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CALCULATION OF EXTRAPOLATION CURVES IN THE $4\pi(\text{LS})\beta\text{-}\gamma$ TECHNIQUE WITH THE MONTE CARLO CODE GEANT4

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Abstract

At LNE-LNHB, a liquid scintillation (LS) detection setup designed for TDCR (Triple to Double Coincidence Ratio) measurements is also used in the β -channel of a $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence system. This LS counter based on 3 photomultipliers was first modeled using the Monte Carlo code Geant4 to enable the simulation of optical photons produced by scintillation and Cerenkov effect. This stochastic modeling was especially designed for the calculation of double and triple coincidences between photomultipliers in TDCR measurements. In the present paper, this TDCR-Geant4 model is extended to $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence counting to enable the simulation of the efficiency-extrapolation technique by the addition of a γ -channel. This simulation tool aims at the prediction of systematic biases in activity determination due to eventual non-linearity of efficiency-extrapolation curves. First results are described in the case of the standardization ^{59}Fe . The variation of the γ -efficiency in the β -channel due to the Cerenkov emission is investigated in the case of the activity

measurements of ^{54}Mn . The problem of the non-linearity between β -efficiencies is featured in the case of the efficiency tracing technique for the activity measurements of ^{14}C using ^{60}Co as a tracer.

1. Introduction

The efficiency-extrapolation technique (Baerg, 1973) is an important measurement procedure associated to the $4\pi\beta\text{-}\gamma$ coincidence method (Campion, 1959) thus making it a powerful primary method in radionuclide metrology. Basically, a detection setup for coincidence counting can be described as two detection β - and γ -channels which are connected through specialized electronics allowing coincidence counting. At LNE-LNHB, the β -channel (where the radioactive source is placed) can be equipped with either a proportional counter (PC) or a liquid scintillation (LS) detector. The respective counting rates obtained in the three channels are usually denoted N_β , N_γ and N_c . The basic classical formula for the activity calculation is $A = N_\beta N_\gamma / N_c$.

Basically, the application of the efficiency-extrapolation technique enables the correction for the excess of counting in the β -channel related to γ -transition events (γ - and x-ray photons, conversion and Auger electrons). It is also implemented to avoid systematic biases on activity determination when the measured efficiency in the β -channel given by N_c/N_γ does not represent the true value expressed by ε_β . In practice, the efficiency-extrapolation technique is carried out by detection-efficiency variation in the β -channel. For each measurement, the calculated activities are generally fitted with a straight-line equation using as a variable an expression depending on N_c/N_γ such as $(1 - N_c/N_\gamma)/(N_c/N_\gamma)$. The true activity is determined by taking the y-intercept corresponding to $N_c/N_\gamma = 1$. Depending on the radionuclide to be standardized and the coincidence detection system used, the conditions for fitting with a straight-line equation cannot be always fulfilled. For instance in the case of complex decay-schemes, an important condition is the invariance of the linear relation between β -inefficiencies (Williams *et al.*, 1968). The general problem of non-linearity which can yield significant biases on activity determination has extensively been addressed using analytical and Monte Carlo calculations (Grigurescu, 1973; Funck and Nylandstedt Larsen, 1983; Chauvenet *et al.*, 1987; Dias *et al.*, 2006).

The present article describes the first simulations of extrapolation curves obtained with the modeling of the $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence system implemented at LNHB using the Monte Carlo code Geant4 (Agostinelli *et al.*, 2003). This setup is characterized by a complete TDCR (Triple to Double Coincidence Ratio) detection system based on 3 photomultiplier

tubes (PMTs) in the β -channel (Broda *et al.*, 2007). The events counted in this channel are double and triple coincidences between PMTs. Ionization quenching and the related non-linearity of light-emission strongly contribute to the detection efficiency. Compared with a proportional counter, the chief drawback is a significant increase of the γ -sensitivity of the β -channel. After-pulses in LS counting due to the scintillation process have also to be taken into account using an appropriate dead-time management based on extendable dead-times (Bouchard, 2000).

