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Development of Portable Liquid Scintillation Counters for On-Site Primary Measurement of Radionuclides Using the Triple-To-Double Coincidence Ratio Method

P. Cassette, M. Capogni, L. Johansson, K. Kossert, O. Nähle, J. Sephton, P. De Felice

I. INTRODUCTION

Abstract— The Triple-to-Double Coincidence Ratio (TDCR) method in Liquid Scintillation counting (LSC) is a primary radionuclide standardization method widely used in National Metrology laboratories and was primarily developed for the activity measurement of beta emitters. It is based on liquid scintillation: the light is detected by three photomultipliers (PM) and the detection efficiency is evaluated by using a model which uses the ratio of triple-to-double coincidences between the PM tubes.

Up to now, most of current TDCR systems were locally-made metrology instruments neither aimed at nor suitable for in-situ measurements. In the framework of the European Metrofission project, a work package was dedicated to the realisation of miniature self-calibrated primary TDCR systems, which are state-of-the-art, for use on-site. The challenge was to develop a versatile portable, table-top designed instrument, from this metrology device. This implied improvements for the miniaturisation of the detection chamber, for the miniaturisation of electronic modules by exploring the possibilities of digital treatment, and for the 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. Four prototypes of counters were built by the Metrofission partners ENEA (Italy), LNHB (France), NPL (UK) and PTB (Germany) using various technical approaches.

The paper describes these prototypes and provides some details on the choice of the technical options concerning the design of the optical chamber, of the photodetectors and of the acquisition system.

Index Terms—Liquid scintillation, Metrology, TDCR counter.

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The European Metrology Research Programme (EMRP) is a coordinated programme that encourages cooperation between the various European metrology laboratories and this paper presents work performed as part of the EMRP Joint Research Project MetroFission that addresses metrology issues relating to the next generation nuclear power plants. One aspect of the project has been concerned with development of instrumentation for on-site measurement of the activity levels of low-energy beta emitters. The essential aim was to develop an instrument suitable for on-site use, starting from a metrology laboratory equipment based on the primary technique, the Triple-to-Double Coincidence Ratio (TDCR) technique. The TDCR technique is an advanced method of liquid scintillation counting that has been adopted at many National Metrology Institutes (NMI's) for the purpose of primary standardizations [1], [2], [3]. The liquid scintillation source is placed in an optical chamber that has a highly reflective inner surface and three PM tubes are used to detect the emission of light from the source. The number of counts that arise from coincident detection by any pair of tubes (the "doubles") and all three tubes (the "triples") are determined. From the knowledge of the ratio of triple to double coincidences, the detection efficiency of the system can be calculated and used for activity determination.

At the onset of this project, TDCR systems established at NMI's tended to be rather bulky and therefore ill-suited for routine on-site use at, for example, a nuclear power plant. Therefore the MetroFission project set out to develop miniaturized TDCR systems that would be portable and easy to use in an industrial environment. Four NMIs; ENEA (Italy), LNHB (France), NPL (UK) and PTB (Germany) have developed miniature TDCR systems, based on their own design.

Current TDCR systems are home-made metrology instruments not suitable for in-situ measurements. They are generally of large size, of heavy weight 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 were pursued in this work package are the realisation of a miniature self-calibrated

primary TDCR system, which is state-of-the-art, for use on-site. The challenge was 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 PMs, 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.

II. THE TDCR METHOD IN LSC

The LS process is the emission of light following the interaction of ionising particles or photons with a liquid scintillator. The radiation–matter interaction eventually creates electrons, whose energy is dissipated within the solvent of the scintillator. Part of this energy is then transferred to the fluorescent molecules, which emit light by radiative de-excitation. Depending on the nature of the scintillator, the light pulse may have a prompt component followed by delayed emission. The duration of the prompt emission is lower than 10 ns for most liquid scintillators.

The light emitted is generally in the blue to near-UV spectrum range. This emission is incoherent and isotropic, but most of the light escapes from the meniscus created at the scintillator–vial wall interface.

The overall energy conversion in the liquid scintillation process is rather low: 1 keV of electronic energy produces only a few photons. Detection efficiency is thus generally lower than 1 and must be calculated.

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

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

These photons are collected on the photocathodes of the PM tubes and photo-electrons are then produced with a quantum efficiency ν . 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 photo-electron for a mean expected value of $m\nu$.

