MMC detector, combined with the appropriate analysis tools, will allow the use of MMCs in more common tasks in radionuclide metrology, namely in primary activity standardisation. The specifications for both types of detectors have been defined and multiple sets of wafers employing the final designs were manufactured. The detectors were thoroughly tested for circuit integrity and the functionality at low temperature was confirmed. MMCs of both detector types were successfully operated in multiple measurements including the use of the re-usable feature.

In parallel to these improvements, the existing spectrometer setups were upgraded to allow measurements of >10 detector channels simultaneously requiring adapted wiring and a new multi-channel readout. All necessary designs have been defined, all components have been manufactured and were installed in the cryostat systems. Multi-channel measurements were performed confirming the functionality of the upgraded systems.

The objective was fully achieved.

Objective 2: Radioactive source preparation and primary activity standardisation

Ion-implantation into the absorbers of a fully optimised multi-pixel MMC detector was achieved at the RISIKO off-line mass separator facility for radioisotopes at Mainz University. Based on preparative laser-spectroscopic studies on stable ^{56}Fe, a monoisotopic ^{55}Fe ion beam with high spatial characteristics was produced to perform the implantation. Well-controlled and quantitative implantation runs were performed by strong focalization as well as precise positioning and scanning of the ion beam onto the MMC detector chip surface. Using optimized process parameters for laser ionization and mass separation, as well as ion beam control and quantification, implantation runs successfully achieved activities of 5 Bq per absorber.

For the composite detectors, the absorber surfaces were nano-structured, reducing the spectrum distortions, that are attributed to the decay radiation scattering in the usually dielectric source material. Fully micro-fabricated absorbers with integrated nanostructured surfaces have successfully been fabricated including counterparts to fully encapsulate the source material by diffusion welding. Iodine-129 was deposited in those absorbers using the iodine self-absorption in silver.

Activity standardisation with MMCs allows to count decays with efficiencies close to 100% and nearly zero energy threshold. This method is less prone to systematic uncertainties and should be able to reach the permille level. Studies to estimate the influence of absorber designs were carried out based on Monte Carlo simulations. This method was used for the electron-capture nuclide ^{55}Fe and alpha decaying nuclide ^{241}Am each and validated against established activity standardisation techniques.

Electrodeposited ^{241}Am sources have been fabricated and the activity was measured independently by four institutes using established, absolute methods. Good agreement between those standardisations was obtained including the measurements using MMCs. In addition, semi-quantitative source preparation by micro-dispensing and direct linking to liquid scintillation methods was used to prepare ^{55}Fe sources that were standardised using MMCs. Uncertainties of a few permille were obtained for validation purposes. However, large discrepancies between the liquid scintillation and MMC methods were observed, which are most likely caused due to sample preparation by micro-dispensing.

Outstanding new source preparation techniques were developed and applied. However, the objective was not fully achieved because it has not yet been possible to determine activities with a clear quantitative link and the desired small uncertainties.

Objective 3: High precision measurement of ^{55}Fe and ^{129}I spectra for accurate determination of decay data

An extensive simulation of ^{129}I was carried out to test analysis routines for determination of live-time, for suppression of pile-up and for count rate estimation. Once validated, the analysis routines were then applied to the measured data.

Upgrading the MMC/SQUID spectrometers of the consortium partners to more than ten read-out channels enabled high-statistics spectrum measurements of ^{55}Fe and ^{129}I with several 10^6 events within a few days. In promising measurements towards the end of the project, structures in the recorded spectra were observed which may be explained by higher-order effects such as electron shake-off and shake-up in the EC of ^{55}Fe. These findings need to be confirmed via future high-statistics measurements. Furthermore, it was shown that spectra obtained from ion-implanted sources are of significantly higher spectral quality compared to spectra from drop-deposited samples. The experimentally determined beta spectrum of ^{129}I showed interesting features. The beta spectrum of the dominant transition was shifted by about 40 keV due to the subsequent gamma transition. This effect was anticipated when proposing this experiment and will allow the study of the beta spectrum shape down to virtually 0 keV. Moreover, the preliminary measurements indicated that the maximum beta energy is about 15% higher than currently adopted in literature, which is an unexpected result that will have large impact.

The objective was not fully achieved within the project duration as some of the measurements and related analyses can only be completed later. However, outstanding new results are expected in the near future.

Objective 4: Theoretical predictions of ^{129}I beta spectrum and ^{55}Fe electron-capture decay

To describe the underlying physics of the decays in more detail, some existing calculation codes were optimised for parallel processing, including higher order atomic transitions. The different model parameters were tested for their influence on the model's accuracy, with a trade-off with respect to the additional computational burden induced. This allowed to identify significant differences in X-ray spectra, when generated from an electron capture or internal conversion.

Technical compatibility issues between the calculations codes of the different partners were identified and fixed, which allowed the validation of the results of the much faster but less accurate atomic framework based on Relativistic Density Functional Theory (RDFT) against the results of the state-of-the-art but slower Multi-Configurational Dirac-Fock (MCDF) approach. Combined with the BetaShape code that was adapted to RDFT and MCDF results, this allowed to calculate fully synthesised EC spectra based on ab initio atomic models.

