Publishable Summary for 15SIB10 MetroBeta
Radionuclide beta spectra metrology

Overview

The project MetroBeta aimed to improve knowledge of beta spectra, through both theoretical and experimental approaches. Existing theoretical knowledge was used to account for the nuclear structure effects on beta spectra and a new code BetaShape was produced. Experiments were performed using four types of devices: i) specifically developed cryogenic metallic magnetic calorimeters (MMCs), operating at very low temperature, ii) solid scintillators, iii) a magnetic spectrometer and iv) silicon lithium (SiLi) detectors. Comparison of the calculated and measured spectra was performed to validate the quality of the spectra and the improved theory. The effect of this improved knowledge on the measurement of the activity (the becquerel) of beta emitting radionuclides was demonstrated.

Need

In the development of new nuclear reactors and in light of the increasing quantity of nuclear waste from existing reactors, the requirements on the diagnostics of nuclear reactor cores and nuclear waste management are becoming more and more stringent. In particular, the calculation of decay heat upon reactor shutdown and in nuclear waste requires improved knowledge of the energy of the beta emissions. The intended monitoring of the isotopic composition of reactor cores via measurement of the anti-neutrino spectrum are dependent on the precise determination of individual beta spectra.

In nuclear medicine, the number of radionuclides being utilised in the fight against cancer is increasing. The available information for these radionuclides can be uncertain, particularly for new applications. In the case of beta emitters, a precise knowledge of the energy spectra is of paramount importance since the physiological impact varies strongly with energy. Thus poorly known beta spectra can lead to large discrepancies in the effective dose.

There is also an increasing demand for precision beta spectrometry from several fields of fundamental research, such as in the direct measurement of the neutrino mass and in nuclear astrophysics.

Finally, the metrology of ionising radiation was another main driver for this project, since the SI derived unit for activity, the becquerel, is determined via very precise primary activity measurements which crucially depend on the knowledge of the shape of the beta spectra.

Objectives

This project aimed to meet the above needs and to improve knowledge of the energy of the particles emitted during the beta decay process, by providing a validated computer code to enable precise predictions for all types of beta spectra, including reliable uncertainties.

This project addressed the following scientific and technical objectives:

1. To improve modern measurement techniques for silicon detectors (Si(Li)), solid scintillator crystals ($LaBr_3$/$CeBr_3$) and magnetic spectrometers for measurements of beta spectra.
2. To optimise beta spectrometers, based on metallic magnetic calorimeters (MMCs), and measure high resolution beta spectra for low (< 100 keV) and intermediate (< 1 MeV) end-point energy pure beta emitters Sm-151, C-14, Tc-99 and Cl-36.
3. To improve theoretical computation methods on the basis of the measured spectra and compare the measured and calculated beta spectra.

4. To investigate the effect of improved beta spectra on absolute activity measurements and measure Bremsstrahlung cross-sections to quantify their effect.
5. To facilitate the take up of the technology and measurement infrastructure developed by the project by the measurement supply chain (accredited laboratories, instrumentation manufacturers) and end users (the nuclear medicine community and the nuclear power industry).

Progress beyond the state of the art

Improvement of modern measurement techniques for silicon detectors, solid scintillator crystals and magnetic spectrometers for measurements of beta spectra

Three experimental methods were used to measure beta spectra in order to validate the results. These improved the estimation of impurity quantities in pure beta emitting radionuclide sources, helped reduce experimental uncertainties and lead to improved detector performance.

Optimisation of beta spectrometers and measurement of new high resolution beta spectra

A third MMC system, as an addition to the previous two MMC systems in existence used for measuring beta spectra, was commissioned. Individual and optimised sources for each individual beta emitter were produced and their beta spectra measured.

Improvement of theoretical methods on the basis of the measured spectra and comparison of the measured and calculated beta spectra

The theoretical component of this project allowed the inclusion of nuclear structure to be incorporated into the calculations. The calculated and measured beta spectra were compared and used to validate the methods put in place.

Investigation of the effect of improved beta spectra on absolute activity measurements and measurement of Bremsstrahlung production.

The absolute activity measurement using the liquid scintillation counting technique, an essential tool in radionuclide metrology, was improved by taking into account the better knowledge of the beta spectra and the uncertainty propagation on the final activity measurement was included. Additionally, the production of Bremsstrahlung photons was measured, since they affect both beta spectra measurements with MMCs and the absolute activity measurements.