When introducing this in the Poisson formula for a 1/3 symmetry counter using three photodetectors with the same quantum efficiency ν , the detection efficiency for one photodetector is

$$R_1 = 1 - e^{-\nu m/3} \quad (2)$$

This formula applies when the PM tube setting makes possible the detection of a single photoelectron.

For two and three photodetectors 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:

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

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 :

$$m(E) = \int_0^E \frac{AdE}{1 + kB \frac{dE}{dx}} \quad (4)$$

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:

$$TDCR = \frac{\int S(E)(1 - e^{-\eta})^3 dE}{\int S(E)(3(1 - e^{-\eta})^2 - 2(1 - e^{-\eta})^3) dE}$$

with

$$\eta = \frac{\nu}{3} \int_0^E \frac{AdE}{1 + kB \frac{dE}{dx}} \quad (5)$$

This simple model assumes that the PMTs have similar efficiency but if this is not the case, a set of 3 equations must be solved instead of equation (5) (see, e.g. Ref [3]).

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 towards 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 those 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.

III. GENERAL DESIGN PRINCIPLES AND OPTIONS

We adopted the following design principles, taking into account the constraints on the portability of the system and also the requirements of the TDCR model described above:

A. Portability and Power Supply

A portable TDCR system should fit into a standard car. If the counter is supposed to be handled by a single person, no part should be heavier than approximately 30 kg.

As it can be expected that electric mains is available in places where the system is intended to be used, a battery is not needed for power supply. The power consumption must be less than 1 kW, depending on the components used.

The implementation of temperature control is not considered to be absolutely necessary, but an optional feature.

The samples are changed manually and the implementation of an automatic sample changer is an optional feature.

Either a standalone system or a computer peripheral system can be considered.

B. Shielding

Passive shielding is heavy and therefore reduces the transportability. An active shielding might lead to a loss of real counts, especially when measuring photon-emitting radionuclides. It might also influence the photon statistics, which is a crucial assumption of the TDCR model. Thus, a potential active shielding should be at least disconnectable. It also increases size and weight, and the evaluation of the guard detector signals complicates both the electronics and the software design.

C. Optical Chamber and Light Collection

As the efficiency of the LS process is low, it is very important to collect the maximum amount of light. As a rule, the uncertainty of the detection efficiency increases as the detection efficiency decreases, because the model has to correct for the missing information. It is advisable to use highly reflective materials in the optical chamber. Specular light reflections must be avoided in the optical chamber in order to increase the isotropy of the reflected light and prevent light trapping by internal reflection. It is advisable to use photodetectors with etched windows and etched glass vials in order to avoid total reflection at the air–glass interface. It is recommended that the counter allows the use of standard 20 ml vials for comparisons with existing non-portable counters.

D. Photodetectors Choice and Discriminator Threshold Adjustment

As the counter must have a non-zero detection probability for single photons, it is essential that the single-photoelectron peak is well resolved in order to allow a precise adjustment of the counter threshold. This implies the use of high-gain photodetectors having a good peak-to-valley ratio of the

single-photoelectron peak. In this case, the discriminator level must be adjusted to a flat region of the spectrum, just before the single-photoelectron peak, in order to make the counter insensitive to small photodetector gain fluctuations. The optimum diameter is 12.5 mm or 25 mm for classical PMTs and is limited by available technology for other photodetectors. Commercial miniature PMTs with square window can also be used. The detection efficiency can be improved by using surface treatment of the photodetector window. A diffusive layer is effective but refractive index adaptation by multilayer coating could be explored. Lower dark count rate can be obtained when the photocathode is at the ground potential, thus by using positive high voltage power supply.

The correct adjustment of the discriminators is mandatory when using the TDCR model, as the correct detection of single photoelectrons is the requirement for taking the detection efficiency as the complement of the non-detection probability (see equation (2)).

E. Efficiency Variation Method

Classical methods are based on optical (e.g. grey filters), mechanical (e.g. diaphragm or variation of the vial position) or electronic methods (e.g. PMT defocusing). Any efficiency variation method implemented in the counter increases size and complexity and hence could have an influence on the portability. Efficiency variation by means of chemical quenching is also possible and does not require special counter modifications.

F. External Gamma-Ray Source

The classical TDCR method does not need the determination of a quenching index, and an external gamma-ray source is not needed. Anyway it could be optional to use an external standard source for comparing with other standardization methods. The price to pay is an increase in complexity of the system and the need to comply with regulations concerning health protection, safety and radioactive material management and transportation.

