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Effect of Birks Constant and Defocusing Parameter on Triple-to-Double Coincidence Ratio Parameter in Monte Carlo Simulation-GEANT4

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Abstract**—**This project concerns with the detection efficiency of the portable Triple-to-Double Coincidence Ratio (TDCR) at the National Institute of Metrology of Ionizing Radiation (INMRI-ENEA) which allows direct activity measurement and radionuclide standardization for pure-beta emitter or pure electron capture radionuclides. The dependency of the simulated detection efficiency of the TDCR, by using Monte Carlo simulation Geant4 code, on the Birks factor (kB) and defocusing parameter has been examined especially for low energy beta-emitter radionuclides such as ^{3}H and ^{14}C, for which this dependency is relevant. The results achieved in this analysis can be used for selecting the best kB factor and the defocusing parameter for computing theoretical TDCR parameter value. The theoretical results were compared with the available ones, measured by the ENEA TDCR portable detector, for some pure-beta emitter radionuclides. This analysis allowed to improve the knowledge of the characteristics of the ENEA TDCR detector that can be used as a traveling instrument for in-situ measurements with particular benefits in many applications in the field of nuclear medicine and in the nuclear energy industry.

Keywords**—**Birks constant, defocusing parameter, GEANT4 code, TDCR parameter.

I. INTRODUCTION

THE applications and developments of nuclear medicine (NM) have been an interest of nuclear science for almost half a century. In recent years, a wide variety of radionuclides have been used in NM with different and complex decay schemes. Therefore, the accuracy for activity measurements significantly requires standardization of radioactivity [1]. Precise and accurate radioactivity measurements are considerably difficult due to the complexity in the kind of radionuclides used and their decay characteristics (decay rates, type of particles emitted and their energies and intensities). In addition, one cannot use the same detector or method for measuring all radionuclides. For that reason, standardization measurement of a particular radionuclide implies a specific study of its decay scheme and the measurement technique for their interaction with matter [2]. 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 standardization of pure electron capture (ec) or pure beta emitting radionuclides [3]. A portable TDCR detector, made up of three photomultipliers (PMT) working in coincidence and symmetrically arranged around a specially designed optical chamber, is available at ENEA-INMRI. It allows for the measurement of radioactive material in solution of liquid scintillator, contained in a vial which is put inside the optical chamber. It is used for standardization of pure beta emitters and electron capture radionuclides [4], [5].

The portable TDCR counter was built in 2013 at ENEA-INMRI [6] with identical performance as the TDCR counter operated for activity primary measurements in metrology [3]. This counter can be used as an on-site device for activity standardization in Primary Metrology Institutes (PMI) or it can be used in environmental applications or in NM laboratories of the hospitals to measure the activity of a radiopharmaceutical injected into patients, such as ^{18}F or ^{11}C. The portable TDCR is remarkably reducing the effort required for metrological assurance in the liquid scintillation (LS) counter; for that reason, CAEN in collaboration with ENEA-INMRI developed a front-end electronic acquisition system for the portable TDCR counter [6]. Following the theory of TDCR technique, the free parameter -which represents the number of emitted photons per unit energy released in the scintillation solution- and the PMT quantum efficiency can be adjusted to achieve the TDCR detection efficiency. The nonlinearity of the light emission is taken into account in the model through the Birks law [7] as:

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

where $m(E)$ is the mean number of photons produced by the energy E, A is a free parameter characterizing the LS-cocktail efficiency, kB is a Birks factor and dE/dx is the linear energy transfer. The non-linearity is a result of the ionization quenching, that describes the linear energy transfer dE/dx of the ionizing radiation in the scintillator solution by using the Birks' factor, kB [7]. The kB factor has a powerful effect on the activity measurement especially for low-energy beta emitters [8]. The detection efficiency variation technique can be used generally to select the value of the kB factor. The activity of the radionuclides can be measured using the triple and double coincidence between PMTs tubes based on the triple and double