The modeling developed for $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements takes advantage of the previous studies concerning the stochastic approach implemented with the TDCR-Geant4 model. The interest of such investigations is a better knowledge of the LS counter properties in order to explain and to prevent potential biases on activity measurements. In particular, the development of the TDCR-Geant4 model reveals the problem of the stochastic dependence of coincidences between PMTs (Bobin *et al.*, 2012) at low energies (lower than 20 keV); it also confirms the existence of coincidence counting due to Cerenkov radiation in the PMT windows resulting from Compton scattering (Thiam *et al.*, 2010). In this article, the comparisons between simulated and experimental extrapolation curves are first presented in the case of the $\beta\text{-}\gamma$ emitter ^{59}Fe . The influence of Cerenkov light contributing to the γ -efficiency in the β -channel and its variation is highlighted with efficiency-extrapolation measurements of ^{54}Mn . Finally, the problem of the non-linearity between β -inefficiencies is illustrated in the case of the efficiency-tracing technique for the activity measurements of ^{14}C using ^{60}Co as a tracer (Steyn, 1973).

2. Extension of the TDCR-Geant4 model to $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence counting

2.1 Description of $4\pi(\text{LS})\beta\text{-}\gamma$ detection system

As already described (Bobin *et al.*, 2007), the LS β -channel is equipped with a complete TDCR setup based on three XP2020Q PMTs (Photonis) arranged symmetrically around an optical cavity containing the LS. The XP2020Q PMTs are designed with fused-silica windows and bialkali photocathodes. The reflective part of the optical chamber is made of Teflon[®]. The γ -channel is equipped with a standard 3"x3" NaI(Tl) scintillator detector (76B76/3M, Scionix Holland) in a 0.5 mm-thick Al housing.

The LS counting in the β -channel is performed in the same manner as for TDCR measurements. For each PMT, the detection threshold is set below the single photoelectron signal. The counting in the β -channel is triggered in a MTR2 module (Bouchard, 2000) when

at least a coincidence between 2 PMTs occurred within a resolving time generally set to 80 ns. Based on extendable dead times (well-adapted for the processing of after-pulses in LS counting), this module is part of a complete instrumentation designed for live-timed anticoincidence measurements using home-made electronics. In the γ -channel, the signals given by a shaping amplifier are used to feed an analogue-to-digital converter (ADC 3512 and its associated memory 3588, LeCroy) in order to register two γ -spectra (coincident and non-coincident). The amplitude analysis is monitored by a MTR2 module to account for the dead-time generated in the γ -channel. The two γ -spectra are used to calculate indirectly coincidence rates as it is commonly performed with an anticoincidence system. The efficiency-extrapolation technique is generally performed by defocusing the PMTs. The variation of the detection efficiency in the β -channel can also be obtained by adding a quenching agent (e.g. nitromethane).

2.2 The Geant4 modeling for $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence

The TDCR-Geant4 modeling for liquid scintillation counting was previously described in the case of the standardization of ^{63}Ni (Thiam *et al.*, 2012) and for the measurements of the scintillation yield (Halter *et al.*, 2014) of the Ultima Gold (UG) scintillator cocktail. The present study on efficiency-extrapolation curves is performed with the addition of a γ -channel to the TDCR-Geant4 model. The main aspects of the whole Monte Carlo modeling are described in the following. The Geant4 code was chosen for its packages enabling the simulation of the transport of ionization radiation as well as light emission (scintillation, Cerenkov). In the present study, the simulations were implemented with the Geant4.10 version using the low-energy electromagnetic physics based on the PENELOPE models (Apostokalis *et al.*, 1999). The model also includes the atomic relaxation using the Livermore Evaluation Atomic Data Library, which contains the radiative and non-radiative transition probabilities for each sub-shell of each element, for $Z=1-100$ (Perkins *et al.*, 1991). Based on the UNIFIED model (Levin and Moisan, 1996), optical properties were defined according to the photon wavelengths to simulate reflection and refraction processes depending on refractive indices of medium boundaries.

The modeling of the LS β -channel aims at simulating optical photons from their creation along the track of a charged particle to the production of photoelectrons at PMT photocathodes. To this end, all the components of the optical cavity are considered in the geometry modeling as well as their optical properties (transmittance, refractive index depending on wavelengths):

- for the UG scintillator cocktail, dispersive refractive index ~ 1.57 at 430 nm (Kossert, 2013),
- for the glass vial (borosilicate, 20 mL), refractive index ~ 1.52 at 430 nm, and low-wavelength transmittance cutoff ~ 0.50 at 290 nm,
- for the reflective part of the optical cavity (Teflon[®]), lambertian-type reflectivity 0.95, for the PMT window (fused silica, \varnothing 52 mm) placed at 17 mm from the center of the optical cavity, dispersive refractive index 1.47 at 400 nm,
- for the PMT photocathode (bialkali), dispersive refractive index ~ 2.5 at 430 nm, and high-wavelength sensitivity cutoff ~ 650 nm.

