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Metrology for New Generation Nuclear Power Plants – MetroFission

Lena Johansson, Jean-Remy Filtz, Pierino DeFelice, Mohamed Sadli, Arjan Plompen, Jan Heyse, Bruno Hay, Alan Dinsdale, Stefaan Pommé, Philippe Cassette, John Keightley

Abstract— This EMRP (European Metrology Research Programme) project, MetroFission, has been looking at solving metrological problems related to a new generation of nuclear power plants. The proposed Generation IV power plants are designed to run safely, make efficient use of natural resources, minimize the waste and maintain proliferation resistance. In order to reach these goals, the reactor operation involves higher temperatures, high-energy neutron fluence, different types of fuel where the minor actinides are included etc. In this multidisciplinary project, which has 12 partners in 10 countries, the work has focused on improved temperature measurements, investigation of thermal properties of advanced materials, determination of new and relevant nuclear data and development of measurement techniques for radionuclides suitable for Generation IV power plants.

The improved temperature measurement for nuclear power plant applications includes the development of a new Fe-C fixed point. Robust, repeatable and versatile cells have been constructed and compared with success among the project participants and their melting temperatures have been determined. Furthermore, the methodology of self-validating thermocouples has proven efficient at several fixed point temperatures (Au, Cu, Co-C, Fe-C) using different designs. A practical acoustic thermometer has been tested at high temperature (1000 °C) with success thanks to the use of innovative signal processing methods. Mo/Nb thermocouples have been obtained with different sheath materials and tested with the aim to achieve for the first time a reference function determined with the best possible uncertainties.

Following reviews of designs and technology proposed for fourth generation nuclear plants effort within this project, with regards to thermal properties of advanced materials for nuclear design, has concentrated on provision of thermodynamic data to

support the development of the sodium cooled fast reactor (ASTRID).

Data has been critically assessed to represent the potential interaction between the Na coolant and the nuclear fuel taken to be based on (U,Pu)O₂ but incorporating minor actinides such as Np and Am. Data for the fission products and containment materials have also been reviewed and new data assessed to represent the interaction between them and the Na coolant, in order to permit calculation of phase and chemical equilibria for severe accident scenarios. The aim is to publish the data developed during this project within a revised version of “Thermochemical data for reactor materials and fission products”, last published in 1990.

Reference metrological facilities and methods have been developed or improved for the measurements of normal spectral emissivity and specific heat up to 1500 °C, and thermal diffusivity up to 2000 °C. The following actions were notably performed:

- Development of two complementary facilities based on different metrological approaches (calorimetric and optical methods) by PTB and LNE for the measurement of emissivity of solid materials at high temperature.
- Modification of the LNE diffusivimeter with the development and integration of a specific inductive furnace, and the adaptation of the associated IR detection systems.

These facilities have now been used for the measurement of thermophysical properties of some materials (Ni, MgO and ZrO₂), because they have thermal properties similar to those encountered in Generation IV reactors.

Regarding neutron cross sections, the current nuclear databases concentrate on thermal energies, but new fast reactor designs will involve materials exposed to higher energy neutron fields. Advantage has been taken of National Measurement Institutes experience in neutron fluence measurements to improve high-energy cross-section measurements. A secondary fluence standard has been established and used to determine the cross sections of actual interest improving nuclear data standards in the form of reduced measurement uncertainties.

The heavier actinides and their decay products have a role in the future development and adoption of nuclear power plants in whose fuels they will be present in controlled amounts. A list of specific needs for improved actinide decay data has recently been outlined in a review by the IAEA. Responding to those needs, an objective of the MetroFission project was to measure the alpha-particle emission probabilities of ²³⁸U with certified isotopic composition. The nuclear data from alpha spectrometry of ²³⁸U sources has been successful, obtaining data that will be provided to the international community and that further lower the uncertainties of the nuclear data. The quality of the alpha spectrum measurement, in terms of energy resolution and peak tailing of the ²³⁸U alpha spectra, was significantly better than in any previous measurement.

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Fission fragments containing more beta-decay isotopes will result from proposed fast reactors. Cryogenic detectors have been developed aimed at improving the measurement of beta spectra and verifying theoretical models. A beta spectrum shape of ^{63}Ni has successfully been obtained. Improvements have been made in the source components and in the source production method in order to lower uncertainties. The exchange effect has been implemented in the calculation of theoretical beta spectra. The measurement of the beta spectrum of ^{63}Ni has for the first time confirmed the exchange effect at very low energy. The energy threshold of the metallic magnetic calorimeter is 180 eV. At this energy, the enhancement of the beta emission due to the exchange effect is larger than 20 %.

