RADIONUCLIDE METROLOGY FOR NEW GENERATION NUCLEAR POWER PLANTS

J. Keightley[1] *P. Cassette*[2], *L. Johansson*[1]

[1] NPL, Teddington, United Kingdom
[2]CEA-LNHB, Saclay, France

E-mail (corresponding author): john.keightley@npl.co.uk

Abstract − The radioactivity measurement techniques being developed within the EMRP "MetroFission" Joint Research Project (JRP) [1], are aimed at performing on-site activity measurements, by the development of a portable Triple to Double Coincidence Ratio (TDCR) system along with associated digital electronics. The TDCR standardisation method for beta-emitting radionuclides (e.g. ^{3}H and ^{241}Pu) produces an estimate of the sample activity without the requirement for a preceding calibration, and has the potential to reduce uncertainties by a factor of two. The digital systems being developed are state of the art, utilising sampling rates of up to 10^{9} s^{-1} and are being employed to implement both two channel ($4\pi\beta$–γ coincidence counting) and three channel (TDCR) detector systems for absolute activity concentration measurements.

Keywords Radionuclide metrology, TDCR, Digital signal processing, Instrumentation, Generation IV

1. CONTEXT

Nuclear energy is one of the energies that 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 [2]. 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 [3], [4].

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 Flammanville, 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 incremental improvements called Generation III+. The purpose of the Metrofission JRP 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.

The overall aim of the work packages on radionuclide metrology is to enhance operation of new generation nuclear power plants (NNPP) by enabling on-site determination of low-energy beta-emitters created in the fuel cycle (e.g. ^{241}Pu) and/or as activation products in the reactor and its enclosure (e.g. ^{35}S, ^{63}Ni, ^{41}Ca, ^{3}H etc.). It is normally a greater scientific challenge to measure pure beta emitters but it should be mentioned that the same system could also be applied to alpha emitters, i.e. most of the minor actinides (which in the NNPP will be part of the fuel cycle and the reprocessing of the fuel).

Addressing the demand of nuclear power plants for on-site activity measurements of pure beta emitters, Work Package 6 (WP6) of the MetroFission JRP plans to develop a miniature Triple to Double Coincidence Ratio (TDCR) system, clearly advancing technology beyond the current state-of-the-art. Present systems are metrology instruments not suitable for use on-site, partly due to their large size and weight. One step in this direction will be the miniaturisation of the instrument's detection chamber itself by selecting and utilising newer, smaller, more efficient photomultiplier tubes [5]. However, Work Package 7 (WP7) will also look at replacing the currently used analogue electronic modules (non-portable) with digital acquisition systems and signal processing in order to significantly improve digital coincidence counting (DCC).

2. SCIENTIFIC AND TECHNICAL OBJECTIVES

2.1. Triple to Double Coincidence Ratio (TDCR): WP6

The TDCR method is relatively well established in European NMIs [6-10] and was primarily developed as an absolute method for the activity measurement of beta emitting radioisotopes. However, the method also has potential to be extended to other nuclides like alpha emitters (without any adjustments to the system), to electron-capture decaying nuclides [11, 12] and to nuclides with complex decay schemes including many gamma-rays (the later two cases with significant updates in the theoretical model that is used for the activity calculation). It is based on liquid scintillation, i.e. the emission of light resulting from the transfer of energy from ionizing radiation emitted by an aliquot of radioactive solution to a solvent and then to fluorescent molecules (scintillator) [13, 14]. The light is collected by three photomultipliers (PM) (Fig. 1) and the detection efficiency is evaluated by using a model that uses the ratio of triple-to-double coincidences between the PM tubes. The counting and signal treatment is carried out using dedicated analogue electronic modules [15], which are not commercially available. The source preparation is simple as it consists in adding a weighted drop of radioactive solution in the flask filled with the scintillating cocktail.

