

JRC TECHNICAL REPORT

JRC – SCK CEN collaboration agreement: Projects 2016 - 2020

Martin Ramos, M., Massaut, V., Schillebeeckx, P.,
Van den Eynde, G.

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Contact information

Name: Peter Schillebeeckx

Address: Retieseweg 11, B – 2440 Geel

Email: peter.schillebeeckx@ec.europa.eu

Tel.: +3214571475

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Contents

Acknowledgements	1
Abstract	2
1 Introduction.....	3
2 Approved projects.....	4
3 Projects	5
3.1 Transmission measurements on Bi at GELINA.....	5
3.2 Improving nuclear data for MYRRHA.....	6
3.3 Uncertainty quantification and propagation.....	7
3.4 Fast neutron detector development for measurements at VENUS-F.....	8
3.5 Release of fission products from lead-bismuth eutectic.....	9
3.6 Volatile radionuclide detection in the MYRRHA beamline.....	10
3.7 Capture cross section measurements for ^{235}U at BR1	11
3.8 Characterisation of Bi-samples by NRCA.....	12
3.9 Study of NDA observables to characterise nuclear fuel.....	13
3.10 Neutron measurements on spent fuel pellets.....	14
3.11 Uranium enrichment and plutonium isotopics	16
3.12 Certification of an environmental reference material.....	17
3.13 Measurements of radon in water	18
3.14 Invariability of decay constants.....	19
3.15 Realisation of computer models of gamma-ray detectors.....	20
3.16 Radionuclide calibrators in nuclear medicine.....	21
3.17 Common organization of workshops, events, topical days.....	23
4 Output: publications.....	24
5 Summary and conclusions	26
List of abbreviations and definitions	27
List of figures	29
List of tables.....	30
Annex Abstracts.....	31
Annex 1. Journal papers.....	31
Annex 2. Contributions to conference proceedings or workshops.....	38
Annex 3. Technical Reports.....	42

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Marc Verwerft	SCK CEN
Jan Wagemans	SCK CEN
Gaspar Žerovnik	EC - JRC

Authors

Vincent Massaut	SCK CEN
Manuel Martin Ramos	EC - JRC
Peter Schillebeeckx	EC - JRC
Gert Van den Eynde	SCK CEN

Abstract

This report presents the activities and main outcomes of the first Collaboration Agreement between JRC and SCK CEN which ran from 2016 to 2020. The collaboration focused on scientific and technological fields common to JRC and SCK CEN: MYRRHA, nuclear data for reactor and other applications, nuclear safeguards and security, nuclear fuel research, emergency preparedness, dosimetry and medical applications. The scope of this collaboration was not only scientific progress, but also education and training and scientific outreach.

Projects were defined in project sheets and each project was assigned a lead researcher from both institutes. The umbrella of the collaboration agreement made it easy for scientists to visit the other institute and use the facilities there available.

Based on the success of this collaboration, measurable in the amount of scientific publications and in an increased awareness among the scientists of both institutes of available know-how and infrastructure, the management of JRC and SCK CEN decided to renew this collaboration and new projects are already running or in the process of being defined.

1 Introduction

The Belgian Nuclear Research Centre (SCK CEN) is a Foundation of Public Utility with a legal mission to perform nuclear research and development, innovate, educate, communicate and provide services within a perspective of sustainable development. In this regard, SCK CEN is performing research in radiation protection issues (including radiation biology, radiation medicine and radiation ecology), nuclear safety and security, waste management and decommissioning.

As the science and knowledge service of the European Commission, the mission of the Joint Research Centre (JRC) is to support EU policies with independent evidence throughout the whole policy cycle. Through its Directorate for Nuclear Safety and Security located in Geel (Belgium), Ispra (Italy) Karlsruhe (Germany), and Petten (the Netherlands), the JRC conducts research in the fields of nuclear reactor and fuel safety; radioactive waste and spent fuel management, nuclear data for reactor and other applications; nuclear safeguards and security; emergency preparedness; radioprotection, dosimetry, decommissioning and nuclear science applications. JRC structures its collaboration with relevant nuclear research organisations in Member States under appropriate collaboration instruments, in order to ensure that JRC's research and training activities are aligned as appropriate with the national ones. Bringing scattered cooperation or joint projects under wider scope, more comprehensive frameworks allows for higher efficiency, easier follow-up, and appropriate visibility.

JRC and SCK CEN have, since the establishment of the Central Bureau for Nuclear Measurements in Geel in 1960, a long-standing scientific collaboration, in particular in nuclear data, infrastructure and nuclear fuel. Due to the proximity between the JRC-Geel site and the SCK CEN site in Mol, and due to the similar scope of activities, there are other aspects of common interest to be explored, such as the disposal of the radioactive waste generated in both sites, storage of nuclear materials, use of emergency rooms, share of site safety resources, etc. of mutual benefit for both organisations by allowing the development of synergies and collaborations in different areas of nuclear research.

The first Collaboration Agreement between the JRC and SCK CEN entered into force on 9 August 2016 for 5 years. The scope of cooperation covers scientific research, education and training, scientific outreach and seminars, in the following areas:

- Specific MYRRHA support programme
- Nuclear data for reactor and other applications
- Nuclear safeguards and security
- Nuclear fuel research
- Emergency preparedness, Radioprotection, Dosimetry and Medical Applications
- Nuclear infrastructure development and access to facilities
- Nuclear reactor safety

The agreement facilitates the access of researchers to the nuclear research infrastructures of the JRC and SCK CEN.

This report summarizes the activities and main outcomes of the first Collaboration Agreement between JRC and SCK CEN.

2 Approved projects

Twenty projects were approved and carried out during the period 2016 – 2020. The methodology of implementation based on specific project descriptions with defined deliverables and time schedules using harmonised project sheets with reference to the scientific topics covered by the agreement has proven to be very effective. The projects together with the JRC Unit and SCK CEN expert group responsible for the project are listed in Table 1. A summary of each project is given in section 3.

The output in terms of number of papers in peer reviewed journals, contributions to conferences and workshops, books and theses are summarised in Table 2. More details about the publications are given in section 4.

Table 1. List of approved project sheets for the period 2016 - 2020

Research area Project	JRC Unit	SCK CEN, Institute/Expert group
<i>Specific MYRRHA support programme</i>		
Transmission measurements on Bi at GELINA	G.2	ANS/NSP
Improving nuclear data for MYRRHA	G.2	ANS/NSP
Uncertainty quantification and propagation	G.2	ANS/NSP
Fast neutron detector development for measurements at VENUS-F	G.2	ANS/NSE
Release of fission products from lead-bismuth eutectic	G.I.3	ANS/RRE
Volatile radionuclide detection in the MYRRHA beamline	G.I.3	ANS/RRE
<i>Nuclear data for reactor and other applications</i>		
Capture cross section measurements for ²³⁵ U at BR1	G.2	ANS/NSE
Characterisation of Bi-samples by NRCA at GELINA	G.2	ANS/NSE
Study of NDA observables to characterise spent fuel	G.2, G.II.7	ANS/NSP, NMS/FMA
Neutron measurements on spent nuclear fuel pellets	G.2	ANS/FMA, NMS/NSP
<i>Nuclear safeguards and security</i>		
Uranium enrichment and plutonium isotopics	G.2	EHS/NST
<i>Emergency preparedness, Radioprotection, Dosimetry and Medical Applications</i>		
Certification of an environmental reference material	G.2	EHS/LRM
Measurements of radon in water	G.2	EHS/LRM
Invariability of decay constants	G.2	EHS/LRM
Realisation of computer models of gamma-ray detectors	G.2	EHS/LRM
Measurements of gross alpha/beta activities in water	G.2	EHS/LRM
Radionuclide calibrators in nuclear medicine	G.2	EHS/LRM
<i>Education & Training and Communication</i>		
Capture cross section measurements for ²³⁵ U at BR1	G.2	SCK CEN Academy
ELINDER	G.2	SCK CEN Academy
STEM support via initiatives for high schools and other outreach activities	G.I.6, G.2, G.10	SCK CEN Academy

Table 2. Output produced as part of the projects listed in Table 1

Output	Number
Papers in peer reviewed journals	14
Contributions to conference proceedings or workshops	9
Technical Reports	4
Contributions to books	1
Master Thesis	1

3 Projects

3.1 Transmission measurements on Bi at GELINA

Scientific/technical contacts

JRC : J. Heyse (JRC.G.2)

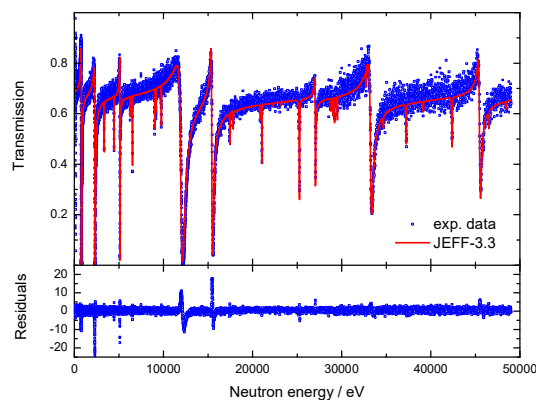
SCK CEN : A. Stankovskiy (ANS/NSP)

Summary

As a result of the CHANDA project and activities within the project “Improving nuclear data for MYRRHA” and “Uncertainty quantification and propagation” recommendations were given to improve the nuclear data for neutron induced interactions with bismuth. The conclusion was to use the JENDL-4.0 library as a basis combined with results of inelastic scattering cross section data of Mihailescu et al. (Nuclear Physics A 799 (2018) 1 - 29) carried out at GELINA. These recommendations were adopted to produce a new data file for JEFF-3.3. To verify the quality of this file, in particular the resonance parameters in the energy region below 100 keV, measurements were carried out at the 50 m transmission station of GELINA. A 14 mm thick sample was produced and characterised at the JRC-Geel. To determine the experimental transmission together with its covariance information the AGS code was used. A resonance shape analysis was performed with REFIT to validate the quality of the resonance parameters file in JEFF-3.3. The results in Fig. 1 reveal shortcomings in particular to describe the resonance profiles of strong s-wave resonances.

To improve the resonance parameters a simultaneous analysis of transmission and capture cross section data is required. Therefore, we propose to extend this task sheet and complement the transmission data with: (1) additional transmission data using a sample optimised for the strong s-wave resonances and (2) capture cross section measurements at GELINA. These measurements are complementary to the cross section measurements that have been carried out recently by JRC-Geel and SCK CEN at the ANNRI facility of J-PARC to improve the cross section data for the $^{209}\text{Bi}(n,\gamma)$ reaction to the ground state and the isomeric state. A new project sheet will be defined to cover the production of a new evaluation for neutron interactions with ^{209}Bi , which is also part of the JRC-Geel and SCK CEN contribution to the SANDA project (<http://www.sanda-nd.eu/>).

Figure 1. Comparison of the experimental and theoretical transmission together with the residuals. The theoretical transmission is calculated using the parameters for ^{209}Bi recommended in JEFF-3.3, which already includes the recommendations resulting from the CHANDA project.



Publications

The data obtained within this project are not yet published. A publication makes only sense after a simultaneous analysis of transmission and capture cross section data. However, a report to submit the transmission data to the EXFOR library is in preparation.

3.2 Improving nuclear data for MYRRHA

Scientific/technical contacts

JRC : G. Žerovnik (JRC.G.2)

SCK CEN : A. Stankovskiy (ANS/NSP)

Summary

The objective of this project is to improve nuclear data that are required for the design and safe operation of MYRRHA. Several tasks have been defined. A sensitivity study of the main reactor safety parameters such as the effective neutron multiplication factor and effective delayed neutron fraction to nuclear data (e.g. energy dependent cross sections, fission spectra and neutron multiplicities) was conducted for the latest MYRRHA designs, including a MOX critical core model. This resulted in a list of nuclides and reactions that are relevant for MYRRHA. Sensitivity profiles, i.e. sensitivities as a function of neutron energy, were derived for both critical and sub-critical cores using codes that are based on different methodologies including stochastic and deterministic calculations (i.e. SCALE, MCNP and XSUN). In a second task, the quality of the present evaluated files (i.e. JEFF, JENDL, ENDF/B and TENDL) was studied using experimental data available in the literature. Additionally, a list of integral benchmarks that can be used for validation of future nuclear data for MYRRHA was produced. Based on these studies recommendations were given to improve the release of the JEFF-3.3 library published by Plompen et al. (European Physics Journal A 56 (2020) 181). In addition, recommendations to improve nuclear data for several nuclides and reactions of interest for MYRRHA were given.

