Outline

- LS methods in radionuclide metrology
- Applications (some examples)
- Research related to LS method
- Current limitations and challenges
LS methods in radionuclide metrology
Radionuclide metrology

Main tasks:
- Determination of activities with high precision and high accuracy
- Provision of activity standards
- Determination of radionuclide decay data
- International comparisons
Radionuclide metrology

Links between activity determination and nuclear decay data:

Activity $A$ (Bq)

$$A = \frac{R}{\varepsilon \cdot P}$$

$$A = \frac{\ln 2 \cdot N}{T_{1/2}}$$

**Note:** Several methods to determine the activity require nuclear decay data as input data.
Activity determination using LSC

\[ a = \frac{R_{net}}{\varepsilon \cdot m} \]

\[ \varepsilon = ? \]
CIEMAT/NIST method (2 PMTs):

\[ \epsilon_2 = \int_{0}^{E_{\text{max}}} S(E) \left(1 - e^{-EQ(E)/2M}\right)^2 dE \]

The free parameter \( M \) is obtained from a measurement of a tracer radionuclide (e.g. \(^3\)H) under same experimental conditions. Usually external quenching indicators are used for the efficiency transfer.
Radionuclide metrology using LSC

K. Kossert
The TDCR method

Radionuclide metrology using LSC
The TDCR method

TDCR value:

\[
\frac{R_T}{R_D} = \frac{E_{\text{max}}}{E_{\text{max}}} \frac{\int S(E)(1 - e^{-\eta})^3 dE}{\int S(E)((3(1 - e^{-\eta})^2 - 2(1 - e^{-\eta})^3))dE}
\]

with

\[
\eta = \frac{\nu}{3} \int_0^E \frac{A dE}{1 + k B} \frac{dE}{dx}
\]

If counting statistics is good enough the ratio of measured counting rates must agree with the computed ratio

\[
\frac{R_T}{R_D} = \frac{T}{D} = \text{TDCR}
\]
The stochastic model

We consider a system with 2 PMTs. The counting efficiency is given by:

\[ \varepsilon_2 = \int_0^{E_{\text{max}}} S(E) \left[ 1 - \exp \left( \frac{-E \cdot Q(E)}{2\lambda} \right) \right]^2 dE \]

But: This only works if we have only one electron per decay.

Problem: Efficiency computation of complex β/γ or electron-capture radionuclides is not possible.
The stochastic model

The “CIEMAT/NIST” counting efficiency

\[ \varepsilon_2(\lambda) = \sum_{i=1}^{N} \left( 1 - \exp \left[ \frac{-\sum_{l=1}^{M_i} E_{il} Q(E_{il})}{2\lambda} \right] \right)^2 / N \]

- \( N \) total number of simulated decay events.
- \( M_i \) number of electrons generated in the decay \( i \).
- \( E_{il} \) energy of the electron \( l \) ejected in the decay \( i \).
The stochastic model

The TDCR counting efficiency

\[
\varepsilon_T(\lambda) = \sum_{i=1}^{N} \left\{ 1 - \exp \left[ -\frac{\sum_{l=1}^{M_i} E_{il} Q(E_{il})}{3\lambda} \right] \right\} / N
\]

\[
\varepsilon_D(\lambda) = \sum_{i=1}^{N} \left\{ 3 \left[ 1 - \exp \left[ -\frac{\sum_{l=1}^{M_i} E_{il} Q(E_{il})}{3\lambda} \right] \right] \right\}^2 - 2 \left[ 1 - \exp \left[ -\frac{\sum_{l=1}^{M_i} E_{il} Q(E_{il})}{3\lambda} \right] \right] \right\} / N
\]

- $N$ total number of simulated decay events.
- $M_i$ number of electrons generated in the decay $i$.
- $E_{il}$ energy of the electron $l$ ejected in the decay $i$.

Details about stochastic model can be found here:
The stochastic model

Extension of the model:

- The model was extended to make computations of beta and EC transitions possible even when they are in coincidence with up to 7 γ/IC transitions.

Advantage: With the current model it is possible to apply the CIEMAT/NIST efficiency tracing and the TDCR method with the same assumptions and input data (nuclear data, dE/dx, etc.).
Radionuclide metrology using LSC

4πβ-γ coincidence counting

\[ N_\beta = A \cdot \varepsilon_\beta \]

\[ N_\gamma = A \cdot \varepsilon_\gamma \]

\[ N_c = A \cdot \varepsilon_\beta \cdot \varepsilon_\gamma \]

\[ A = \frac{N_\beta \cdot N_\gamma}{N_c} \]
4πβ-γ coincidence counting

Efficiency variation and extrapolation

- The β counter can also detect events due to γ interaction
- γ transitions can lead to the ejection of electrons due to internal conversion (IC)

\[ N_\beta = A \left( \epsilon_\beta + (1 - \epsilon_\beta) \cdot \left( \frac{\alpha}{1 + \alpha} \epsilon_{ce} + \frac{1}{1 + \alpha} \epsilon_{\beta\gamma} \right) \right) \]

\[ N_\gamma = A \cdot \frac{1}{1 + \alpha} \cdot \epsilon_\gamma \]

\[ \frac{N_c}{N_\gamma} = \epsilon_\beta \]

\[ \frac{N_\beta \cdot N_\gamma}{N_c} = A \cdot \left[ 1 + \frac{1 - \epsilon_\beta}{\epsilon_\beta} \cdot \left( \frac{\alpha}{1 + \alpha} \epsilon_{ce} + \frac{1}{1 + \alpha} \epsilon_{\beta\gamma} \right) \right] = A + k \cdot \frac{1 - \epsilon_\beta}{\epsilon_\beta} = A + k \cdot x \]

\[ \chi^2 / \text{ndf} \quad 37.06 / 41 \]
\[ A \quad 1052.98 \pm 0.59 \]
\[ k \quad 2.43 \pm 0.14 \]
**4πβ-γ coincidence counting**

