Preliminary TDCR measurements at low energies using a miniature x-ray tube

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Preliminary TDCR measurements at low energies using a miniature x-ray tube

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Abstract

Developed for radionuclide standardization using liquid scintillation, the TDCR method (Triple to Double Coincidence Ratio) is applied using coincidence counting obtained with a detection system composed of three photomultiplier tubes. For activity determination, a statistical model of light emission is classically used to establish a relation between the detection efficiency and the experimental *TDCR* value. Among the usual assumptions specified for the standard TDCR model, the condition of stochastic independence between PMTs is not explicitly mentioned. The validity of this condition can be tested using a stochastic approach of the TDCR modeling based on the Geant4 simulation code. The interest of this TDCR-Geant4 model is the possibility to simulate the propagation of optical photons from their creation in the scintillation vial to the production of photoelectrons in PMTs. Previous investigations revealed the existence of stochastic dependence between PMTs of geometrical origin in the case of low-energy depositions (lower than 20 keV). Not considered in the standard model, this effect can entail non-negligible deviations on calculated detection efficiencies and in turn on activity determination.

As an alternative to the use of radionuclide sources, preliminary TDCR measurements are presented using a miniature x-ray tube closely coupled to the scintillation vial. The first objective of this new set-up was to enable experimental studies with low-energy depositions for both time and geometrical dependence between PMTs. As for the statistical TDCR model, the non-linearity of light emission is implemented in the TDCR-Geant4 model using the Birks formula which depends on the *kB* factor and the scintillation yield. These parameters are assessed from TDCR measurements obtained using the x-ray tube. They are tested afterwards in the TDCR-Geant4 model for activity measurements of ^{3}H.

Keywords: TDCR method, radionuclide metrology, x-ray tube, Geant4 simulation code

1. Introduction

The TDCR (Triple to Double Coincidence Ratio) method is widely applied in National Metrology Institutes for primary radionuclide standardization (Broda et al., 2007). Based on a specific LS (Liquid Scintillation) counter equipped with three photomultipliers tube (PMTs), the activity is determined using the experimental *TDCR* ratio given by double and triple coincidences between PMTs. To this end, a statistical model of light emission is implemented according to several assumptions such as stochastic independence between PMTs (Bobin et al., 2012a), Poisson distribution of photoelectrons in PMTs, etc. In the statistical modeling, double and triple coincidences between PMTs are calculated using an analytical expression of the probability to have at least one PMT count as a result of an energy deposition. However, the statistical approach does not account for the optical and geometrical properties of the detector in terms of refraction and reflection processes of optical photons. At LNE-LNHB (Laboratoire National Henri Becquerel), an alternative to the statistical TDCR model is studied using the Geant4 simulation toolkit (Agostinelli et al., 2003) in order to simulate the transport of charged particles and optical photons resulting from scintillation and Cerenkov effect. With the TDCR-Geant4 model, optical photons are simulated from their creation in the optical cavity to the generation of photoelectrons in PMTs leading to double and triple coincidences. From previous studies, several results were obtained from the application of the TDCR-Geant4 model in the case of low-energy depositions (lower than 20 keV). In particular, simulations carried out for mono-energetic depositions (5 keV, 8 keV and 12 keV) revealed the existence of a stochastic dependence between PMTs of geometrical origin (Bobin et al., 2012a). This effect is due to the sensitivity of the photon distribution between PMTs to the location of light emission inside the scintillator volume combined with reflection and refraction processes occurring at the different interfaces of the optical cavity. As shown with the TDCR-Geant4 model for discrete-energy emitters, the consequence is an overestimation of detection efficiencies obtained with the statistical standard model. These simulation results were experimentally confirmed in the case of the ^{51}Cr standardization (this radionuclide disintegrates by electron capture with maximum energies of x-ray photons and Auger electrons mainly comprised between 4 keV and 6 keV). The influence of the geometrical stochastic dependence on detection efficiencies is directly observed by a shift of the counting rates for the same experimental *TDCR* value when using diffusive polyethylene vials instead of glass vials. This effect leading to an increase of the activity calculation with polyethylene vials has also been mentioned by Simpson et al. (2010) in the case of ^{55}Fe measurements. Another effect of stochastic dependence between PMTs is a deviation between the variance and the mean number of photoelectrons detected in each PMT (Bobin et al., 2012a). It has to be noted that this modification of the Poisson distribution was already investigated in previous studies for low-energy radionuclides but without actual physical interpretation (Broda et al., 2007).

In the present paper, preliminary TDCR measurements using a miniature x-ray tube are presented as an alternative to radionuclide sources. In order to obtain interactions of low-energy x-rays in the liquid scintillator, the end-window transmission-target x-ray tube (MAGNUM® 40 kV manufactured by Moxtek) can easily be coupled to a scintillation vial. From previous studies, two types of stochastic dependence were identified: on the one hand, when the coincidence resolving time is too short with regards to the time distribution of photons between PMTs, on the other hand, due to geometrical and optical effects as previously discussed. These two effects are first investigated experimentally for a low-energy deposition set to 2.7 keV. The influence of the liquid scintillator on the evolution of the counting rates according to the resolving time is checked. The modification of the relation between the detection efficiency of double coincidences and *TDCR* values is also tested using glass and polyethylene vials. As for the statistical model, the Birks formula is implemented in the TDCR-Geant4 model to account for the non-linearity of light emission as a consequence of ionization quenching. The associated parameters (*kB* factor, scintillation yield) are measured using the x-ray tube at 3 different energies (2.7 keV, 8.7 keV and 17.3 keV). These experimental values are tested afterwards in the case of the standardization of ^{3}H with the TDCR-Geant4 model.