A previously conducted MMC measurement of the β/γ-decay of ^{151}Sm was re-analysed and matching-updated theoretical calculations were performed. Many of the tools developed are beneficial to the case of ^{129}I because of similarities in the decay scheme. A corresponding article has been published in a peer-reviewed journal. The analysis tools and theoretical improvements developed during the project were applied to an existing dataset of ^{99}Tc measured with the MMC-spectrometers of the consortium that describes the beta-spectrum with unprecedented accuracy. Those results were submitted to a high-ranking journal and are already available as preprint.

The BetaShape code, which accurately calculates beta and EC transitions, was used to describe several other decay spectra, such as ^{6}He, ^{176}Lu and ^{99}Tc. It was recently adopted by the Nuclear Structure and Decay Data (NSDD) IAEA network for future ENSDF evaluations.

The objective was fully achieved.

Impact

A web page (https://prima-ltd.net/) was set up to give interested parties an overview of the activities and scientific goals the consortium was addressing within the project. Contact details for interested parties were given and a stakeholder registration platform was established and advertised. A concept for keeping the stakeholders up to date was developed. Information about recent publications, conferences and training courses was disseminated using these platforms. In total 18 stakeholders from 15 organizations have registered. Two well-attended stakeholder workshops as well as two training courses were held. The stakeholder workshops covered all aspects of the project starting with a project overview followed by presentations of the work packages and discussions. In the two training courses, recent progress in nuclear counting using MMCs as well as in MMC measurements and analysis in metrology was presented. While the focus was set on the project, also external speakers were invited and contributed to the training courses. About 35 participants from universities, research institutes and national metrology institutes not involved in the project joined the training courses

In total, the project and its progresses were presented at several conferences with a total of 25 talks and posters. Outcomes of the project resulted in 11 open access, peer-reviewed publications. Six technical visits – two of them virtual – took place within the consortium for in-depth discussion on specific technical aspects.

Impact on industrial and other user communities

Many users of radioactive materials will benefit from improved nuclear decay data and related more accurate activity determination. The nuclear power industry uses decay data to determine the residual heat and its evolution with time in nuclear reactors and in nuclear waste management. For example, the average energy per ^{129}I decay depends on the shape of the beta spectrum and the maximum beta energy. Thanks to the project, this data will be available with significantly improved accuracy. The preliminary experimental results of the maximum beta energy of ^{129}I suggests a higher value, which will be of enormous importance. Results of environmental monitoring of radioactivity will also be able to reduce uncertainties by using decay data with smaller uncertainties.

Nuclear medicine will also benefit because, for example, the improved beta spectrum calculation methods from this project can be used. This plays a role in some radiopharmaceuticals that can be used for diagnosis or therapy. More accurate decay data allow a more accurate calculation of the dose per administered activity, whose determination will also become more accurate with the results of this project.

While the nuclides investigated in this project are not of specific interest in industry or medicine, the newly developed and validated theoretical models covered nuclides in these applications. Experience from previous projects prove that newly available nuclide data are taken up by NMIs/DIs soon resulting in a better service and better-quality radioactive sources for commercial customers.

Impact on the metrology and scientific communities

The development of a completely new method for primary activity determination will be a great benefit for radionuclide metrology, since it will lower the achievable uncertainties by up to an order of magnitude. Some nuclides, such as ^{7}Be, that have not been standardised yet might also be accessible with this method.

Advanced theoretical calculations of decay spectra and their validation with high-resolution and high-statistics data of the ^{55}Fe electron capture and ^{129}I beta decay down to lowest energies, will benefit the accuracy of decay data in general. This decay data, based on the broadly applicable theoretical models, is not only used in metrology, but, for example, also in the calculation of neutrino spectra, especially from nuclear reactors for the determination of neutrino oscillation parameters or the search of non-weak interacting (sterile) neutrinos. Background estimation of low-background experiments, such as the search for dark matter reaching beyond the standard model of particle physics, also rely on accurate decay data. It should be noted again that the preliminary experimental results of the maximum beta energy of ^{129}I show unexpected deviations, which will be of high importance for metrology and other fields.

A paper on the study of the beta spectra and decay scheme parameters of ^{151}Sm has been published and the article was already cited by other researchers and it triggered new proposals for verification experiments to determine the maximum beta energy by means of Penning traps. Corresponding information for public relation purposes (https://www.ptb.de/cms/index.php?id=samarium-151) has also been published as an early outcome of the work carried out in the framework of objective 4. The developments in MMC technology, from fabrication to data acquisition and data processing of multi-channel spectrometers, will also not only benefit the contributing partners, but also the LTD community as whole.

Another paper, that has recently been published, presents the K- and L- shell fluorescence yield values of the full isonuclear sequence of Fe ions, using a state-of-the-art multiconfiguration Dirac–Fock approach.

