Adaptation of PTB’s analytical modelling for TDCR-Cherenkov activity measurements at LNE-LNHB

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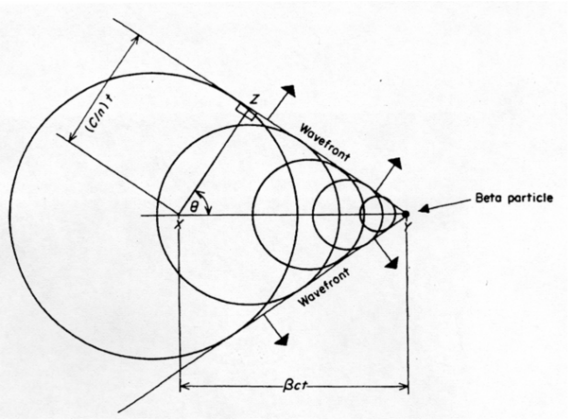
TDCR method (Triple to Double Coincidence Ratio) based on Cherenkov counting

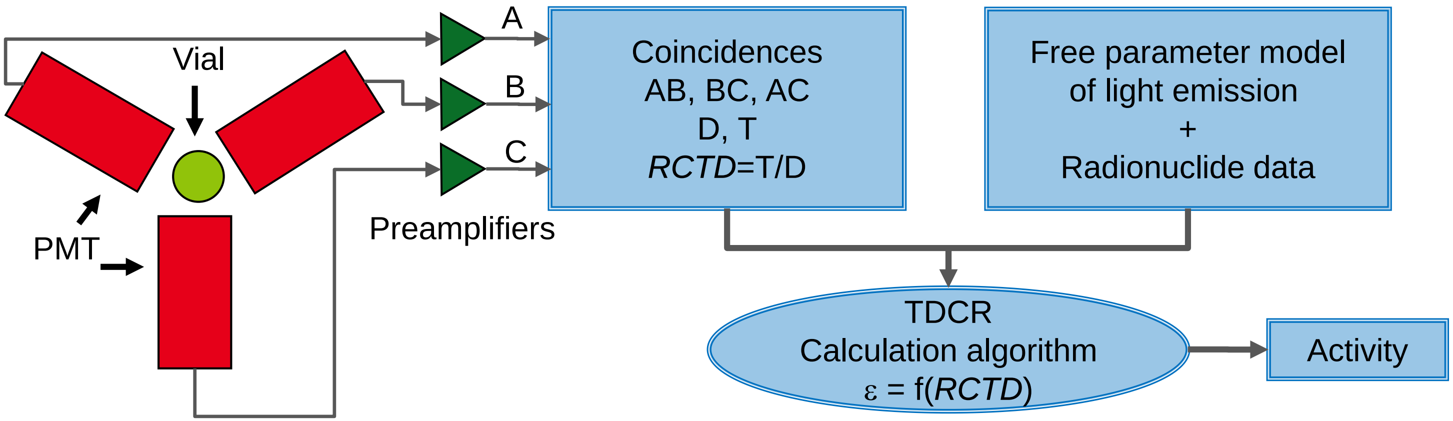
- Cherenkov radiation: discovery by Cerenkov (1937). *Phys. Rev. 52, 378.*
- Theoretical interpretation by Frank and Tamm (1937) *J. Phys. 1, 439–454.*
  - ✓ Electromagnetic shockwave resulting from a charged particle moving in a material faster than the velocity of light in that medium
  - ✓ In radionuclide metrology only electron or positron can produce Cherenkov light
  - ✓ Photons emitted as a cone with spanning angle θ: $\cos\theta = \frac{1}{n\beta}$
  - ✓ Condition of Cherenkov emission: $n\beta \geq 1 \; where \; \beta = {}^{v}/_{c}$



- Main physical proprieties:
  - ✓ Threshold effect (for electron in water: n ~ 1.33; E_{th} ~ 260 keV)
  - ✓ Directional character (not isotropic emission)
  - ✓ Large spectral bandwidth (comprised between UV and visible wavelengths)