G. Spectrometry

Spectrometry is not strictly necessary for the TDCR method but offers two useful features: first it facilitates the adjustment of the discriminator thresholds, and second it allows the verification of the radionuclide spectrum and sometimes the detection of impurities. A difficulty appears when using saturated photodetectors like channel PM tubes. In this case, spectrometry is only possible by the addition of a specific linear photodetector.

For a counter using classical PM tubes, spectrometry can be easily implemented by processing the signal of one of the PM dynodes through a charge amplifier. The minimum implementation in a portable TDCR counter could be an output which can be connected to a traditional spectrometry unit like the ones used for gamma spectrometry.

H. Dead-time Unit

Prompt light emission is followed by afterpulses correlated to the initial emission. This results in pulse clusters and spoils the stationary nature of the observed process. Moreover, the noise of some photodetectors, like PMTs, can appear as burst of pulses and cannot be considered as stationary. It is thus necessary to add a dead time period after each detected event in order to avoid false coincidences of photocathode noise and delayed light emission. The base duration of this dead time is typically a few tens of microseconds. The use of extending-type dead time ensures that every detected event is preceded by an event-free period: false-coincidence probability is thus minimised. The efficient and simpler evaluation of live-time when using an extending-type dead time is based on the use of a live-time clock.

I. System Symmetry

The use of matched photodetectors in a symmetrical arrangement is the only design that allows the use of the simple TDCR model previously described. Nevertheless, as it is expected to have variability in the photodetector responses, it is necessary to have access to individual double coincidences and to use a TDCR model taking asymmetry into account.

J. Balance Equations

The logical relation between the coincident and non-coincident pulses allows the calculation of balance equations:

$$A+B+C=T+D+S$$

$$AB+BC+CA=2T+D$$

To calculate these equations, it is necessary to acquire the signals (A, B, C) from the three PMTs, the logical sum of these signals (S), the three double-coincidence signals (AB, BC, CA), the logical sum of the double-coincidence signals (D) and the triple-coincidence signals (T).

It is useful to verify that these balance equations are fulfilled during each data acquisition. This is an indicator of the correct operation of the counter.

K. Coincidence Unit

Considering that the prompt light pulse duration is less than 10 ns and that this pulse is followed by spurious after-pulses it is necessary to use fast detectors, fast preamplifiers and fast-coincidence units in order to acquire clean signals and to minimize false-coincidence corrections. An adjustment of the coincidence resolving time in the 20 ns to 200 ns range is desirable.

The use of an FPGA-based system for digital pulse processing is desirable, as it is very flexible and may help to reduce the size of the system and to optimize its robustness.

Asymmetries of the photodetector efficiencies must be taken into account [3] and at least the counting in channels AB, BC, AC and T must be recorded. Even if D can be deduced from the previous signals, its acquisition is interesting

as it allows the verification of the fulfilment of balance equations. Other features, such as the counting of the single channels or gamma coincidences when using an additional gamma counter, can be considered as optional.

L. Detection Efficiency Variation Methods

As for some nuclides, equation (5) might have several solutions, it is necessary to change the detection efficiency of the system to solve this ambiguity.

The TDCR model also requires an external parameter: the kB coefficient. Several detection efficiency conditions give the possibility to find out the value of this kB coefficient for which the source activity remains the same for all counting conditions. Detection efficiency variation is possible by defocusing the photodetectors, by using coaxial grey filters around the source or by changing the position of the source in the optical chamber.

IV. DESCRIPTION OF PROTOTYPES

Each laboratory applied these design principles to develop their own prototypes which are described hereafter.

A. ENEA Counter

The optical chamber of the prototype developed by ENEA is made of PTFE, with a prismatic shape (Fig.1). The photomultiplier tubes used are Hamamatsu R7600U-200 square miniature PMTs, with a high quantum efficiency photocathode. The sample holder is composed of a lift to introduce the vial and there is an optical shutter to separate the sample holder and the optical chamber, so the PM tubes can be continuously powered, in order to increase the stability of the system (Fig. 2).

Anode signals are directly processed by a CAEN digitizer, acting as level discriminator and fast ADC. The amplitudes of the pulses are recorded in a file, associated to timestamps, and the coincidence calculation is made off-line by software. This allows the possibility to re-analyze the measurement with different parameters like the coincidence resolving time and the dead-time type and duration. In practice, this off-line treatment is transparent, as the processing delay is a few seconds. More details on this instrument can be found in [4] and [5].

