# 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. The project will close this gap by developing new primary techniques for activity standardisation using low-temperature calorimeters. The combination of high-resolution spectrometry for radioactive decays with sophisticated novel theoretical calculations of the spectrum shape will also increase knowledge of the fundamental decay data.


## Need
The composition of radioactive sources used in industry, or for nuclear medicine is not easy 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 use Low Temperature Detectors (LTDs) due to extremely low energy thresholds (down to a few 10 eV) and versatility with all decays. 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, such as the 2nd forbidden non-unique decay of $^{129}$I. Additionally, the theory needs accurate experimental data for validation. LTDs can deliver validation data, by high-resolution, high statistics measurements in 4π geometry.

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

The specific objectives of the project are:

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 should aim 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 should include determining L-subshell probabilities and a precise study of shake-up and shake-off effects. In addition, this approach should be 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**

Activity standardisation heavily relies on established techniques, such as liquid scintillation counting or coincidence counting, and even the same instruments are being used for decades, to keep the results reliable and traceable. Nevertheless, steady re-evaluations and improvements are necessary to keep up with the demand of customers, e.g., to establish new radionuclides, that have not been standardised before or to improve on the precision of the measurements and reduce the uncertainties.

In the last few years, new spectroscopic measurements performed with low temperature detectors, such as metallic magnetic calorimeters (MMCs), in conjunction with improved theoretical calculations were used to improve nuclear data. This data is used as the basis for many primary and secondary activity standardisation techniques and the improvements helped to reduce uncertainties of the measurements.

Usually, a single MMC detector is prepared and used for a single spectroscopic measurement of one radionuclide, and is often discarded after a successful measurement, since they are not intended to be re-used for different radionuclides. This project aims to develop and fabricate new MMC detectors intended to be used directly for activity standardisation for the first time and improve upon the proven spectroscopy capabilities, by increasing the achievable statistics by one order of magnitude to about $10^8$ events / spectrum. Both applications require the use of multi-detector setups, therefore re-useable detectors become more favourable because of limited fabrication capabilities, and the measurement infrastructure needs to be upgraded as well. (Objective 1)

Previous measurements have shown that the limiting factor in spectrum measurements is often the source preparation rather than the detector technology. Unfortunately, the most convenient techniques, such as drop deposition of aqueous solutions, often yield sources with lowest quality. Therefore, this project investigates other source preparation techniques, such as ion-implantation to be used with MMCs. These samples will be used to standardise the activity of several radionuclides with MMCs and compare the results with established techniques. (Objective 2)

High-resolution spectroscopy of radioactive decays has been a regular application for MMCs in radionuclide metrology, and both beta- and electron capture decay have been investigated in recent EMPIR projects. To improve upon the status quo, new nuclides, namely $^{55}$Fe and $^{129}$I will be measured more accurately than any other nuclide before. Improving resolution and statistics will allow us to observe and characterise higher-order processes, such as shake-up and shake-off in the electron capture of $^{55}$Fe. The measurement of the 2$^{nd}$ forbidden decay of $^{129}$I is expected to become the most accurate measurement of this type of decay. The fact that the decay is measured in coincidence with the 40 keV gamma-transition of $^{129}$I, will enable us to observe the beta-spectrum at very low energies, without being impacted by technical limitations, such as detector threshold. (Objective 3)

Also, the theoretical description of decay spectra has been much improved in previous EMPIR projects, by including more and more effects and corrections from nuclear and atomic physics in the calculations. Nevertheless, the higher order atomic physics to describe shake-up and shake-off effects still must be worked out as well as the nuclear physics needed to describe 2$^{nd}$ forbidden decays. In addition, by using different and independent models for the nuclear physics, theoretical calculations will be validated against each other, in addition to the 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 improving the already good energy resolution. The former can only be achieved in a reasonable time by using a multiple MMC-pixels, while the latter rests on a further optimized MMC design. Another generation of re-useable MMCs, where the radionuclide sources are non-permanently attached, combined with the appropriate analysis tools, allows 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 a final design was agreed with fabrication starting soon.

In parallel to these improvements, the existing spectrometer setups are being upgraded to allow measurements of at least 10 detector channels simultaneously requiring adapted wiring and a new multi-channel readout.

*Objective 2: Radioactive source preparation and primary activity standardisation*

Performance will be enhanced by employing a mass separator to selectively ion-implant only the wanted nuclide into the fully optimised multi-pixel MMC design and enclosing it afterwards. For the composite detectors, the absorber surface will be nano-structured, reducing the spectrum distortions, that are attributed to the decay radiation scattering in the usually dielectric source material. Nanostructured surfaces have been fabricated and techniques to encapsule the surface investigated.

Activity standardisation with MMCs will allow to count decays with efficiencies close to one and virtually zero energy threshold. This method will be less prone to systematic uncertainties and should be able to reach uncertainties in the order of 1 permille. Studies to estimate the influence of absorber design were done based on Monte Carlo simulations. This method will be used for one beta emitter ($^{129}$I), electron capture nuclide ($^{55}$Fe) and alpha decaying nuclide emitter ($^{241}$Am) each and validated against established activity standardisation techniques. Radioactive solutions were selected and have been characterized.

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

By employing multi-pixel MMC arrays with at least 10 read-out channels, the statistics of the spectrum measurements of $^{55}$Fe and $^{129}$I will be increased to approximately $10^8$ events. This in turn will allow to study the spectrum shape in more detail by looking at low probability effects, such as shake-up and shake-off.

*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, the computational load is expected to increase significantly. Therefore, some existing simulation codes were optimised for parallel processing and the influence of different model parameters were tested for their influence on the model's accuracy compared to the additional computational burden. Technical compatibility issues between simulation codes of different partners were identified and fixed.

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 now been published in a peer-reviewed journal.

**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 is addressing within the project. Contact details for interested parties are given and a stakeholder registration platform was established and advertised. A concept for keeping the stakeholders up to date was developed, in particular with a regular newsletter. Information about recent publications, conferences and training courses will be disseminated using these platforms.

The activities and scientific approaches of the consortium were presented at the EURAMET TC-IR meeting in 2022.

*Impact on industrial and other user communities*

Many users of radioactive materials will benefit from improved nuclear decay data and 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. Results of environmental monitoring of radioactivity will also be able to reduce uncertainties by using decay data with smaller uncertainties.

Nuclear medicine and the use of radiopharmaceuticals will also profit. 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 will cover nuclides in these applications.

*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, 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.

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. Corresponding information for public relation purposes (link to webpage) has also been published (so far only in German).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.

*Impact on relevant standards*

The project will lead to improved nuclear decay data by direct measurements and by improving the theoretical calculation techniques. Hence, the outcome of this project will be 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.

*Longer-term economic, social and environmental impacts*

The wider dissemination of the 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 benefit in nuclear power industry, environmental monitoring and nuclear medicine, will also continue on a long-term, both from the improved decay data and better activity standardisation with the inevitable wider adoption of LTD technology.

**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.

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

| Project start date and duration: | | June 2021, 36 months |
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| Coordinator: Ole Jens Nähle, PTB | Tel: +49 531 592 6110 | E-mail: ole.j.naehle@ptb.de |
| Project website address: https://prima-ltd.net/ | | |
| Internal Funded Partners:<br>1. PTB, Germany<br>2. CEA, France<br>3. CHUV, Switzerland<br>4. CIEMAT, Spain | External Funded Partners:<br>5. CNRS, France<br>6. IPSA, France<br>7. JGU, Germany<br>8. KIT, Germany<br>9. UHEI, Germany<br>10. UNL, Portugal | Unfunded Partners: |
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