Standardisation of ^{18}F using new portable $4\pi\beta(LS)-\gamma$ coincidence detection system at ENEA-INMRI

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Abstract. This project concerned with the development at ENEA-INMRI of new in-situ $4\pi\beta(LS)-\gamma$ coincidence detection system for activity measurement of the short half-life radionuclides used in Nuclear Medicine. The hardware of the new portable $4\pi\beta(LS)-\gamma$ coincidence detection system was implemented at ENEA-INMRI, in collaboration with Catania University and INFN, by adding a gamma channel on the existing TDCR portable detector available at ENEA-INMRI. A new data analysis software was developed at CAEN, independently from an existing one elaborated at ENEA.INMRI, in order to analyse the data recorded in list-mode by the new detector equipped with the CAEN desktop digitizer DT5720. The activity for short half-life radionuclides used in nuclear medicine can be then computed. Two primary activity measurement TDCR and $4\pi\beta(LS)-\gamma$ coincidence methods were then used to determine the activity of ^{18}F at ENEA-INMRI. The TDCR parameter is measured for the ^{18}F standard solution using both CAEN and ENEA-INMRI data analysis software.

1 Introduction The $4\pi\beta-\gamma$ coincidence counting technique has been a powerful and widely used method for radionuclide standardisation and to measure the absolute activity concentration of radionuclides [1, 2, 3]. Radionuclide activity can be measured using a direct activity measurement method, which doesn't require any quenching-indicating parameters or reference standards to determine detection efficiency. [4]. Among the currently available methods of radionuclides activity measurement, the coincidence counting technique is one of the most widely utilised in any of its variants [5, 2, 3]. The direct measurement of activity concentration can be performed for radionuclides decaying via pure β, electron capture (EC), $\beta-\gamma$, $EC-\gamma$, and $X-\gamma$ without making any assumptions about the values of the detector efficiencies, branching ratios, or decay scheme parameters [6]. The principles and theory of the $4\pi\beta-\gamma$ coincidence counting method are well-established and thoroughly understood. Nevertheless, it's difficult to apply these principles to actual measurements. Attempts to improve the accuracy and the precision of $4\pi\beta-\gamma$ coincidence counting has been ongoing for decades. Grigorescu studied the accurate fulfilment of the $\gamma-$ray channel linearity conditions [7]. Campion authored the first and most renowned paper on the $4\pi\beta-\gamma$ coincidence counting [2]. Campion showed that a $4\pi\beta-\gamma$ coincidence system is principally made up of two detectors; beta and gamma detectors and an electronic unit to generate a coincidence pulse by detecting synchronised gamma and beta pulses which come from the same nuclear disintegration.

He demonstrated that precise measurements of $\beta-\gamma$ emitter radionuclides can be accomplished by employing a $\beta-$detector with 4π geometry for the coincidence method. In this configuration, many of the necessary corrections are minimal, ensuring accurate results. It is expected that the $\gamma-$detector responds only to $\gamma-$rays, the $\beta-$detector responds only to $\beta-$particles, and the coincidence channel records only the simultaneous pulses come from the two detectors (true coincidences) [2]. This technique has been the focus of many papers and comprehensive reports [8, 6] since the publications of Dunworth [1] and Campion [2]. The fundamental principle of coincidence counting can be illustrated through the prototypical case of the activity measurement of a radionuclide with a simple decay-scheme consisting of $\beta-$emission followed directly by a $\gamma-$ray from the de-excitation of the daughter radionuclide or annihilation process. The basis of the coincidence technique comes from the additional coincidence channel. This additional channel records disintegration pulses that emanate from both $\beta-$ and $\gamma-$counters. The standardisation of a wide range of radionuclides can be achieved using the original $4\pi\beta-\gamma$ coincidence counting and its extended techniques [9]. Most laboratories have used a proportional counter (PC) in the $\beta-$channel for a long time to standardise radionuclides using the $4\pi\beta(PC)-\gamma$ coincidence method such as Laboratoire National Henri Becquerel (LNHB) and Physikalisch-Technische Bundesanstalt: National Metrology Institute (PTB) [10, 11]. Afterwards, $4\pi\beta-\gamma$ coincidence system has been updated by replacing the proportional counter with the liquid scintillation (LS) counter as a beta counter. This new coincidence counting system has been developed and improved in metrology laboratories. $4\pi\beta(LS)-\gamma$ coincidence counting systems had already been implemented [12, 13, 14], but the goal was to have a complete TDCR (Triple to Double Coincidence System) counter as the $\beta-$channel [15, 16]. The

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TDCR system has been specially developed for the purpose of standardising pure beta and pure electron capture emitters, and it can be extended to standardise simple $\beta-\gamma$ and $EC-\gamma$ emitter radionuclides.