All the physical processes generating optical photons are considered in the simulation of the β -channel. The number of scintillation photons is calculated from the energy released in the LS cocktail by the various interactions that can occur after a disintegration (electrons, x-ray and γ -photons). In the case of electron capture or internal conversion, the KLM rearrangement model is applied (Kossert and Grau Carles, 2006). The calculation of the number of scintillation photons is implemented with a Poisson law using the energy released during the step length of a charged particle. The emission is spread uniformly along track segment in 4π sr. The non-linearity of light emission due to ionization quenching in the scintillation process is taken into account with Birks' formula. The kB value (comprised between 0.1 and 0.13 mm/MeV) and the scintillation yield (comprised between 8 and 9 photons/keV) are drawn from previous studies (Thiam *et al.*, 2012; Halter *et al.*, 2014)

Cerenkov photons can be emitted in each transparent elements of the optical chamber (LS cocktail, borosilicate vial, PMT windows) by electrons initially produced in the LS vial or created by Compton scattering (e.g. in PMT windows). For each component, the Cerenkov threshold depends on the respective refractive index. Cerenkov photons are emitted on the surface of a cone with an angle that opens as charged particles slow down in the medium. The number of Cerenkov photons is calculated for each step length using a Poisson distribution with a mean value determined from the Frank and Tamm theory (Tamm and Frank, 1937). At the end, only optical photons (scintillation and Cerenkov) refracted at the bialkali photocathode are considered for the following step which consists in their conversion into photoelectrons. For that purpose, the calculation is performed using binomial trials implemented with the PMT quantum efficiency depending on wavelengths (~ 0.24 in the 300-400 nm) given by Araújo *et al.* (1998)

Regarding the modeling of the γ -channel, the detector geometry consists in the crystal and the close surroundings in front of the β -channel. The housing around the crystal has a

total thickness of 2 mm with 1.5 mm for the reflector (alumina) and 0.5 mm of aluminum. The front of the γ -detector is placed at a distance of 7 cm from the center of the optical cavity.

For each disintegration occurring randomly in the LS volume (UG, 10 mL), the simulation reports the number of photoelectrons produced in each photomultiplier according to the origin of light emission in the optical cavity (LS cocktail, borosilicate vial, PMT window) and linked to the energy released in the γ -channel. As expected, light emission from PMT windows is only composed of Cerenkov photons. These results are obtained by taking into account the ionizing radiation emitted from each decay-scheme pathway depending on radionuclides. The energy resolution for the NaI(Tl) scintillator (7 % at 662 keV and 15 % at 60 keV) is taken into account afterwards by a convolution using a Gaussian distribution. In order to simulate the efficiency-extrapolation technique, the PMT defocusing is implemented by a second binomial trial using a focusing parameter. The purpose is to simulate the decrease of the detection efficiency in the β -channel by reducing the number of photoelectrons reaching the first dynode. For each disintegration, a photoelectron is counted in a PMT when a binomial success has occurred. As for TDCR measurements, at least 2 binomial successes in two different PMTs are required to trigger a count in the β -channel.

The calculation of beta spectra used in the simulations is implemented with a computation code developed at LNHB (Mougeot *et al.*, 2011).

3. Activity measurements of ^{59}Fe

Fe-59 is a β -emitter with a half-life of 44.494 (12) d used as a tracer for the study of iron metabolism (Bé *et al.*, 2004). This radionuclide mainly decays through three β^- transitions, $\beta_{0,3}^-$ (energy: 130.7 keV, probability: 1.25(3) %, allowed), $\beta_{0,2}^-$ (energy: 273.4 keV, probability: 45.19(34) %, allowed) and $\beta_{0,1}^-$ (energy: 465.7 keV, probability: 53.30(31) %, allowed). The disintegration of ^{59}Fe mainly feeds two excited levels of ^{59}Co that can be used for coincidence counting: the $\gamma_{1,0}$ -transition at 1099.2 keV and the $\gamma_{2,0}$ -transition at 1291.6 keV. At these energies, the detection efficiency of γ -photons in the β -channel also includes Cerenkov photons produced in PMT windows (fused silica, Cerenkov threshold \sim 180 keV) due to Compton scattering (Thiam *et al.*, 2010).