TDCR method (Triple to Double Coincidence Ratio) based on Cherenkov counting

- Applied for β-emitting radionuclides, use existing LS counters:
  - ✓ Direct measurements with aqueous solutions (easy source preparation)
  - ✓ Drawback: counting efficiencies lower than LS counting (due to the threshold effect)
  - ✓ Natural discrimination for low-energy β particles and for alpha particles



- As for LS, the free parameter is adjusted for the calculation of the detection of double coincidences using experimental *TDCR* values
  - ✓ The model has to take into account the physical properties of Cherenkov emission

Towards model of Cherenkov counting for radionuclides standardization:

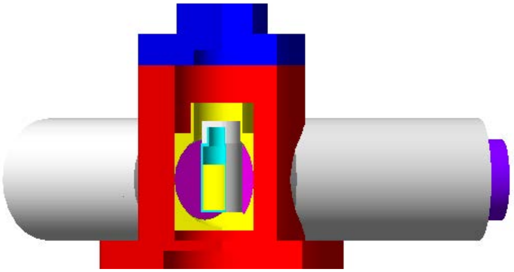
- A free parameter model of light emission (to compute Cherenkov counting efficiencies) first proposed by Grau Carles and Grau Malonda (2006)
  - ✓ Developed for Cherenkov counting standardization with detection systems using two PMTs

- Kossert (2010) and Kossert et *al.* (2014): extends this model for standardizations with TDCR detection systems
  - ✓ Based on adaptation the statistical model usually carries our for LS counting
  - ✓ New empirical formula to account for anisotropy of Cherenkov emission

- Bobin al.(2010) and Thiam et al.(2011) describe a stochastic approach based on Monte Carlo simulation using Geant4 code

Monte Carlo Model using Geant4 code: Comprehensive modelling of detection setup

- Simulates interactions of ionizing radiation and propagation of Cherenkov photons from their creation to the production of photoelectrons in PMTs
  - ✓ Geometry modelling, EM physics
  - ✓ Optical processes: refraction, reflection, transmission and absorption
  - ✓ Model works for measurements with glass vials

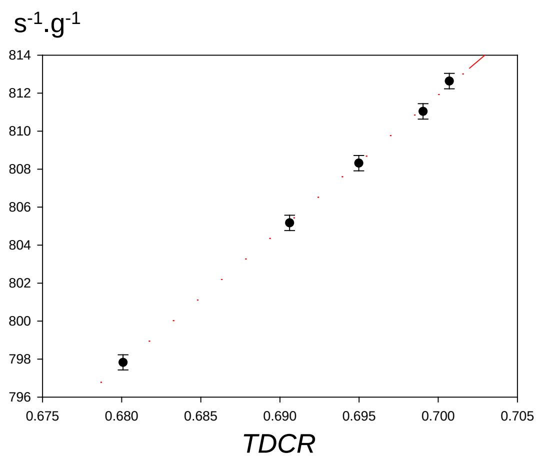


Geometry model with Geant4

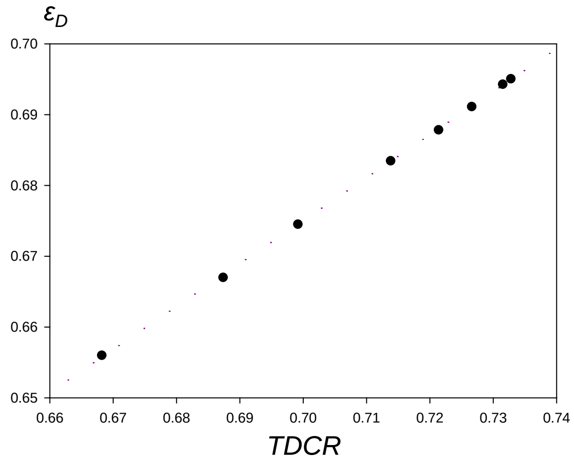
Detector parameter	Material	Optical parameter
PMT-window (Ø = 52 mm)	Fused silica	Dispersive refractive index (DRI) ~ 1.47 at 400 nm and 1.64 at 160 nm Surface type (ST): dielectric-dielectric. Skin: polished
PMT-photocathode (Ø = 46 mm)	Bi-alkali (K_2CsSb)	DRI ~ 2.5 at 430 nm, Harmer et al. (2006). ST: dielectric-dielectric
Optical chamber	Teflon®	ST: dielectric-metal, Lambertien-type reflectivity 95%
Vial (1 mm layer)	Borosilicate	DRI ~ 1.52 at 430 nm. ST: dielectric-dielectric, Skin: polished
Aqueous solution (15 mL)	Water	DRI ~ 1.33 at 2600 nm