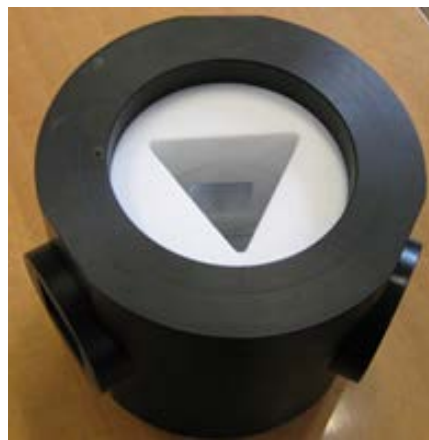


Fig.1. Optical chamber of the ENEA portable TDCR counter.



Fig. 2. ENEA TDCR counter

B. NPL Counter

The optical chamber of the NPL prototype is made of aluminium, internally coated with a Spectrafect® reflective paint. The first PM tubes tested were 29 mm diameter ET 9142A but they were replaced by Hamamatsu R6095 28 mm diameter tubes with lower noise and better resolution of the single-electron peak. The system is equipped with piston type sample loading and an efficiency-varying system. The installation of an electrically-operated shutter is planned like the installation of a stepper motor to set the vertical position of the vial (Fig. 3 and Fig. 4). Anode signals from the PMTs are amplified by Ortec 474 fast amplifiers and processed by the MAC3 coincidence unit [6]. More details on this instrument can be found in [7].

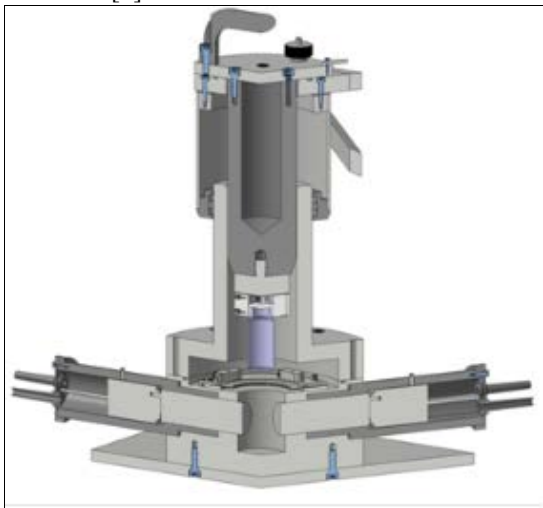


Fig. 3. Internal view of the optical chamber of the NPL counter



Fig. 4. Optical chamber of the NPL TDCR counter

C. PTB Counter

The PTB prototype uses 3 channel PM tubes from Perkin Elmer, in an optical chamber made of highly reflective material, OP.DI.MA ODM98 from Gigahertz-Optik (Fig. 5). The optical chamber and high-voltage modules are installed in a compact box (Fig. 6). Signal processing is made by an FPGA-based coincidence module 4KAM, specifically developed by PTB. This module emulates the MAC3 module but with additional features like an adjustable coincidence resolving time, and includes counters for coincident and non-coincident information and time signals. The processing is made on-line and information is transferred to a personal computer through a serial interface.

More information on this instrument can be found in [8].

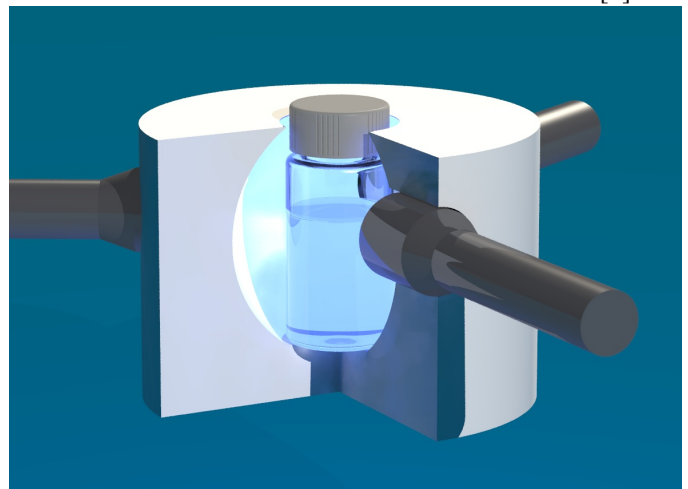


Fig. 5. Artist's view of the optical chamber of PTB counter



Fig. 6. The PTB TDCR counter with optical chamber, PM tubes and HV supplies.