We propose to keep this project open to keep MYRRHA a point of interest for the nuclear data community, in particular the JEFF community, which is currently working on the development of the JEFF-4 release of the library.

Publications

Journal papers

Romero et al., “Nuclear data sensitivity and uncertainty analysis of effective neutron multiplication factor in various MYRRHA core configurations”,
Annals of Nuclear Energy 101 (2017) 330

Plompen et al., “The Joint Evaluated Fission and Fusion Nuclear Data Library, JEFF-3.3”,
European Physics Journal A 56 (2020) 181

Contributions to conference proceedings and workshops

Romero et al., “Neutron-induced nuclear data for the MYRRHA fast spectrum facility”,
European Physics Journal Web of Conferences 146 (2017) 09007

Žerovnik et al., “Systematic effects on cross section data derived from reaction rates in reactor spectra and a re-analysis of ^{241}Am reactor activation measurements”,
Nuclear Instruments and Methods A 877 (2017) 300

Technical Reports

Žerovnik et al., “Assessment of evaluations for MYRRHA”,
JRC Technical Reports, EUR 28240 EN (2016)

Žerovnik et al., “Recommendations for MYRRHA relevant cross section data to the JEFF project”,
JRC Technical Reports, EUR 28957 EN (2017)

3.3 Uncertainty quantification and propagation

Scientific/technical contacts

JRC : G. Žerovnik (JRC.G.2)

SCK CEN : L. Fiorito (ANS/NSP)

Summary

SANDY is a nuclear data sampling code which was developed at SCK CEN. The code is compatible with nuclear data files in ENDF-6 format. Exploiting the basic theory of stochastic sampling, SANDY generates random nuclear data files to produce covariance information stored in the ENDF-6 files. The use of the code has been tested at JRC Geel.

SANDY was tested for the generation of random ENDF-6 files with perturbed cross sections and resonance parameters. These files were used for an uncertainty quantification of integral responses, such as one-group integrated cross sections and resonance integrals of neutron induced reaction cross sections, and to test its capabilities for sensitivity studies. Results obtained with SANDY were fully consistent with those from the ERRORR module of NJOY, which relies on a deterministic methodology. The results of this exercise are described by Fiorito et al. (Annals of Nuclear Energy 101 (2017) 359).

In addition, the code was used to assess the uncertainty associated with the ^{210}Po production in MYRRHA. The impact of using different nuclear data libraries was evaluated. The uncertainty of the amount of ^{210}Po was determined by sampling from the nuclear data covariance matrices with SANDY. In addition, estimates of the sensitivity profiles were obtained with a linear regression approach. This study revealed a large source of uncertainty of the ^{210}Po production related to nuclear data for the $^{209}\text{Bi}(n,\gamma)$ reaction to the ground state. This is mainly due to a lack of experimental data for the cross section of the $^{209}\text{Bi}(n,\gamma)$ reaction, in particular the total capture cross section and the branching ratio for the $^{209}\text{Bi}(n,\gamma)$ reaction to the ground state and the isomeric state. Therefore, new capture cross section data as function of energy for this reaction are required.

The SANDY code was used for a sensitivity/uncertainty analysis applied to reactor criticality and nuclear fuel depletion problems. A sensitivity analysis was performed to identify what role the actinides' reaction cross sections play on the evolution of the nuclear fuel composition in a neutron irradiation environment representative of a typical PWR. The results are published in a peer-reviewed journal authored by Fiorito et al. (Annals of Nuclear Energy 161 (2021) 108415).

The results of this project show that the SANDY code is a powerful tool for sensitivity and uncertainty analysis. This project is finalised.

Publications

Journal papers

Fiorito et al., "Nuclear data uncertainty propagation to integral responses using SANDY",
Annals of Nuclear Energy, 101 (2017) 359

Fiorito et al., "Nuclear data uncertainty analysis for the Po-210 production in MYRRHA",
European Physics Journal Nuclear Science and Technology, 4 (2018) 48

Fiorito et al., "On the use of criticality and depletion benchmarks for verification of nuclear data",
Annals for Nuclear Energy 161 (2021) 108415

3.4 Fast neutron detector development for measurements at VENUS-F

Scientific/technical contacts

JRC : J. Heyse (JRC.G.2)

SCK CEN : J. Wagemans (ANS/NSE)

Summary

The objective of this project was to manufacture and calibrate a fast neutron detector, using high-purity ^{238}U base material available at JRC-Geel and combining the different fields of expertise available at JRC-Geel and SCK CEN. This detector can be used for experiments at the VENUS-F fast neutron facility of the SCK CEN to optimise the subcriticality measurement technique that has been developed for the licensing of MYRRHA. These experiments help also to validate nuclear data needed for modelling key parameters of the fast reactor systems like VENUS-F and MYRRHA.

A fission chamber was manufactured using high-purity ^{238}U base material provided by the JRC Geel. The relative amount of ^{238}U in the original material was 0.99998. The fission chamber consists of an aluminium body with a ^{238}U deposit length of 21.1 cm. A gas mixture of Ar + 4% N_2 is used as filling gas. The chamber has been calibrated in the standard irradiation fields of the BR1 reactor at SCK CEN.

The data obtained in the thermal and prompt fission neutron field suggest that the final deposit does not correspond with the characteristics of the original base material. Therefore, the material used for the construction of the detector has been re-analysed at the JRC-Geel. The results of this analysis confirm the observation of a higher amount of ^{235}U content than originally considered.

In any case, the purity of the new fission chamber is better than other chambers available at the VENUS-F facility. The newly manufactured fission chamber has been used for the subcriticality measurements at VENUS-F. The experimental results show an improved detector response, enabling a more accurate determination of the reactor reactivity.

This project is finalised.

Publications

Contributions to conference proceedings and workshops

Wagemans et al., "Fast neutron detector development for measurements at the VENUS-F reactor", EUFRAT user meeting, JRC Geel, 4 – 7 December 2017, pp. 89 – 91 (2017)

3.5 Release of fission products from lead-bismuth eutectic

Scientific/technical contacts

JRC : O. Benes (JRC.I.3)

SCK CEN : A. Aerts (ANS/RRE)

Summary

The objective of this project is to determine the migration of fission products (Cs, I, Te and Mo) from lead bismuth eutectic (LBE) coolant. The results of these studies will serve as input for the safety assessment of LBE cooled reactors such as MYRRHA.

At SCK CEN, it was foreseen to construct a Knudsen Effusion Mass Spectrometer (KEMS) with electrochemical cell for *in situ* oxygen control/measurement. However, it was decided to use the Molecular Beam Mass Spectrometry (MBMS) technique instead. This method allows measurement of release of fission products at ambient pressure, and is therefore complementary to the KEMS experiments at JRC, which are performed in vacuum. The MBMS installation has been designed and built, and initial tests are ongoing.

At JRC, fission product containing LBE samples will be measured by KEMS for determination of release mechanism of selected fission products from the LBE coolant. The work will include investigation of various fission product concentrations. The vapour pressure of LBE will be determined simultaneously.

The activities proposed in this collaboration are ongoing and included in the recently started projects PATRICIA and PASCAL in the Horizon 2020 EU Framework Programme.

3.6 Volatile radionuclide detection in the MYRRHA beamline

Scientific/technical contacts

JRC : J.-Y. Colle (JRC.I.3)

SCK CEN : A. Aerts (ANS/RRE)

Summary

The objective of this project is to determine transport and detection of mercury in an evacuated steel tube in conditions relevant to the MYRRHA proton beam line. The results of these studies will serve as input for the safety assessment of ADS such as MYRRHA, specifically for a beam window leak accident scenario.

A scaled MYRRHA beamline was designed, and constructed at JRC. A first series of experiments has been performed, using pure mercury as well as mercury-doped LBE as source, in both pulsed and continuous source modes. An SCK CEN scientist visited JRC Karlsruhe to help performing the experiments. Detailed MOLFLOW simulations have started.

The activities proposed in this collaboration are ongoing and included in the recently started project PATRICIA in the Horizon 2020 EU Framework Programme.

3.7 Capture cross section measurements for ^{235}U at BR1

Scientific/technical contacts

JRC : P. Schillebeeckx (JRC.G.2)

SCK CEN : J. Wagemans (ANS/NSE)

Summary

The objective is to determine the $^{235}\text{U}(n,\gamma)$ cross section in a thermal neutron field. This measurement is part of a series of experiments to determine the $^{235}\text{U}(n,\gamma)$ cross section as a function of incident neutron energy.

An experimental determination of the $^{235}\text{U}(n,\gamma)$ cross section is extremely difficult. Methods based on the detection of prompt γ -rays are complex due to the competition between gamma-rays originating from the $^{235}\text{U}(n,\gamma)$ and $^{235}\text{U}(n,f)$ reactions. One of the most accurate methods is a combination of activation measurements and atom counting of the reaction products using Accelerator Mass Spectrometry (AMS). By combining measurements at facilities with different neutron spectra the energy dependence of the cross section can be obtained. Therefore, the results of previous measurements at a beam with a Maxwellian spectrum with $kT = 25$ keV and 426 keV will be combined with results from similar measurements at a cold neutron beam of the ILL Grenoble, FRM II Munich and the thermal neutron field in the BR1 reactor at SCK CEN.

The experiments at BR1 are finalised:

- samples were prepared at JRC-Geel
- irradiation of a ^{235}U sample and ^{197}Au foils were carried out at the BR1 reactor
- the ^{197}Au foils were analysed by γ -ray spectrometry at SCK CEN and JRC-Geel to determine the neutron fluence during the irradiation period
- the irradiated ^{235}U sample together with additional reference materials from JRC Geel to calibrate the mass spectrometers were sent to the University of Vienna for an AMS analysis.

The project sheet will be closed once the AMS results are available and the final results are reported in a journal paper.

Publications

Contributions to conference proceedings and workshops

Wallner et al., "Precise measurement of the neutron capture cross section and capture-to-fission ratio of ^{235}U at thermal and sub-thermal neutron energies",
Intern. Conf. on Nuclear Data for Science and Technology, Beijing (China), 19 – 24 May 2019

3.8 Characterisation of Bi-samples by NRCA

Scientific/technical contacts

JRC : J. Heyse (JRC.G.2)

SCK CEN : A. Krása (ANS/NSE)

Summary

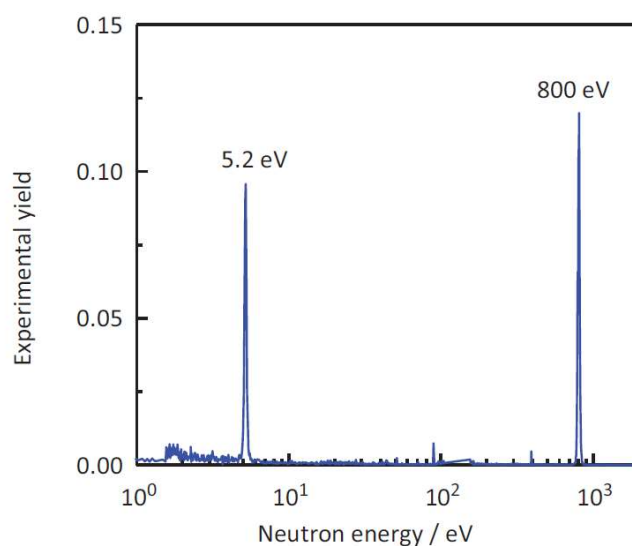
The objective is to characterize bismuth samples by NRCA (Neutron Resonance Capture Analysis) at the GELINA facility of the JRC-Geel. The samples are representative for the Bi-material that has been used for integral benchmark experiments at the VENUS-F facility of the SCK CEN at Mol. The Bi-material was already characterised by Neutron Activation Analysis (NAA) at SCK CEN and by Inductively Coupled Plasma Mass Spectrometer (ICP-MS) by the provider of the material. For some elements with a relatively high probability for neutron absorption a substantial difference was observed.