Monte Carlo Model using Geant4 code

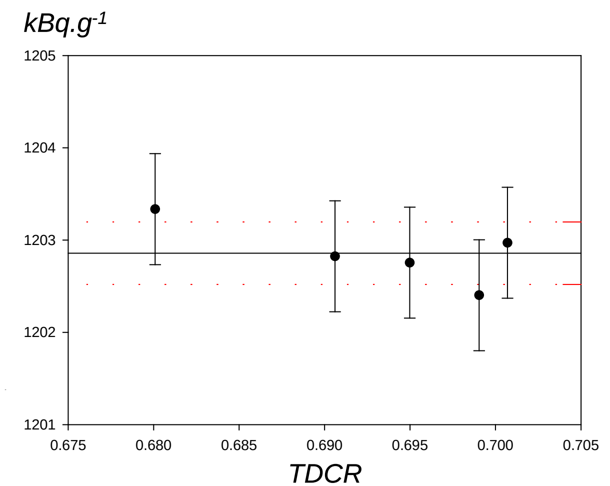
- Detection efficiency variation implemented using the PMT-defocusing technique
- Activity calculated with TDCR-Geant4 model validated for an aqueous solution of ^{90}Y



Double coincidence rate according to *TDCR*



ε_D=f(*TDCR*) given by Geant4



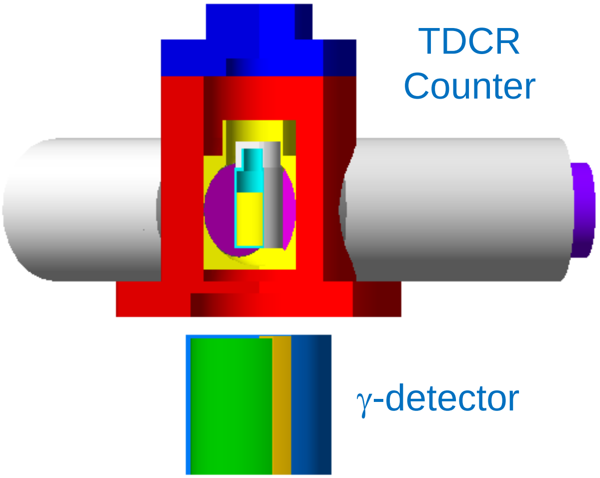
Activity concentration according to *TDCR*
No significant trend

More details on: Appl. Radiat. Isot. 68 (2010) 2366–2371.