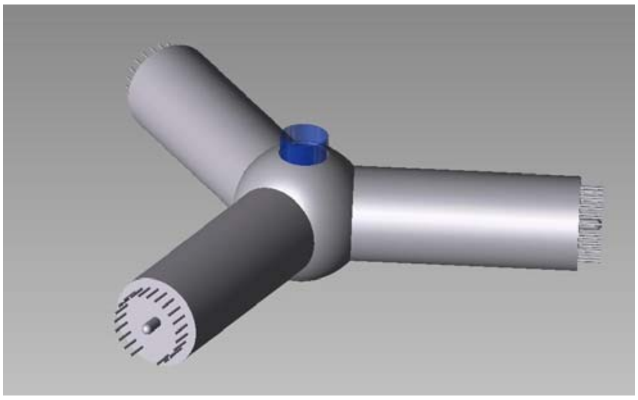

Fig. 1. Schematic TDCR system with three photon detectors.

The TDCR method is based on the free parameter model in LSC. Under some physical and statistical assumptions concerning the light emitted by the scintillator, the detection efficiency can be calculated from the knowledge of a free parameter linked to the intrinsic light efficiency of the scintillating source in a specific counter. The TDCR method is a way, among others, to calculate this free parameter. The basic physical and statistical models used are described hereafter.

If it is assumed that the energy E of an electron is released in the scintillator, light is emitted with a mean number of photons m. The statistics of the number of photons emitted by the scintillating source can be described by a Poisson distribution: the probability of the emission of x photons for a mean value m is:

$$P\left(x/m\right)=\frac{m^{x}e^{-m}}{x!} \tag{1}$$

The detection efficiency can be derived from the detection probability, which is the complement of the non-detection probability. The non-detection probability is the probability to get zero photons for a mean expected value of m. When introducing this in the Poisson formula for a 1/3 symmetry counter using three photo-detectors with the same quantum efficiency ν, the detection efficiency for one photo-detector is:

$$R_1 = 1-e^{-\nu m/3} \tag{2}$$

For two and three photo-detectors in coincidence, we get, respectively, Eq. (2) to the powers of 2 and 3. The detection efficiency for the logical sum of the double coincidences is:

$$R_D = 3\left(1-e^{-\nu m/3}\right)^2 - 2\left(1-e^{-\nu m/3}\right)^3 \tag{3}$$

The light emission process is not linear and the mean number of photons emitted is known not to be proportional to the energy released in the scintillator by the ionising radiation. A semi-empirical relation, described by Birks, gives the mean number m of photons emitted as a function of the energy E released in the scintillator, and the linear energy transfer dE/dx:

$$m\left(E\right)=\int_0^E \frac{AdE}{1+kB\,dE/dx} \tag{4}$$

A is a free parameter characterising the LS-cocktail efficiency and kB is a semi-empirical parameter.

If the energy spectrum emitted by the radionuclide is described by a normalised density function $S(E)$, the ratio of the probability of the triple coincidence to the probability of the double coincidence, TDCR, is:

$$TDCR=\frac{\int_{spectrum} S\left(E\right)\left(1-e^{-\eta}\right)^3 dE}{\int_{spectrum} S\left(E\right)\left(\left(3\left(1-e^{-\eta}\right)^2-2\left(1-e^{-\eta}\right)^3\right)\right)dE} \tag{5}$$

with:

$$\eta=\frac{\nu}{3}\int_0^E \frac{AdE}{1+kB\,dE/dx}$$

The triple and double coincidences are recorded by the LS counter. For a large number of events, the ratio of experimentally determined counting rates converges toward the ratio of calculated efficiencies. The detection efficiency calculation algorithm is used to determine the free parameter νA (figure of merit as the number of photons emitted per unit of energy released in the scintillator), for which the experimental frequency ratio equals the calculated ratio. It can be shown that for pure beta spectra, there is a unique solution, but for other kinds of spectra, including that obtained for nuclides that decay by electron capture, one TDCR value can correspond to up to three detection efficiency values. In that case, the use of several counting conditions for a single sample can solve the ambiguity.