2. Experimental TDCR set-up with a miniature x-ray generator

As already described in the framework of previous studies (Thiam et al., 2011), the detection system is composed of three XP2020Q photomultipliers equipped with a fused silica window. Counting vials (glass or polyethylene) are hung inside a spherical cavity made of Teflon®. For coincidence counting, the electronic chain is composed of a fast amplifier (Phillips scientific model 777) and a Constant Fraction Discriminator (CFD) module (Canberra Quad CFD 454). The logical signals provided by this front-end electronics are used to feed either a MAC3 module (Bouchard and Cassette, 2000) modified in order to set variable resolving times or a FPGA-based digital system which is also used as time-to-digital converter combined to the processing of counting losses according to the live-time technique using extendable dead-times (Bobin et al., 2012b).

The interest of integrating a miniature x-ray tube into the detection set-up is the ability to perform TDCR measurements with low-energy x-ray photons in the liquid scintillator as an alternative to radionuclide sources. Coincidence counting can be carried out to study the TDCR modeling. As depicted in Fig. 1, this small-size x-ray generator (~ 55 mm length, ~ 29 mm diameter) is directly coupled to the counting vial (for the x-ray beam, a small hole is machined in the plastic cap). The x-ray tube is designed to be a transmission-target end-window configuration with tungsten for the anode material. Three energies have been selected for this study: 2.7 keV, 8.7 keV and 17.3 keV. The x-ray-beam energy is tuned by applying an appropriate high-voltage potential and by using an attenuation film placed at the output of the x-ray tube. The upper-energy cutoff is defined by the film component and the energy spectrum is sharpened using a suitable attenuation thickness (see table 1 for the settings used for the 3 x-ray energies). For instance, the 2.7 keV-energy emission is obtained using the following settings: high voltage set to 3 kV, PVC film of ~ 100 μm thickness (cutoff energy of chlorine equal to ~ 2.8 keV). The spectrum shapes at the end-window were measured with a Silicon Drift Detector (AXAS-M manufactured by KETEK) calibrated using ^{55}Fe and ^{241}Am sources.

3. Experimental results

The sensitivity of coincidence counting with the coincidence resolving time for low-energy radionuclides has already been observed by several laboratories (Steele et al., 2009; Bobin et al., 2010a). According to complementary investigations, this effect is at the origin of stochastic time dependence between PMTs when the coincidence resolving time is too short compared with the time-arrival distribution of photoelectrons between PMTs. As observed for the standardization of ^{3}H, the consequence can be a deviation of detection efficiencies calculated with the statistical TDCR model (Bobin et al., 2010a).

The evolution of double-coincidence rates according to *TDCR* values has been compared for a 2.7 keV-energy emission between two liquid scintillators, Hionic Fluor (HF) and Ultima Gold (UG). The experimental results given by the time-to-digital converter especially designed for TDCR measurements are plotted in Fig. 2, taking the counting rates for a coincidence resolving time set at 40 ns as a reference (~ 220 s^{-1} for HF; ~ 400 s^{-1} for UG). It can be observed that the counting rates are less sensitive to increasing coincidence resolving times for the HF scintillator. This difference of behaviour between the two scintillators is in agreement with the hypothesis given previously in Bobin al. (2010a), *i.e.* when the number of emitted photons is low, genuine double and triple-coincidences can be lost when the coincidence resolving time is too short compared with scintillation lifetimes that depend on the liquid scintillator used. Complex processes such as triplet-triplet annihilation which contributes to the slow component in the scintillation emission could be involved in this phenomenon.

Additional measurements carried out with the UG scintillator have shown that the type of vial (glass or polyethylene) has no significant impact on counting rates with regards to the influence of coincidence resolving times. On the contrary, as displayed in Fig. 3, a significant shift is observed for the relation between the double coincidence rates and the experimental *TDCR* value. As previously studied with the TDCR-Geant4 model, this effect reflects the influence of the geometrical stochastic dependence between PMTs on triple and double coincidence counting (Bobin et al., 2012a).