Nuclear power plants would benefit from on-site activity measurements of radionuclides including low-energy beta emitters that are abundant but can prove difficult to assay. Within this project, a primary liquid scintillation coincidence counting method called Triple to Double Coincidence Ratio (TDCR) has been modified for this purpose. Five partners have built miniature TDCR systems, miniaturising the instrument's detection chamber by using newer, smaller, more efficient photomultiplier tubes. The system also includes replacing the analogue electronic modules with digital acquisition systems and signal processing in order to significantly improve digital coincidence counting (DCC).

Index Terms— Digital Signal Processing, Generation IV reactor, Instrumentation, Liquid scintillation, Metrology, Neutron cross-section, Nuclear data, Temperature measurements, Thermal properties, Thermochemical data modelling.

I. INTRODUCTION

Nuclear energy is one of the energies which emit the least greenhouse gases - such as CO_2 - during its lifecycle. With 31% of Europe's electricity produced from nuclear at the moment, this is the most important low carbon technology in Europe's energy mix [1]. Europe can maintain the current level of nuclear energy by long-term operation of existing plants and an ambitious programme of new build. Some European countries have already decided to build new nuclear reactors; Other EU countries are actively considering whether new nuclear power plants should form part of their energy strategy [2], [3].

The nuclear power plants currently in use are first generation Magnox and second generation advanced gas cooled and light water reactors. Generation III type light water reactors of the European Pressurized Water type are currently being deployed, e.g. in Olkiluoto in Finland and Flamanville, France. The near term build will use designs that were essentially developed 30 years ago, because there has been little nuclear build in the last two decades. In terms of safety these designs are significant improvements over second-generation reactors. New builds will involve

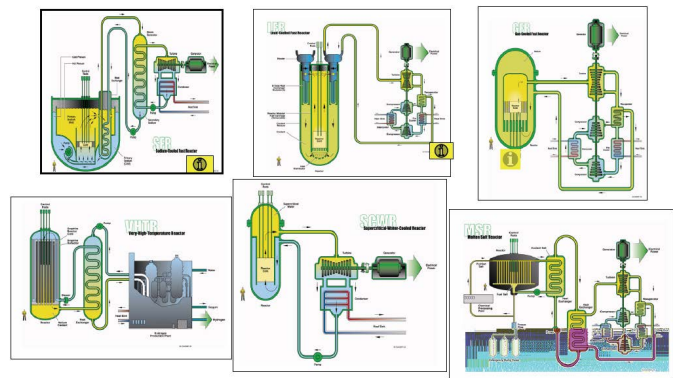


Fig. 1. The proposed types of reactors for the next Generation of Nuclear Power Plants

Incremental improvements called Generation III+. The purpose of MetroFission is to provide the metrological infrastructure necessary for the design of Generation IV (Gen IV) nuclear power plant. There are, however, some areas where this project will improve Generation III+ power plants.

GenIV aims to improve nuclear safety, improve proliferation resistance, minimize waste and natural resource utilization, and to decrease the cost to build and run such plants. Amongst other, these Gen IV reactors pose the following key challenges [4]:

1. How can the temperatures be measured, since existing thermocouple-based methods will not work in the envisioned temperatures and environments?
2. What materials should be used in these reactors and what are their thermal properties?
3. What are the nuclear data and radionuclide and neutron metrology techniques that will be needed?

In this multidisciplinary project, which includes twelve National Measurement Institutes (NMIs) in 10 countries, we have been working from the point of view of developing novel metrology with traceability to the SI units in order to address these challenges.

II. SCIENTIFIC AND TECHNICAL OBJECTIVES

The present EMRP project [5] aims to address the mentioned challenges by the following specific scientific and technological objectives:

- A. Contribute to improved temperature measurements for nuclear power plant applications.
- B. Contribute to improved thermo-chemical data and modelling for nuclear design.
- C. Development of reference metrological setups and methods for the measurement of thermal properties of advanced materials at high and very high temperatures. Characterisation of reference materials for high temperature thermal properties measurements.
- D. Contribute towards improved neutron cross section data through neutron fluence measurements and standards

- E.
- F. Measurement of nuclear decay parameters and emission probabilities of priority nuclides.
- G. Development the Triple-to-Double Coincidence Ratio (TDCR) method for radionuclide standardisation of radionuclides relevant to nuclear power and
- H. Incorporate Digital Coincidence Counting (DCC) to make these systems mobile and suitable for on-site use.