TDCR-Cherenkov model

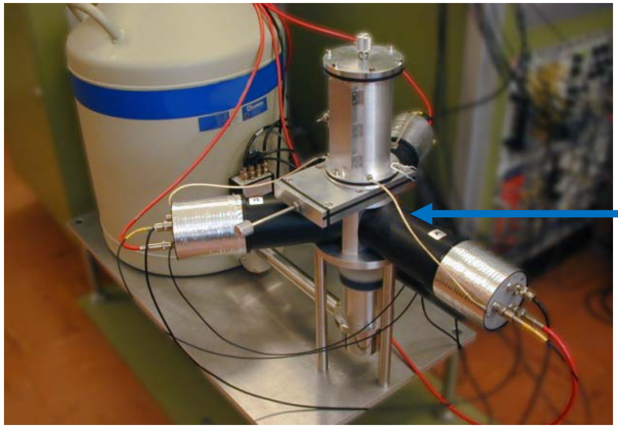
Monte Carlo Model using Geant4 code

- Well validated for several radionuclides
  - ✓ ^{90}Y, ^{32}P, ^{11}C, ^{68}Ga/^{68}Ge. Appl. Radiat. Isot. 68 (2010) 2366–2371
- Applicable also for liquid scintillation counting
  - ✓ ^{3}H, ^{63}Ni… Appl. Radiat. Isot. 70 (2012) 2195–2199
- $4\pi\beta$(LS)γ coincidences method
  - ✓ ^{56}Fe, ^{54}Mn, ^{14}C. Appl. Radiat. Isot. 109 (2016) 319–324
- Useful to study the contribution of several parameters and physical effects in counting
  - ✓ Volume effect of the source and vial
  - ✓ Contribution of bremsstrahlung, 511 γ-annihilation
  - ✓ Variation of optical parameters
- C++ programing, long computing time

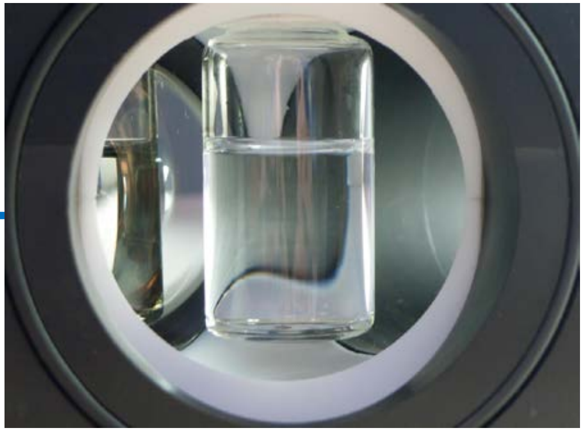


Adaptation of PTB's analytical model at LNHB

- Objective: test an extension of PTB model for TDCR-Cherenkov measurements by considering physical properties of the detection setup in use at LNHB
- Calculation implemented using MATLAB computing environment
- The three-PMTs counter used for measurements: equipped with X2020Q PMTs (Photonis)
  - ✓ Fused silica window, large spectral sensitivity (160 to 600 nm)
  - ✓ Quantum efficiency ~ 24% (300 to 400 nm)



3-PMT TDCR at LNHB



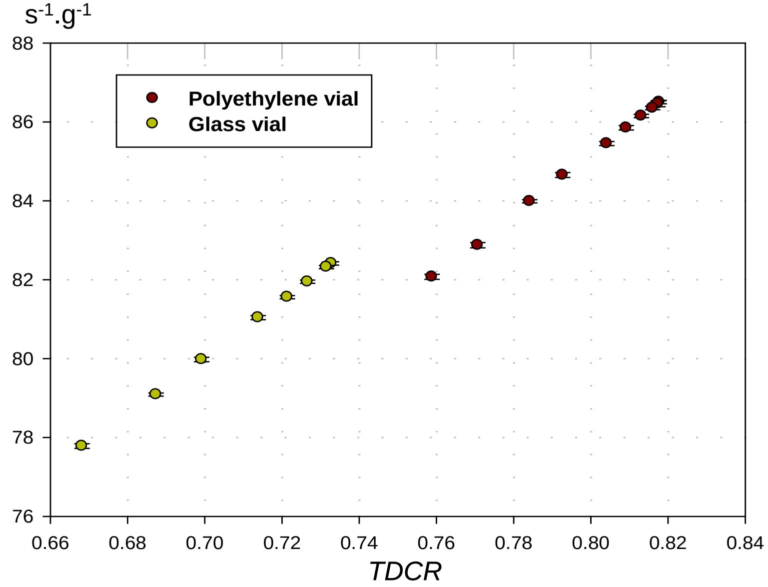
Spherical optical chamber made of Teflon® (~ 95 % Lambertian reflexion)