4. The TDCR-Geant4 modeling

The Monte Carlo Geant4 code was chosen for its capability to simulate the transport of ionizing and electromagnetic radiations (Agostinelli et al., 2003). This code allows the construction of a geometrical model of the LS counter including the optical properties attached to each element of the optical cavity (glass vial, PMT windows, etc.). As already described in previous studies (Bobin et al., 2010b; Thiam et al., 2011), this new modeling has been first investigated with the application of the TDCR-Cerenkov technique. In that case, the TDCR-Geant4 benchmark is able to simulate the transport of ionizing particles emitted in aqueous solutions and subsequent Cerenkov photons. Taking into account refraction and reflection processes at boundaries and spectral transmittance of materials (borosilicate vial, PMT windows), optical photons are tracked from their creation (including the anisotropy of Cerenkov emission) inside the optical chamber (aqueous solution, glass vial, PMT windows) to their conversion into photoelectrons at the PMT photocathode. For each energy deposition following a disintegration, a binomial trial is applied to all the photoelectrons produced in the PMTs in order to know if they are detected or not, and then to calculate double and triple coincidences between PMTs. This trial represents the probability for a photoelectron to reach the first PMT dynode and it is used to simulate the PMT defocusing for the detection efficiency variation. The probability to count at least one photoelectron in a PMT calculated with the TDCR-Geant4 model differs from the statistical modeling by taking into account potential stochastic dependence between PMTs. Activity measurements based on Cerenkov emission were carried out for various radionuclides: ^{90}Y, ^{11}C, ^{32}P (Bobin et al., 2010b; Thiam et al., 2011). Because of the anisotropy of Cerenkov emission, these results are considered as a validation step of the geometrical modeling. As for the classical statistical model, the extension of the TDCR-Geant4 model to scintillation counting is based on the Birks formula with regards to the non-linearity of light emission as a consequence of ionization quenching. In that case, the TDCR-Geant4 model has been applied to activity measurements of ^{63}Ni, ^{51}Cr and ^{60}Co in UG (Bobin et al., 2012b; Thiam et al., 2012).

The simulation results obtained in the present study are performed with the Geant4.9.4 version using the low-energy electromagnetic physics based on PENELOPE models (Apostolakis et al., 1999). The model includes also the atomic relaxation using the Livermore Evaluation Atomic Data

Library (EADL), which contains the radiative and non-radiative transition probabilities for each sub-shell of each element, for Z=1 to 100 (Perkins et al., 1991).

5. Estimation of the *kB* factor and scintillation yield used in the Birks formula

When the statistical TDCR method is implemented, the activity is usually determined from the experimental *TDCR* values obtained by variation of the detection efficiency. An optimal *kB* factor is estimated in order to obtain the most consistent activities over a range of experimental *TDCR* values. As it is included in a free parameter, the value of the scintillation yield (characterising the liquid scintillator) is not explicitly specified for the activity determination (Broda et al., 2007). In the case of TDCR-Geant4 model, the scintillation process is also implemented using the Birks formula but the scintillation yield has to be defined in terms of photons emitted per keV. The measurement of this parameter was previously carried out using 4π(LS)β–γ measurements applied with radionuclides (Bobin at al., 2012a).

For given liquid scintillator and x-ray energy, pairs of *kB* factor and scintillation yield are calculated with the TDCR-Geant4 model using the *TDCR* measurement obtained with the x-ray generator coupled to the LS counter. For the preliminary tests of this system, the measurements were carried out using a glass vial containing 10 mL of UG. The size of the x-ray beam has been reduced using a Delrin collimator (Ø = 8 mm) in order to decrease the diameter of the volume where x-ray photons interact in the liquid scintillator. In this configuration, the geometrical stochastic dependence on TDCR measurements resulting from the photon distribution between PMTs is minimized (Bobin et al., 2012a). The experimental *TDCR* values obtained for x-ray energies equal to 2.7 keV, 8.7 keV and 17.3 keV are respectively 0.137 (2), 0.646 (2) and 0.928 (2). As expected with the TDCR-Geant4 model, it has been observed that the collimation increases the measured *TDCR* values: for instance, at 2.7 keV *TDCR*=0.128 (2) without the collimator.

In the TDCR-Geant4 modeling, the x-ray emission is implemented according to a beam flux with energies randomly generated using the spectra measured with the SDD for each energy. The spectrum obtained in the case of 2.7 keV x-ray photons is displayed in Fig. 4. The atomic rearrangement resulting from x-ray interactions with the main components of the UG scintillator (~79 % carbon, ~10 % hydrogen, ~ 9 % oxygen and ~ 1.4 % phosphorous) is also considered Geant4 simulations. The photoelectron spectrum resulting from 2.7 keV x-ray interactions in UG is shown in Fig. 5. The energy distribution is mainly comprised between 2 and 2.7 keV. The photoelectron spectrum features also a non-negligible component at about 600 eV (~ 12 %) due to photoelectric effect with phosphorous.

For each photon energy obtained with the x-ray tube, several simulations with the TDCR-Geant4 model were carried out for *kB* factors ranging from 0.07 to 0.13 (corresponding to usual values drawn from the literature) and scintillation yields (ranging from 7 to 10 photons.keV^{-1}). The pairs of *kB* factor and scintillation yield displayed in Fig. 6 are obtained by minimizing the difference between measured and calculated *TDCR* values (*e.g.* in the case of the 2.7 keV x-ray energy, each pair of *kB* factor and scintillation yield corresponds to a *TDCR* value equal to 0.137 (2)). In the range of *kB* factors and scintillation yields investigated, it appears that the plots follow a straight line with a slope which decreases when x-ray energy increases. Considering that the pair of *kB* factor and scintillation yield is constant with energy in the Birks formula, a coherent pair can be obtained: *kB* = 0.13 mm.MeV^{-1} associated with a scintillation yield equal to 8.9 (1) photons.keV^{-1}. In the future, the range of the *kB* factor will be extended above 0.13 mm.MeV^{-1} for further simulations.