2 Experimental setup of ENEA $4\pi\beta(LS)-\gamma$ coincidence counting system

At the Italian National Institute of Ionizing Radiation Metrology (INMRI), belonging to ENEA and located in Casaccia Research Center, the TDCR method is used for absolute activity measurements and standardisation of pure electron capture (EC) or pure beta emitting radionuclides [17, 18]. A portable TDCR detector consists of three photomultipliers (PMTs) operating in coincidence. These PMTs are symmetrically positioned around a specially designed optical chamber [19]. It allows for the measurement of radioactive material in solution of liquid scintillator, contained in a vial which is put inside the optical chamber [17].
ENEA-INMRI in collaboration with Catania University, Istituto Nazionale di Fisica Nucleare (INFN)- Sezione di Catania section and Costruzioni Apparecchiature Elettroniche Nucleari (CAEN) developed a new in-situ coincidence system for activity measurement for short half-life radionuclides used in nuclear medicine which is called $4\pi\beta(LS)-\gamma$ coincidence detection system. The portable TDCR counter [19] at ENEA-INMRI is extended to $4\pi\beta(LS)-\gamma$ coincidence system by implementing a cylindrical-type Scionix Standard 51B51/2M NaI(Tl) scintillation detector as a gamma counter. The NaI(Tl) scintillation detector consists of a scintillation material integrally coupled to a photomultiplier tube. The NaI(Tl) scintillation detector with the (3”×3”) crystal dimension includes a 14 pins photomultiplier (0.4 mm) thick aluminium housing inserted into the bottom of the TDCR counter as shown in Fig.1.

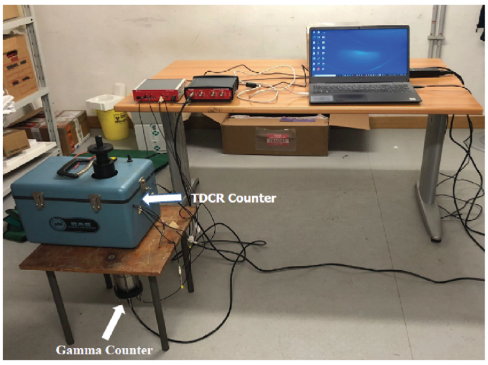

Fig. 1. Experimental setup of ENEA-INMRI portable $4\pi\beta$ (LS) $-\gamma$ coincidence system.

The new ENEA-INMRI portable $4\pi\beta(LS)-\gamma$ coincidence system can be used in environmental applications and in nuclear medicine laboratories of the hospitals to measure the activity of a radiopharmaceutical injected into patients, such as ^{18}F, ^{11}C or ^{131}I. The schematic configuration of the new ENEA-INMRI portable $4\pi\beta(LS)-\gamma$ coincidence counting system is shown in Fig. 2.

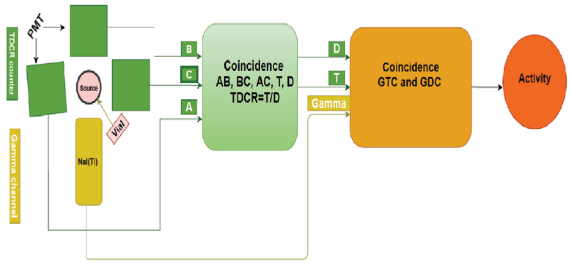

Fig. 2. The schematic configuration of $4\pi\beta(LS)-\gamma$ coincidence system available at ENEA-INMRI.

