Preliminary TDCR Measurements Using a X-ray Tube at Low-Energies (Lower Than 20 keV)

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SUMMARY

• About the TDCR method and the classical model

• Short description of the new TDCR-Geant4 model

• TDCR countings with miniature x-ray tube at low-energies (<20 keV)

• Estimation of the kB and Scintillation Yield values with the TDCR-Geant4 model

• Application to the case of 3H activity measurements using the TDCR-Geant4 model

• Discussion and perspectives
THE TDCR METHOD

- TDCR technique (Triple to Double Coincidence Ratio): primary activity measurement method based on liquid scintillation counting.
  - Scintillation vial (containing a LSC cocktail) placed inside a specific detection system using three PMTs.
  - A disintegration may be detected by 2 (or 3) PMTs: double (or triple) coincidence.
  - Activity classically determined in NMIs with a statistical model of light emission using the TDCR value as an indicator of the detection efficiency.
ABOUT THE STATISTICAL MODEL

• **Free parameter** statistical model built to obtain the relation: \( \varepsilon_D = f(TDCR) \).

• Based on an **analytical expression of the probability** \( P_E(x) \) to obtain at least one photoelectron in a PMT for a **mean number of photons** \( m(E) \) (Poisson distribution) following an energy deposition \( E \) in the LSC.

\[
P_E(x) = 1 - \exp(-m(E)v/3)
\]

• Decrease of light emission due to ionization quenching taken into account by the semi-empirical Birks relation:

\[
m(E) = A \int_0^E \frac{dE}{1 + kB \frac{dE}{dx}}
\]

  Scintillation yield  Birks factor

• \( kB \) factor: largest influence for low-energy radionuclides measurements (below 20 keV, e.g. \(^3\)H); in that case, the \( kB \) factor is used as an additional adjustment parameter for the activity determination.

• PMTs independence condition has to be fulfilled for the expression of coincidence probabilities (i.e. no correlation on coincidence counting)
New approach of the TDCR modeling: TDCR-Geant4 model includes a detailed description of the geometry and optical properties of the detector to obtain a more realistic description of the counting system.

Geant4 code enables the transport of ionizing particles and optical photons (scintillation and Cerenkov) from their creation to the production of photoelectrons in PMTs.

Optical modeling of TDCR-Geant4

<table>
<thead>
<tr>
<th>Detector parameter</th>
<th>Material</th>
<th>Optical parameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>PMT-window (Ø=52 mm)</td>
<td>Fused silica</td>
<td>Dispersive refractive index (1.47 at 400 nm and 1.64 at 160 nm)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Surface type: dielectric–dielectric, Skin: polished</td>
</tr>
<tr>
<td>PMT-photocathode (Ø=46 mm)</td>
<td>Bialkali (K₂CsSb)</td>
<td>Dispersive refractive index ~2.5 at 430 nm</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Surface type: dielectric–dielectric</td>
</tr>
<tr>
<td>Optical chamber</td>
<td>Teflon®</td>
<td>Surface type: dielectric–metal, lambertien-type reflectivity 95%</td>
</tr>
<tr>
<td>Vial (1 mm layer)</td>
<td>Borosilicate</td>
<td>Dispersive refractive index ~1.52 at 430 nm</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Surface type: dielectric–dielectric</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Skin: polished</td>
</tr>
<tr>
<td>LS cocktail (10 mL), including</td>
<td>Ultima Gold</td>
<td>Refractive index 1.5 with spectral response set at 430 nm (corresponding to the</td>
</tr>
<tr>
<td>meniscus zone</td>
<td></td>
<td>bis-MSB fluorophore</td>
</tr>
</tbody>
</table>
TDCR MODEL BASED ON THE MONTE CARLO GEANT4 CODE / 2

Description of the different steps of the TDCR-Geant4 model

1. Primary emissions randomly generated in LS volume.

2. Optical photons emitted along the electron path according to a Poisson distribution: mean value calculated using the Birks formula using a coupled Scintillation yield (A) and kB value.