A. *New High-Temperature References, Sensors and Validation Methods*

New temperature sensors and methods for in-situ measurements in nuclear power plants have been investigated and developed with an emphasis on extending the measurement range to higher temperatures and characterising, limiting, or completely eliminating the sensor drift in a high temperature, high-neutron-flux environment.

Reference fixed points above the copper point have been developed to perform post-irradiation studies of thermocouples. This project has achieved the realisation of the Fe-C fixed point (1154 °C); in a joint effort between NPL and LNE-Cnam, different combinations of materials and designs have been tested and a comparison of different realisations has been performed. The results show a promising repeatability within $\pm 0.2^\circ\text{C}$ of this fixed point. It has been observed that there was no influence from the metal origin or difference between the two different designs. ITS-90 temperatures have been assigned by both NMIs.

Suitable thermocouples from the Mo/Nb family were investigated. These thermocouples are showing negligible neutronic drift (low neutron activation). The thermocouples were identified, constructed, and studied with respect to their reference functions, stability, and thermoelectric homogeneity [6]. The results show that the homogeneity of the thermo couples (supplied by Thermocoax) at 200°C was better than 0.2°C in all cases. However, the measurements at silver fixed point showed instabilities, even after long annealing. Hence, a decision was made to discontinue the work with Mo/Nb thermocouples within this project but might still be pursued in the future.

The practicality of self-validation methods for thermocouples has been examined as a means to reduce or eliminate drift related problems. Self-validating methods suitable for thermocouples was identified and feasibility demonstrated to high temperatures. Two methods were developed by both LNE-Cnam (France) and NPL (UK). The mini-cells were based on graphite crucibles, embedded on thermocouples. The methods were compared to the copper point (1084.62 °C) and measurements give an agreement within 0.8 °C.

In order to achieve a proof of concept of an innovative driftless thermodynamic method of temperature measurement, an acoustic thermometer was studied for possible application in an ionising environment at temperatures up to 1000 °C. A prototype

The aim of this task was to construct a demonstrator of a driftless primary thermometry method, by speed of sound measurements in a gas, in the temperature range of generation

3+ reactors. A prototype acoustic thermometer was designed and constructed (Fig 2), applicable to at least 1000°C. It was demonstrated that the drift was less than 2°C at 1000°C of this primary acoustic thermometer. The drift of the temperature sensors was performed by comparison with thermocouples through extended exposure in a high temperature environment at NPL.



Fig 2. A PAT thermometer made from Inconel. The exposed length of Inconel is the temperature sensitive region.

B. *Improved Thermo-Chemical Data and Modelling*

Following a review of the fuel and coolants of potential use in Gen IV nuclear plant it was decided that the work on thermodynamic modelling would concentrate on the development of data needed to support the development of the sodium cooled fast reactor (Astrid) in order to best target available resources. There have already been a number of well-funded successful projects concerned with the development of data for the nuclear fuel and its potential interaction with a clad. However there was very little information available concerning the liquid sodium coolant and its interactions with the fuel, containment materials and the environment and in Metrofission it was decided to target this area. The use of chemical thermodynamic data is well established in the nuclear sector and the various compilations of data thus far have been mainly concerned with the properties of stoichiometric crystalline materials and gas phase species. However to model complex chemical processes involving nuclear materials it is necessary to model the thermodynamic properties of phases over ranges of both temperature and composition. This requires use of the CALPHAD technique where critically assessed thermodynamic data for component binary and ternary systems can be extrapolated to multicomponent systems and then software such as MTDATA [7] used to calculate chemical and phase equilibria. During this project a thermodynamic database has been developed through critical assessment of all related experimental thermodynamic and phase diagram data for a number of systems and data gleaned from the scientific literature.

The nuclear fuel was taken to be based on $(U,Pu)O_2$ but incorporating minor actinides such as Np and Am. Data for the fuel, fission products and containment materials have been reviewed and new data assessed to represent their interaction with the Na coolant, in order to permit calculation of phase and chemical equilibria for severe accident scenarios. The ultimate aim is to publish the data developed during this project within a revised version of “Thermochemical data for reactor materials and fission products”, last published in 1990.