Based on detection efficiencies measured with $4\pi(LS)\beta-\gamma$ coincidence counting (Bobin et al., 2012a), a [*kB*/scintillation yield] pair equal to [0.1 mm.MeV^{-1}/ 8.2 (3) photons.keV^{-1}] was previously assessed from ^{54}Mn measurements (energy mainly comprised between 4.5 and 6 keV).

This first result is in agreement with those reported at 8.7 keV photons given by the x-ray generator: [0.1 mm.MeV^{-1} / 8.3 (1)].

6. Application of the TDCR-Geant4 model to the standardization of ^{3}H

The results obtained with the x-ray generator (*kB* = 0.13 mm.MeV^{-1}; scintillation yield = 8.9 (1) photons.keV^{-1}) were used in the TDCR-Geant4 model for activity measurements of ^{3}H in UG. Due to the low energies of emitted electrons, the standardization of this β^{-}-emitter (E_{max}=18.6 keV) is sensitive to the *kB* factor. Activity concentrations were calculated from measurements obtained by PMT defocusing (*TDCR* values ranging from 0.49 to 0.54) using both the TDCR-Geant4 and statistical models. As observed in Fig. 7, the activity concentrations are about 1.3 % higher in the case of the TDCR-Geant4. Moreover, the slope featuring activity concentrations as a function of *TDCR* values is lower for the results given by the statistical model. Additional plots in Fig. 7 were obtained with the TDCR-Geant4 model by limiting the electron emission at the center of the scintillator volume. This configuration of the Geant4 calculations can be considered as closer to the statistical model (Bobin et al., 2012a): the slope difference between both models vanishes. In the case of the statistical model, the slope can be modified by changing the value of the *kB* factor. This practice is generally applied to minimize the fitting slope of the function giving the activity versus *TDCR* in order to obtain an optimized *kB* factor for the activity determination. For the present study, the optimal *kB* factor given by the statistical model is equal to 0.10 (1) mm.MeV^{-1}. In the case of the TDCR-Geant4, the magnitude of the slope (activity concentration versus *TDCR*) is unexpected because the observed trend was not encountered for the standardization of ^{51}Cr (low-energy discrete emitter). In addition, as for activity measurements of the β^{-}-emitter ^{63}Ni (Thiam et al., 2012), the slope is not sensitive to the modification of the *kB* factor. Further studies are needed to understand the differences observed between both TDCR-Geant4 and statistical models for the standardization of ^{3}H. In particular, the hypothesis can be proposed that the stochastic geometrical dependence between PMTs could have an influence on the estimation of the *kB* factor in the case of the statistical model.

7. Discussion and perspectives

Preliminary investigations using an x-ray generator coupled with a TDCR detection set-up were presented. The first objective of this new system was the possibility to reproduce with low-energy x-ray photons (less than 20 keV) the observations previously obtained with radionuclides (*e.g.* discrete low-energy emitters like ^{51}Cr). The aim was also to have an experimental set-up specifically designed to test and to refine the TDCR-Geant4 model for activity measurements of low-energy emitters. The influence of the vial type (glass and polyethylene) observed by a shift of the relation between double-coincidence rates versus *TDCR* values was confirmed with an x-ray beam tuned at 2.7 keV. By using the same configuration, a difference on the evolution of double-coincidence rates with coincidence resolving time was also shown when comparing two commercial liquid scintillators (Ultima Gold and Hionic Fluor).

A technique using the x-ray tube has been tested to assess the pair of *kB* factor and scintillation yield used in the Birks formula for a given liquid scintillator. Calculations are based on the minimization of the difference between an experimental *TDCR* value obtained for a given x-ray energy and calculations performed with the TDCR-Geant4. From measurements obtained with three x-ray energies (2.7 keV, 8.7 keV and 17.3 keV), a preliminary estimation of a pair of *kB* factor and scintillation yield is proposed: [0.13 mm.MeV^{-1}/ 8.9 (1) photons.keV^{-1}]. The estimated *kB* factor is higher compared with the value given by the statistical model (*kB*=0.10 mm.MeV^{-1}) obtained by minimizing the fitting slope of the function giving the activity concentration of ^{3}H versus *TDCR* values. The origin of the higher value given by the TDCR-Geant4 model is not well understood. This result could be due to the fact that the TDCR-Geant4 model includes geometrical and optical properties of the LS counter. As a result, the variation of the slope of activity versus *TDCR* obtained with the statistical modeling could include more complex processes than only the non-linearity of light emission. Finally, further investigations are planned to understand the difference between both the TDCR on activity measurement of ^{3}H by using other scintillation cocktail (e.g. Insta-gel and Pico-fluor, containing no phosphorous) or by measuring other low-energy emitters (^{55}Fe, ^{241}Pu).