Regarding $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements of ^{59}Fe , 5 radioactive sources were prepared, each containing 10 mL of UG cocktail plus aliquots of the master solution (10 $\mu\text{g/g}$ of Fe in the chemical form FeCl_2 in 0.1 M HCl). As shown in Fig. 1, the efficiency-extrapolation technique was implemented by PMT defocusing. For each plot, coincidence

counting was obtained using a γ -window centered on the two γ -photon energies (1099.2 keV and 1291.6 keV). Using this setting, the values of the experimental N_c/N_γ ratios are comprised between 93 % and 97 %. The fitting of the plots in Fig. 1 was implemented by means of the following expression:

$$N_\beta = A[1 - K_{\beta\gamma}(1 - N_c/N_\gamma)] \quad (1)$$

The experimental slope $K_{\beta\gamma}$ was equal to 0.920 (12) ($R^2 \sim 0.998$). The relative standard uncertainty associated with the activity concentration determined from the 5 sources was estimated at 0.2 %. The main components that contribute to the uncertainty budget are related to the efficiency extrapolation, statistics and weighing.

During the exercise, the result obtained with $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence counting was compared with the activity concentration given by $4\pi(\text{PC})\beta\text{-}\gamma$ coincidence using a proportional counter (CH_4 , atmospheric pressure). Uncertainties obtained with both measurement techniques were comparable as well as the maximum N_c/N_γ values. But in the case of $4\pi(\text{PC})\beta\text{-}\gamma$ coincidence counting, the extrapolation slope $K_{\beta\gamma}$ was equal to 1.16 (2) indicating a significant difference on the behaviour between both the LS detector and the proportional counter.

The calculation with the Geant4 modeling was implemented with the simulations of 5×10^6 events corresponding to disintegrations in the LS volume. Both scintillation and Cerenkov radiations were considered for the calculation of coincidences between PMTs. Regarding the emission of scintillation photons, a kB value of 0.01 cm/MeV and a scintillation yield equal to 8.5 photons/keV were applied. The plots depicted in Fig. 2 were obtained using the same γ -window as for experimental data and by decreasing the focusing parameter from 1 down to 0.35. Both the experimental and calculated maximum values of N_c/N_γ are approximately equal to 0.97. The slope $K_{\beta\gamma}$ calculated from the fitting of the plots in Fig. 2 is equal to 0.920 (5) which is consistent with the experimental value. The same calculation performed by only considering the scintillation process (without Cerenkov effect) yields a slope slightly different: $K_{\beta\gamma} = 0.929$ (5). In addition, simulations were also implemented by changing the scintillation features ($kB = 0.013$ cm/MeV and a scintillation yield equal to 9.0 photons/keV) including Cerenkov effect; in that case the variation of the calculated slope is more significant: $K_{\beta\gamma} = 0.903$ (5). No problem of bias on the activity determination was predicted.

4. Influence of Cerenkov radiation due to Compton scattering in the efficiency-extrapolation of ^{54}Mn

Mn-54 disintegrates with a half-life of 312.19 (3) d through electron capture to the 834.8 keV excited level of ^{54}Cr (Bé *et al.*, 2004) leading to the emission of a single γ -photon (the internal conversion is negligible). The ionizing radiation emitted following the electron capture is mainly constituted of x-ray photons and Auger electrons having energies comprised between 4 keV and 6 keV. The standardization of ^{54}Mn does not raise any specific difficulty when using a $4\pi(\text{PC})\beta\text{-}\gamma$ coincidence system. On the contrary, an unexpected bias in the activity determination (more than 1 %) was observed in the case of $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements during the participation to the key comparison CCRI(II)-K2.Mn-54 in 2003. Additional measurements revealed a non-equivalence of the β -efficiency variations carried out by PMT defocusing and by adding quenching agent in TDCR measurements (Thiam *et al.*, 2010).