3 $4\pi\beta(LS)-\gamma$ data analysis software

The $4\pi\beta(LS)-\gamma$ coincidence system is directly linked to the CAEN DT5720 digitizer. The CAEN DPP-CI Control Software remotely controls the digitizer [20]. The acquisition parameters such as gate length, DC offset, pulse polarity, threshold level, etc. can be set independently for each channel to configure the hardware, and perform the data readout. In order to perform a $4\pi\beta(LS)-\gamma$ analysis on the collected event lists coming from the TDCR detector and the gamma detector, a customised TDCR DAQ software has been updated and modified. The software is written in C language and designed for off-line implementation of $4\pi\beta-\gamma$ coincidence counting method. Since all the acquisition channels of a digitizer are synchronised, it is possible to correlate events from different input channels comparing their Trigger Time Tag (TTT)s. Once the run is over, the CAEN TDCR DAQ software scans the resulting event lists looking for coincidences between the events come from the $\beta-$channels in the TDCR counter. Afterwards, the two output files are created for both Triple N_T and Double N_D coincidences data inside the CAEN TDCR code and they are saved as a ASCII file or binary file. These two output files are used as input files for the CAEN TDCRGamma code. Next, the $4\pi\beta(LS)-\gamma$ (CAEN TDCRGamma) DAQ software scans the event lists generated, searching for coincidences between the gamma channel and beta channel. It specifically looks for coincidences between gamma event comes form gamma counter and the triple coincidences and the logical sum of double coincidences output files within the TDCR counter, as illustrated in Fig. 3. This data subset from TDCR counter is used together with the set of data coming from the $\gamma-$channel. The set of events in the $\beta-$channels which are time correlated with the gamma event are taken into account. One can select the $\gamma-$energy window close to the full-energy peak of the $\gamma-$ray for the $\gamma-$channel. Furthermore, the code will look for the event coming from the beta counter and gamma counter by looking to the minimum TTT of each event in double or triple coincidences data output files in $\beta-$counter and $\gamma-$counter to create a new coincidence window. In other word, let's assume τ_γ and τ_β are the minimum TTT of gamma and beta events respectively, and ω is the coincidence window. Moreover, if $\tau_\gamma < (\tau_\beta + \omega)$, this means that there is a coincidence between $\gamma-$channel and $\beta-$channel, otherwise there is no coincidence between the events coming from the both detectors.

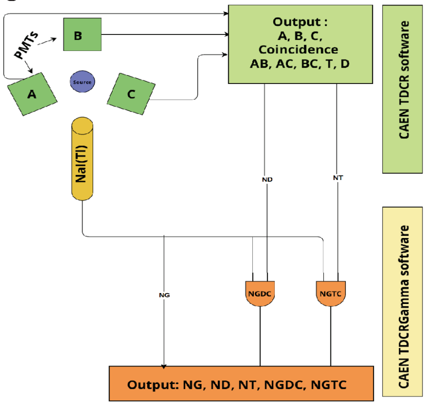

Fig. 3. $4\pi\beta\ (LS)\ -\ \gamma$ coincidence system configuration.

Following this, the TDCRGamma software includes the minimum TTT event as a dataset within the beta coincidence window in the triple and double coincidence data files. Thus, it starts looking for coincidences with events in the gamma counter and extending the coincidence window until the event is found in the gamma channel. Finally, TDCRGamma software will compare the TTT for all the acquisition from the three input channels (γ −channel and the two output files from β −channel). Subsequently, the TTT for each selected event in coincidence in the TDCR counter and gamma counter will be saved. This intimates that the coincidence window for the new $4\pi\beta(LS) - \gamma$ coincidence system starts from the minimum time of measuring the first event in files in the β −channel until the event in the γ −channel is measured. The new time coincidence window for $4\pi\beta(LS)\ - \gamma$ coincidence system is longer than the time coincidence window of the TDCR system. The dead-time of the TDCR counter is longer than the selected dead time for the gamma channel, in order to eliminate the after-pulses in the liquid scintillation counter. The recorded data stream is then analysed by the CAEN TDCRGamma analysis software. Moreover, the DAQ software implements the same algorithms of the well-known French MAC3 analogic module emulating its operations in terms of coincidence definition and extendable-type dead time management [21]. In particular, the CAEN TDCRGamma software ignores all those events found in the lists whose TTTs lie in a dead time window; the dead time is then extended according to the MAC3 logic [20]. As a result of this analysis, the new CAEN TDCRGamma DAQ software can provide the counting from the three channels: The single count rate for each channel, gamma count rate N_G, from γ −channel. The triple coincidence N_T and the logical sum of the double coincidence N_D, from β −channels. Moreover, the coincidences between gamma channel with double coincidence in beta channel N_{GDC}, and the coincidences between gamma channel with triple coincidence in beta channel N_{TDC} will be measured. In addition, real-, dead-, and live-times can be evaluated as shown in Fig. 3. The activity of the radionuclides can be measured, at the "zero order approximation", by using the conventional formula of the $4\pi\beta(LS) - \gamma$:

$$A = \frac{N_G \cdot N_D}{N_{GDC}} = \frac{N_G \cdot N_T}{N_{TDC}} \qquad (1)$$

Independently from CAEN, another TDCR analysis software was developed at ENEA-INMRI by implementing the MAC3 philosophy in the CERN ROOT framework [20], an object-oriented package for physics analysis. The algorithm for the TDCR coincidence analysis was then developed by following the main idea described by Bouchard and Cassette [20] and by taking into account the powerful CERN ROOT resources for analysing data coming from complex detectors. Both codes can run either on Windows or Linux machines. The results obtained by the two software have been compared by measuring a standard solution of ^{18}F with TDCR parameters by using CAEN software and ENEA software. In Table 1, one can observe the typical TDCR value acquired from these measurements along with the corresponding efficiencies for the logical sum of double coincidences and triple coincidences, as analysed using both CAEN and ENEA software. A coincidence window of t_c = 140 ns and a dead-time 50 μs were applied on the set of recorded data to perform TDCR analysis for ^{18}F. The deviation in percent between the TDCR parameter TDCRCAEN computed by CAEN code and TDCRENEA computed by ENEA code for ^{18}F sources are also reported in Table 1.

Table 1. TDCR values for radionuclide ^{18}F by using CAEN software and ENEA software code.

Radionuclide	Software	N_D	N_T	TDCR
^{18}F	CAEN	2693230	2673821	0.9928±0.03
	ENEA	2693096	2673695	0.9928±0.03
	Δ%	0.7126	0.005	0.00

At the same time, the activity of ^{18}F is calculated using conventional Eq. 1 without any knowledge of the detection efficiency of the detectors. The γ −energy window close to the full-energy peak about 511 keV of ^{18}F for the gamma channel is selected. A coincidence window of t_c = 2000 ns and a dead-time 10 μs were applied on the set of recorded data to perform the $4\pi\beta$ (LS) − γ analysis for ^{18}F. Table 2 shows that the ^{18}F activity obtained by both CAEN TDCR and $4\pi\beta(LS) - \gamma$ DQA methods, and the minimum deviation less than 0.1% between the two methods is measured. In the TDCR method one can determine the activity of ^{18}F source by using the following equation:

$$A = \frac{R_c D}{\varepsilon_D} = \frac{R_c T}{\varepsilon_T} \qquad (2)$$

Where $R_c D$ and $R_c T$ are logical sum of double and triple corrected count rates for ^{18}F respectively. Corrected count rate can be calculated by multiply the net count rate of logical sum of double count $R_n D$ and triple count $R_n T$ by the correct Factor. Another important parameters are the logical sum of double efficiency ε_D and triple efficiency ε_T which can be computed theoretically by using Monte Carlo Simulation GEANT4 code [22]. The measurement conducted with TDCR achieves a level of uncertainty at 0.5%, considering all aspects of uncertainty. This includes not only statistical uncertainty but also uncertainties associated with factors such as the decay model, background measurements, coincidence resolving time, dead-time, and decay during measurement, as well as decay at the reference date. The good agreements between the two methods confirms that CAEN $4\pi\beta(LS)-\gamma$ (TDCRGamma) data analysis software developed for in-situ $4\pi\beta(LS)-\gamma$ coincidence system at ENEA operates correctly.

Table 2. Activity measurements of radionuclides ^{18}F using TDCR and $4\pi\beta(LS)-\gamma$ methods.

Radionuclide	Method	Activity (Bq)
^{18}F	TDCR	41379.69 ± 0.15
	$4\pi\beta(LS)-\gamma$	41777.06 ± 0.13
	Δ%	0.1

4 Conclusion

The $4\pi\beta(LS)-\gamma$ coincidence counting technique is the most powerful technique for the absolute activity measurements of $\beta-\gamma$ or $EC-\gamma$ emitting nuclides. The $4\pi\beta(LS)-\gamma$ coincidence method allows for the direct standardisation of short-lived pure β, α, EC, $\beta-\gamma$, and $EC-\gamma$ radionuclides directly on-site. Furthermore, it can be used to calibrate the devices used in the Nuclear Medicine Departments, such as activimeters, PET, SPECT, Gamma camera, etc, without moving the radioactive sources between the Hospitals and the Metrology Institutes. The activity of ^{18}F is determined using two different methods, such as TDCR and $4\pi\beta\,(LS)-\gamma$ coincidence methods. The standardisation in situ of short half-lived radionuclides is advisable to avoid the transport of radioactive sources between the hospital and metrology institutes.