NRCA experiments at GELINA were carried out at the 12.5 m capture station of GELINA. The total energy detection principles in combination with the pulse height weighting technique was applied. The data reduction and analysis is finalized. Figure 1 shows the experimental yield as a function of incident neutron energy. These data confirm that the relative amount of Ag in the Bi samples is more than the 0.5 ppm declared by the provider. A resonance shape analysis of the data results in an abundance of 30 (5) ppm.

The results have been reported at the EUFRAT workshop organized in 2017 at JRC Geel and at the 16th International Symposium on Reactor Dosimetry.

The project is finalised.

Figure 2. Experimental capture yield as a function of incident neutron energy resulting from NRCA measurements with a Bi sample.



Publications

Contributions to conference proceedings and workshops

Krásá et al., "Characterisation of Bi-samples by Neutron Resonance Capture Analysis", EUFRAT user meeting, JRC Geel, 4 – 7 December 2017, pp. 53 – 55 (2017)

3.9 Study of NDA observables to characterise nuclear fuel

Scientific/technical contacts

JRC : P. Schillebeeckx (JRC.G.2)

SCK CEN : L. Fiorito (ANS/NSP)

Summary

The main objective of this project was to study the main nuclear data that have an impact on the results of Non-Destructive Analysis (NDA) methods to characterise Spent Nuclear Fuel (SNF) material. NDA measurements performed on samples with a well-known irradiation history can be used to verify the quality of depletion code calculations. They can also be applied to verify the declared history of a SNF assembly prior to final disposal.

The main observables or source terms of interest are: the decay heat, the neutron emission rate, the gamma-ray emission spectra, and the nuclide inventory for a criticality safety analysis. In this project the nuclides contributing to these observables were identified. The list was established as a function of cooling time. Estimates of confidence limits due to nuclear data uncertainties were given. For this study the SERPENT code was used. The results of this work were summarised in a report and a conference contribution.

To finalise the work a more extensive sensitivity analysis is required. Such a study is part of the activities that are foreseen in Task 2 of the work package “Spent Fuel Characterisation and Evolution Until Disposal” (WP SFC) of the EURAD Horizon 2020 project. Therefore, we propose to close this project sheet and define a new project sheets to reflect the JRC and SCK CEN contribution to the SFC WP.

Publications

Contributions to conference proceedings and workshops

Žerovnik et al., “Characterisation of spent pressurized water reactor (PWR) fuel for decay heat, neutron and gamma-ray emission: code comparison”,
Proceeding of the International Conference on Mathematics and Computational Methods applied to Nuclear Science and Engineering (M&C 2019), pp. 2736 – 2745 (2019)

Schillebeeckx et al., “Characterisation of spent nuclear fuel by theoretical calculations and non-destructive analysis”,
Proceedings of the International Workshop on Numerical Modelling of NDA Instrumentation and Methods for Nuclear Safeguards, Luxembourg, 16 – 17 May 2018, pp. 101 – 113 (2018)

Technical Reports

Žerovnik et al., “Observables of interest for the characterisation of Spent Nuclear Fuel”,
JRC Technical Reports, EUR 29301 EN (2018)

3.10 Neutron measurements on spent fuel pellets

Scientific/technical contacts

JRC : P. Schillebeeckx (JRC.G.2)

SCK CEN : M. Verwerft (ANS/FMA)

Summary

The main objective was to directly determine the neutron emission rate of spent nuclear fuel (SNF) by a direct Non-Destructive Analysis (NDA) method with high accuracy, and to investigate which would be the way forward in the analysis strategy if further improvements were to be aimed at. In practice, a target was set to determine the neutron emission rate by NDA with an uncertainty of 2% at the 1 sigma level, and to use an existing detector for which an additional shielding was to be developed to enable the handling of highly active spent nuclear fuel.

Special procedures were developed to avoid any contamination and excess dose rate related to the handling of spent nuclear fuel. The method relies on a transfer procedure that is adapted to the hot-cell facilities at the LHMA laboratory of the SCK CEN. A sample capsule was designed and fabricated at SCK CEN to seal a short segment of a SNF rod. The capsule together with an additional DENSIMET® shielding was inserted into an existing lead transport container of SCK CEN, modified for insertion in the neutron detection device of the AWCC type available at JRC-Geel.

To verify the dose rate and performance characteristics of the detection system a series of Monte Carlo simulations were carried out. The results of the dose rate calculations were confirmed by experiments at JRC Ispra using a 37 GBq ¹³⁷Cs source. The results of the efficiency estimations were verified by measurements with a ²⁵²Cf and ²⁴¹AmBe radionuclide source at JRC Ispra. The measured detection efficiency proved to be lower compared to the simulated one (ratio experimental/calculated ≈ 0.85). To verify the reason for the difference between calculated and measured detection efficiency, the impact of the source position, polyethylene density of the device, detector performance, electronics, HV settings and stability was verified. None of these effects could explain the difference.

In October 2018 measurements were performed at the LHMA facility of SCK CEN. The sample was a segment taken from a SNF rod that was irradiated in the Tihange 1 PWR reactor to a burnup of around 50 GWd/t. The composition and design specifications of this fuel rod and the irradiation conditions are well documented. The experiments with the SNF sample were successful. They confirm a slightly lower detection efficiency for spontaneous fission neutrons from ²⁴⁴Cm. The emission rate of prompt fission neutrons and α -ratio derived from the measured totals and reals rate resulted in a neutron emission rate for spontaneous fission of $S_{sf} = 24505 (375) \text{ s}^{-1}$ and a ratio between neutrons due to (α ,n) reactions and spontaneous fission of $\alpha = 0.036 (15)$, respectively. Additional measurements using certified ²⁵²Cf-sources, which were calibrated in the Mn-bath at NPL (UK), were carried out at JRC-Ispra to determine the detection efficiency. An analysis of these data together with a comparison of results of depletion calculations using ALEPH2, SCALE and SERPENT is in progress.

Based on the positive outcome of these measurements a new optimized system with a higher detection efficiency, improved detection efficiency profile, shorter dead time and better gamma-ray shielding is being designed. We expect to obtain good quality measurements for a broad range of spent fuels, including high neutron emitting MOX fuels or fuels with low neutron emission and comparatively high gamma-ray emission (e.g. spent fuel with short cooling time, low burnup fuels, UO₂-Gd₂O₃, ...).

Once a journal paper is produced that describes the result of the measurement together with results from depletion code calculations, we consider this task sheet as finalised. Future work with the new optimised measurement system will be part of a new task sheet that covers the experimental activities which are aligned with the contribution of SCK CEN and JRC-Geel to Task 2 of the work package "Spent Fuel Characterisation and Evolution Until Disposal" (WP SFC) of the EURAD Horizon 2020 project.

Publications

Contributions to conference proceedings and workshops

Schillebeeckx et al., "Characterisation of spent nuclear fuel by theoretical calculations and non-destructive analysis",
Proceedings International Workshop on Numerical Modelling of NDA Instrumentation and Methods for Nuclear Safeguards, Luxembourg, 16 – 17 May 2018, pp. 101 – 113 (2018)

Technical Reports

Schillebeeckx et al., "A non-destructive method to determine the neutron production rate of a sample of spent nuclear fuel under standard controlled area conditions",
JRC Technical Report, EUR 30379 EN (2020)

3.11 Uranium enrichment and plutonium isotopics

Scientific/technical contacts

JRC : P. Schillebeeckx (JRC.G.2)

SCK CEN : A. Borella (EHS/NST)

Summary

The objective was to produce experimental gamma-ray spectroscopy data from measurements of uranium and plutonium reference standards using different types of gamma-ray detectors. The U_3O_8 reference material set EC NRM 171 and PuO_2 reference material set EC NRM 271 were used in the project. The detector types included: high-purity Ge-detector, CdZnTe (CZT) solid state detectors and $LaBr_3$ and CeBr scintillators. In addition, spectroscopic data with calibrated point sources were used to determine experimental response functions.

Spectra were taken at JRC-Geel with a HPGe coaxial detector and CZT solid state detector and with a BGO; $LaBr_3$ and CeBr scintillator. For these measurements special collimators were constructed by JRC-Geel. The scintillators and HPGe detector were coupled to a conventional spectroscopic chain using standard analog electronics and the DAQ2000 system of the JRC-Geel.

A 10 mm x 10 mm x 5 mm quasi-hemispherical CZT detector was coupled to a digitizer produced by GBS-Elektronik. The spectra were analysed using response functions that were determined separately from measurements with radionuclide sources. Monte Carlo simulations were used to determine the energy dependence of the intrinsic gamma-ray detection efficiency and the gamma-ray attenuation in the sample. The enrichment was determined relying on a multi-peak spectrum analysis. A detailed description of this work is given in Borella et al. (Nuclear Instruments and Methods A 986 (2021) 164718). Triggered by the results described in this paper, we were invited to provide a chapter on “CZT detectors for safeguards applications” in a book entitled “Semiconductor Radiation Detection Systems” published by CRC Press/Routledge.

The project is finalised. We intend to open a new project sheet related to the development and use of innovative detection systems for nuclear safeguards applications.

Publications

Journal papers

Borella et al., “Peak shape calibration of a Cadmium Zinc Telluride detector and its application for the determination of uranium enrichment”,

Nuclear Instrument and Methods A986 (2020) 164718

Contributions to conference proceedings and workshops

Meleshenskii et al., “Measurements on U and Pu standards with medium resolution gamma-ray spectrometers”,

EUFRAT user meeting, JRC Geel, 4 – 7 December 2017, pp. 92 – 94 (2017)

Borella et al. “Peak Shape Characterization of a 500 mm³ Cadmium Zinc Telluride Detector and Analysis of Spectroscopic Measurement Data for Uranium Samples”,

Proceedings of the International Workshop on Isotopic Analysis of Uranium and Plutonium by Nondestructive Assay Techniques for Nuclear Safeguards, IAEA, 16-19 February 2021

Contributions to a book

Schillebeeckx et al., “Cadmium Zinc Telluride detectors for safeguards applications”,

Chapter in Semiconductor Radiation Detection Systems, 2nd Edition, edited by CRC Press/Routledge

3.12 Certification of an environmental reference material

Scientific/technical contacts

JRC : V. Jobbágy (JRC.G.2)

SCK CEN : M. Bruggeman and M. Vasile (EHS/LRM)

Summary

The objective of the collaboration was to give support to JRC-Geel to be able to do a certification work of an IAEA reference material for radionuclides in sediment collected from the Baltic Sea. JRC-Geel had no available environmental tracers to start the work at that time and the transport of these materials was predicted to be too long. Therefore, there was a risk to miss the deadline for result submission. SCK CEN kindly provided the necessary tracer materials, thus JRC-Geel was able to start the nuclide specific analytical work in time and deliver the results before the submission deadline. The radionuclides that were determined by alpha-spectrometry were: ^{210}Po , ^{234}U , ^{235}U , ^{238}U , ^{238}Pu , $^{239+240}\text{Pu}$, ^{228}Th , ^{230}Th and ^{232}Th .

The final deliverable will be an IAEA certified reference material for environmental radioactivity analysis which helps to maintain and improve the quality assurance of laboratories in this field.

Publications

The production of a certification report is in progress

3.13 Measurements of radon in water

Scientific/technical contacts

JRC : V. Jobbágy (JRC.G.2)

SCK CEN : M. Bruggeman and M. Vasile (EHS/LRM)

Summary

The objective of the joint project was to collaborate in radioactivity measurements of environmental samples, i.e. the European Proficiency Test (PT) exercise focusing on radon in water measurements. With the PTs the Euratom Treaty Article 35 (and 39) are supported by comparing measurement results and verifying data submitted by EU Member States (following Article 36). SCK CEN was involved in the following tasks:

- Pilot-PT 2017 participant as expert laboratory,
- REM2018 PT participant,
- Determination of radon massic activity in the PT water samples using three most common radon in water measurement methods (liquid scintillation counting, gamma-ray spectrometry and emanometry),
- Contributing to the JRC radon in water workshop in March 2019.