Current TDCR systems are home-made metrology instruments neither aimed at nor suitable for in-situ measurements. They are generally of large size (larger than 1,5 m x 1,5 m x 1 m), of heavy weight (about 100 kg) and not transportable. The activity determination results from a calculation relying on a model specific to the decay scheme of each measured nuclide. The new scientific areas that will be pursued in this work package will be the realisation of a miniature self-calibrated primary TDCR system, which is state-of-the-art, for use on-site. The challenge is then to develop a versatile portable, table-top designed instrument, from this metrology device without losing its characteristics. This implies three major improvements:

- Miniaturisation of the detection chamber by studying new types of photomultipliers, efficient and smaller,
- Miniaturisation of electronic modules by exploring the possibilities of digital treatment,
- Validation of models and extension of them to nuclides with special beta spectrum shapes, to nuclides with complex decay schemes including many gamma- rays and to nuclides with higher atomic number decaying by electron capture.

The task includes the study and choice between different types of photomultipliers (channeltrons, silicon photomultiplier tubes, multi-anode PM tubes, etc), the study of the implementation of digital signal treatment and the study of theoretical models (validation, extension). The design protocol will also consider aspects such as facilitating a self-calibration by adding a source of Compton electrons of defined energy [16]. The implementation of the system will be tested including its portability.

Only one commercial company has up to now started the production of TDCR devices. The JRP will also test the measurement capabilities of this currently available commercial TDCR system (Hidex), with associated validation measurements.

2.2. Digital Coincidence Counting (DCC): WP7

Work Package 7 (WP7) will develop a high-sampling-speed digital system for the purpose of performing activity measurements by coincidence counting in order to:

- Enable an in-situ detector system (analogue electronics would not apply to being portable), as proposed in WP6.
- Extend the analysis software to perform radioisotope standardisations for a range of beta-gamma emitters via the $4\pi\beta{-}\gamma$ coincidence counting technique.
- Create a cost-effective system that is less labour intensive – coincidence techniques for activity determination normally requires manual variation of parameters whereas a digital system offers the possibilities for automatic parameter variation.
- Facilitate some form of basic pulse shape discrimination which in turn can be used to discriminate between different types of radiation (e.g. neutrons and gamma-rays)
- Allow for high count-rates of activity, which would give a more dynamic range of radioactive samples that can currently be measured.

By taking advantage of the new possibilities offered by digital signal processors (DSP), FPGA based systems (field programmable gate arrays) and the performances in terms of sampling frequencies at relatively low cost, manufacturers of nuclear instrumentation are or will be developing counting systems based on digital rather than analogue treatment of pulse trains from radiation detector systems.

However, national metrology laboratories still mainly use conventional analogue systems. It is now a necessity for them to obtain experience and expertise in digital treatment to be able to conceive and develop such systems for metrological applications that meet the metrological needs. They must take the lead and gain recognised expertise to accompany the evolution in the nuclear industry and be able to assess the potential performance of such systems.

Digital acquisition systems developed at NMIs for the purpose of digital coincidence counting have up until now utilised signals from radiation detectors and associated shaping amplifiers to determine estimates of the pulse arrival times and peak amplitude of the signal [17]. The limited sampling rate of existing ADCs has not allowed fast pulses to be sampled accurately; hence the pulse shape is determined largely by the amplifier integration of output pulses. Those systems that have attempted to sample and store the entire pulse shapes have exhibited a limited dynamic range of source activities (due to the large data throughput rates), and those which have attempted to store only estimates of the pulse arrival times and amplitudes necessarily reject relevant information.

Recent advances in the field of high-speed digital sampling and digital signal processing along with the advent of user-configurable FPGA devices can significantly improve digital coincidence counting in the provision of primary standards of radioactivity. State-of-the-art technology nowadays provides 12-bit cards working at Giga-samples-per-second (GSPS) rates, with on-board FPGA devices, which greatly enhances the application of digital signal processing for the implementation of digital coincidence counting.