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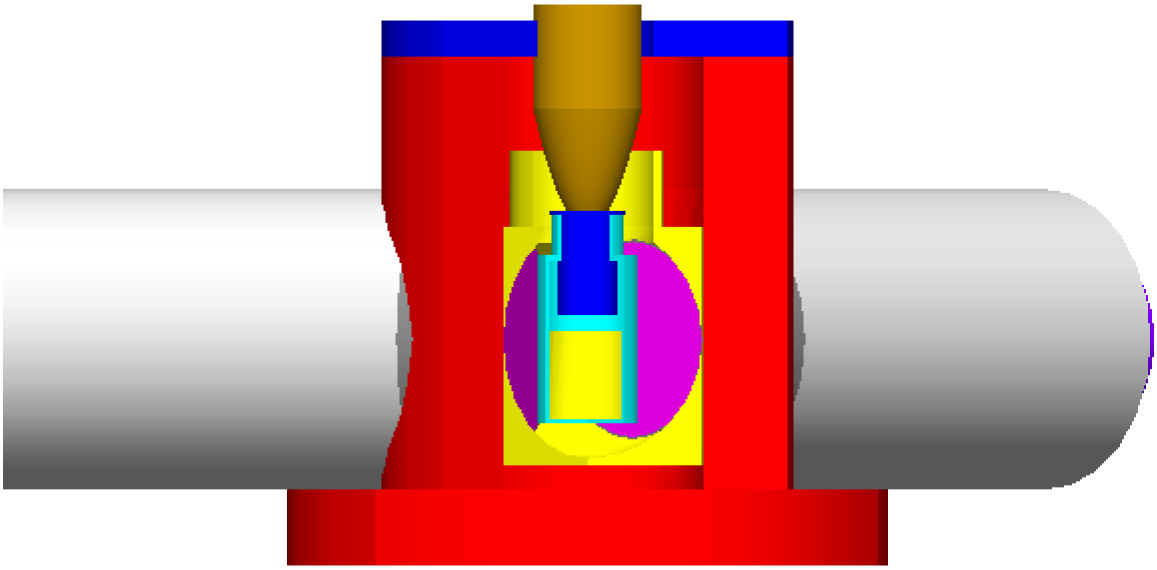
Fig. 7. Activity concentrations versus *TDCR* values in the case of ^{3}H in UG obtained with both statistical and TDCR-Geant4 models (*kB* equal to 0.13 mm.MeV^{-1}).

Table 1

X-ray energy	**Attenuation film**
2.7 keV	Polyvinyl chloride (100 µm) – cutoff energy: 2.8 keV
8.7 keV	Pure copper (300 µm) – cutoff energy: 8.9 keV
17.3 keV	Pure zirconium (800 µm) – cutoff energy: 17.7 keV