SCK CEN submitted measurement results from all three routine methods so that all the common methods included in the ISO standards were covered. The additional benefit of the SCK CEN measurements was that they identified some method specific pitfalls related to the radon in water measurement methods. The measurement results from SCK CEN were not included in the reference value but used to verify the JRC measurement results. We would like to keep the collaboration open and involve SCK CEN in other environmental proficiency tests where complimentary measurements and expertise are needed.

Publications

Contributions to conference proceedings and workshops

Presentations at the Workshop on “EC REM 2018 Radon-in-Water Proficiency test” at JRC-Geel, Belgium, 26 – 29 March 2019:

- Michel Bruggeman, “Gamma-ray spectrometry of radon-in-water”
- Edmond Dupuis, “Radon via emanation method-new developments”
- Mirela Vasile, “Validation of the ISO13164-4 method for the determination of ^{222}Rn in water: the LRM approach”
- Viktor Jobbágy, “Results of the REM2018 PT”
- Viktor Jobbágy, “Pilot-PT2017”

3.14 Invariability of decay constants

Scientific/technical contacts

JRC : S. Pommé (JRC.G.2)

SCK CEN : M. Bruggeman and L. Verheyen (EHS/LRM)

Summary

The objective was to demonstrate that decay constants are invariable, contrary to claims in literature that solar neutrinos induce beta decay and that variations in decay rates can be used as a neutrino flux monitor.

In the last decade, controversy arose about claimed violations of the exponential-decay law, since experimental decay curves showed unexplained cyclic deviations at the permille level. New theories with high aspirations – involving a 5th force! – were proposed to explain that decay can be induced by solar and cosmic neutrinos. Consequently, decay constants would not be fixed values – as believed for more than a century – but variables sensitive to changes in neutrino flux. Patents were issued to use repeated beta decay measurements as an alternative to elaborate solar neutrino flux monitoring experiments. Others patented methods to reduce radioactive waste by shortening the half-lives of long-lived nuclides. Questions were raised about the validity of nuclear dating in archaeology and geo- & cosmochronology. The very foundation of radioactivity measurements was affected, since direct comparison of measurements is only possible through normalisation to a reference date via the exponential-decay law. International equivalence of standards for the SI unit becquerel implicitly assumes the invariability of the decay constants, therefore would require an adjustment if 'neutrino-induced radioactivity' would be sensitive to variations in neutrino flux, e.g. through seasonal changes in the distance Earth-Sun.

The JRC called for an international collaboration through the Consultative Committee for Ionizing Radiation, CCRI(II), to collect the most extensive set of long-term activity measurements. The SCK CEN participated with long-term data sets of activity measurements on 8 different HPGe gamma-ray spectrometers. In phase I of the project, these data were used to demonstrate that small annual cyclic effects in the measurements were caused by environmental influences and not by variations in the solar neutrino flux resulting from the annual variations in the Earth-Sun distance.

In phase II, counterevidence was provided with the same data sets of new claims of monthly cycles in residuals from exponential decay supposedly being related with rotation of the solar interior. A paper was presented in the opening session of ICRM-2017 and published in ARI. It is the most downloaded paper of the journal in 2019. The project is finalised, since the counterevidence provided by JRC-Geel, SCK CEN and others was of superior quality and could not be ignored by those making unsubstantiated claims.

Publications

Journal papers

Pommé et al., "Evidence against solar influence on nuclear decay constants",
Physics Letters B 761 (2016) 281-286

Pommé et al., "On decay constants and orbital distance to the sun – part I: alpha decay",
Metrologia 54 (2017) 1-18

Pommé et al., "On decay constants and orbital distance to the sun – part II: beta minus decay",
Metrologia 54 (2017) 19-35

Pommé et al., "On decay constants and orbital distance to the sun – part III: beta plus and electron capture decay",
Metrologia 54 (2017) 36_50

Pommé et al., "Is decay constant?",
Applied Radiation and Isotopes 134 (2018) 6-12

3.15 Realisation of computer models of gamma-ray detectors

Scientific/technical contacts

JRC : M. Hult (JRC.G.2)

SCK CEN : M. Bruggeman (EHS/LRM)

Summary

Both SCK CEN and JRC-Geel are using numerous HPGe-detectors for gamma-ray spectrometry. Today, many correction factors (e.g. for coincidence summing and efficiency transfer) are calculated using advanced computer algorithms involving Monte Carlo simulations. Such simulations require that a good model of the detector is realized in the computer code. The aim of this work was to better characterize a few detectors at SCK CEN and JRC-Geel and to build and test the computer codes of the models created in EGS-nrc and EFFTRAN. The project was extended by using the scanning-facility in HADES to measure the thickness-variations of the top deadlayer in three detectors. The work resulted in a Master-Thesis and a scientific article. The key conclusion is quite revolutionary as it can show that the Li-diffused deadlayers significantly change over time when the detector is kept at room-temperature. One impact of this work is that the detector producer Mirion (Olen) now sends a recommendation to users to keep the HPGe-detector cold at all times to avoid Li-diffusion from contacts. The alternative is to perform a new efficiency calibration after a detector has been kept warm for a significant time.

Publications

Journal papers

Hult et al., "Determination of homogeneity of the top surface deadlayer in an old HPGe detector", Applied Radiation and Isotopes, 147 (2019) 182-188

Master Thesis

Stef Geelen, "Study of parameters influencing the response of HPGe-detectors", Master Thesis, Hasselt University, June 2018

3.16 Radionuclide calibrators in nuclear medicine

Scientific/technical contacts

JRC : S. Pommé (JRC.G.2)

SCK CEN : C. Saldaria Vargis (EHS/LRM)

Summary

The objective was to provide reference values for proficiency tests on the accuracy of radionuclide calibrators in nuclear medicine.

Nuclear medicine is an invaluable tool for diagnostic and therapeutic purposes. The most widely used radionuclide is ^{99m}Tc , accounting for approximately 80% of all nuclear medicine examinations and about 90% of those used for diagnostic purposes. In 2008, the world total number of procedures performed with ^{99m}Tc was estimated to range between 25 and 30 million annually, with 6–7 million of them taking place in Europe. Other frequently used radionuclides for nuclear medicine in Europe are ^{18}F , ^{201}Tl , ^{123}I , ^{131}I , ^{67}Ga and ^{111}In and the future holds an increasing interest in radionuclides for targeted therapy and theranostics, such as *e.g.* ^{177}Lu , ^{90}Y , ^{223}Ra , ^{225}Ac and Tb isotopes.

In the last decades, the application of personalized molecular radiotherapy using theragnostics has gained a lot of interest in nuclear medicine. Theragnostic approaches aim to optimize molecular radiotherapy for individual patients using pre-therapeutic diagnostic imaging. In particular, assessment of the therapeutic absorbed dose to malignant tissue and to organs at risk based on these images facilitates a personalized therapeutic activity approach. These approaches require accurate quantification of the activity administered to patients both in diagnostic and therapeutic applications. Accurate activity calibration of radionuclide imaging equipment such as SPECT and PET cameras is also essential in theragnostics, to enable an accurate estimation of radiopharmaceutical uptake in patient tissues.

Administration of the correct activity of these radiopharmaceuticals is necessary for the optimization of patient healthcare. When handling radioactive substances, one applies the long-accepted ALARA principle to keep the doses “as low as reasonably achievable”. Articles 55, 56 and 60 of the European Council Directive 2013/59/EURATOM on the principles of justification and optimization of medical exposures imply proper calibration of all sources giving rise to medical exposure. After all, the use of diagnostic reference levels and the study of dose-effect relationships in therapeutic nuclear medicine procedures depend on the accuracy of the measurement of the activity to be administered.

The activity of radiopharmaceuticals is quantified prior to patient administration by means of a radionuclide calibrator (RC). This instrument consists of a well-type gas-filled ionisation chamber coupled to a high voltage supply, an electrometer, and a display unit. When a radioactive sample is introduced inside the chamber, an ionisation current is produced from the interactions between ionising radiation and the filling gas, the amount of which is proportional to the activity of the sample. The measured current is converted to an activity value by applying a calibration factor (CF) [$\text{pA}\cdot\text{MBq}^{-1}$]. Already at purchase, the RCs are provided with factory-set CFs for different radionuclides. The manufacturer uses radioactive sources in standard containers with specific volumes of solution, thus establishing traceability to primary standards for those specific measurement conditions.

A first study addressed the current status of performance of radiopharmaceutical activity measurements using radionuclide calibrators in Belgium. An intercomparison exercise was performed among 15 hospitals to test the accuracy of ^{99m}Tc , ^{18}F and ^{111}In activity measurements by means of radionuclide calibrators. Four sessions were held in different geographical regions between December 2013 and February 2015. The data set includes measurements from 38 calibrators, yielding 36 calibrations for ^{99m}Tc and ^{111}In , and 21 calibrations for ^{18}F . The reference value for the massic activity of the radioactive solutions was determined by means of primary and secondary standardisation techniques at the radionuclide metrology laboratory of the JRC. The overall results of the intercomparison were satisfactory for ^{99m}Tc and ^{18}F , since most radionuclide calibrators (> 70%) were accurate within $\pm 5\%$ of the reference value. Nevertheless, some devices underestimated the activity by 10–20%. Conversely, ^{111}In measurements were strongly affected by source geometry effects. Large overestimations (up to 72%) were observed. The results of this exercise encourage the hospitals to perform corrective actions to improve the calibration of their devices where needed.

In a new study, the first international proficiency test was held on the accuracy of radionuclide calibrators in nuclear medicine theragnostics. In total, 32 radionuclide calibrators from 8 hospitals located in the Netherlands, Belgium, and Germany were tested. For each radionuclide, a set of four samples comprising two clinical containers (10-mL glass vial and 3-mL syringe) with two filling volumes were measured. The reference value of each sample was determined by two certified radioactivity calibration centers (SCK CEN and JRC) using two secondary standard ionization chambers. The deviation in measured activity with respect to the reference value was determined for each radionuclide and each measurement geometry. In addition, the combined systematic deviation of activity measurements in a theragnostic setting was evaluated for 5 clinically relevant theragnostic pairs: $^{131}\text{I}/^{123}\text{I}$, $^{131}\text{I}/^{124}\text{I}$, $^{177}\text{Lu}/^{111}\text{In}$, $^{90}\text{Y}/^{99m}\text{Tc}$, and $^{90}\text{Y}/^{111}\text{In}$.

For ^{99m}Tc , ^{131}I , and ^{177}Lu , a small minority of measurements were not within $\pm 5\%$ range from the reference activity (percentage of measurements not within range: ^{99m}Tc , 6%; ^{131}I , 14%; ^{177}Lu , 24%) and almost none were outside $\pm 10\%$ range. However, for ^{111}In , ^{123}I , ^{124}I , and ^{90}Y , more than half of all measurements were not accurate within $\pm 5\%$ range (^{111}In , 51%; ^{123}I , 83%; ^{124}I , 63%; ^{90}Y , 61%) and not all were within $\pm 10\%$ margin (^{111}In , 22%; ^{123}I , 35%; ^{124}I , 15%; ^{90}Y , 25%). A large variability in measurement accuracy was observed between radionuclide calibrator systems, type of sample container (vial vs syringe), and source-geometry calibration/correction settings used. Consequently, we observed large combined deviations (percentage deviation $> \pm 10\%$) for the investigated theragnostic pairs, in particular for $^{90}\text{Y}/^{111}\text{In}$, $^{131}\text{I}/^{123}\text{I}$, and $^{90}\text{Y}/^{99m}\text{Tc}$.

Our study shows that substantial over- or underestimation of therapeutic patient doses is likely to occur in a theragnostic setting due to errors in the assessment of radioactivity with radionuclide calibrators. These findings underline the importance of thorough validation of radionuclide calibrator systems for each clinically relevant radionuclide and sample geometry.