This resulted in a thermodynamic database. Data currently in the MetroFission thermodynamic database represent the systems:

- O-U-Pu
- Am-Fe-Np-Pu-U-Zr (some binary systems)
- Mo-Pd-Rh-Ru-Tc (metallic fission products)

which have been validated, modified and/or reassessed with consistent reference states for elements.

Further critical assessments for the systems Na-O, Na-I, Cs-I, Na-Te, Cs-Te, Zr-Te, and Zr-O are currently being carried out using available experimental data supplemented by calculations using *ab-initio* quantum mechanics. This will be extended in any follow on project with the systems: Na-U-O, Na-Pu-O, Na-Fe-O, Na-Zr-O, Na-Cs, and Zr-I.

The thermodynamic database(s) arising from this work will be compatible with the major software packages used to calculate phase equilibria from thermodynamic data.

Using the combination of models and data assessed in the project it will be possible to make predictions related to the high-temperature phase equilibria (such as phase transformation temperatures, vapour pressures or the potential interaction between the fuel and coolant), which can partly be validated by comparison with experimental observations.

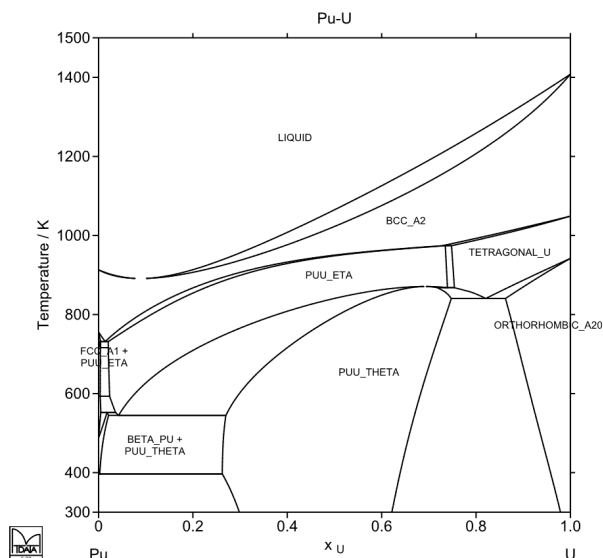


Fig 3. Calculated phase diagram for the Pu-U system

C. Thermophysical Properties of Advanced Materials

This work package of the Metrofission project aimed to implement a European metrological infrastructure for the study of thermophysical properties at high temperature [8] [9]

[10] for materials having thermal properties similar to those used in fission reactors. The main objective was to enable the nuclear research laboratories to insure the traceability of the measurements that they carry out with their own apparatuses on irradiated materials and nuclear fuels in some cases up to 2000 °C. For that, the measurement capabilities of European National Metrology Institutes (LNE, PTB and NPL) for the characterization of thermal properties have been improved, with in particular the development of new metrological facilities. Two complementary apparatuses have been designed by PTB and LNE for the measurement of normal spectral emissivity of solid materials up to 1500 °C, and the reference diffusivimeter of LNE has been modified to perform thermal diffusivity measurements up to 2000 °C.



Fig 4. New diffusivimeter of LNE

Figure 4 shows the new configuration of the high temperature diffusivimeter of LNE, and Figure 5 presents a scheme of the lamp image furnace designed to heat the specimens up to 1500 °C in the new high temperature emissometer of LNE. More information about the equipments developed in the framework of this project by PTB and LNE can be found in [11] and [12]. These apparatuses have been validated in their operating temperature ranges by measuring emissivity and thermal diffusivity of two homogeneous materials (tungsten and graphite) [13]. Thermal diffusivity measurements were also performed on these materials by JRC-ITU with two homemade facilities as well as by NPL and PTB with commercial equipments. Obtained results are in agreement with measurements performed with the new high temperature diffusivimeter developed for this project and with data available in literature. These new metrological facilities will be applied to the characterisation of thermophysical properties of some materials (Ni, MgO and ZrO₂) that could be used as “transfer reference materials” by nuclear research laboratories for high temperature thermal properties measurements.

D. Improved Cross Sections through Neutron Metrology

New fast reactor designs will involve materials exposed to higher-energy neutron fields. The current nuclear databases concentrate on thermal energies. Work will need to be done to determine and/or achieve lower uncertainties of nuclear data at higher energies. Advantage was taken of the experience of National Metrology Institutes (NMIs) in neutron metrology, particularly fluence measurements. Practically the project aims at improving neutron interaction data measurements beyond the present state-of-the-art, which is set by self-normalizing methods and the neutron data standards used at laboratories where the data are measured [14] [15].