3. Each emitted photon is followed to account for optical processes inside the optical chamber (refraction and reflection).

4. Photoelectrons generated in each PMT given by a binomial trial applied on photons refracted at the bialkali photocathode.

5. To simulate the PMT defocusing used to reduce the detection efficiency, counting in PMTs obtained using a binomial trial applied to each photoelectron (using a parameter corresponding to the probability to reach the first dynode).


- TDCR-Geant4 model: stochastic approach to calculate coincidences probabilities from each photon created inside the optical cavity following a primary emission in the LS volume.

- TDCR-Geant4 model first validated in the case of primary measurements using Cerenkov emission: $^90$Y, $^{11}$C, $^{32}$P (Because of the anisotropy of Cerenkov emission these measurements are considered as a validation of the geometrical and optical modeling).

- Applied for LSC counting to the standardization in Ultima Gold (UG) of $^{63}$Ni, $^{51}$Cr, $^{60}$Co.
COMPARISON BETWEEN THE TDCR-GEANT4 AND CLASSICAL MODELS AT LOW ENERGY

- Standardization of the low-energy emitters $^{51}$Cr (electron-capture emitter, mainly 4 to 6 keV)
  - Significant difference between the TDCR-Geant4 and classical models (~ 4%).
  - Coherent activity values obtained with the TDCR-Geant4 model and the $4\pi\beta-\gamma$ coincidence method.

- Investigations lead to the interpretation that this problem is the consequence of a geometric dependence between PMTs occurring for low-energy emitters (below 20 keV).
  - Correlations between PMTs are due to the influence of the position of light emission in the vial combined to refraction and reflection processes in the optical chamber.
  - Consequence: classical model calculation overestimates the detection efficiency.

- Correlations between PMTs are shown when comparing polyethylene (more diffusing) and glass vials ($^{51}$Cr standardization):
  - Better detection efficiency obtained.
  - Significant shift of the TDCR curve not taken into account by the « classical » statistical model.
TDCR COUNTINGS WITH MINIATURE X-RAY TUBE AT LOW-ENERGIES (<20 KEV)

- Interest of x-ray generator coupled to the TDCR counter:
  - Relation between the TDCR-Geant4 model and experiments without radionuclides
  - Geometric dependence between PMTs below 20 keV.
  - $kB$ and Scintillation Yield parameters of liquid scintillation cocktails.
Continuous spectra: filtered to be quasi-monoenergetic
Three energies have been chosen: 2.7, 8.7 and 17.3 keV (50 to 5000 emissions/sec)
For each energy, the real spectrum has been acquired with a Silicon Drift Detector
ESTIMATION OF THE $KB$ AND $A$ USING THE X-RAY GENERATOR / 1

- TDCR experimental values obtained with 8 mm collimator

<table>
<thead>
<tr>
<th>X-ray energy</th>
<th>TDCR</th>
</tr>
</thead>
<tbody>
<tr>
<td>2.7 keV</td>
<td>0.137 (2)</td>
</tr>
<tr>
<td>8.7 keV</td>
<td>0.646 (2)</td>
</tr>
<tr>
<td>17.3 keV</td>
<td>0.928 (2)</td>
</tr>
</tbody>
</table>

- Remark: collimation of the x-ray beam increases measured $TDCR$ values as expected with the TDCR-Geant4 model ($TDCR=0.128 (2)$ without the collimator).

- Methodology for $kB$ and $A$ estimation with the TDCR-Geant4 model
  - Calculations of $TDCR$ values for various $kB$ values (0.07 to 0.13) and Scintillation Yields (7 to 10 photons/keV).
  - $kB$ and $A$ obtained by minimizing the difference between the measured and simulated $TDCR$ values
• X-ray tube emissions are implemented as a 2D beam flux (theta = 15°) from the end-window (located to 40 mm from the surface of LS volume).