Representatives of CEA, PTB and KIT took part in a technical meeting of the AMoRE Double Beta Decay Experiment. This experiment also employs MMC detector technology. The PrimA-LTD representatives informed specifically about the detector technology developed within the project (obj. 1) as well as MMC readout strategies (obj. 3) as an early outcome.

Impact on relevant standards

The project led to improved nuclear decay data by direct measurements and by improving the theoretical calculation techniques. Hence, the outcome of this project is a valuable contribution for nuclear decay data evaluations. Publications and tables with recommended data play a key role for research and many applications and are also a basis for international standards. Presentations were given during each EURAMET TC-IR meeting highlighting the research activities, presenting the latest developments and the projected output. The uptake of new decay data (obj. 4) and awareness for the importance of decay data for activity determination (obj. 3 and 4) are proof of the early impact from this project. Results of the high-precision measurements on ^{151}Sm have already been incorporated into an evaluation of the Decay Data Evaluation Project (DDEP) and results are publicly available (http://www.lnhb.fr/nuclides/Sm-151_tables.pdf and http://www.lnhb.fr/nuclides/Sm-151.BetaShape.zip).

Longer-term economic, social and environmental impacts

The wider dissemination of MMCs and similar LTD technologies can and will have large impact on research, medicine and industry. In X-ray spectrometry, LTDs can combine high resolution and high energy bandwidth, that classically require two different detector systems. Nuclear forensics and nuclear safeguards can be made more accessible, since the high resolution of LTDs allows radiochemical preparations before measurements to be omitted and can combine the capabilities usually obtained separately from alpha- and mass-spectrometry in a single measurement.

The use and benefits in nuclear power industry, environmental monitoring and nuclear medicine, will also continue in the long-term, both from the improved decay data and better activity standardisation with the inevitable wider adoption of LTD technology. All these end-users base their developments on nuclear decay data that come either from the DDEP (Decay Data Evaluation Project) collaboration, designed for and coordinated by the metrology community, or from the ENSDF (Evaluated Nuclear Structure Data File) community. Both have adopted the BetaShape code, which has been significantly improved in this project, for their evaluations, ensuring significant knowledge transfer from this project to the most applicative communities (obj. 4).

List of publications

1. Kossert, K., Loidl, M., Mougeot, X., Paulsen, M., Ranitzsch, P., Rodrigues, M.: High precision measurement of the ^{151}Sm beta decay by means of a metallic magnetic calorimeter. In: Applied Radiation and Isotopes 185 (2022), 110237, https://doi.org/10.1016/j.apradiso.2022.110237.
2. Pinheiro, D., Fernandes, A., Godinho, C., Machado, J, Baptista, G., Grilo, F., Sustelo, L., Sampaio, J.M., Amaro, P., Leitão, R.G., Marques, J.P., Parente, F., Indelicato, P., de Avillez, M., Santos, J.P., Guerra, M.: K- and L-shell theoretical fluorescence yields for the Fe isonuclear sequence. In: Radiation Physics and Chemistry A 203 (2022), 110594, https://doi.org/10.1016/j.radphyschem.2022.110594.
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4. Agnes, P. et al.: Search for low-mass dark matter WIMPs with 12 ton-day exposure of DarkSide-50. In: Physical Review D 107 (2023), 063001, https://doi.org/10.1103/PhysRevD.107.063001.
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8. Andoche, A., Mouawad, L., Hervieux, P.-A., Mougeot, X., Machado, J., Santos, J.P.: Influence of atomic modeling on electron capture and shaking processes. In: Physical Review A 109 (2024), 032826, https://doi.org/10.1103/PhysRevA.109.032826.
9. Paulsen, M. et al.: High precision measurement of the ^{99}Tc β spectrum, Physical Review C, [submitted]; Preprint available: http://arxiv.org/abs/2309.14014.
10. Müller, M., Rodrigues, M., Beyer, J., Loidl, M., Kempf, S.: Magnetic Microcalorimeters for Primary Activity Standardization Within the EMPIR Project PrimA-LTD, Journal of Low Temperature Physics 214 (2024), https://doi.org/10.1007/s10909-024-03048-7.
11. Niemeyer, T. et al.: Ion implantation of the electron-capture nuclide Fe-55 for measurements by means of metallic microcalorimeters, Applied Radiation and Isotopes [submitted].

This list is also available here: https://www.euramet.org/repository/research-publications-repository-link

<table>
<tr><td colspan="2">Project start date and duration:</td><td>1 June 2021, 36 months</td></tr>
<tr><td colspan="3">Coordinator: Ole Jens Nähle, PTB   Tel: +49 531 592 6110   E-mail: ole.j.naehle@ptb.de
Project website address: https://prima-ltd.net/</td></tr>
<tr><td>Internal Funded Partners:
1. PTB, Germany
2. CEA, France
3. CHUV, Switzerland
4. CIEMAT, Spain</td><td>External Funded Partners:
5. CNRS, France
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7. JGU, Germany
8. KIT, Germany
9. UHEI, Germany
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