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coincidence detection efficiency of the TDCR. To find the detection efficiencies of double and triple coincidences, a statistical model of the light emission and the experimental value of TDCR are needed. Indeed, a stochastic approach is explored as an alternative to this classical model, by implementing a complete representation and modeling of the TDCR counter drawn on the Monte Carlo simulation Geant4 code [9], [10]. Geant4 code has the ability to simulate different physical processes associated with the charged particle or photons transportation inside matter and generate optical photons resulting from the interaction between charged particles with the scintillator in the liquid scintillation counter (LSC). This approach allows to obtain TDCR model, taking into account the different reflection and refraction processes which take place at the interfaces inside the optical chamber such as: vial to air, air to PMT-window, etc. Practically, there are different techniques to achieve detection efficiency variation such as: use of coaxial grey filters, chemical quenching, and PMT defocusing [8]; while the simulated TDCR parameter depends on the kB factor and defocusing parameter especially for low energy beta emitter radionuclides (^{3}H). For this purpose, it is important to know how the theoretical detection efficiency of TDCR varies with the kB factor using Geant4 code for Monte Carlo simulation (MCS). In this paper the simulated detection efficiency variation of TDCR counter has been examined for low-energy (^{3}H and ^{14}C) and high-energy (^{90}Y) pure-beta emitting radionuclides by using two different methods. The detection efficiency of TDCR counting system for different pure-beta emitters such as (^{3}H, ^{14}C, ^{90}Sr, ^{18}F and ^{90}Y), experimentally measured, is compared with the simulated TDCR parameter by using GEANT4.

II. BRIEF DESCRIPTION OF THE PORTABLE TDCR COUNTER

TDCR method is considered a powerful method for absolute activity measurement for pure-beta and pure electron-capture radionuclides in LSC. In this project the portable TDCR at ENEA-INMRI is used [11]. It consists of an optical chamber counter which has an inner prismatic shape with equilateral base (H = 73 mm and L = 60 mm). The optical chamber is made from white PTFE (Teflon) and it is surrounded by a black Teflon outer cylindrical box shape of ($\emptyset = 150\ mm\ and\ H = 150\ mm$). The three Hamamatsu R7600U-200 square package type PMTs arranged symmetrically 120° around the LS vial in the optical chamber [6]. The selected PMTs have a high quantum efficiency, high gain, short time of response, comparatively wide-range wavelength of about 300–650 nm, with relatively low supply voltage (about 900 V) with the grounded cathode [11]. The PMTs operated in photon counting mode [11]. The high-power electronics device is necessary to record the fast signals for the TDCR applications. For such a reason, the DT5720 CAEN digitizer is connected directly without any preamplifier to the output signals of the PMTs. The DT5720 CAEN digitizer provides a fast and precise 12 bit and sampling rate Analog Digital convertor (ADC) 250 MSample/s within the time acquisition window. The CAEN digitizer is PC remote-controlled by the CAEN Control Software which runs on Windows or Linux operating systems; the data acquisition parameter (gate length, DC offset, pulse polarity, threshold level, etc.) for each channel can be set independently by the software [6].

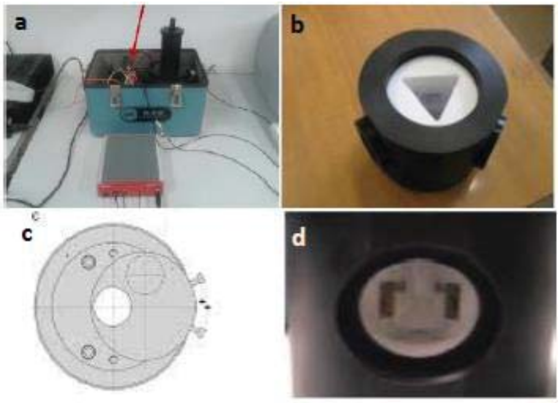

Fig. 1 (a) The portable TDCR system at ENEA. (b), (d) The external and the internal view (up and down) respectively of the optical chamber (c) Optical shutter design