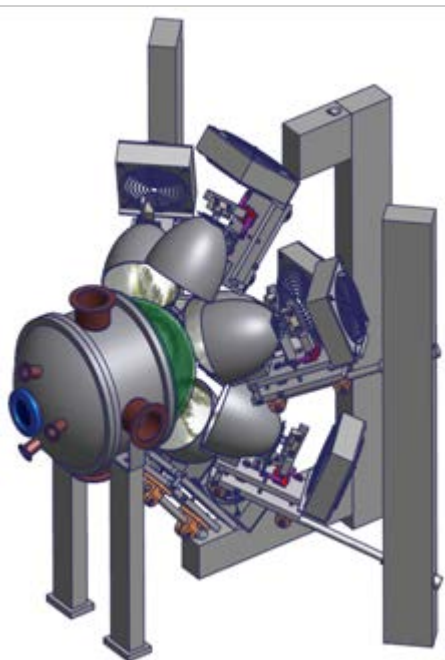


Fig 5. Schematic of the lamp image furnace of the high temperature emissometer of LNE

The program of work included the establishment of an easy-to-use secondary fluence standard at the EC-JRC-IRMM GELINA facility along with a procedure for reliable determination of the efficiency of fluence measurement devices used in neutron data measurements at EC-JRC-IRMM and elsewhere. The set-up consists of a fixed flux monitor, consisting of a double Frisch-gridded ionization chamber loaded with B-10 layers, and a reference chamber, consisting of a double Frisch-gridded ionization chamber loaded with a well characterized U-235 layer on one side and a B-10 layer on the other side. This reference chamber was installed at one of GELINA's flight paths and allowed determining the neutron flux as a function of neutron energy in an absolute way. The reference chamber can be replaced with any other fluence monitor in order to calibrate this device. A multilayer parallel plate transfer chamber consisting of 8 layers of U-235 was calibrated in this way and used for fluence measurements at other neutron beams. All U-235 layers were prepared and carefully characterized at EC-JRC-IRMM.

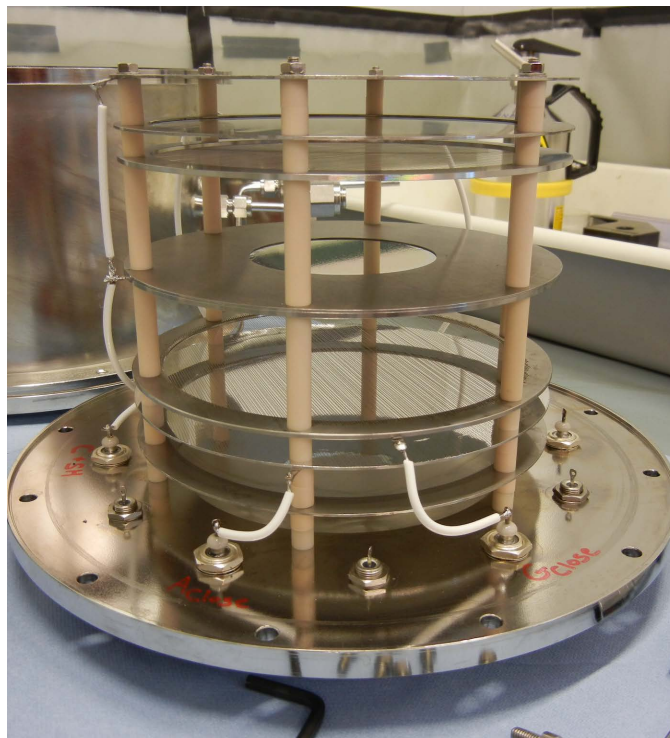


Fig 6. A double Frisch-gridded ionization chamber loaded with U-235 and B-10 was used as a reference chamber for neutron fluence measurements.

The potential of these fluence measurement devices was demonstrated by a number of neutron induced fission cross section measurements on Pu-240 and Pu-242 with monoenergetic neutron beams produced at two different NMIs, i.e. at the Van de Graaff accelerator and the cyclotron of the Physikalisch-Technische Bundesanstalt (PTB) in Braunschweig (D) and at the Van de Graaff accelerator of the National Physical Laboratory (NPL) in Teddington (UK). The measurements were performed using gas filled detectors, consisting of a Frisch-gridded ionization chamber containing the Pu sample, mounted back to back to a parallel plate chamber containing a U-238 sample. During the experiments the neutron fluence was measured in parallel with the U-238 parallel plate chamber and with the fixed neutron fluence monitors available at the respective institutes.