The MetroFission JRP partners will develop high performance 2-channel (DCC) and 3-channel (TDCR) acquisition systems with associated pulse characterisation and software that will be tested for both spectrometric and coincidence measurements with regard to:

- count-rate dependence
- pulse-height resolution
- pulse timing resolution,
- off-line processing of coincidence counting with variable resolving times.

Optimal design of a DCC system requires previous knowledge of the pulse characteristics. Therefore, pulses from at least two types of proportional counters (atmospheric and pressurized) will be digitized with a high speed sampling card at a rate in the order of 1 GSPS. The digitized shapes will be analysed by a set of numerical algorithms to establish the parameters that best characterise the pulses depending on their origin (alpha particles, beta particles, neutron and gamma-rays) and to identify piled-up pulses at high count rates. A numerical database of digitized pulses will be made available to the JRP partners to help in the design of the algorithms to be implemented in the final hardware and DSP system.

This digital system will be developed in co-ordination with WP6, but designed so that it can be applied to other detectors of ionising radiation and other detector configurations with up to three channels. The work also involves validation measurements of the systems by comparison with analogue system and by inter-laboratory comparison of radioactive sample. The final report will include the description of the DSP system along with recommendations regarding the feasibility of digital sampling of pre-amplifier pulses, pile-up discrimination and the benefits of off-line versus on-line digital systems.

3. CONCLUSION

It can be concluded that each MetroFission JRP participant will build their own portable TDCR LS counter prototype based on a common protocol. The lead laboratory of WP6, CEA-LNHB, has completed a document defining the characteristics of the transportable TDCR counter in progress at the four partner institutes. This report included a description of the TDCR method, a commented list of criteria, which must be fulfilled by a TDCR counter, a bibliography and a review of available photo-detectors that can be used in the project.

Regarding the Digital Signal Processing, following the dissemination of a literature review [17-19], discussions between the participants have led to the following design specifications of the Digital Counting System (as a minimum):

	Minimum Specification	Recommended Specification
analog capture channels	2	4
Sampling rate per channel	100 MSPS	≥ 100 MSPS
ADC Coding depth	10 bit	≥ 12 bit

Other specifications such as the number of minimum number of FPGA logic cells, DSP slices and on-board memory, were left up to the individual NMI's discretion. It was decided that it is not necessary to log all sampled data-points above a user-defined threshold. Time stamping of individual pulses is a necessity, and sufficient information to reconstruct pulses (rise time, fall-time, pulse widths, pulse pile-up flags) is required if data-logging for off-line analysis is desirable. A common data format for comparison of off-line analysis routines is being devised.

The NMIs are presently familiarising themselves with the capabilities of their respective equipment, particularly with regards to FPGA implementation of on-board logic for counting, dead-time, live-timing etc. Initial data collection tests have been made on pulses from NaI(Tl), Ge and $LaBr_3$ detector systems, using as inputs both the direct pulses from PM bases, and those following pre-amplification and shaping amplifiers. The need for specific circuitry in order to adapt the voltage range of the signals provided by the shaping amplifiers to the voltage range of the ADC inputs (typically 0 to 2 V) has been discussed in depth.

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Authors:

[1] NPL Management Ltd, Hampton Road, Teddington, TW11 0LW, United Kingdom
Dr. J. Keightley[1],phone +44(0)2089436398,
john.keightley@npl.co.uk
Dr. L. Johansson[1], phone +44 (0)2089438587, fax +44 (0)2089438700, lena.johansson@npl.co.uk

[2] Laboratoire National Henri Becquerel, LNE-LNHB
CEA-Saclay, France
P. Cassette[2], phone +33 1 69 08 48 68, fax +33 1 69 08 26 19,
philippe.cassette@cea.fr