III. THE TDCR SYSTEM SETTING AND ANALYSIS SOFTWARE

The precise adjustment of the discrimination above the noise for each one of the three PMTs and threshold under the single-electron peak (SEP) are important settings of the TDCR method [12]. The SEP signal is generated by each PMT in the portable TDCR counter. The setting of the data acquisition parameters is established and recorded by the CAEN digitizer [11]. For each individual channel, the control software of the digitizer is used to fix the energy discrimination threshold and the width of the ADC gate (typically 24 ns). This setting allows to minimize the noise and process the entire analog pulse which arrives from the PMT tubes, and which is covered fully in time by the gate pulse [11]. In order to have the same gain of the three PMTs in different channels, one can check the SEP for each PMT in order to be located at the same histogram channel. To achieve this goal, the DT5720 digitizer records a typical SEP spectrum from each PMT of the TDCR counter without a vial inside the optical chamber. The CAEN digitizer was used in histogram mode [11]. The CAEN digitizers DT5720 are dead-timeless acquisition devices; the software is written and customized in C++ [6]. The dead-time is managed by the digitizer which is provided by CAEN [6]. In the portable TDCR system, signals come from each PMT, and the digitizer manages the off-line coincidence analysis between them by emulating the MAC3 logic [13]. For the off-line analysis of the portable TDCR system, a software was implemented in CERN ROOT frame [13]. When the CAEN digitizer records and organizes data, they are read event by event by the ENEA-INMRI software as “leaves" of a tree in the ROOT output file. In fact, by using the DT5720 digitizer it is possible to read the Trigger Time Tag (TTT) and integrated Charge (Q) values for each event. In addition, for defined dead-time t_{dead} (μs) and coincidence resolving time t_c (ns) values, one can compute the off-line TCDR analysis for the single counts of each PMT (A, B, and

C), the coincidence counts (AB, BC, AC, D and T), the sum of them (S), the live time (t_{live}), the real time (t_{real}) and the TDCR parameter [11].

IV. Experimental Measurements with TDCR

TABLE I
EXPERIMENTAL TDCR PARAMETERS OF DIFFERENT RADIONUCLIDE SOURCES

Radionuclides	E_β(keV)	Mean E_β(keV)	TDCR parameter
^{3}H	18.591	5.68	0.3106
^{14}C	156.48	49.16	0.8903
^{90}Sr	545.9	196	0.9757
^{18}F	633.9	249.5	0.9917
^{90}Y	2278.7	926.7	0.9910

The TDCR parameter can be measured by using the ENEA portable TDCR detector linked with DT5720 digitizer for low-energy pure-beta (^{3}H and ^{14}C) and for high-energy pure-beta (^{90}Sr, ^{18}F and ^{90}Y) emitting sources. The TDCR parameters for those beta emitting radionuclides measured by TDCR technique are interesting for radionuclide standardization and activity measurement in NM applications and for other kinds of applications, such measurements on nuclear site or in environment. The TDCR analysis was performed by recording the Q value with an ADC gate equal to 24 ns. The sources were prepared at the Radiochemistry laboratory at the ENEA-INMRI for other past experiments; the master solutions were checked by a high energy resolution HPGe detector – owned by ENEA-INMRI - to perform a preliminary gamma-impurity check. The sources were made, as usual, of 10 ml Ultima Gold (UG) as liquid scintillator and approximately 10 mg of radioactive solution (with different aliquots of CCl_4 as a quenching agent) in 20 ml borosilicate glass vials. At the first, for background measurements one blank source was prepared containing only 10 ml of UG in the previously mentioned geometry. A customized data acquisition software was implemented in order to perform a TDCR analysis on the events which are collected in a list-mode file; corresponding events from different input channels can then be compared to their TTTs when each of the acquisition channels of a digitizer are synchronized. After the run ends up, the data acquisition software scans the consequent event lists checking for coincidences. The recorded data stream is then analyzed by the ENEA-INMRI analysis software and values of dead time $(t_{dead} = 50\,\mu s)$ and coincidence resolving time $t_c = 140\,ns$ are applied. The ratio of $R_T/R_{AC}, R_T/R_{BC}\ and\ R_T/R_{AB}$ are calculated for each set of data from the net count rates of the double coincidences $(R_{AC} = AC/t_{live}, R_{AB} = AB/t_{live}, and\ R_{BC} = BC/t_{live})$ and the net count rates of the triple coincidences R_T from the three pairs of PMTs. They used to compute the efficiencies for the triple coincidences ε_T and for the logical sum of the double coincidences ε_D. TDCR parameter can be calculated experimentally by taking into account the ratio of the net count rate (background subtracted) of triple coincidence to the logical sum of double coincidence and it is equal to the ratio of the efficiency of triple coincidence ε_T to the efficiencies for the logical sum of the double coincidences ε_D for high statistics conditions of light emissions (i.e., for high-energy beta emitters) [14]. Different TDCR parameters, obtained for different radionuclides - measured in the portable TDCR counter - with a mean energy E_β of the beta spectrum known with the uncertainty about 1%, are shown in Table I.