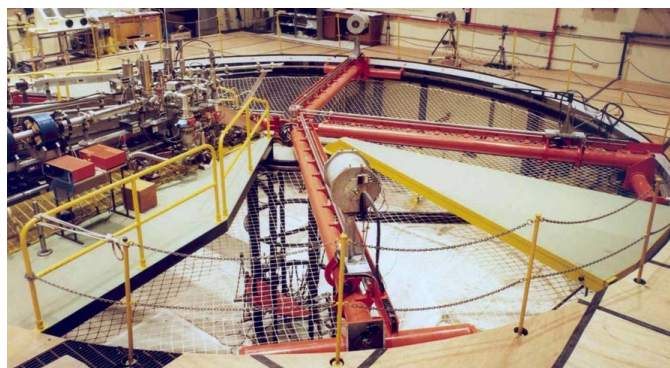


Fig 7. The 3.5 MV Van de Graaff accelerator at NPL (UK) produces monoenergetic neutrons in a low-scatter environment.

E. Nuclear Decay Data

The minor actinides and their decay products have generated increased interest in recent years because of their significant role in the foreseen development and adoption of nuclear fuel. A list of specific needs for improved actinide decay data has recently been outlined in a review by the IAEA [16] [17]. Responding to those needs, an objective of this project was to measure the alpha-particle emission probabilities of U-238, using highly enriched material with certified isotopic composition. Furthermore, as fission fragments containing more beta-decaying radionuclides will result from proposed fast reactors, developing beta spectrometry by cryogenic detectors was another objective aimed at improving the measurement of beta spectra and verifying theoretical models, particularly for those of the forbidden type. This will lead to improved calculations of decay heat produced by nuclear fission, a quantity important for safe reactor operation (reactor shutdown, post irradiation handling of nuclear fuels).

Thus two major objectives steer this part of the research at improving

1) Our knowledge of decay data for radionuclides plays a major role in power-related fields. It was proposed to measure some nuclear data that have been identified as incomplete or inconsistent, and in which NMIs could provide a major impact with their dedicated facilities for primary standardization of radioactivity. NMIs have a proven record of providing accurate decay data such as half-lives and alpha-particle emission probabilities. It was decided to perform alpha-particle emission probability measurements in the decay of ^{238}U , for which decay data were very scarce and an explicit demand for better data had been expressed [16] [17]. To achieve this goal, extensive tests were needed to optimize the source preparation by electrodeposition [18], the sources had to be measured for 1-2 years in several detectors [19] and a magnet system had to be developed to eliminate interfering conversion electrons that distorted the alpha spectrum [20]. The quality of the alpha spectrum obtained, in terms of accuracy, statistical precision and energy resolution was significantly better than in any previous measurement. The alpha-particle emission probabilities of ^{238}U have been improved by an order of magnitude [19]. The newly obtained decay data will be made publicly available to the international community.

2) The use of a cryogenic detector - and in particular a metallic magnetic calorimeter - to measure beta energy spectra is quite innovative. It will help us to improve our knowledge of the shape of beta spectra through experiment and implement this into theoretical models. The new technology of cryogenic detectors offers exceptional characteristics in terms of energy resolution and detection efficiency in a wide energy range. In particular, it can obtain information also at the low-energy side (threshold as low as 180 eV), as it is less prone to energy losses and electronic noise compared to other methods. Measurements have been performed on a beta spectrum of the allowed type, ^{63}Ni , and of the first forbidden type, ^{241}Pu . The tests have taught us how sources should best be prepared for this method and has confirmed some theoretical models of the shape of an allowed beta spectrum. It was also demonstrated

to be applicable to ^{241}Pu , since for this nuclide certain special conditions are met. One of the most important conclusions following from these measurements is the clear evidence for an 'exchange effect' occurring in beta decay, which has as effect a clear increase of the beta spectrum at low energy. No other measurement has ever demonstrated this effect so clearly, as the threshold level in this experiment was exceptionally low. A correction factor for the exchange effect has been implemented in the theoretical calculation of beta spectra, which correctly predicts the enhancement by 20% of the beta spectrum of ^{63}Ni at 180 eV. This improvement will have an impact in several fields, such as primary standardization of radioactivity and decay heat calculations.