Publications

Journal papers

Saldarriaga Vargas et al., "Intercomparison of ^{99m}Tc , ^{18}F and ^{111}In activity measurements with radionuclide calibrators in Belgian hospitals",
Physica Medica 45 (2018) 134-142

Saldarriaga Vargas et al., "An international multi-center investigation on the accuracy of radionuclide calibrators in nuclear medicine theragnostics",
European Journal of Nuclear Medicine and Molecular Imaging, Physics 7 (2020) 69

3.17 Common organization of workshops, events, topical days

Scientific/technical contacts

JRC : P. Schillebeeckx (JRC.G.2)

SCK CEN : M. Coeck (SCK CEN Academy)

Summary

This project sheet formalises and intends to fortify the common organisation of events (coursed/workshops/topical days) and participation of experts of both institutions in each other's events.

The events that were jointly organised during the period 2016 – 2020 are:

- Nuclear career day at SCK CEN on 28/02/2017 and 13/02/2019
- Topical day “From nuclear data to a reliable estimate of spent fuel decay heat” at SCK CEN on 01/06/2018
- Scientific JRC/SCK CEN progress meeting at JRC-Geel on 01/06/2018
- ELINDER-Training course on “Decommissioning of nuclear installations” at SCK CEN from 03/12/2018 to 07/12/2018
- Training course on “Nuclear safeguards: Pu-DA and NDA measurements” at JRC-Geel from 04/02/2019 to 06/02/2019
- Workshop on “Detection of neutrinos” at JRC-Geel on 23/05/2019

In addition during 2017 – 2018 a Nuclear Game Challenge was organised by SCK CEN with support of the JRC-Geel and JRC-Karlsruhe. Nearly 100 students from 17 different schools gathered for the Nuclear Game Challenge to create an interactive and creative game on the topic of nuclear science and applications.

This is an open project sheet that will continue.

4 Output: publications

The journal papers, contributions to conference proceedings or workshops and technical reports are listed in Table 3 – Table 5. The abstracts and related projects are specified in the ANNEX.

Table 3. Papers published in peer reviewed journals

Author	Title	Reference
Pommé et al.	“Evidence against solar influence on nuclear decay constants”	Physics Letters B, 761 (2016) 281-286
Pommé et al.	“On decay constants and orbital distance to the sun – part I: alpha decay”	Metrologia, 54 (2017) 1
Pommé et al.	“On decay constants and orbital distance to the sun – part II: beta minus decay”	Metrologia, 54 (2017) 19
Pommé et al.	“On decay constants and orbital distance to the sun – part III: beta plus and electron capture decay”	Metrologia, 54 (2017) 36
Romojaro et al.	“Nuclear data sensitivity and uncertainty analysis of effective neutron multiplication factor in various MYRRHA core configurations”	Annals of Nuclear Energy, 101 (2017) 330-338
Fiorito et al.	“Nuclear data uncertainty propagation to integral responses using SANDY”	Annals of Nuclear Energy, 101 (2017) 359
Žerovnik et al.,	“Systematic effects on cross section data derived from reaction rates in reactor spectra and a re-analysis of ^{241}Am reactor activation measurements”	Nuclear Instruments and Methods, A 877 (2018) 300
Fiorito et al.,	“Nuclear data uncertainty analysis for the Po-210 production in MYRRHA”	European Physics Journal Nuclear Science and Technology, 4 (2018) 48
Pommé, et al.	“Is decay constant?”	Applied Radiation and Isotopes, 134 (2018) 6
Saldarriaga Vargas et al.	“Intercomparison of $^{99\text{m}}\text{Tc}$, ^{18}F and ^{111}In activity measurements with radionuclide calibrators in Belgian hospitals”	Physica Medica, 45 (2018) 134
Borella et al.	Peak shape calibration of a Cadmium Zinc Telluride detector and its application for the determination of uranium enrichment”	Nuclear Instrument and Methods, A986 (2020) 164718
Plompen et al.,	“The Joint Evaluated Fission and Fusion Nuclear Data Library, JEFF-3.3”	European Physics Journal A 56 (2020) 181
Saldarriaga Vargas et al.	“An international multi-center investigation on the accuracy of radionuclide calibrators in nuclear medicine theragnostics”	European Journal of Nuclear Medicine and Molecular Imaging, Physics 7 (2020) 69

Table 4. Contributions to conference proceedings and workshops

Author	Title	Reference
Romojaro et al.	“Neutron-induced nuclear data for the MYRRHA fast spectrum facility”	Eur. Phys. J. Web of Conferences 146 (2017) 09007
Krásá et al.	“Characterisation of Bi-samples by Neutron Resonance Capture Analysis”	EUFRAT user meeting, JRC Geel, 4 – 7 December 2017, pp. 53 – 55 (2017)
Wagemans et al.	Fast neutron detector development for measurements at the VENUS-F reactor	EUFRAT user meeting, JRC Geel, 4 – 7 December 2017, pp. 89 – 91 (2017)
Meleshkovskii et al.	Measurements on U and Pu standards with medium resolution gamma-ray spectrometers	EUFRAT user meeting, JRC Geel, 4 – 7 December 2017, pp. 92 – 94 (2017)
Schillebeeckx et al.	Characterisation of spent nuclear fuel by theoretical calculations and non-destructive analysis	Proc. Intern. Workshop on Numerical Modelling of NDA Instrumentation and Methods for Nuclear Safeguards, Luxembourg, 16 – 17 May, pp. 101 – 113 (2018)
Žerovnik et al.	Characterisation of spent pressurized water reactor (PWR) fuel for decay heat, neutron and gamma-ray emission: code comparison	Proc. of the Intern. Conf. on Mathematics and Computational Methods applied to Nuclear Science and Engineering (M&C 2019), pp. 2736 – 2745 (2019)
Wallner et al.	Precise measurement of the capture cross section and capture-to-fission ratio of ^{235}U at thermal and sub-thermal neutron energies	Intern. Conf. on Nuclear Data for Science and Technology, Beijing (China), 19 – 24 May 2019
Borella et al.,	Peak Shape Characterization of a 500 mm ³ Cadmium Zinc Telluride Detector and Analysis of Spectroscopic Measurement Data for Uranium Samples	Proc. Intern. Workshop on Isotopic Analysis of Uranium and Plutonium by Nondestructive Assay Techniques for Nuclear Safeguards, IAEA, 16-19 February 2021

Table 5. Technical reports

Author	Title	Reference
Žerovnik et al.	“Assessment of evaluations for MYRRHA”	JRC Technical Reports, EUR 28240 EN (2016)
Žerovnik et al.,	“Recommendations for MYRRHA relevant cross section data to the JEFF project”	JRC Technical Reports, EUR 28957 EN (2017)
Žerovnik et al.	Observables of interest for the characterisation of Spent Nuclear Fuel	JRC Technical Reports, EUR 29301 EN (2018)
Schillebeeckx et al.	A non-destructive method to determine the neutron production rate of a sample of spent nuclear fuel under standard controlled area conditions	JRC Technical Report, EUR 30379 EN (2020)

5 Summary and conclusions

Thanks to a comprehensive collaboration agreement between JRC and SCK CEN, researchers from both institutes could profit from the complementary nature of the competences and infrastructure of JRC and SCK CEN. The definition of clear and concise project sheets allowed researchers a quick start of their projects and the steering committee an easy way to keep oversight. The clear definition of the goals in the project sheets allows a measurable success rate, which, as one can observe, is very high. Many projects have reached their end-goal in the five year program and those project sheets are formally closed.

The scientific output generated by the seventeen scientific projects during this five-year period is very high. In total, there were 14 articles published in peer-reviewed journals, 9 contributions to conference proceedings or workshops and 4 technical reports. All of these carry the names of the researchers from both institutes, highlighting the success of the collaboration.

By pooling scientific resources from both JRC and SCK CEN and with the help of the SCK CEN Academy for Nuclear Science and Technology, a number of workshops, seminars and other scientific outreach activities like the “Nuclear Game Challenge” could be organized. This increased the visibility of both JRC and SCK CEN in the scientific field but also for the young generation (hopefully) on their way to a career in nuclear science or engineering.

The collaboration agreement made it also easier for researchers from both institutes to reach out to each other and we believe that the awareness of availabilities in know-how and infrastructure at both parties has increased.

A point of attention is the limited involvement of master thesis and PhD students. The idea of common PhD proposals and/or common master thesis subjects could be promoted more with researchers from both institutes.

Based on the successful outcome of this first collaboration agreement, the management of JRC and SCK CEN have decided to renew this agreement. However, for practical reasons as to simplify the text and better take into account the current organizational structure of the JRC, it was decided that the renewal of the Collaboration Agreement would be done using a newly drafted agreement text and not as an amendment. This will also allow to incorporate the lessons learned from the first edition and define a broad framework that will provide flexibility to define the areas of work.

We, the authors of this report, are convinced that this strategic collaboration between the JRC and SCK CEN has proven to be very successful for both partners. We therefore welcome the next edition and will do all what lies in our possibilities to make it again a success story.

List of abbreviations and definitions

AGS	Analysis of Geel spectra
ALARA	As low as reasonably achievable
ALEPH	A Monte Carlo burnup code
AMS	Accelerator mass spectrometry
ANNRI	Accurate neutron-nucleus reaction measurement instruments
ANS	Advanced nuclear systems
ARIEL	Accelerator and research reactor infrastructures for education and learning
AWCC	Active well coincidence counter
BR1	Belgian reactor 1
BROND	Russian evaluated neutron <i>data</i> library
BUC	Burnup credit
BWR	Boiling water reactor
CBNM	Central bureau for nuclear measurements
CCRI	Consultative committee for ionizing radiation
CEA	Commissariat à l'Énergie atomique et aux Énergies alternatives
CERN	Organisation Européenne pour la Recherche Nucléaire
CF	Calibration factor
COMMARA	Neutron cross section covariance library
CHANDA	Solving challenges in nuclear data
CZT	Cadmium zinc telloride
DA	Destructive analysis
EC	European Commission
EHS	Environment, health and safety
ELINDER	European learning initiatives for nuclear decommissioning and environmental remediation
ENDF/B	Evaluated nuclear data file/B-version
EU	European Union
EUFRAT	European facility for innovative reactor and transmutation data
EURAD	European joint programme on radioactive waste management
EXFOR	0045change format for experimental numerical nuclear reaction data
FMA	Fuel materials
FP	Fission product
FRM II	Forschungs-neutronenquell Heinz Maier-Leibnitz
GELINA	Geel electron linear accelerator
HPGe	High purity germanium
ICRM	Institute for certified reference materials
ICP-MS	Inductively coupled plasma mass spectrometry
ILL	Institut Laue Langevin
JEFF	Joint evaluated fission and fusion file
JENDL	Japan evaluated nuclear data library
J-PARC	Japan proton accelerator research complex
JRC	Joint research centre
KEMS	Kundsen effusion mass spectrometer
LBE	Lead bismuth eutectic
LHMA	Laboratory for high and medium level activity
LRM	Low-level radioactivity measurements
MBMS	Molecular beam mass spectrometry
MCNP	Monte Carlo n-particle code
MOLFLOW	Monte Carlo code for simulating molecular flows
MONNET	Mono-energetic Neutron Tower
MOX	Mixed oxide
MYRRHA	Multi-purpose hybrid research reactor for high-tech applications
NAA	Neutron activation analysis
NDA	Non-destructive analysis
NJOY	Nuclear data processing system
NMS	Nuclear materials sciences
NSE	Nuclear systems exploitation
NSP	Nuclear system physics
NST	Nuclear science and technology

NRCA	Neutron resonance capture analysis
PASCAL	Proof of augmented safety conditions in advanced liquid-metal-cooled systems
PATRICIA	Partitioning and transmuter research initiative in a collaborative innovation action
PENDF	Point wise evaluated nuclear data file
PET	Positron emission tomography
PT	Proficiency test
PWR	Pressurized water reactor
RRE	Reactor research and engineering
SANDA	Solving challenges in nuclear data for the safety of European nuclear facilities
SANDY	Sampler of nuclear data and uncertainty
SCALE	Standardized Computer Analyses for Licensing Evaluation
SCK CEN	Belgian Nuclear Research Centre
SERPENT	A continuous-energy Monte Carlo reactor physics burnup calculation code
SFC	Spent fuel characterisation and evolution until disposal
SNF	Spent nuclear fuel
SPECT	single-photon emission computerized tomography
STEM	science, technology, engineering and mathematics
TENDL	TALYS-based evaluated nuclear data library
VENUS-F	A fast, lead-bismuth cooled reactor/accelerator-driven system
WP	Work package
XSUN	Windows interface environment for transport and sensitivity-uncertainty