D. LNHB Counter

The optical chamber of the LNHB prototype is made of polymer coated with TiO_2 paint. Six PerkinElmer channel PM tubes are used which are operated two by two in parallel in order to increase the photocathode surface (Fig; 7). A miniature Hamamatsu PMT is also used for spectrometry purposes, as the channel PM tubes operate in saturated mode. The system is also equipped with a CdTe gamma detector and a ^{241}Am source, to allow its use as a Compton spectrometer (Fig. 8) [9]. Acquisition electronics is based on a locally developed FPGA module, including on-line coincidence and dead-time processing, and counters for coincident, non-coincident and time signals. The acquisition module is connected to a personal computer through a USB interface.

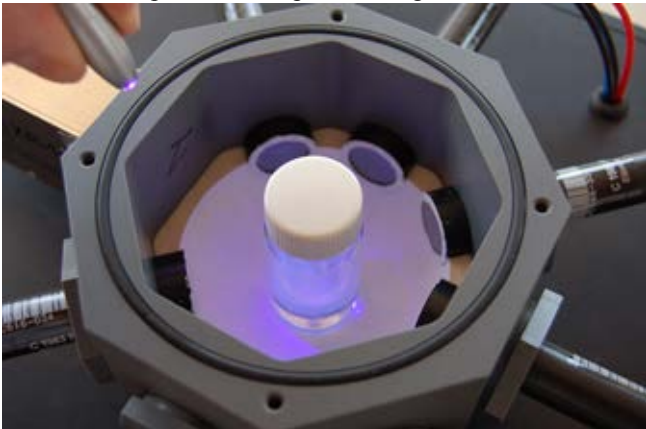


Fig. 7. Internal view of the optical chamber of the LNHB counter

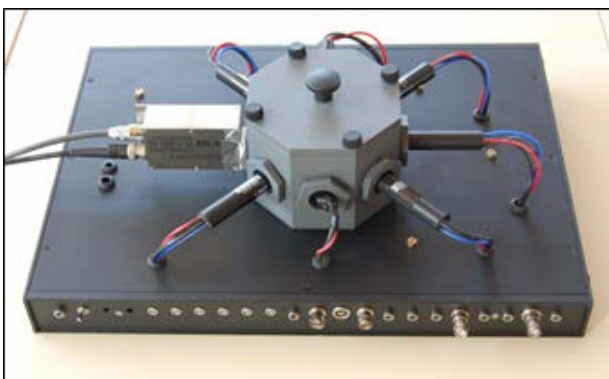


Fig.8. LNHB TDCR counter

V. SOFTWARE DEVELOPMENT AND VALIDATION

Specific software was developed for data acquisition and for the calculation of detection efficiencies of various radionuclides. ENEA and NPL developed programs to calculate coincidence counting rates for the TDCR system, from the raw acquisition files generated by the CAEN digitizer, using respectively the ROOT toolkit [10], [11], and C++. Specific VHDL programs were developed by LNHB and PTB for on-line data acquisition and processing units.

NPL developed Excel macros to calculate the detection efficiency of pure beta radionuclides. The extension of the TDCR model to radionuclides with complex decay scheme was also undertaken during this project and is reported in [12].

The validation of these prototypes is ongoing in the developing laboratories. It was observed that the detection efficiencies of portable counters of ENEA and NPL using dynode PM tubes are similar or slightly lower than the ones of traditional TDCR counters used in these NMI. The counters of LNHB and PTB using channel PMs exhibit a significantly lower detection efficiency than counters using dynode PM tubes. Nevertheless the first measurement results of low-energy emitters like ^3H are in very good agreement with the ones obtained using conventional TDCR counters, which is a good indicator of the quality of these portable instruments. A measurement comparison of the activity of ^3H and ^{241}Pu solutions is underway and the results will be available in June 2013.

VI. CONCLUSIONS

Portable TDCR counters were developed at ENEA, LNHB, NPL and PTB, for the purpose of in-situ measurements of low-energy beta emitters at a nuclear site. These systems will also be useful in other contexts, like for the on-site measurement of short half-life radionuclides in production centers, or as a traveling metrological instrument for radionuclide activity intercomparisons. First validation measurements of these instruments show that they are adequate for the purpose and yield activity values similar to the ones obtained with primary measurement systems in use in NMIs. This action gave also the opportunity to extend the use of the TDCR method to more complicated decay-scheme radionuclides than pure beta emitters.

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