Adaptation of PTB's analytical model at LNHB

- Measurements carried out with plastic vials to attenuate the geometry dependence of coincidence counting

Comparison between glass-vials and plastic-vials for measurements of ^{90}Y

➔ Significant shift observed



- Because of geometrical effect due to their diffusive wall, reflection and refraction process taking place at the wall/air boundary are modified

➔ Better detection efficiencies are obtained with usual LS plastics vials

Description of the analytical modelling

Using Frank and Tamm (1937) theory; the number of Cherenkov photons dk emitted by an e^- or e^+ along a path dx for an energy E and λ is given by:

$$\frac{dk}{dx d\lambda} = 2\pi\alpha_{FS}\frac{1}{\lambda^2}\left(1 - \frac{1}{\beta^2 n^2(\lambda)}\right)$$

α_{FS}: fine structure constant

$n(\lambda)$: refractive-index of transparent medium

In term of energy variation (dE/dx from ESTAR database) the equation became:

$$\frac{dk(E,\lambda)}{dE d\lambda} = 2\pi\alpha_{FS}\frac{1}{\lambda^2}\left(1 - \frac{1}{\beta^2 n^2(\lambda)}\right)\frac{1}{\rho\, dE/dx}$$

The mean number of photoelectrons distributed in PMTs is obtained by the integration of the equation between E_c and the energy released in the aqueous solution.

- ✓ Considering PMT-spectral response
- ✓ Taking into account the variation of the Cherenkov threshold $\mathrm{E_c}$ with decreasing photon wavelengths

NOTE: Optical properties of vials are not taken into account

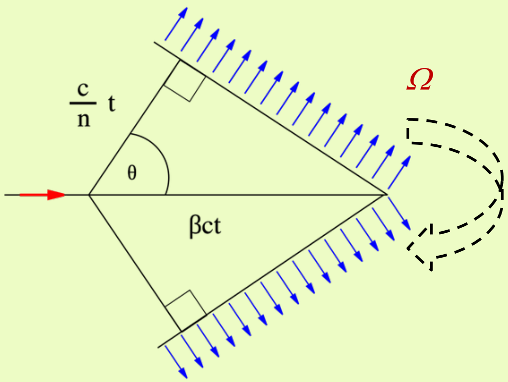
Description of the analytical modelling

The empirical probabilities R_1, R_2 and R_3 to have at least one count obtained as for LS counting i.e. we can apply an adjusting free parameter (q) (based on a Poisson distribution assumption)

$$R_1(E_{\text{el}}) = 1 - e^{-q\alpha_1 k(E_{\text{el}})} \qquad \alpha_1 = \alpha(E_{\text{el}})$$

$$R_2(E_{\text{el}}) = 1 - e^{-q\alpha_2 k(E_{\text{el}})} \qquad \alpha_2 = \frac{3}{2}\left(1 - \alpha(E_{\text{el}})\right)\alpha(E_{\text{el}})$$

$$R_3(E_{\text{el}}) = 1 - e^{-q\alpha_3 k(E_{\text{el}})} \qquad \alpha_3 = 1 - \alpha_1 - \alpha_2$$



Anisotropy described with $\alpha(E_{\text{el}})$, expressed as a function of Ω

PTB	LNHB
$\alpha(E_{\text{el}}) = x\left(\frac{2}{3} - \frac{1}{3}\frac{\Omega}{4\pi}\right) = x\left(\frac{1}{2} + \frac{1}{6}\cos\theta\right)$	$\Omega = 2\pi\,(1+\sin\theta)$ $\alpha(E_{\text{el}}) = x\left(\frac{2}{3} - \frac{1}{3}\frac{\Omega}{4\pi}\right) = x\left(\frac{1}{2} - \frac{1}{6}\sin\theta\right)$
if $\theta \Rightarrow 0, \Omega = 0$	if $\theta \Rightarrow 0, \Omega = 2\pi$