Monte Carlo simulations have confirmed that coincidences in the LS counter can be obtained with Cerenkov photons emitted from PMT windows due to electrons produced by Compton scattering (Thiam *et al.*, 2010). The existence of this effect was also observed in TDCR measurements by introducing solid sources of γ -photon emitters (^{137}Cs , ^{54}Mn , ^{60}Co) inside the optical cavity of the LS counter (without liquid scintillator). This light emission can be considered as a part of the γ -efficiency in the β -channel in the case of $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence counting. Due to the low amount of Cerenkov photons, this contribution to the extrapolation slope can be sensitive to the variation of the detection efficiency in the β -channel. In order to look into this problem, simulations were carried out to confirm the non-linearity observed in the case of the standardization of ^{54}Mn with the $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements. Simulations of monoenergetic γ -photons emitted from the LS volume were also performed to investigate the contribution of the Cerenkov emission to the γ -efficiency of the β -channel.

Monte Carlo simulations of efficiency extrapolation were compared to $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements carried out with LS vials filled with 10 mL of UG cocktail. For the measurements, a resolving time was set to 100 ns in the LS system for the counting of coincidences between PMTs. Using a γ -window centered on the single full-energy peak at 835 keV for coincidence counting, the values of the detection efficiency N_c/N_γ obtained by PMT defocusing were comprised between 18 % and 53 %. As shown in previous studies on TDCR measurements (Bobin *et al.*, 2012), the maximum detection efficiency N_c/N_γ can significantly change by a few percent depending on the resolving time applied for PMT

coincidences in the case of low-energy emitters like ^{54}Mn . The fitting procedure with expression (1) was applied on 18 coincidence measurements ($R^2 \sim 0.999$). A bias of 1.6 (4) % with the expected activity was assessed and the experimental slope $K_{\beta\gamma}$ was equal to 0.930 (6).

The Monte Carlo simulations were implemented in the same manner as described in the case of ^{59}Fe (5×10^6 events). As already mentioned, the KLM rearrangement model was applied (Kossert and Grau Carles, 2006) to account for the various pathways occurring after an electron capture. Several calculations were implemented by varying the scintillation yield between 8 and 9 photons/keV and by using kB values equal to 0.01 and 0.013 cm/MeV. The maximum calculated efficiency N_c/N_γ ranged within 55 % and 58 % when using a focusing parameter equal to 1 (γ -window centered on the single full-energy peak at 835 keV). In order to match the calculations with experimental N_c/N_γ values, focusing parameters ranging from 0.95 down to 0.3 were applied to simulate the efficiency-extrapolation technique by PMT defocusing. The fitting of the calculated detection efficiency in the β -channel versus the inefficiency $1 - N_c/N_\gamma$ was performed using expression (1). The calculated bias with the expected activity was found equal to 1.0 (3) % with a calculated slope $K_{\beta\gamma}$ equal to 0.927 (3). A slight non-linearity is clearly observed in the calculated residuals displayed in Fig. 3. Simulations were also performed by only considering the scintillation process (*i.e.* without Cerenkov photons). In that case, the bias with the expected activity vanishes and the slope changed slightly ($K_{\beta\gamma} = 0.935$ (3)).

The calculated and experimental values of the slopes $K_{\beta\gamma}$ are consistent. The existence of a deviation with the expected activity is confirmed by the simulation of $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements. The difference between the experimental and calculated biases reflects the difficulty to simulate accurately the small amount of Cerenkov photons created in PMT windows. Indeed the measurements carried out with a solid source of ^{54}Mn placed inside the optical cavity yield a detection efficiency due to Compton scattering in PMT windows equal to 1 % (Thiam *et al.*, 2010). In addition, the physical process leading to coincidences is highly anisotropic because Cerenkov radiation is first created in a PMT window. But this effect cannot be neglected in the case of the standardization of low-energy emitters such as ^{54}Mn when using $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence counting. Indeed, the shape of the residuals displayed in Fig. 3 is the result of the variation of the Cerenkov contribution to the γ -efficiency in the LS counter. This bias with the expected activity is also due to the sensitivity of the efficiency-extrapolation technique when it is applied to emitters having low detection efficiency such as ^{54}Mn .