• Energies are randomly generated using spectra measured with Silicon Drift Detector for each energy investigated (2.7, 8.7 and 17.3 keV).

• Electronic emission due to atomic rearrangement following the x-ray interaction with the main LSC components (for UG: C, H, O, P) is modeled.

• For 2.7 keV x-rays, energy distribution of primary ejected electrons:
  
  ➢ mainly between 2 and 2.7 keV.
  
  ➢ 12% of emission at ~ 600 eV corresponding to interactions with phosphorous.
ESTIMATION OF THE $kB$ AND $A$ USING THE X-RAY GENERATOR / 3

- For each x-ray energy (2.7, 8.7 and 17.3 keV), $(kB, A)$ values are obtained when the simulated and measured $TDCR$ results are coherent.
- Usual $kB$ factors comprised between 0.07 and 0.13 used for simulation.

Based on the hypothesis that the $kB$ factor is constant with energies, coherent $(kB, A)$ values for the 3 excitation energies are obtained for $kB = 0.13$; further simulations are in progress for higher $kB$ values.

$(kB, A) = (0.1, 8.3(1))$ at 8.7 keV: coherent with results obtained with $^{54}\text{Mn}$ (mainly between 4.5 to 6 keV) using another procedure based on detection efficiencies measured with $4\pi(\text{LS})\beta-\gamma$ coincidence method (0.1, 8.2 (3)).
APPLICATION OF THE TDCR-GEANT4 MODEL IN THE CASE OF $^3$H MEASUREMENTS: EXPERIMENTAL RESULTS

- $^3$H: $\beta$-emitter with $E_{\text{max}} \sim 18.6$ keV
- TDCR measurements carried out in Ultima Gold
- Detection efficiency decreasing implemented by defocusing PMTs

- As for $^{51}$Cr measurements, a shift of the evolution of double-coincidence counting rates with TDCR values is observed when using diffusing vial wall
  
  - $^{51}$Cr (electron capture; discrete emission spectrum): **upward** shift
  - $^3$H (continuous spectrum): **downward** shift
Activity concentration as a function of $T_{DCR}$

Comments:

✓ Activity concentration as a function of $T_{DCR}$: the observed trend not expected.

✓ Unlike in the classical model, changing the $kB$ value has no influence on the slope.

✓ When the TDCR-Geant4 model was applied using Cerenkov counting (e.g. $^{11}$C and $^{90}$Y):
  - No systematic trends were observed.
  - Calculation of Cerenkov light emission based on electromagnetic processes.

✓ In the case of $^{63}$Ni and $^{51}$Cr standardization with TDCR-Geant4 (LS), no slope was also observed.

FURTHER INVESTIGATIONS ARE REQUIRED TO UNDERSTAND THIS BEHAVIOUR...

- For instance, modification of Birks formula at low energies
DISCUSSION AND PROSPECTS

- First results obtained with a x-ray generator coupled to the TDCR counter are promising.

- Experimental results obtained at low-energy depositions (< 20 keV) with the new set-up can be linked to the TDCR-Geant4 model:
  - The influence of measurement conditions on TDCR values has been investigated, for instance:
    - Comparison between glass and plastic vials.
    - The increase of the TDCR value when the x-ray beam is collimated as predicted by the TDCR-Geant4 model.
  - A methodology to estimate the $k_B$ value for the TDCR method without the use a radionuclide has been described.

- Application of the TDCR-Geant4 model in the case $^3$H measurements
  - Further investigations are in progress to understand the slope observed on the TDCR curve.

- Other investigations using the TDCR-Geant4 model:
  - Scintillation cocktails: Hionic Fluor for $^3$H measurements
  - Low-energy emitters: $^{55}$Fe (electron capture), $^{241}$Pu (continuous beta spectrum) $E_{\text{max}}$ 20.8 keV
Thank you