Results

Availability of beta spectra

The BetaShape code, which has been further developed within this project, has been used to provide beta spectra for two major international nuclear decay data evaluation projects. The International Atomic Energy Agency's (IAEA) *International Network of Nuclear Structure and Decay Data Evaluators* (NSDD), provides both plots and tabulated beta spectra calculated with the BetaShape code. These spectra are available from the *Live Chart of Nuclides* tool available at: https://www-nds.iaea.org/relnsd/vcharthtml/VChartHTML.html. A second international collaboration, the Decay Data Evaluation Project (DDEP), who provide decay scheme data mainly to the metrology community, have also made available the tabulated data for the beta spectra calculated with the BetaShape code at: http://www.lnhb.fr/nuclear-data/nuclear-data-table/.

Improvement of modern measurement techniques for silicon detectors (Si(Li)), solid scintillator crystals ($LaBr_3$/$CeBr_3$) and magnetic spectrometers for measurements of beta spectra.

Improvements in the use of three current measurement techniques have been implemented and more precise beta spectra have been measured.

The precise characterisation of the physical dimensions of the Si(Li) detector was achieved by X-raying the detector and housing, and a detailed and accurate Monte Carlo model was created. The model was then used to characterise the detector response prior to the measurement of the beta spectra for seven radionuclides: P-32, Cl-36, Sr-89, Sr-90, Y-90, Pm-147 and Tl-204 and the beta spectra of a variety of Y-90/Sr-90 mixtures. A specially designed and purpose-built collimator was used in order to reduce the distortion to the measured spectra.

The solid scintillator crystal development work has successfully measured the Lu-176 and Rb-87 beta spectra through developments in the data acquisition system, for which a dedicated lead-castle-based acquisition set up was implemented and characterised.

The magnetic spectrometer was commissioned and a precise energy calibration was performed, prior to being used to measure the beta spectra for six radionuclides: Cl-36, Tc-99, Co-60, Cs-134, Cs-137 and Tl-204.

Optimisation of beta spectrometers, based on metallic magnetic calorimeters (MMCs), and measurement of new high resolution beta spectra.

Extensive work was undertaken on the commissioning of the new metallic magnetic calorimeter based beta spectrometer, resulting in an operating device capable of measuring high resolution beta spectra. Specifically designed MMC devices were tailored to the needs for the project and were used in the two measurement systems. Extensive development of the data acquisition system and analysis routines was carried out allowing high resolution beta spectra to be measured for four radionuclides: Sm-151, C-14, Tc-99 and Cl-36. The Sm-151 measurement also allowed an assessment to be made of the intensities of the two possible decay branches.

As an addition to the original project proposal, novel nano-composite gold absorbers, i.e. a gold nano-foam with pore sizes sufficiently small so that nano-constrained crystallisation occurs, were developed and tested.

Improvement of theoretical methods on the basis of the measured spectra and comparison of the measured and calculated beta spectra.

The code, BetaShape, used for the calculation of theoretical beta spectra was further developed and tested. A simplified model to determine the nucleon wave functions using either a relativistic or non-relativistic harmonic oscillator was included, as well as a phenomenological nuclear mean-field approach employing the so-called Woods-Saxon potential. The intrinsic model in the code was re-implemented during this project owing to inconsistencies seen through the coupling with the nuclear structure component. The propagation of the uncertainties in the calculation of the nuclear structure components has now been included into the relevant part of the calculations. The new parameterisation of the Woods-Saxon potential of the mean-field Hamiltonian and the spin-orbital potential was used to generate the realistic nucleonic wave-functions for calculating the beta spectra.

Investigation of the effect of improved beta spectra on absolute activity measurements and measurement of Bremsstrahlung production.

Comprehensive studies were made on the absolute activity determination of Co-60 and Tc-99 using two primary activity measurement methods, based on liquid scintillation counting. Results have already been published for Co-60, and they underline the importance of using accurate beta spectra. Further studies were carried out using the TDCR-Cerenkov technique for Cl-36.

Simulations of the Bremsstrahlung cross sections were performed and the measurement device was fully commissioned and calibrated in terms of energy and efficiency. Unfortunately, the radioactive source available for these measurements, a Sr-90 source with an activity of 37 MBq, produced a simulated Bremsstrahlung production rate of only 100 photons per day, meaning that no measurements were feasible. A new source of 10 GBq was ordered, but was not delivered before the end of the project.

Impact

Information on the project and various results has been presented at many national and international meetings. The following three ICRM Working Groups: Nuclear Decay Data, Beta Spectrometry and Life Sciences in 2017, 2018 and 2019; the JEFF Working Group of the OECD/NEA at a number of their biannual meetings; the Technical Committee for Ionising Radiation of the European Metrology Organisation, as well as at two meetings of the International Atomic Energy Agency's Nuclear Structure and Decay Data Network (2017 and 2019) and the German Physical Society Spring 2018 Meeting on Condensed Matter Physics.