List of figures

Figure 1. Comparison of the experimental and theoretical transmission together with the residuals. The theoretical transmission is calculated using the parameters for ^{209}Bi recommended in JEFF-3.3, which already includes the recommendations resulting from the CHANDA project.	5
Figure 2. Experimental capture yield as a function of incident neutron energy resulting from NRCA measurements with a Bi sample.	12

List of tables

Table 1. List of approved project sheets for the period 2016 - 2020	4
Table 2. Output produced as part of the projects listed in Table 1	4
Table 3. Papers published in peer reviewed journals.....	24
Table 4. Contributions to conference proceedings and workshops	25
Table 5. Technical reports.....	25

Annex Abstracts

Annex 1. Journal papers

Evidence against solar influence on nuclear decay constants	
Principal author JRC	Stefaan Pommé
Principal author SCK CEN	Leen Verheyen
Publication type	Journal paper
Reference	Physics Letters B 761 (2016) 281
Relevant project sheet	Invariability of decay constants
<p>The hypothesis that proximity to the Sun causes variation of decay constants at permille level has been tested and disproved. Repeated activity measurements of mono-radionuclide sources were performed over periods from 200 days up to four decades at 14 laboratories across the globe. Residuals from the exponential nuclear decay curves were inspected for annual oscillations. Systematic deviations from a purely exponential decay curve differ from one data set to another and are attributable to instabilities in the instrumentation and measurement conditions. The most stable activity measurements of alpha, beta-minus, electron capture, and beta-plus decaying sources set an upper limit of 0.0006% to 0.008% to the amplitude of annual oscillations in the decay rate. Oscillations in phase with Earth's orbital distance to the Sun could not be observed within a 10^{-6} to 10^{-5} range of precision. There are also no apparent modulations over periods of weeks or months. Consequently, there is no indication of a natural impediment against sub-permille accuracy in half-life determinations, renormalisation of activity to a distant reference date, application of nuclear dating for archaeology, geo- and cosmochronology, nor in establishing the SI unit becquerel and seeking international equivalence of activity standards.</p>	

On decay constants and orbital distance to the Sun—part I: alpha decay	
Principal author JRC	Stefaan Pommé
Principal author SCK CEN	Leen Verheyen
Publication type	Journal paper
Reference	Metrologia 54 (2017) 1
Relevant project sheet	Invariability of decay constants
<p>Claims that proximity to the Sun causes variation of decay constants at permille level have been investigated for alpha decaying nuclides. Repeated decay rate measurements of ^{209}Po, ^{226}Ra, ^{228}Th, ^{230}U, and ^{241}Am sources were performed over periods of 200 d up to two decades at various nuclear metrology institutes around the globe. Residuals from the exponential decay curves were inspected for annual oscillations. Systematic deviations from a purely exponential decay curve differ in amplitude and phase from one data set to another and appear attributable to instabilities in the instrumentation and measurement conditions. The most stable activity measurements of α decaying sources set an upper limit between 0.0006% and 0.006% to the amplitude of annual oscillations in the decay rate. There are no apparent indications for systematic oscillations at a level of weeks or months. Oscillations in phase with Earth's orbital distance to the sun could not be observed within 10^{-5}–10^{-6} range precision.</p>	

On decay constants and orbital distance to the Sun—part II: beta minus decay

Principal author JRC	Stefaan Pommé
Principal author SCK CEN	Leen Verheyen
Publication type	Journal paper
Reference	Metrologia 54 (2017) 19
Relevant project sheet	Invariability of decay constants

Claims that proximity to the Sun causes variations of decay constants at the permille level have been investigated for beta-minus decaying nuclides. Repeated activity measurements of ^3H , ^{14}C , ^{60}Co , ^{85}Kr , ^{90}Sr , ^{124}Sb , ^{134}Cs , ^{137}Cs , and ^{154}Eu sources were performed over periods of 259 d up to 5 decades at various nuclear metrology institutes. Residuals from the exponential decay curves were inspected for annual oscillations. Systematic deviations from a purely exponential decay curve differ in amplitude and phase from one data set to another and appear attributable to instabilities in the instrumentation and measurement conditions. Oscillations in phase with Earth's orbital distance to the Sun could not be observed within 10^{-4} – 10^{-5} range precision. The most stable activity measurements of β^- decaying sources set an upper limit of 0.003%–0.007% to the amplitude of annual oscillations in the decay rate. There are no apparent indications for systematic oscillations at a level of weeks or months.

On decay constants and orbital distance to the Sun—part III: beta plus and electron capture decay

Principal author JRC	Stefaan Pommé
Principal author SCK CEN	Leen Verheyen
Publication type	Journal paper
Reference	Metrologia 54 (2017) 36
Relevant project sheet	Invariability of decay constants

The hypothesis that seasonal changes in proximity to the Sun cause variation of decay constants at permille level has been tested for radionuclides disintegrating through electron capture and beta plus decay. Activity measurements of ^{22}Na , ^{54}Mn , ^{55}Fe , ^{57}Co , ^{65}Zn , $^{82+85}\text{Sr}$, ^{90}Sr , ^{109}Cd , ^{124}Sb , ^{133}Ba , ^{152}Eu , and ^{207}Bi sources were repeated over periods from 200 d up to more than four decades at 14 laboratories across the globe. Residuals from the exponential nuclear decay curves were inspected for annual oscillations. Systematic deviations from a purely exponential decay curve differ from one data set to another and appear attributable to instabilities in the instrumentation and measurement conditions. Oscillations in phase with Earth's orbital distance to the sun could not be observed within 10^{-4} – 10^{-5} range precision. The most stable activity measurements of β^+ and EC decaying sources set an upper limit of 0.006% or less to the amplitude of annual oscillations in the decay rate. There are no apparent indications for systematic oscillations at a level of weeks or months.

Nuclear data sensitivity and uncertainty analysis of effective neutron multiplication factor in various MYRRHA core configurations

Principal author JRC	Gašper Žerovnik
Principal author SCK CEN	Alexey Stankovski
Publication type	Journal paper
Reference	Annals of Nuclear Energy 101 (2017) 330
Relevant project sheet	Improving nuclear data for MYRRHA

A sensitivity and uncertainty analysis was carried out to estimate the uncertainty in the neutron multiplication factor k_{eff} and to identify the most important nuclear data for neutron induced reactions for criticality calculations of the latest MYRRHA designs. Sensitivity profiles, i.e. sensitivity to the nuclear data as a function of incoming neutron energy, were derived for both a critical and sub-critical core. They were calculated using codes that are based on different methodologies including stochastic and deterministic calculations (i.e. SCALE, MCNP and XSUN). The neutron induced nuclear data sensitivity analysis outlined the following quantities to be of special importance for the MYRRHA reactor concept: $^{239}\text{Pu}(n,\gamma)$ both in resonance and fast energy region, (n,f) fast, χ and ν fast; $^{238}\text{U}(n,n')$ fast, (n,γ) resonance and fast, (n,n) resonance and fast; ^{240}Pu ν fast; $^{238}\text{Pu}(n,f)$ both resonance and fast; $^{56}\text{Fe}(n,\gamma)$ both resonance and fast.

Differences of less than 4% between codes were obtained for these quantities, with few exceptions ($^{238}\text{Pu}(n,f)$, $^{238}\text{U}(n,n)$ and $^{56}\text{Fe}(n,\gamma)$ reactions). Nuclear data covariance matrices of different libraries (SCALE-6, COMMARA-2 and JENDL-4.0m) were used to derive the uncertainty in k_{eff} based on the calculated sensitivities. This study reveals that the largest contributions to k_{eff} uncertainty result from the uncertainty in the average prompt neutron fission multiplicity of ^{239}Pu , in the ^{238}U inelastic scattering cross section and ^{239}Pu fission cross section, using the covariances from SCALE-6, COMMARA-2 and JENDL-4.0m, respectively.

Nuclear data uncertainty propagation to integral responses using SANDY

Principal author JRC	Gašper Žerovnik
Principal author SCK CEN	Luca Fiorito
Publication type	Journal paper
Reference	Annals of Nuclear Energy 101 (2017) 359
Relevant project sheet	Uncertainty quantification and propagation

SANDY is a nuclear data sampling code compatible with nuclear data files in ENDF-6 format. Exploiting the basic theory of stochastic sampling, SANDY generates random nuclear data samples that reproduce the covariance information stored in the ENDF-6 files. Such random data are rewritten in perturbed ENDF-6 or PENDF files and can be used as nuclear code inputs to produce perturbed responses. After the statistical analysis of the sampled responses, the sample mean and variance are quantified. Not only can SANDY be used for the study of the response sample variance, but it can also estimate global sensitivity indices for correlated and uncorrelated input parameters using a variance-based decomposition method.

SANDY was tested for the generation of random ENDF-6 files with perturbed cross sections and resonance parameters. These files were used for the uncertainty quantification of integral responses, such as the one-group integrated cross section and the resonance integral of several isotopes. Then, the response variance was apportioned to the several inputs and the parameters producing the largest impact were identified.

Systematic effects on cross section data derived from reaction rates in reactor spectra and a re-analysis of ^{241}Am reactor activation measurements

Principal author JRC	Gašper Žerovnik
Principal author SCK CEN	Luca Fiorito
Publication type	Journal Paper
Reference	Nuclear Instruments and Methods A 877 (2018) 300
Relevant project sheet	Improving nuclear data for MYRRHA

Methodologies to derive cross section data from spectrum integrated reaction rates were studied. The Westcott convention and some of its approximations were considered. Mostly measurements without and with transmission filter are combined to determine the reaction cross section at thermal energy together with the resonance integral. The accuracy of the results strongly depends on the assumptions that are made about the neutron energy distribution, which is mostly parameterised as a sum of a thermal and an epi-thermal component. Resonance integrals derived from such data can be strongly biased and should only be used in case no other data are available. The cross section at thermal energy can be biased for reaction cross sections which are dominated by low energy resonances. The amplitude of the effect is related to the lower energy limit that is used for the epi-thermal component of the neutron energy distribution. It is less affected by the assumptions on the shape of the energy distribution. When the energy dependence of the cross section is known and information about the neutron energy distribution is available, a method to correct for a bias on the cross section at thermal energy is proposed. Reactor activation measurements to determine the thermal $^{241}\text{Am}(n,\gamma)$ cross section reported in the literature were reviewed. In case enough information was available, the results were corrected to account for possible biases and included in a least squares fit. These data combined with results of time-of-flight measurements give a capture cross section of 720 (14) b for $^{241}\text{Am}(n,\gamma)$ at thermal energy.

Nuclear data uncertainty analysis for the Po-210 production in MYRRHA

Principal author JRC	Gasper Žerovnik
Principal author SCK CEN	Luca Fiorito
Publication type	Journal paper
Reference	European Physics Journal Nuclear Science and Technologies, 4 (2018) 48
Relevant project sheets	Uncertainty quantification and propagation Improving nuclear data for MYRRHA

MYRRHA is a multi-purpose research reactor able to operate in sub-critical and critical modes and currently in the design phase at SCK CEN. The choice of LBE was driven by its chemical stability, low melting temperature, high boiling point, low chemical reactivity with water and air and a good neutronic performance. As a drawback, the neutron capture in ^{209}Bi results in the production of ^{210}Po , a highly radiotoxic alpha emitter with relatively short half-lives (~138 d). The ^{210}Po production represents a major safety concern that has to be addressed for the reactor licensing. In this work we used the ALEPH-2 burnup code to accurately calculate the ^{210}Po production in a MYRRHA operating cycle. The impact of using different nuclear data libraries was evaluated and the reliability of the results was determined by quantifying the uncertainty of the ^{210}Po concentration. The uncertainty quantification was carried out sampling the currently available nuclear data covariance matrices with the SANDY code. Also, estimates of the sensitivity profiles were obtained with a linear regression approach. The activation yield of the ^{209}Bi neutron capture reaction was assessed as the largest nuclear data source of uncertainty, however the lack of covariances for such data represent a capital drawback for the ^{210}Po content prediction.