Figure 1



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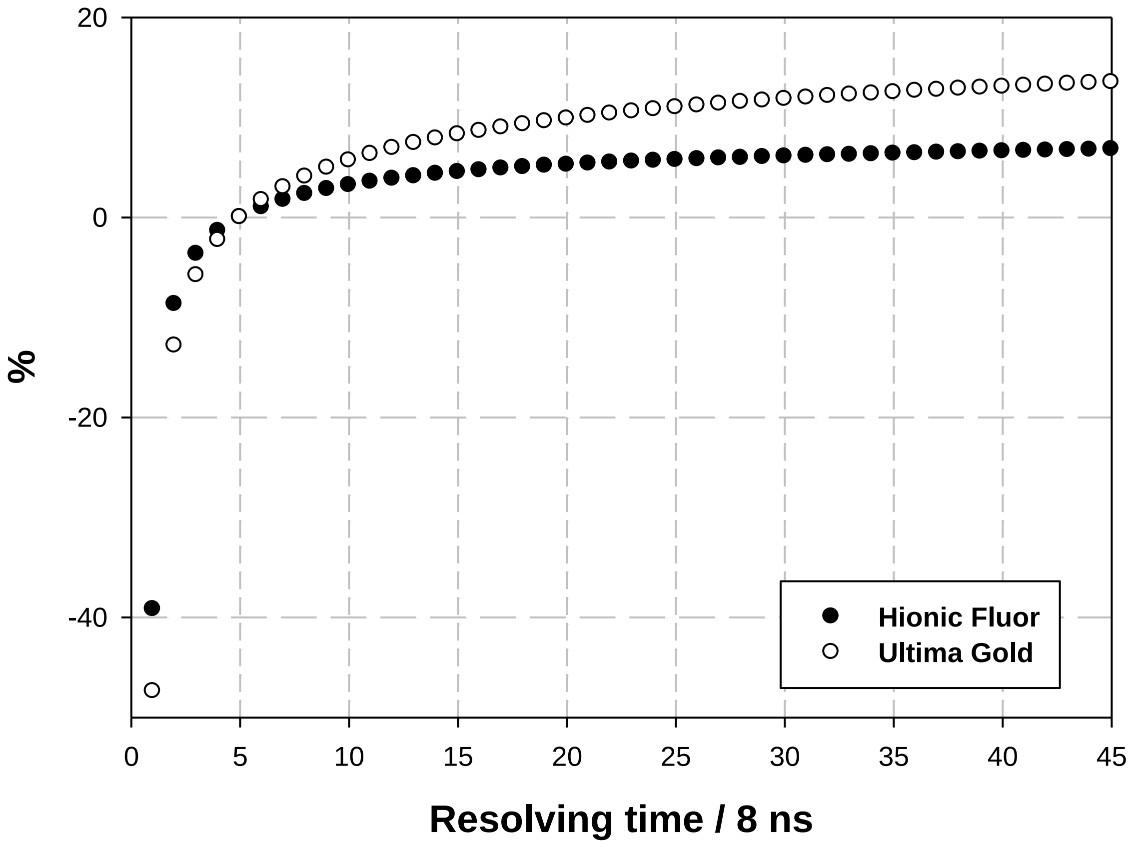
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0
-20
-40
%
0
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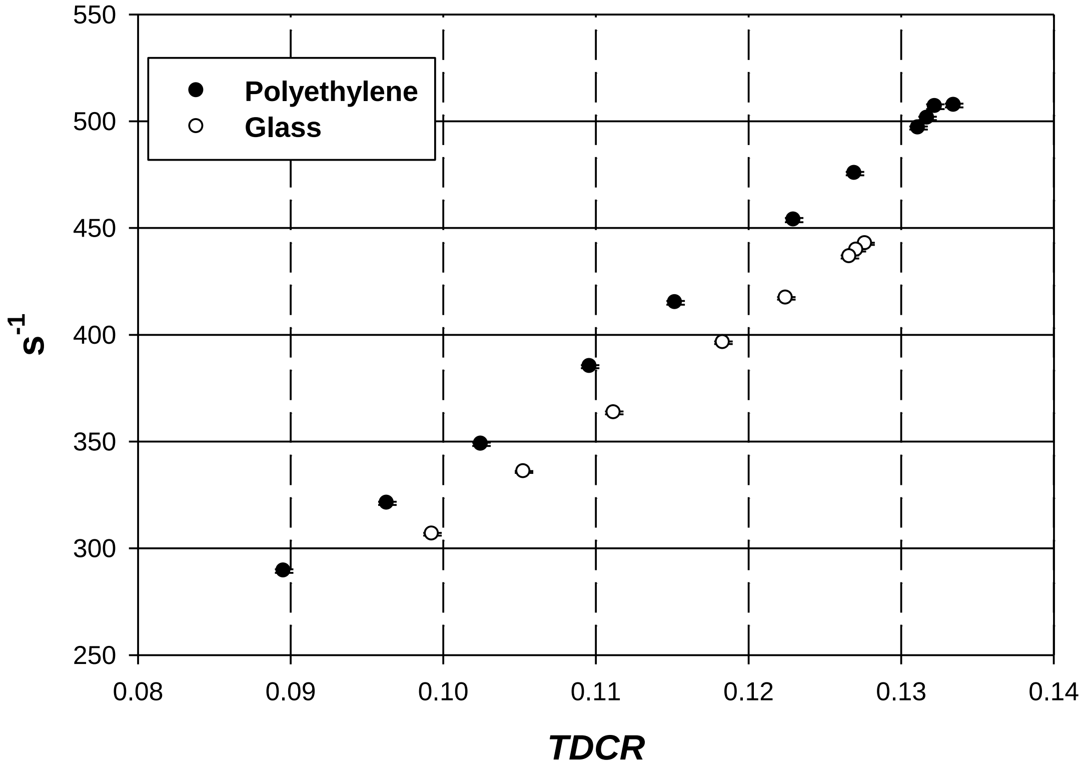
10
15
20
25
30
35
40
45
Resolving time / 8 ns
● Hionic Fluor
○ Ultima Gold

Figure 3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○ Glass
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500
450
400
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300
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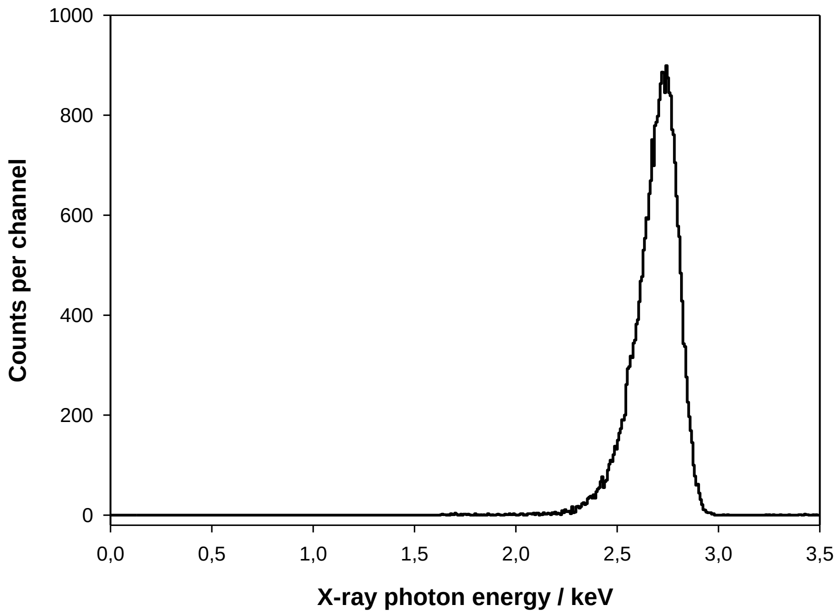
0.13
0.14
TDCR