V. The TDCR-GEANT4 Modeling Based on LS

The MCS technique has overcome the shortcoming of the numerical method used for simple geometries based on the radiation transport equations [15]. Although MCS has been used for a few decades in many areas of experimental physics such as elementary particles physics and high energy physics, it has only gained interest in radiation physics in recent decades due to the great advance of computing power. MCS in radiation physics simulates the transport of electrons, positrons, and photons in materials with arbitrary compositions [16]. In this project, Monte Carlo Geant4.9.6 p04 code was selected for its complete set of tools permitting accomplish modeling of the TDCR counter, simulating the different physical processes ongoing in the optical chamber and determining the TDCR parameters for different radionuclides sources in LSC. In LS counting, Monte Carlo can be used to calculate the probability for the interaction of beta or electron capture particles in a given scintillator and the absorbed energy distribution [17]. The photon absorbed energy distribution is mostly deliberated in discrete energy bins and the quenching function is applied to each bin midpoint [15]. The "low energy" package on the basis of Livermore data was selected for simulating photons and electrons which are created by Bremsstrahlung [13]. In order to achieve a realistic simulation, an explanation of the optical properties related to the materials of the optical chamber is carried out comprehensively: the ultra bialkali photocathodes, the borosilicate vial glass loaded with liquid scintillator, the fused silica window of the PMTs, and the reflecting cavity of Teflon is about 95% [18]. Regarding the TDCR detection efficiency calculation, the extension to LS counting needs the exact description of the borosilicate vial loaded with 10 ml of Ultima Gold LS cocktail. The Ultima Gold atomic composition and the dimensions of the vial are taken from the experimental set up. The borosilicate glass vial is used and the dispersive refractive index for the borosilicate material is 1.52 at wavelength of 400 nm [18]. The borosilicate vial is placed inside the optical chamber, the model of dielectric – dielectric applied for photon reflection and refraction at the inner and outer scintillation vial (borosilicate) vial surfaces [13]. In order to calculate the production of photoelectrons each time when the photons reflect at the interface with the ultra bialkali photocathode, one can consider the various optical processes happening within the fused silica of the PMT window. The center of the optical chamber is positioned 17 mm from the outer surface of the PMT window; for that reason, the dielectric–dielectric boundary model is used. The range of PMT spectral sensitivity from 300 nm to 650 nm has a peak at 400 nm. The fused silica window is covered by the ultra bialkali photocathode and the model of dielectric–dielectric boundary is used [18]. Quantum efficiency is provided by using binomial trials for converted photons to photoelectrons in the photocathode [18]. The maximum quantum efficiency is ~ 43% at 430 nm [19]. The double- and triple- coincidences are determined for each PMT tube by counting the numbers of photoelectrons. The probability for photoelectrons to reach the first dynode depends on the defocusing parameter; it permits the detection-efficiency variation to be simulated as experimentally found by PMTs defocusing [18]. The relationship between triple- and double-coincidence detection efficiencies and TDCR value can be determined by applying the defocusing parameter, and the activity of the source can be calculated.

In this paper, we carried out two different computations to obtain the TDCR parameter of ^{3}H, ^{14}C and ^{90}Y. We used CEA and G4RadioctiveDecay class in Geant4 energy spectrum data for pure beta emitting radionuclides above in order to compute the theoretical Monte Carlo simulated TDCR detection efficiency. The radioactive decay simulation is a standard task in MCS Geant4 code. The simulation of the overall process uses some physical models with unknown theoretical parameters; for this reason, the Monte Carlo code is necessary to fix these parameters by comparing the simulated results against the experimental ones.