Is decay constant?	
Principal author JRC	Stefaan Pommé
Principal author SCK CEN	Leen Verheyen
Publication type	Journal paper
Reference	Applied Radiation and Isotopes 134 (2018) 6
Relevant project sheet	Invariability of decay constants
<p>Some authors have raised doubt about the invariability of decay constants, which would invalidate the exponential-decay law and the foundation on which the common measurement system for radioactivity is based. Claims were made about a new interaction - the fifth force - by which neutrinos could affect decay constants, thus predicting changes in decay rates in correlation with the variations of the solar neutrino flux. Their argument is based on the observation of permille-sized annual modulations in particular decay rate measurements, as well as transient oscillations at frequencies near 11 year⁻¹ and 12.7 year⁻¹ which they speculatively associate with dynamics of the solar interior. In this work, 12 data sets of precise long-term decay rate measurements have been investigated for the presence of systematic modulations at frequencies between 0.08 and 20 year⁻¹. Besides small annual effects, no common oscillations could be observed among α, β^-, β^+ or EC decaying nuclides. The amplitudes of fitted oscillations to residuals from exponential decay do not exceed 3 times their standard uncertainty, which varies from 0.00023 % to 0.023 %. This contradicts the assertion that 'neutrino-induced' beta decay provides information about the deep solar interior.</p>	

Intercomparison of ^{99m} Tc, ¹⁸ F and ¹¹¹ In activity measurements with radionuclide calibrators in Belgian hospitals	
Principal author JRC	Stefaan Pommé
Principal author SCK CEN	Clarita Saldarriaga Vargas
Publication type	Journal paper
Reference	Physica Medica 45 (2018) 134
Relevant project sheet	Radionuclide calibrators in nuclear medicine
<p>An intercomparison exercise was performed among 15 Belgian hospitals to test the accuracy of ^{99m}Tc, ¹⁸F and ¹¹¹In activity measurements by means of radionuclide calibrators. The data set includes measurements from 38 calibrators, yielding 36 calibrations for ^{99m}Tc and ¹¹¹In, and 21 calibrations for ¹⁸F. For each radionuclide, 3 ml of stock solution was measured in two clinical geometries: a 10 ml glass vial and a 10 ml syringe. The initial activity was typically 100 MBq for ^{99m}Tc, 15 MBq for ¹¹¹In and 115 MBq for ¹⁸F. The reference value for the massic activity of the radioactive solutions was determined by means of primary and secondary standardisation techniques at the radionuclide metrology laboratory of the JRC.</p> <p>The overall results of the intercomparison were satisfactory for ^{99m}Tc and ¹⁸F, since most radionuclide calibrators (>70%) were accurate within $\pm 5\%$ of the reference value. Nevertheless, some devices underestimated the activity by 10–20%. Conversely, ¹¹¹In measurements were strongly affected by source geometry effects and this had a negative impact on the accuracy of the measurements, in particular for the syringe sample. Large overestimations (up to 72%) were observed, even when taking into account the corrections and uncertainties supplied by the manufacturers for container effects. The results of this exercise encourage the hospitals to perform corrective actions to improve the calibration of their devices where needed.</p>	

Determination of homogeneity of the top surface deadlayer in an old HPGe detector	
Principal author JRC	Mikael Hult
Principal author SCK CEN	Michel Bruggeman
Publication type	Journal paper
Reference	Applied Radiation and Isotopes , 147 (2019) 182
Relevant project sheet	Realisation of computer models of gamma-ray detectors
<p>A collimated source of ^{241}Am was scanned over the endcap of a 21 year old coaxial HPGe-detector that had spent about 75% of its life at room temperature (and the remaining time at 77 K). The detector response was recorded and used as a measure of the relative thickness of the top deadlayer. This thickness was not homogeneous and was thicker near to the outer surface of the crystal compared to the centre, which could be a result of increased diffusion of Li atoms during times the detector was kept at room temperature. The results were compared with two newer HPGe-detectors that proved to have homogeneous top deadlayers.</p>	

Peak shape calibration of a Cadmium Zinc Telluride detector and its application for the determination of uranium enrichment	
Principal author JRC	Peter Schillebeeckx
Principal author SCK CEN	Alessandro Borella
Publication type	Journal paper
Reference	Nuclear Instruments and Methods A 986 (2021) 164718
Relevant project sheet	Uranium enrichment and plutonium isotopics
<p>Cadmium Zinc Telluride (CZT) detectors are portable, room temperature serviceable, medium-resolution gamma-ray spectrometers. Their full-energy peak shape exhibits a low energy tail which complicates the analysis of spectra with overlapping peaks. In this paper, we determined the peak shape parameters of a CZT detector from measurements with calibrated point sources from 0.06 MeV up to 1.332 MeV. The peak shape parameters were obtained by applying a peak fitting algorithm that includes a Gaussian and a tail with energy dependent parameters in the region around the gamma-ray peak. The net peak areas were used to verify the absolute detection efficiency obtained with a Monte Carlo model of the CZT detector and the agreement in absolute terms was within 10% over the considered energy range. The peak fitting algorithm was then applied to determine the net peak areas of the full energy peak in spectra recorded with certified uranium standards. The enrichment was then determined by using the so-called 'peak ratio' method. We observed a systematic bias in the net peak areas of the 0.258 MeV gamma-ray which therefore was not included in the analysis. Hence, the enrichment was underestimated by about 10%.</p>	

The Joint Evaluated Fission and Fusion Nuclear Data Library, JEFF-3.3	
Principal author JRC	Arjan Plompen
Principal author SCK CEN	Alexey Stankovskiy
Publication type	Journal paper
Reference	European Physics Journal A 56 (2020) 181
Relevant project sheet	Improving nuclear data for MYRRHA
<p>The joint evaluated fission and fusion nuclear data library 3.3 is described. New evaluations for neutron-induced interactions with the major actinides ^{235}U, ^{238}U and ^{239}Pu, on ^{241}Am and ^{23}Na, ^{59}Ni, Cr, Cu, Zr, Cd, Hf, W, Au, Pb and Bi are presented. It includes new fission yields, prompt fission neutron spectra and average number of neutrons per fission. In addition, new data for radioactive decay, thermal neutron scattering, gamma-ray emission, neutron activation, delayed neutrons and displacement damage are presented. JEFF-3.3 was complemented by files from the TENDL project. The libraries for photon, proton, deuteron, triton, helion and alpha-particle induced reactions are from TENDL-2017. The demands for uncertainty quantification in modelling led to many new covariance data for the evaluations. A comparison between results from model calculations using the JEFF-3.3 library and those from benchmark experiments for criticality, delayed neutron yields, shielding and decay heat, reveals that JEFF-3.3 performs very well for a wide range of nuclear technology applications, in particular nuclear energy.</p>	

An international multi-center investigation on the accuracy of radionuclide calibrators in nuclear medicine theragnostics	
Principal author JRC	Stefaan Pommé
Principal author SCK CEN	Clarita Saldarriaga Vargas
Publication type	Journal paper
Reference	European Journal of Nuclear Medicine and Molecular Imaging, Physics 7 (2020) 69
Relevant project sheet	Radionuclide calibrators in nuclear medicine
<p>This international multi-center study aimed to investigate the clinical measurement accuracy of radionuclide calibrators for 7 radionuclides used in theragnostics: $^{99\text{m}}\text{Tc}$, ^{111}In, ^{123}I, ^{124}I, ^{131}I, ^{177}Lu, and ^{90}Y. In total, 32 radionuclide calibrators from 8 hospitals located in the Netherlands, Belgium, and Germany were tested. For each radionuclide, a set of four samples comprising two clinical containers (10-mL glass vial and 3-mL syringe) with two filling volumes were measured. The reference value of each sample was determined by two certified radioactivity calibration centers (SCK CEN and JRC) using two secondary standard ionization chambers. The deviation in measured activity with respect to the reference value was determined for each radionuclide and each measurement geometry. For $^{99\text{m}}\text{Tc}$, ^{131}I, and ^{177}Lu, a small minority of measurements were not within $\pm 5\%$ range from the reference activity (percentage of measurements not within range: $^{99\text{m}}\text{Tc}$, 6%; ^{131}I, 14%; ^{177}Lu, 24%) and almost none were outside $\pm 10\%$ range. However, for ^{111}In, ^{123}I, ^{124}I, and ^{90}Y, more than half of all measurements were not accurate within $\pm 5\%$ range (^{111}In, 51%; ^{123}I, 83%; ^{124}I, 63%; ^{90}Y, 61%) and not all were within $\pm 10\%$ margin (^{111}In, 22%; ^{123}I, 35%; ^{124}I, 15%; ^{90}Y, 25%). A large variability in measurement accuracy was observed between radionuclide calibrator systems, type of sample container (vial vs syringe), and source-geometry calibration/correction settings used. Consequently, we observed large combined deviations (percentage deviation $> \pm 10\%$) for the investigated theranostic pairs, in particular for $^{90}\text{Y}/^{111}\text{In}$, $^{131}\text{I}/^{123}\text{I}$, and $^{90}\text{Y}/^{99\text{m}}\text{Tc}$.</p> <p>Our study shows that substantial over- or underestimation of therapeutic patient doses is likely to occur in a theranostic setting due to errors in the assessment of radioactivity with radionuclide calibrators. These findings underline the importance of thorough validation of radionuclide calibrator systems for each clinically relevant radionuclide and sample geometry.</p>	

Annex 2. Contributions to conference proceedings or workshops

Neutron-induced nuclear data for the MYRRHA fast spectrum facility	
Principal author JRC	Gašper Žerovnik
Principal author SCK CEN	Alexey Stankovskiy
Publication type	Conference contribution
Reference	European Physics Journal Web of Conferences 146 (2017) 09007
Relevant project sheet(s)	Improving nuclear data for MYRRHA
<p>The MYRRHA (Multi-purpose hYbrid Research Reactor for High-tech Applications) concept is a flexible experimental lead-bismuth cooled and mixed-oxide (MOX) fueled fast spectrum facility designed to operate both in sub-critical (accelerator driven) and critical modes. One of the key issues for the safe operation of the reactor is the uncertainty assessment during the design works. The main objective of Work Package 10 of the European project CHANDA (solving CHAllenges in Nuclear DATA) is to improve MYRRHA relevant nuclear data in order to reduce the reactor parameter uncertainties derived from them. In order to achieve this goal, several tasks have been undertaken. First, a sensitivity study of MYRRHA integral parameters, such as energy dependent cross sections, fission spectra and neutron multiplicities, to nuclear data has been conducted resulting in a list of MYRRHA relevant quantities (nuclides and reactions). On the second task, an analysis of the existing experimental data and evaluations for the quantities included in the list has been carried out. In this framework, the impact on the multiplication factor of quantities from different nuclear data libraries for different nuclides, reactions and energy regions has been investigated on the MYRRHA MOX critical core model. As the next step, new experiments and evaluations will be performed in order to improve existing nuclear data libraries.</p>	

Characterisation of Bi-samples by Neutron Resonance Capture Analysis	
Principal author JRC	Stefan Kopecky
Principal author SCK CEN	Antonin Krása
Publication type	Workshop contribution
Reference	EUFRAT user meeting, JRC Geel, 4 – 7 December 2017, pp. 53 – 55 (2017)
Relevant project sheet(s)	Characterisation of Bi-samples by NRCA at GELINA
<p>It has been noticed that a provider's assessment of the elemental composition of materials used in validation experiments can be unreliable. Follow-on verification studies are important when the impurities have a significant impact on parameters of interest. Complementary analysis methods including standard neutron activation analysis, mass spectrometry and more advanced methods like neutron resonance capture analysis (available at the GELINA facility of JRC-Geel) are useful tools for that purpose.</p>	