As a complement to this study, the contribution of the Cerenkov radiation to the γ -efficiency of the LS counter was also evaluated by Monte Carlo simulation for increasing monoenergetic photon energies. As observed in the results displayed in Fig. 4, the contribution of Cerenkov light emission becomes significant for energies greater than 600 keV. The uncertainties attached to each plot were assessed with a conservative procedure: for instance, calculations were implemented by changing the distance of the PMT windows to the optical cavity centre from 17 mm to 16 mm.

5. Application to the efficiency-tracing technique

The $4\pi\beta\text{-}\gamma$ coincidence method can be extended to the measurements of pure β -emitters with the application of the efficiency-tracing technique (Campion *et al.*, 1960). This method was more utilized in the past in radionuclide metrology before the advent of primary measurement techniques such as TDCR (Broda *et al.* 2007). For instance the efficiency-tracing technique can be applied for the standardization of ^{137}Cs using ^{134}Cs as a $\beta\text{-}\gamma$ emitter tracer by mixing both radionuclides in the same radioactive source. In order to improve the chemical mixture, efficiency-tracing measurements can be carried out with LS sources as an alternative to classical dry sources (Steyn, 1973). This method relies on the efficiency-extrapolation technique using coincidences measured with the $\beta\text{-}\gamma$ emitter. As mentioned before, no systematic bias on the activity determination is observed as long as the linear relation between β -inefficiencies of the pure-beta emitter and the tracer remains unaltered (depending on respective β -shapes). The case of the activity measurements ^{14}C was investigated with the Geant4 modeling using ^{60}Co as a tracer mixed in a LS vial filled with 10 mL of UG. ^{14}C disintegrates with a half-life of 5700 (30) y through a β - transition to the ground state of ^{14}N (energy of 156.5 keV, allowed transition). ^{60}Co disintegrates with a half-life of 5.2711 (8) y to excited states of ^{60}Ni mainly through two β - transitions (Bé *et al.*, 2006), $\beta_{0,3}^-$ (energy: 317.3 keV, probability: 99.88 (3) %, allowed transition) and $\beta_{0,1}^-$ (energy: 1490.6 keV, probability: 0.12 (3) %, unique 2^{nd} forbidden transition).

The simulation of the efficiency-extrapolation curve was first carried out for $4\pi(\text{LS})\beta\text{-}\gamma$ measurements of ^{60}Co . Considering the scintillation process, two configurations (scintillation yield, kB value) were considered: 8 photons/keV with $kB=0.01$ cm/MeV and 9 photons/keV with $kB=0.013$ cm/MeV. The ratio N_c/N_γ was calculated using a γ -window centred on the two γ -photons (1173.2 keV and 1332.5 keV). The maximum value corresponding to a focusing parameter set to 1 was equal to 0.971 which was slightly greater than the experimental value (0.965). For the simulation of the extrapolation curve, the N_c/N_γ ratios were calculated with

focusing parameters down to 0.3 ($N_c/N_\gamma = 0.925$). The fitting procedure using expression (1) gives $K_{\beta\gamma}$ equal to 0.917 (6) in the case of a scintillation yield of 8 photons/keV and $K_{\beta\gamma}$ equal to 0.923 (7) with 9 photons/keV. In both cases, the y-intercept is close to 1, indicating no problem of bias on the activity determination. The experimental $K_{\beta\gamma}$ value was equal to 0.927 (10).

Regarding the application of the efficiency-tracing technique for activity measurements of ^{14}C , the simulation of the detection efficiency of ^{14}C in the LS counter was carried out with the same settings implemented for ^{60}Co . In that case, the fitting equation was applied using calculated β -efficiencies for ^{14}C versus the inefficiencies ($1 - N_c/N_\gamma$) given by ^{60}Co calculations (each computed coordinate corresponding to the same focusing parameter). From the simulations implemented with both scintillation configurations, the fitting results yield: $K_{\beta\gamma}$ equal to 1.80 (2) and the y-intercept equal to 1.003 (1), *i.e.* a significant bias on the activity determination of 0.3 %. These results were compared with experimental measurements performed with three scintillation vials containing a mixture of ^{60}Co and ^{14}C in 10 mL of UG. The application of the efficiency-tracing technique leads to an overestimation of 0.6 (3) % on the expected activity concentration (previously obtained with TDCR measurements) and $K_{\beta\gamma}$ equal to 1.70 (4). Despite the slight difference observed between experimental and calculated results, the simulation with the Geant4 code was able to predict a bias on the activity determination of ^{14}C based on the efficiency-tracing technique using ^{60}C as a tracer.