Figure 4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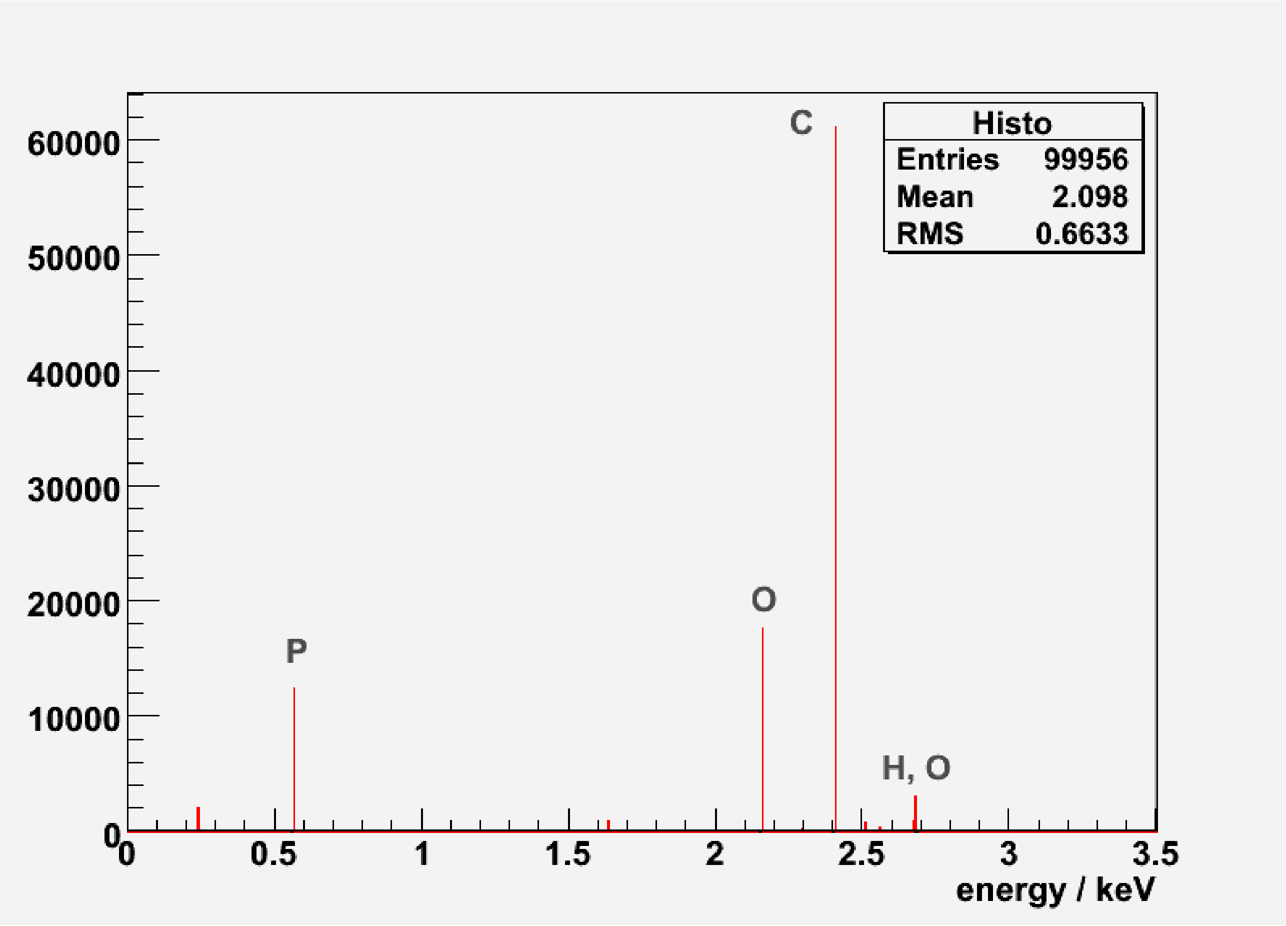
3,5
X-ray photon energy / keV