F. TDCR: Metrology for In-situ Activity Measurements

The general aim of this work is to enhance operation of new generation nuclear power plants (NNPP) by enabling on-site measurement of low-energy beta-emitters created in the fuel cycle (e.g. Pu-241) and/or as activation products in the reactor and its enclosure (e.g. S-35, Ni-63, Ca-41, H-3 etc.).

Addressing the demand of nuclear power plants for on-site activity measurements of pure beta emitters, the objective for this part of the project was therefore to develop a miniature size Triple to Double Coincidence Ratio (TDCR) system, clearly advancing technology beyond the current state-of-the-art [21] [22] [23]. The aim was to develop metrology instruments suitable for use on-site, with lowered measurement uncertainties and traceability to SI units by applying the primary technique of TDCR. Five partners have built miniature TDCR systems, miniaturising the instrument's detection chamber by using newer, smaller, efficient photomultiplier tubes. One example is shown in Figure 8. The systems [24, 25, 26, 27, 28, 29] have been validated with previously standardised radioactive solutions and inter-compared using solutions of H-3 and Pu-241.

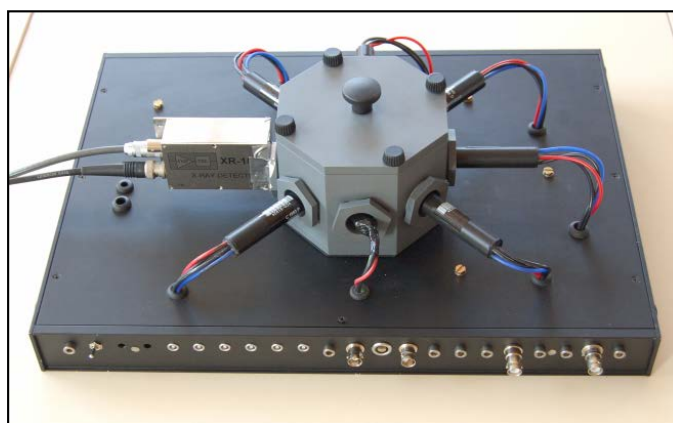


Fig 8. An example of a miniature TDCR system (this specific prototype has been developed at CEA-LNHB in France).

G. Digital Coincidence Counting for Radionuclide Metrology

The work has focused on the provision of a means of on-site activity standardisations for various photon-emitting radionuclides that may be readily standardised using the 4pb-g

coincidence counting technique [30]. The data acquisition systems utilised are based on high-speed digitizers, where the A/D conversion is performed as close as possible to the outputs of the detectors/preamplifiers. Critical examinations on optimisation of the coupling of ADCs to the detector systems (input impedance, bandwidth and voltage ranges, signal to noise ratio etc.), as well as the analysis of the digitized data streams (both “on-line” and via *list-mode* data sets) in order to realise robust 4pb-g Coincidence Counting systems were performed. An international effort is now underway to develop a standard data format for list-mode data sets used in radionuclide metrology.

III. CONCLUSION

This paper presents the various work completed within an EMRP Joint Research Project MetroFission, which started in September 2010. This project has aimed at providing solutions to metrological challenges in relation to nuclear new build and specifically Gen IV reactors so that these reactors can be designed, built and operate safely. The project proposes innovative temperature measurements in order to satisfy the need for measurements at the higher temperatures experienced in GenIV reactors and possible solutions for long-term stability of such equipment. The project has delivered thermo physical and thermo chemical data by experimental methods as well as modelling in order for new nuclear power plants to have suitable construction materials. It has also addressed the issue of nuclear fuel and how accurate and traceable methods of measurements of neutron flux and nuclear data can improve cross-section measurements, crucial for the calculation of the fission yield and the reactor efficiency. The nuclear data will furthermore benefit the operation of the power plant, once realised, as such data is required for calculations of for example the radiation heat. With regards to methods for radionuclide metrology that previously was designated to specialist primary conditions, this project has been investigating the possibility of realising an in-situ measurement facility that will enable accurate determination of difficult to measure radionuclides, in the fuel and during operation. In summary, this project has provided a range of metrology solutions, with a focusing on traceability to SI units that are working towards the realisation of a low-carbon emission source of energy that will provide Europe with a source of sustainable energy.

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