A. Measurement of Tritium (3H)

^{3}H emits 100% β^{-} particles with a maximum energy of 18,591 (1) keV and half-life of $T_{1/2}$ = 12,312 (25) years [20]. The simulated TDCR parameter value for the ^{3}H radionuclide changes in the 0.2 to 0.4 range. The sets of data are corresponding to nine different kB factor values as a function of defocusing parameters used in calculation, namely 0.07, 0.08, 0.09, 0.1, 0.11, 0.12, 0.13, and 0.14 mm/MeV as shown in Fig. 3. From frequent processing of data for different kB, it was established that the best kB value is around kB = 0.09 mm/MeV and the defocusing parameter = 0.78 as shown in Fig. 2. The percentage of deviation (Δ%) of the TDCR parameter, between the experimental $TDCR_{exp.}$ and the simulated one $TDCR_{sim}$, written as $\Delta\% \, TDCR = \frac{|(TDCR_{sim} - TDCR_{exp.})|}{TDCR_{exp.}}$ at that point it is 3.06%. In fact, this is true only for low energy beta emitter radionuclides especially for TDCR values around 0.3. The deviation of the TDCR parameter of ^{3}H for a different kB factor as a function of defocusing parameter is plotted in Fig. 3. It must be observed that the kB value has a dramatic influence on the TDCR efficiency of a ^{3}H source.

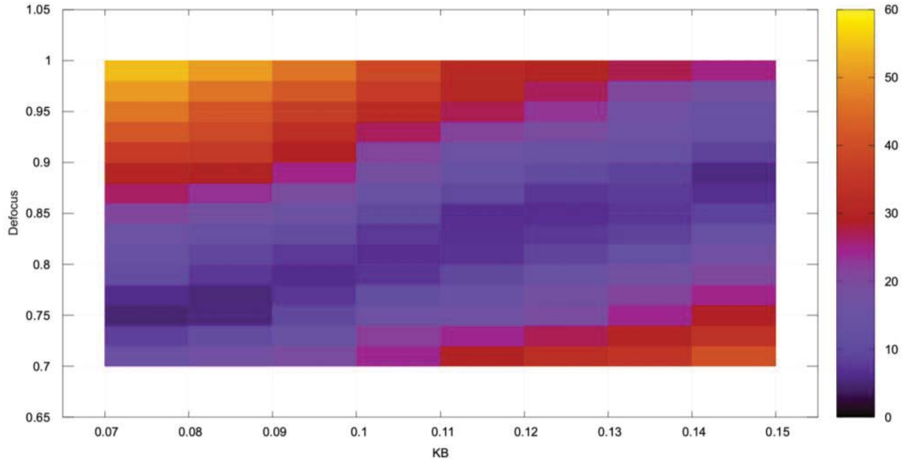

Fig. 2 Relationship between the kB factor and defocusing parameter for ^{3}H

B. Measurements of Carbon-14 (14C)

Carbon-14 (^{14}C) disintegrates 100% by β^{-} transition with a maximum energy of 156.479 (4) keV and half-life of $T_{1/2}$ = 5700 (30) years [21]. The deviation of the TDCR parameter for the ^{14}C radionuclide for different kB factors as a function of defocusing parameter is shown in Fig. 3. The TDCR value is in the range of 0.80 to 0.94. The sets of data are corresponding to nine different kB factor values as a function of defocusing parameters used in calculation, namely 0.07, 0.08, 0.09, 0.1, 0.11, 0.12, 0.13, and 0.14 mm/MeV. The processing of data is repeated with different kB values. It was found that the kB value is a nearly constant for different kB factors and different defocusing parameters. Fig. 3 showed that the deviation of the TDCR parameter for different value of kB factor as a function of defocusing parameter is very low, and it demonstrated that, in the situation of low energy radionuclides, the effect of kB on the TDCR model can be neglected when the TDCR parameter is close to 0.85.