➔ Formula slightly different so as to get an equi-probable distribution of photons between PMTs by setting the free parameter $x = 1$

Description of the analytical modelling

The calculation of detection efficiencies of triple and double coincidences ε_T and ε_D

➔ Implemented as for LS measurements using the probabilities R_1, R_2 and R_3 and the probability density function $S(E_{\text{el}})$ related to the energy distribution of particle

$$\varepsilon_T = \int_{E_c}^{E_{\text{max}}} S(E_{\text{el}}) R_1 R_2 R_3 \, dE_{\text{el}}$$

$$\varepsilon_D = \int_{E_c}^{E_{\text{max}}} S(E_{\text{el}}) (R_1 R_2 + R_2 R_3 + R_1 R_3 - 2. R_1 R_2 R_3)$$

➔ The detection efficiency determined by adjusting the free parameter to match the measured *TDCR* values

Related publications:

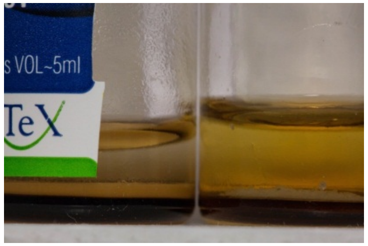
Kossert, Appl. Radiat. Isot. 68 (2010) 1116–1120.

Kossert, Grau Carles, Nähle, Appl. Radiat. Isot. 86 (2014) 7–12.

Application of TDCR-Cherenkov modelling

Activity measurements of solution of dissolved ^{90}Y-labelled microspheres

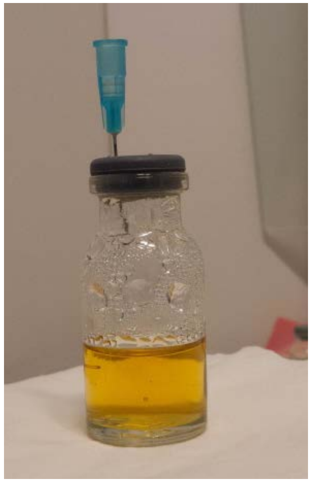
- Standardization of ^{90}Y-labeled microspheres (SIR-Spheres, Sirtex)
  - ✓ Medical device used in Selective Internal Radiation Therapy (SIRT)
  - ✓ Biocompatible microspheres (20 - 60 μm) containing ^{90}Y
  - ✓ high-energy β^- ; main branch (99.98%); Emax ~ 2.28 MeV, Emean ~ 927 keV



- Measurements carried out after dissolution of microspheres
  - ✓ Avoid problems of non-homogeneity
  - ✓ Dissolved solution subsequently diluted to reduce colour quenching

NOTE: More details in Lourenço et al. (2015) Appl. Radiat. Isot. 97, 170-176.

- Sources preparation with plastic vials
  - ✓ 15 mL of carrier solution (25 mg/g of Y in 0.04 M HCl)
  - ✓ 9 mg of dissolved and diluted solution of ^{90}Y-microspheres
- Results compared to classical TDCR LS counting with glass vials
  - ✓ 10 mL of Hionic-Fluor +9 mg of dissolved solution of ^{90}Y-microspheres



After dissolution (no residues)

Application of TDCR-Cherenkov modelling

Activity measurements of solution of dissolved ^{90}Y-labelled resign microspheres

- Three sources measured in standard plastic vials (with out coated Teflon in wall)
  - ✓ The TDCR values: comprised between 0.8 - 0.815, corresponding detection efficiencies : ~ 72%
- Two sources measured using Teflon coated plastic vials
  - ✓ The TDCR values: ~ 0.8, corresponding detection efficiencies : ~ 71.5
  - ✓ The activity concentration with TDCR-Cherenkov consistent with classical TDCR LS

  Relative difference: 0.2% with standard plastic vials; 0.26% with Teflon coated plastic vials
- Uncertainty budget of ^{90}Y measurements using the analytical modelling