6. Discussion

For more than 10 years (Bobin *et al.*, 2006), an experience feedback has been acquired at LNHB on the use of a $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence system based on a complete TDCR setup. Compared to a classical $4\pi(\text{PC})\beta\text{-}\gamma$ coincidence system, LS counting presents some interesting advantages. Source preparation is implemented by direct mixing of the radioactive aliquot into the LS cocktail. Counting losses are not due to self-absorption in dry sources which is sensitive to source preparation. Instead, LS detection efficiency rather depends on physico-chemical effects such as ionization quenching. The variation of the LS efficiency is generally implemented at LNHB by PMT defocusing or by adding quenching agent. Other techniques described in the literature like geometry-efficiency variation (Hwang *et al.*, 2006) were also investigated in radionuclide metrology. The main drawback is a significant increase of the detection efficiency of γ -photons due to, on one hand, direct interactions in the LS volume and, on the other hand, scattering in the surroundings. In the

latter case, the extension of the TDCR-Geant4 modeling to the simulation of $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements confirmed that the contribution of Cerenkov emission in the γ -efficiency in the β -channel cannot be neglected. For γ -photon energies greater than 800 keV, the proportion due to Cerenkov emission is greater than 10 %. Furthermore, the variation of this contribution when the efficiency-extrapolation technique is applied can entail a systematic bias on the activity determination as it was observed in the case of the standardization of the low-energy emitter ^{54}Mn . The Geant4 modeling for $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence simulation reveals to be a useful tool for a better understanding of physical processes in the LS counter and for the prediction of potential biases in activity measurements.

The interest of the $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence system is also the possibility to carry out measurements directly with aqueous solutions and based only on Cerenkov emission and detection (no LS cocktail used). Actually, it is possible to take advantage of the Cerenkov threshold for discrimination purpose in the measurement of radionuclide mixtures such as $^{68}\text{Ge}/^{68}\text{Ga}$. In that particular case, only interactions resulting from ^{68}Ga disintegrations (β^+ events, maximal energy of 1899.1 keV) are counted in the β -channel due to the fact that electron-capture events from ^{68}Ge disintegrations have lower energies (maximal values of about 10 keV) than the Cerenkov threshold in water (~ 260 keV).

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Captions

Fig. 1: Efficiency-extrapolation curve obtained by PMT defocusing in the case of the standardization of ^{59}Fe with $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements.

Fig. 2: Simulation with the Monte Carlo Geant4 code of efficiency-extrapolation curve in the case with $4\pi(\text{LS})\beta\text{-}\gamma$ coincidence measurements of ^{59}Fe .

Fig. 3: Distribution of the residuals resulting from the fitting of the simulated efficiency-extrapolation curve obtained in the case of $4\pi(\text{LS})\beta\text{-}\gamma$ counting of ^{54}Mn . The shape of the residuals is due to the variation of the Cerenkov-emission contribution to the γ -efficiency of the LS counter.

Fig. 4: Simulated detection efficiency of γ -photon interactions in the LS counter obtained with the Geant4 modeling (scintillation vial filled with 10 mL of Ultima Gold). The results are displayed with and without the contribution of Cerenkov emission resulting from Compton scattering in PMT windows.