C. Measurements of Yttrium-90 (90Y)

^{90}Y emits β-particles throughout a 99.983(6)% branch equivalent to the maximum energy of 2278.7 (16) keV and the half-life $T_{1/2}$= 2,6684 (13) days [22]. The TDCR detection efficiency for the pure beta emitter radionuclide ^{90}Y is roughly constant in the range 0.998 to 1.0. The sets of data are corresponding to nine different kB factor values as a function of defocusing parameters used in calculation, namely 0.07, 0.08, 0.09, 0.1, 0.11, 0.12, 0.13, and 0.14 mm/MeV. The processing of data is repeated with different kB values. It was found that the kB value is approximately constant for different kB factors and different defocusing parameters. It is clear from the graphs in Fig. 3 that the deviation of the TDCR parameter for different value of kB factor as a function of defocusing parameter is very low, and it is verifying that, in the case of high energy pure-beta radionuclides, the effect of kB on the TDCR model can be neglected.

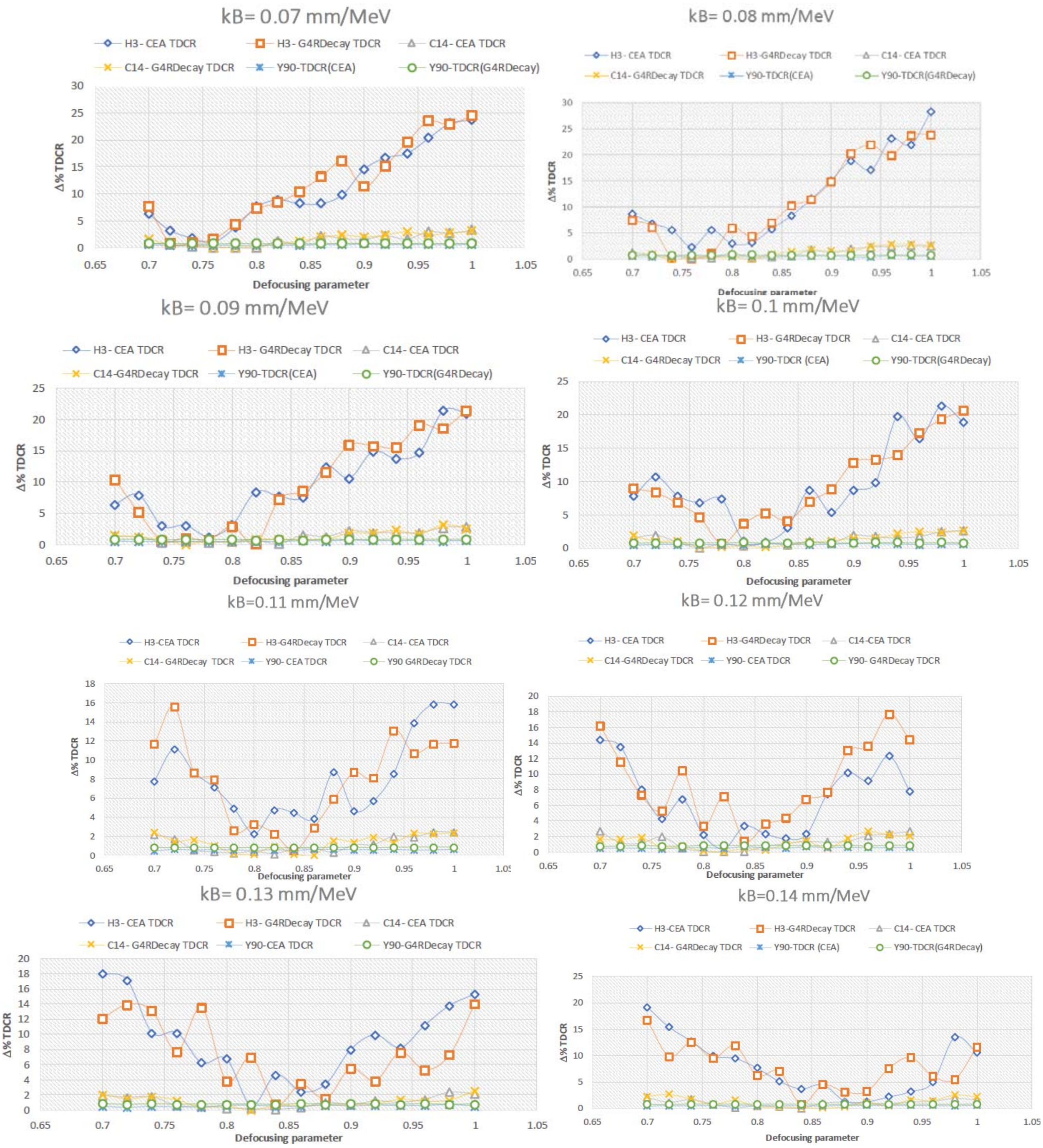

Fig. 3 TDCR deviation for three radionuclides (^3H and ^{14}C as a low energy and ^{90}Y as high energy beta emitter) as a function of defocusing parameters for different kB factors

VI. COMPARISON BETWEEN THE EXPERIMENTAL AND SIMULATED TDCR PARAMETERS

On the basis of the above consideration, we looked at the deviation, Δ%, of the TDCR parameter for each studied radionuclides and for different kB factors and defocusing parameters. So, we were able to report in Table II the selected TDCR parameter, corresponding to the minimum of the Δ%, as a function of the mean beta energy E_β.

TABLE II
SIMULATED TDCR PARAMETERS OF DIFFERENT RADIONUCLIDES SOURCES

Radionuclides	E_β(keV)	Mean E_β(keV)	TDCR parameter
^{3}H	18.6	5.68	0.3145 ± 0.1122
^{63}Ni	66.98	17.434	0.7066 ± 0.1309
^{14}C	157.0	49.16	0.8847 ± 0.1309
^{90}Sr	546.0	196	0.97071 ± 0.0142
^{18}F	634.365	249.5	0.96050 ± 0.0013
^{90}Y	2278.7	926.7	0.9996 ± 0.0141