Fast neutron detector development for measurements at the VENUS-F reactor	
Principal author JRC	Jan Heyse
Principal author SCK CEN	Jan Wagemans
Publication type	Workshop contribution
Reference	EUFRAT user meeting, JRC Geel, 4 – 7 December 2017, pp. 89 – 91 (2017)
Relevant project sheet(s)	Fast neutron detector development for measurements at VENUS-F
<p>Combining the different fields of expertise available at JRC-Geel and SCK CEN, a new fast neutron detector was developed. Using high-purity ²³⁸U base material available at JRC-Geel, a fission chamber was manufactured, calibrated at the standard irradiation field at the BR1 reactor and is currently applied to optimize the sub-criticality measurement technique at the GUINEVERE ADS.</p>	

Measurements on U and Pu standards with medium resolution gamma-ray spectrometers	
Principal author JRC	Peter Schillebeeckx
Principal author SCK CEN	Iaroslav Meleshenkovskii
Publication type	Workshop contribution
Reference	EUFRAT user meeting, JRC Geel, 4 – 7 December 2017, pp. 92 – 94 (2017)
Relevant project sheet(s)	Uranium enrichment and plutonium isotopics
<p>Measurements of uranium and plutonium certified reference materials were conducted at the nuclear infrastructure of JRC-GEEL using two detectors – a 500 mm³ CZT semiconductor detector and a 2×2 inch LaBr₃ scintillator. The stability of the measurement chains during the measurements with the CZT and LaBr₃ detectors was evaluated by determining the main parameters of intense full energy peaks: peak centroid, FWHM and net peak area. The results of these experiments will be used for the development of a uranium and plutonium isotopic composition determination algorithms dedicated for CZT and LaBr₃ detectors. Such algorithms are of particular interest for safeguards applications.</p>	

Characterisation of spent nuclear fuel by theoretical calculations and non-destructive analysis	
Principal author JRC	Peter Schillebeeckx
Principal author SCK CEN	Marc Verwerft
Publication type	Workshop contribution
Reference	Proc. International Workshop on Numerical Modelling of NDA Instrumentation and Methods for Nuclear Safeguards, Luxembourg, 16 – 17 May, pp. 101 – 113 (2018)
Relevant project sheet(s)	Study of NDA observables to characterise spent fuel Neutron measurements on spent nuclear fuel pellets
<p>The characterisation of spent nuclear fuel assemblies in view of transport, intermediate storage and final disposal is discussed. The observables of interest that need to be determined are the decay heat, neutron and gamma-ray emission rate spectra together with the inventory of specific nuclides that are important for criticality safety analysis and to verify the fuel history. Unfortunately, all these observables cannot be determined during routine operation. Hence, the characterisation will be based on theoretical calculations combined with results of Non-Destructive Analysis (NDA). In this work the input parameters affecting most the results of theoretical calculations are identified. In addition, an NDA system to determine the neutron emission rate of a spent nuclear fuel sample is presented. Results of such measurements can be used to validate the results of theoretical calculations. This system will also be used to demonstrate that spent nuclear fuel samples can be characterised by NDA in a conventional controlled area outside a hot cell environment.</p>	

Characterisation of spent pressurized water reactor (PWR) fuel for decay heat, neutron and gamma-ray emission: code comparison	
Principal author JRC	Gašper Žerovnik
Principal author SCK CEN	Luca Fiorito
Publication type	Conference contribution
Reference	Proceedings of the International Conference on Mathematics and Computational Methods applied to Nuclear Science and Engineering (M&C 2019), pp. 2736 – 2745 (2019)
Relevant project sheet(s)	Study of NDA observables to characterise spent fuel
<p>The characterisation of Spent Nuclear Fuel (SNF) in view of intermediate storage and final disposal is discussed. The main observables of interest that need to be determined are the decay heat, neutron and γ-ray emission spectra. In addition, the inventory of specific nuclides that are important for criticality safety analysis and to verify the fuel history has to be determined. Some of the observables such as the decay heat and neutron and γ-ray emission rate can be determined by Non-Destructive Analysis (NDA) methods. Unfortunately, this is not always possible especially during routine operation. Hence, a characterisation of SNF will rely on theoretical calculations combined with results of NDA methods. In this work the observables of interest, also referred to as source terms, are discussed based on theoretical calculations starting from fresh UO_2 and MOX fuel. The irradiation conditions are representative for PWR. The Serpent code is used to define the nuclides which have an important contribution to the observables. The emphasis is on cooling times between 1 a and 1000 a.</p>	

Precise measurement of the neutron capture cross section and capture-to-fission ratio of ^{235}U at thermal and sub-thermal neutron energies	
Principal author JRC	Peter Schillebeeckx
Principal author SCK CEN	Jan Wagemans
Publication type	Conference contribution
Reference	International Conference on Nuclear Data for Science and Technology, 19 – 24 May, 2019, Beijing (China)
Relevant project sheet(s)	Capture cross section measurements for ^{235}U at BR1
<p>The recommended cross-section value for ^{235}U neutron-capture at thermal energies is largely based on the difference from total and competing cross-sections of ^{235}U. Despite its importance and high value of 100 b, direct measurements of (n,γ) are rare (only two exist for thermal energies) and exhibit large uncertainties. The reason is the difficulty to measure the characteristic radiation of the reaction product ^{236}U within a dominant fission background (^{236}U has a long half-life of $23.4 \cdot 10^6$ a).</p> <p>Capture of ^{235}U may exhibit a deviation from a pure $1/v$-behaviour around thermal energies. Additional energy-dependent data are required to provide information about the cross section and of the capture-to-fission ratio. Measurements at different beam temperatures provide information about the energy dependence. For this reasons, we started a project with a new method utilizing different neutron fields to evaluate its energy dependence in the low energy region. We use a combination of neutron activation and subsequent accelerator-mass-spectrometry (AMS) for direct atom counting of the reaction product ^{236}U.</p> <p>Activations of several ~50mg pellets (natural uranium-oxide powder with well-known stoichiometry and low ^{236}U content) have been performed: with an almost pure Maxwellian spectrum at room temperature at BR1 (Mol) and with cold neutrons at MLZ (Munich); more irradiations are planned at ILL (Grenoble) for lower energies.</p> <p>The main quantity measured in AMS is the isotope ratio of the reaction product relative to the target nuclide, this is $^{236}\text{U}/^{235}\text{U}$ for $^{235}\text{U}(n,\gamma)^{236}\text{U}$. Using AMS, the capture cross-section is then simply this isotope ratio divided by the neutron fluence. Simultaneously, we measure the $^{236}\text{U}/^{239}\text{Pu}$ ratio in the irradiated ^{235}U sample giving directly the cross-section ratio of $^{235}\text{U}(n,\gamma)$ relative to $^{238}\text{U}(n,\gamma)$. A measurement relative to fission is possible, too. The latter cases are completely independent of the neutron fluence.</p> <p>AMS will allow for an accurate determination of $^{235}\text{U}(n,\gamma)^{236}\text{U}$. Based on our experience in neutron cross-section measurements and by combining the results from different AMS labs, we expect an AMS-uncertainty <3%.</p>	

Peak Shape Characterization of a 500 mm³ Cadmium Zinc Telluride Detector and Analysis of Spectroscopic Measurement Data for Uranium Samples

Principal author JRC	Peter Schillebeeckx
Principal author SCK CEN	Alessandro Borella
Publication type	Conference contribution
Reference	Proceedings of the International Workshop on Isotopic Analysis of Uranium and Plutonium by Nondestructive Assay Techniques for Nuclear Safeguards, IAEA, 16-19 February 2021
Relevant project sheet(s)	Uranium enrichment and plutonium isotopics

Gamma-ray spectra of calibrated sources and reference standard samples of uranium and plutonium were recorded with a 500 mm³ Cadmium Zinc Telluride (CZT) detector. The measurements with calibrated sources were used to set up the peak-shape calibration of CZT detectors as a function of the deposited energy and the obtained net peak areas were compared with the ones obtained with Monte Carlo modelling. The obtained calibrations were then used to determine unbiased net peak areas in spectra measured on the low-enriched uranium standard samples with the aim to determine their uranium enrichment. Results of analyses on different samples show that both the net peak area of 258 keV gamma-ray and the chosen model for the efficiency have a strong impact on the determined enrichment. The developed procedure was also used to verify the enrichment in high-enriched uranium samples for which the gamma-ray lines of the decay of ²³⁸U were less pronounced both due to the higher ²³⁵U relative content and the small mass of the samples. General recommendations deduced from our experiments on how to analyse complex spectra such as the ones of Pu bearing materials are given.

Annex 3. Technical Reports

Assessment of evaluations for MYRRHA	
Principal author JRC	Gasper Žerovnik
Principal author SCK CEN	Alexey Stankovskiy
Publication type	Report
Reference	JRC Technical Reports, EUR 28240 EN (2016)
Relevant project sheet(s)	Improving nuclear data for MYRRHA
<p>Nuclear data of importance to the operation of MYRRHA, a research infrastructure supporting the development of Generation-IV reactors, were identified by sensitivity analysis. The data recommended in the main nuclear data libraries (i.e. JEFF, JENDL, ENDF/B, and TENDL) were compared and validated using experimental data reported in the literature and results of recent measurements. Recommendations for new experiments to solve inconsistencies between recommended and experimental data were specified. In addition, an assessment was made of the discrepancies and the means of improvement of nuclear data libraries for design, safety assessment and operation of new nuclear reactor systems such as MYRRHA.</p>	

Recommendations for MYRRHA relevant cross section data to the JEFF project	
Principal author JRC	Gašper Žerovnik
Principal author SCK CEN	Alexey Stankovskiy
Publication type	Report
Reference	JRC Technical Reports, EUR 28957 EN (2017)
Relevant project sheet(s)	Improving nuclear data for MYRRHA
<p>Within the framework of Work Package 10 of the EC FP7 CHANDA project, nuclear data of importance for the operation of MYRRHA, a lead-bismuth cooled accelerator driven reactor under development at SCK CEN (BE), were studied. Based on data in the main nuclear data libraries, i.e. JEFF, JENDL, ENDF/B and BROND, and in the TENDL and CIELO libraries and on experimental data reported in the literature, recommendations to the JEFF project were made for several nuclides of interest to the MYRRHA reactor.</p>	

Observables of interest for the characterisation of Spent Nuclear Fuel

Principal author JRC	Gašper Žerovnik
Principal author SCK CEN	Marc Verwerft
Publication type	Report
Reference	JRC Technical Report, EUR 29301 EN (2018)
Relevant project sheet(s)	Study of NDA observables to characterise spent fuel

The characterisation of Spent Nuclear Fuel (SNF) in view of intermediate storage and final disposal is discussed. The main observables of interest that need to be determined are the decay heat, neutron and γ -ray emission spectra. In addition, the inventory of specific nuclides that are important for criticality safety analysis and for verification of the fuel history has to be determined. Some of the observables such as the decay heat and neutron and γ -ray emission rate can be determined by Non-Destructive Analysis (NDA) methods. Unfortunately, this is not always possible especially during routine operation. Hence, a characterisation of SNF will rely on theoretical calculations combined with results of NDA methods. In this work the observables of interest, also referred to as source terms, are discussed based on theoretical calculations starting from fresh UO_2 and MOX fuel. The irradiation conditions are representative for PWR. The Serpent code is used to define the nuclides which have an important contribution to the observables. The emphasis is on cooling times between 1 a and 1000 a.

A non-destructive method to determine the neutron production rate of a sample of spent nuclear fuel under standard controlled area conditions

Principal author JRC	Peter Schillebeeckx
Principal author SCK CEN	Marc Verwerft
Publication type	Report
Reference	JRC Technical Report, EUR 30379 EN (2020)
Relevant project sheet(s)	Neutron measurements on spent nuclear fuel pellets

A method to determine the neutron production rate of a sample of spent nuclear fuel by means of non-destructive analysis conducted under controlled-area conditions is described, validated and demonstrated. A standard neutron well-counter designed for routine nuclear safeguards applications is applied. The method relies on a transfer procedure that is adapted to the hot-cell facilities at the Laboratory for High and Medium level Activity of the SCK CEN. The sample transfer and measurement procedures are described together with results of Monte Carlo simulations. Experiments with radionuclide sources were carried out at the Joint Research Centre to test the procedures and to determine the performance characteristics of the detection device. Finally, measurements of a segment of a spent nuclear fuel rod were carried out at the SCK CEN to validate and demonstrate the method.

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