ENEA-INMRI and CAEN developed TDCR software independently, to perform the TDCR analysis by implementing the MAC3 philosophy. Then, the TDCR parameters are measured for a ^{18}F standard solution by using CAEN and ENEA software. Both codes are compatible and work properly. The activity measurement of the radionuclides is independent on the detection efficiency for either detectors using the $4\pi\beta\,(LS)-\gamma$ method. On the other hand, in TDCR method the detection efficiency has a crucial role in the activity measurement of the radionuclides. For that reason, one can compute the detection efficiency of the detectors by using the Monte Carlo Simulation; in this project GEANT4 Code is used. Another important feature of DAQ TDCRGamma software is that the new software is able to run the $4\pi\beta\,(LS)-\gamma$ detection analysis both online or off real-time measurements and previously acquired data. This makes it possible to perform a parameter sweep analysis repeating the $4\pi\beta\,(LS)-\gamma$ detection technique on the same data set changing automatically for example the coincidence resolving time and/or dead time. It is possible, therefore, to study the behaviour of the results obtained with different parameter values applying them on the same set of data, without repeating the measurements several times and just by changing the parameter values in the software configuration, with no hardware operations.

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References

1. J. V. Dunworth, Rev. Sci. Instr. **11**, 167 (1940).
2. P. J. Campion, Int J. Appl. Radiat. Isotopes **4**, 232 (1959).
3. A. P. Baerg, Metrologia **2**, 23 (1966).
4. C. Bobin, J. Bouchard, S. Pierre, and et al., Appl. Radiat. Isot. **70**, 2012 (2012).
5. J. V. Dunworth, Rev. Sci. Instr. **11**, 167 (1940).
6. Particle Counting in Radioactivity Measurements: International Commission on Radiation Units and Measurements (ICRU), ISBN 0-913394-51-3 **Report 52**, (1994).
7. L. Grigorescu, Int. J. Appl. Radiat. Isot **34:8**, p1151 (1982).
8. R. Resnick, D. Halliday, and K. S. Krane, A handbook of radioactivity measurements procedures (National Council on Radiation Protection and Measurements (PUBLISHER, Washington, 1985), No. 58.
9. P. J. Campion, J. G. V. Taylor, and J. S. Merritt, Int. J. Appl. Radiat. Isot. **8**, 8 (1960).
10. Ch. Bobin and J. Bouchard, Appl. Radiat. Isot. **64**, 124 (2006).
11. E. Sch¨ofeld, H. Janssen, R. Klein, JC. Hardy, V. Iacob, M. Sanchezvega, HC. Griffin, and Ludington MA., Appl. Radiat. Isot. **56**, 215 (2002).
12. B. R. S. Simpson and B. R. Meyer, Nucl. Instr. and Meth. A **14**, 339 (1994).
13. P. Cassette, M. M. Be, F. Jaubert, and M. C. Lepy, Appl Radiat. Isot. **60**, 439 (2004).
14. A. Chylinski and T. Radoszewski, Nucl. Instrum. Meth. A **369**, 336 (1996).
15. K. Pochwalski, R. Broda, and T.Radoszewski, Appl. Radiat. Isot. **39**, 165–172 (1988).
16. J. Bouchard and P. Cassette, Nucl. Instrum. Meth **A 505**, 72–75 (2003).
17. R. Vatin, Mesure absolue de l'activite des radionucleides emetteurs beta purs par scintillateur liquide a l'aide d'un detecteur a 3 photomultiplicateurs. In: Magana, J.F. (Ed.), Mesure de l'activite des radionucleides emetteurs beta purs (Bulletin BNM, Paris, 1991), No. 85 BNM, p. 13–21.
18. Ziemek, T., Broda, R., Listkowska, A. et al. Standardization of an 55Fe solution using the TDCR method in POLATOM as part of the CCRI (II)-K2.Fe-55.2019 key comparison. J Radioanal Nucl Chem **331**, 3241–3248 (2022).
19. Capogni M, De Felice P. A prototype of a portable TDCR system at ENEA. *Appl Radiat Isot*. **93**:45-51(2014)
20. Giuliano Mini, Francesco Pepe, Carlo Tintori, and Marco Capogni, Applied Radiation and Isotopes **87**, 166–170 (2014).
21. J. Bouchard and P. Cassette, Applied Radiation and Isotopes **52**, 669 (2000).

22. C. Thiam, C. Bobin, B. Chauvenet, and J. Bouchard, Applied Radiation and Isotopes **70**, 2195 (2012).