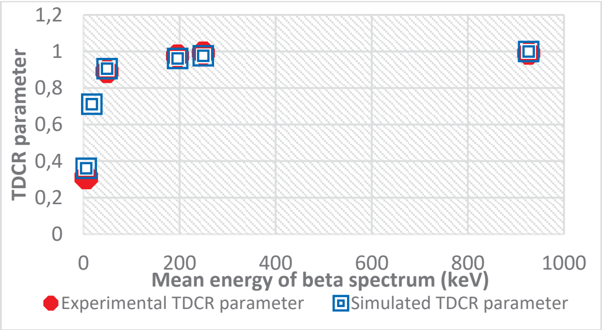

Fig. 4 Experimental and simulated TDCR detection efficiency

By taking into account the results reported both in Tables I and II, we were able to represent in Fig. 4 the experimental ($TDCR_{exp.}$) simulated ($TDCR_{sim}$)TDCR parameters.

Fig. 4 illustrates, in particular, that the TDCR parameter depends on the energy spectrum of beta-emitting radionuclides. The relative uncertainty on a TDCR parameter increases with the decreasing of the beta energy of the radionuclides measured; in fact, for high-energy beta emitters the counting efficiency approaches unity, this means that the uncertainty is very low for these kinds of pure-beta emitters, such as (^{90}Sr and ^{90}Y).

VII. CONCLUSION

In this work, we performed simulations to obtain the TDCR parameter of two low-energy pure-beta (^{3}H and ^{14}C) and one high-energy pure-beta (^{90}Y) emitter. For this purpose, we carried out two different computations by using Geant4 toolkit in which we implemented both CEA energy spectrum and G4RadioactiveDecay class for the three radionuclides above, and in order to find the theoretical TDCR efficiency.

The same code was used to study the relationship between the deviation, Δ%, of TDCR parameters (as previously defined) for ^{3}H, ^{14}C, and ^{90}Y as a function of defocusing parameters for different kB factors. This study highlights that this deviation is very strong for the very low-energy beta radionuclides such as ^{3}H. Furthermore, it can allow to select the best kB factor for different defocusing parameters looking at the minimum of the Δ% deviation.

The comparison between the simulated and experimental TDCR parameters for the studied radionuclides shows that, in many cases, the computed counting efficiencies fit well with the experimental ones, especially for the high-energy ^{90}Y beta emitter, measured by using the ENEA-INMRI portable TDCR detector.

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