Figure 1

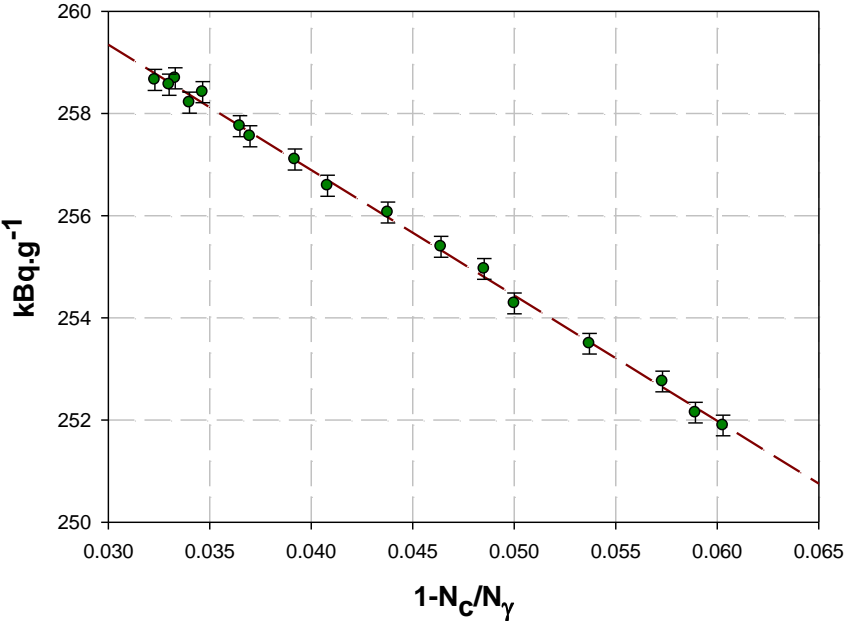


Figure 2

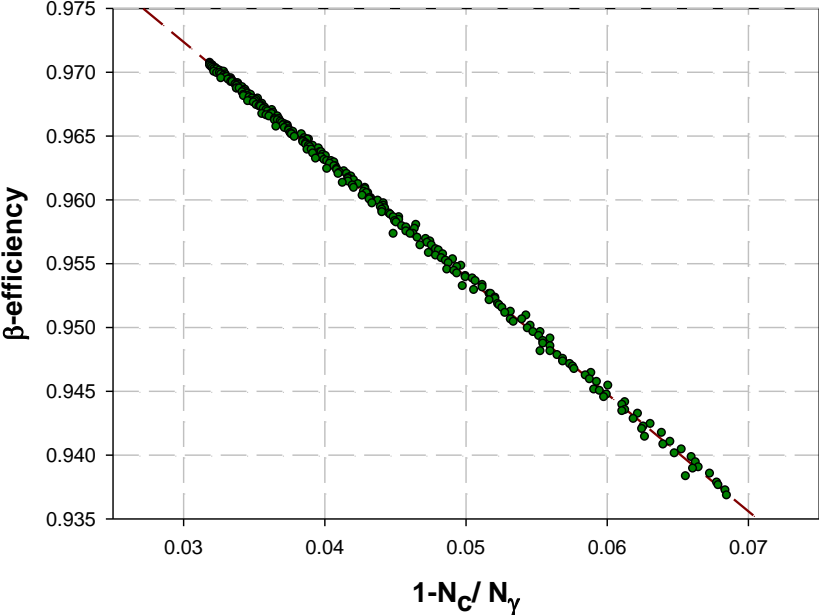


Figure 3

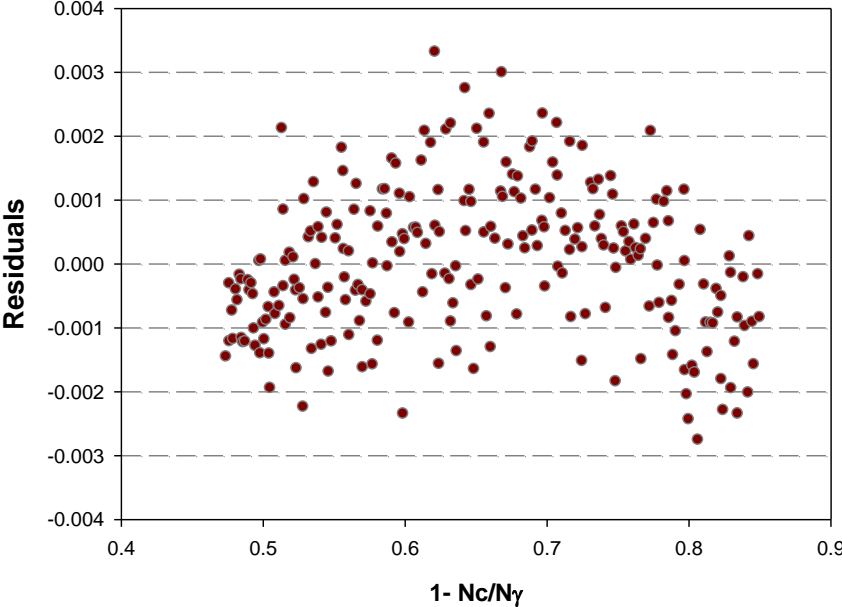


Figure 4