Uncertainty component	Comments	*u* (%)
Measurement variability	Standard deviation of the measured sources	0.1
Weighing	Gravimetric measurements (pycnometer method)	0.1
Live time	1-MHz frequency clock used for the live-time	0.01
Decay correction	Half-life of ^{90}Y: 2.6684 (13) d	0.05
Analytical modelling	Conservative estimation	0.5
Background		0.05
Relative combined standard uncertainty		**0.52**

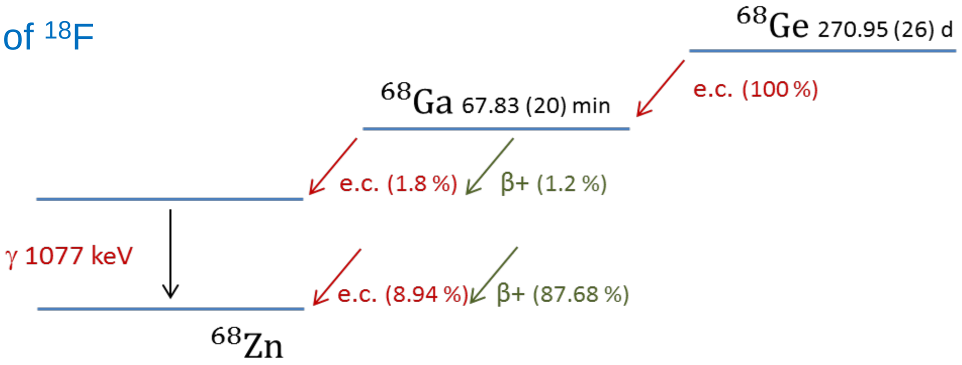
Activity measurements of ^{89}Sr

- Suited radionuclide for Cherenkov measurements
  - ✓ High-energy β^-; main branch (~ 99.99%) with E_{max} ~ 1495 keV, E_{mean} ~ 585 keV
- Sources preparation
  - ✓ Four sources measured in standard plastic vials
  - ✓ 15 mL of carrier solution (10 mg/g of Sr in HCl 0.1 M) + 30 to 100 mg of ^{89}Sr solution
- Results:
  - ✓ Maximum *TDCR* values: ~ 0.665
  - ✓ Corresponding detection efficiencies computed with the analytical modelling: ~ 53%
  - ✓ Activity concentration given by TDCR-Cherenkov: 0.26% lower than the result with TDCR LS
  - ✓ Main uncertainty component due to the analytical modelling: 0.5%

Application of TDCR-Cherenkov modelling

Activity measurements of ^{68}Ga in a solution of $^{68}Ge/^{68}Ga$ in equilibrium

- The activity measurements carried out in the framework of a BIPM international comparison using a radioactive solution prepared at NIST
- $^{68}Ge/^{68}Ga$: favorable to standardization by Cherenkov measurements via ^{68}Ga
  - ✓ Mainly decaying by β^+ emissions ~ 88.9%; E_{max} ~ 1899 keV; $T_{1/2}$ ~ 67.83 min
  - ✓ Take advantage of the Cherenkov threshold to avoid the contribution of EC emissions of ^{68}Ge
  - ✓ Suitable for PET imaging as a surrogate of ^{18}F
  - ➔ Simplified decay scheme of ^{68}Ge - ^{68}Ga



- Sources preparation
  - ✓ Six sources in Teflon coated plastic vials
  - ✓ 15 mL of carrier solution (65 mg/g of Ge^{4+} and Ga^{3+} in 0.5 M HCl) + 10 mg of $^{68}Ge/^{68}Ga$ aliquot

Application of TDCR-Cherenkov modelling

Activity measurements of ^{68}Ga in a solution of $^{68}Ge/^{68}Ga$ in equilibrium

- Results:

  - ✓ Efficiency calculation with analytical model take into account the two β^+ spectra and related branching ratios ($\beta^+_{0,0}$, E_{max} ~ 1899 keV; ~ 87.7 %; $\beta^+_{0,1}$, E_{max} ~ 821.7 keV, 1.2 %)
  - ✓ The contribution of 511 keV annihilation photons also considered using the resulting energy spectrum in the aqueous solution (obtained by Monte Carlo simulation)

  - ✓ Maximum *TDCR* value: 0.746
  - ✓ Corresponding detection efficiency computed with the analytical modelling: 73.4%
  - ✓ Contribution of 511 keV γ-photon interactions: ~ 0.2%

➔ TDCR-Cherenkov compared with $4\pi(LS)\beta-\gamma$ anticoincidence measurements

  - ✓ Both results are in good agreement with 0.16% relative difference

Discussion and conclusion

- First developed at PTB for TDCR-Cherenkov measurements, the analytical model has been well adapted to operate with a TDCR counter in use at LNHB
  - ✓ Implementation inspired from the classical statistical model used for LS counting

- The modelling is based on several approximations
  - ✓ Optical properties of the vials are not taken into account (optical transmittance, refractive indices)
  - ✓ The anisotropy of emission is considered by means of empirical expressions and by using an additional free parameter
  - ✓ Depending on $E(e^-/e^+)$, x and q, the mean number of photoelectrons (therefore probability to obtain a count in PMTs) is based on Poisson-distribution
  - ✓ The contribution of different physical effects may compensated by additional diffusing-effect when using plastics vials

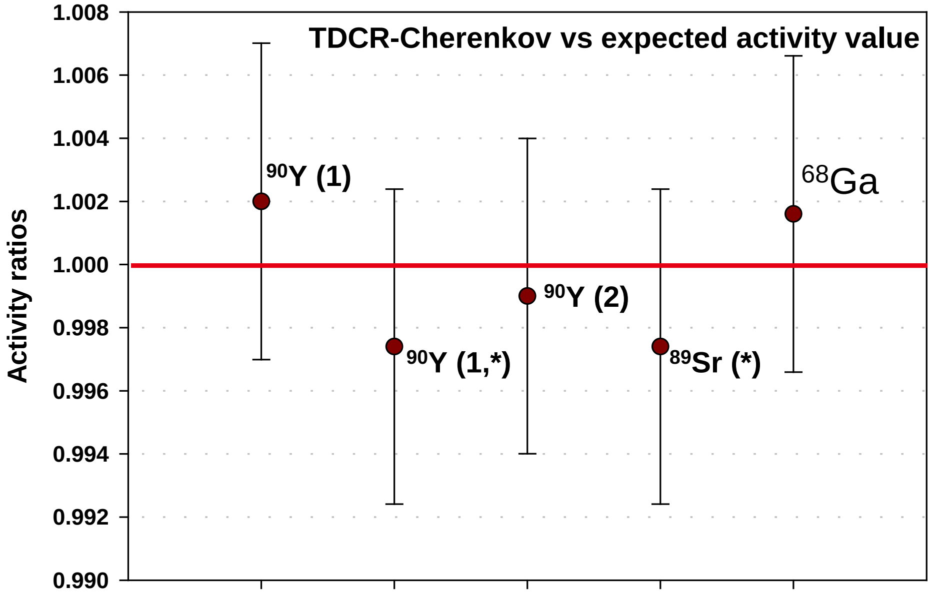
- Despite approximations, Cherenkov measurements using the analytical model give consistent results with a conservative model-uncertainty of 0.5%

➔ The model successfully tested at LNHB for three standardizations of three radiopharmaceuticals:

^{90}Y, ^{89}Sr and ^{68}Ge

➔ Reliable activity measurements already reported by Kossert et al. (2014):

^{32}P, ^{89}Sr, ^{90}Y, ^{204}Tl, ^{106}Rh…



(1) dissolved and diluted solution of ^{90}Y microspheres
(2) aqueous solution of ^{90}Y
(*) plastic-vial without Teflon coating in inner wall

Thank you ….

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