Presentations were also given at several International Conferences: Advances in Liquid Scintillation Counting (LSC 2017), Denmark, May 2017; Radionuclide Metrology (ICRM 2017), Argentina, May 2017 and Radionuclide Metrology (ICRM 2019), Spain, May 2019; International Conference on Nuclear Data and Its Applications, China, May 2019; Low Temperature Detectors (LTD-17), Japan, July 2017 and Low Temperature Detectors (LTD-18), Italy, July 2019; Ultra Low Temperature Physics (ULT-2017), Germany, August 2017 and the 4th International Workshop on Superconducting Sensors and Detectors, Australia, July 2018.

A number of peer-reviewed conference proceeding articles have been published resulting from these presentations.

Impact on industrial and other user communities

Precisely determined mean beta energies, deduced from the spectral shape and with the end-point energies, are important to the nuclear power industry. The data made available from this project will help to reduce the uncertainties on the calculation of the residual decay heat in nuclear reactors that is in the large part due to beta emission after reactor shutdown. In this way, post-irradiation fuel management will be improved and made more cost-efficient. The Joint Evaluated Fission and Fusion (JEFF) project of the OECD Nuclear Energy Agency produces the reference nuclear data libraries used within Europe. The next release of the Radioactive Decay Data sub-library will contain evaluations, which have used the BetaShape code to calculate the beta spectra and associated mean energies.

The precise beta spectra from this project are also being used to reduce unnecessary doses being delivered when beta-emitting radiopharmaceuticals are used for diagnosis or treatment in nuclear medicine, particularly in the treatment of cancer.

The environmental measurement community are also benefitting from the better knowledge of beta spectra.

Impact on the metrology and scientific communities

Significant development work related to the metallic magnetic calorimeter (MMC) detection systems has been carried out and a completely new cryogenic system has been installed and commissioned. The lessons learnt have been documented and made available as a “Good Practice Guide”, ensuring knowledge transfer to other laboratories wishing to establish an equivalent system. This Guide has already been supplied to Los Alamos National Laboratory and Lawrence Livermore National Laboratory upon their request. Specifically designed chips for this project were manufactured and subsequent design modelling work showed the expected energy resolution to be better than originally expected, i.e. 20 eV (instead of 60 eV) and 90 eV (instead of 200 eV), for the two types originally specified for measuring the different energies. In reality, five different chips, have been produced, meaning that a more refined choice for each radionuclide (and endpoint energy) could be made. A number of these new chips were used for the measurements made during the project, for example, C-14, Tc-99 and Sm-151. An additional source preparation technique, not originally planned in the project, using gold nano-foams, was studied.

A first version of the BetaShape code, which can be used to calculate improved beta spectra, has been made available from the CEA website: http://www.lnhb.fr/rd-activities/spectrum-processing-software/. The code was used to calculate the Co-60 beta spectrum and these initial results from the project have already demonstrated the impact on the determination of the becquerel through activity measurements carried out using liquid scintillation counting for Co-60.

Measured spectra have been obtained for a number of radionuclides using a variety of different techniques, as already reported in this summary. These spectra will be useful in constraining further theoretical developments to be included in future versions of the BetaShape code, and also for ensuring accurate absolute activity measurements can be performed for the radionuclides in question, since these spectra are required for the liquid scintillation counting method – a robust method used regularly in many National Metrology Institutes (NMIs).

Impact on relevant standards

The project has had a direct and demonstrated impact on the primary activity standard measurements for pure beta emitters, carried out using the liquid scintillation counting technique, as has clearly been demonstrated for Co-60. This technique is the main method for activity measurements in most National Metrology Institutes (NMIs) and so adoption of the beta spectra calculated and/or measured in this project will be paramount. Their use has been promoted to the relevant bodies, e.g. CCRI(II) of the BIPM, EURAMET Technical Committee on Ionising Radiation, and working groups of the ICRM, etc.

Longer-term economic, social and environmental impacts

A number of “Good Practice Guides” have been produced describing the various developments to the experimental techniques used in this project. In addition, the precisely measured beta spectra, and their associated uncertainties, are now available, to allow their use in metrology laboratories and the relevant industries. A “Validation Report” on the effect of improved beta spectra on absolute activity measurements has been produced and will help improve the quantification of the associated uncertainty component in the measured activity standard. The project has provided stakeholders and end users in research establishments, the nuclear industry and the medical community with new nuclear data and methods for more precise activity measurements (the becquerel) of beta-emitting radionuclides using different measurement techniques, e.g. liquid scintillation counting and Si(Li) detectors. Targeted stakeholder groups have been preferentially addressed, including the broader scientific community, National Metrology Institutes, nuclear power plant operators, radioactive waste agencies, nuclear medicine clinics, environmental agencies and food and water measuring laboratories.

List of publications

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