Figure 5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Mean 2.098
RMS 0.6633
C
O
P
H, O
0
10000
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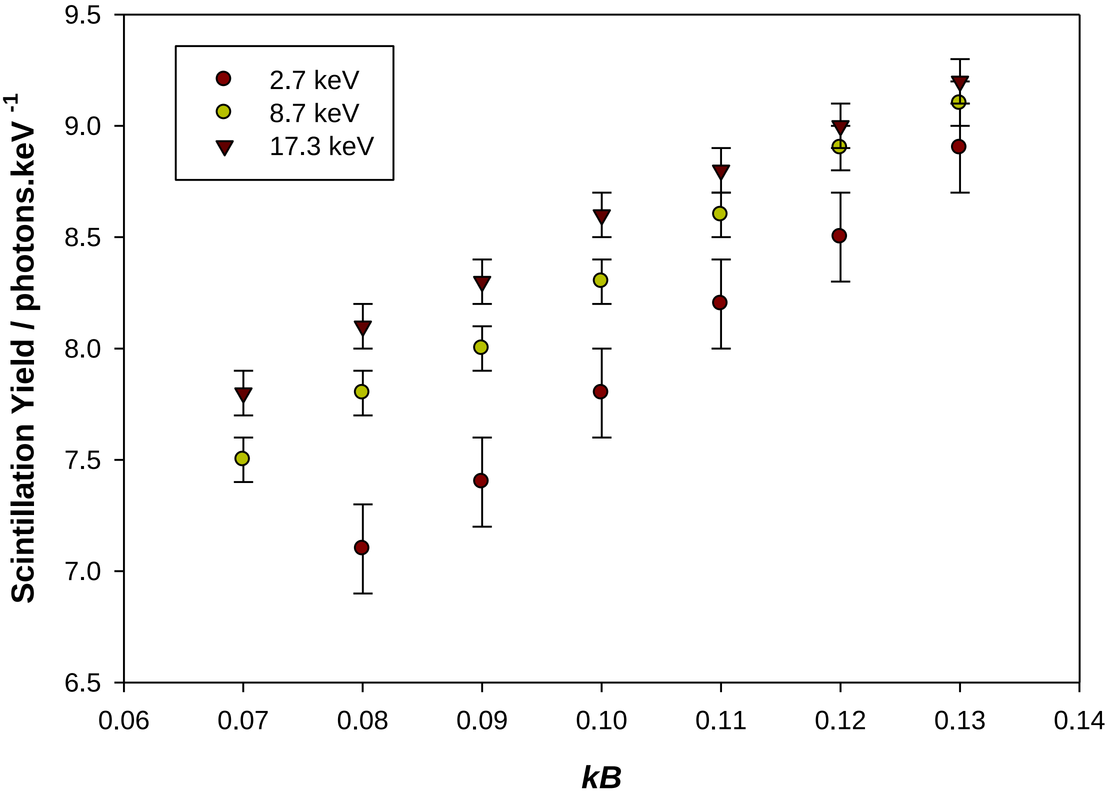
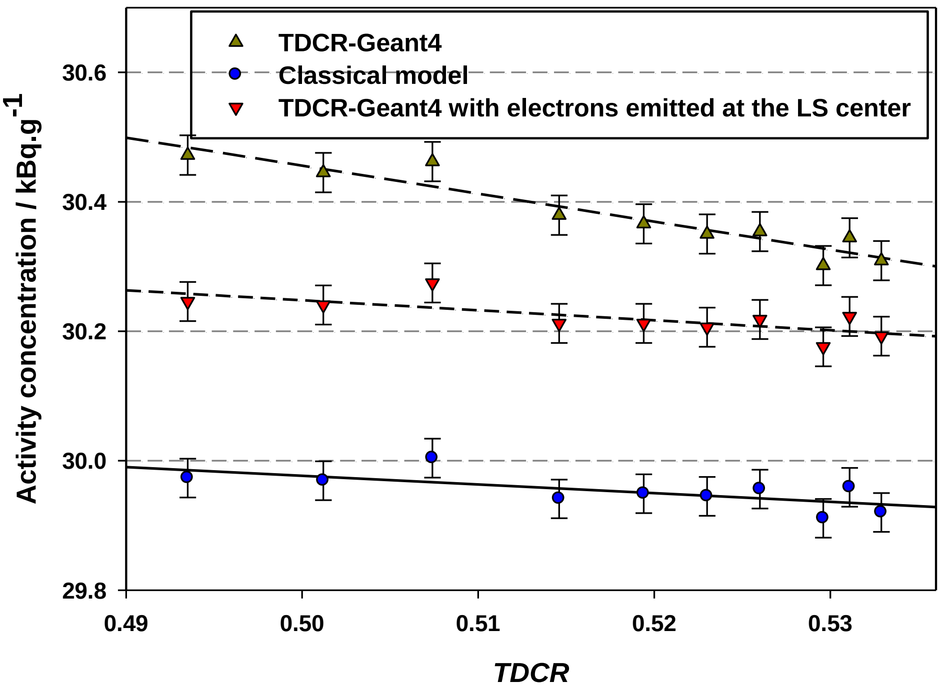
30000
40000
50000
60000
0
0.5
1
1.5
2
2.5
3
3.5
energy / keV

Figure 6



9.5
9.0
8.5
8.0
7.5
7.0
6.5
Scintillation Yield / photons.keV $^{-1}$
0.06
0.07
0.08
0.09
0.10
0.11
0.12
0.13
0.14
kB
2.7 keV
8.7 keV
17.3 keV

Figure 7


TDCR-Geant4
Classical model
TDCR-Geant4 with electrons emitted at the LS center
Activity concentration / kBq.g^{-1}
30.6
30.4
30.2
30.0
29.8
0.49
0.50
0.51
0.52
0.53
TDCR