4πβ-γ CC with LSC in the β channel

- **β-double coincidence**: \( D = AB \) or \( BC \) or \( AC \)
- **β-triple coincidence**: \( T = ABC \)
- **β-γ-coincidence**: \( DG = (AB \) or \( BC \) or \( AC)G \)
  - \( TG = ABCG \)

**Radionuclide metrology using LSC**
4πβ-γ coincidence counting

Radionuclide metrology using LSC
In some cases $4\pi\beta(LS)$-$\gamma$ coincidence counting can be replaced with $4\pi\beta(\check{C})$-$\gamma$ coincidence counting.

Example: $^{68}\text{Ge}/^{68}\text{Ga}$
The TDCR-Čerenkov method
The TDCR-Čerenkov method


Number of photons is computed by numerical integration

\[ k(E) = \int_{E_{\text{th}}}^{E} \frac{dk}{dx} \frac{1}{\rho dE/dX} dE \]

where

\[ \frac{dk}{dx} = 2\pi \alpha_{FS} \left( \frac{1}{\lambda_1} - \frac{1}{\lambda_2} \right) \left( 1 - \frac{1}{n^2 \beta^2} \right) \]

The electron stopping powers \( dE/dX \) are taken from the ESTAR database (NIST).
The TDCR-Čerenkov method

When assuming that the number of created photoelectrons follows the Poisson statistics, we can apply a free parameter model:

\[
\varepsilon_T = \int_{E_{\text{th}}}^{E_0} N(E) \left( 1 - e^{-qk(E)\alpha_1} \right) \left( 1 - e^{-qk(E)\alpha_2} \right) \left( 1 - e^{-qk(E)\alpha_3} \right) dE
\]

\[
\varepsilon_D = \int_{E_{\text{th}}}^{E_0} N(E) \left[ \left( 1 - e^{-qk(E)\alpha_1} \right) \left( 1 - e^{-qk(E)\alpha_2} \right) \left( 1 - e^{-qk(E)\alpha_3} \right) \right. \\
+ \left( 1 - e^{-qk(E)\alpha_1} \right) \left( 1 - e^{-qk(E)\alpha_3} \right) + \left( 1 - e^{-qk(E)\alpha_2} \right) \left( 1 - e^{-qk(E)\alpha_3} \right) \\
- 2 \left( 1 - e^{-qk(E)\alpha_1} \right) \left( 1 - e^{-qk(E)\alpha_2} \right) \left( 1 - e^{-qk(E)\alpha_3} \right) \left] dE \right.
\]
The TDCR-Čerenkov method


\[
\frac{dk}{dxd\lambda} = 2\pi z^2 \alpha_{FS} \left( \frac{1}{\lambda^2} \left( 1 - \frac{1}{\beta^2 n^2(\lambda)} \right) \right)
\]

- Wavelength-dependent refractive index is taken into account
- PMT asymmetry is taken into account
- Wavelength-dependent PMT response is taken into account

See also contribution #105 from Cheick Thiam et al. with alternative MC approach
Applications
Radionuclides

Radionuclides measured at PTB since 2002 using CIEMAT/NIST + … and CIEMAT/NIST + TDCR + …

# Measurement of long half-lives using LSC

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Details</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{10}\text{Be}$</td>
<td>Chmeleff et al., Determination of the $^{10}\text{Be}$ half-life by multicollector ICP-MS and liquid scintillation counting. Nucl. Instrum. Meth B, 268 (2009) 192</td>
</tr>
<tr>
<td>$^{40}\text{K}$</td>
<td>Kossert, Günther, LSC measurements of the half-life of $^{40}\text{K}$. Appl. Radiat. &amp; Isot. 60 (2004) 459</td>
</tr>
<tr>
<td>$^{151}\text{Sm}$</td>
<td>Bé et al., Determination of the $^{151}\text{Sm}$ half-life. Radiochimica Acta 103 (2015) 619-626</td>
</tr>
<tr>
<td>$^{176}\text{Lu}$</td>
<td>Kossert et al., Experimental half-life determination of $^{176}\text{Lu}$. Appl. Radiat. &amp; Isot. accepted (ICRM-LLRMT)</td>
</tr>
</tbody>
</table>

**In progress:** $^{129}\text{I}$, $^{40}\text{K}$ (improved attempt with ANU)
Complex decay series

\(^{228}\text{Th}^\): Kossert, Nähle: Activity determination of \(^{228}\text{Th}\) by means of liquid scintillation counting. LSC2010, Advances in Liquid Scintillation Spectrometry: Proceedings of the 2010 International Conference on Liquid Scintillation Spectrometry, Paris, France, 6-10 September 2010


\(^{227}\text{Ac}, \(^{223}\text{Ra}^\): Kossert et al.: Activity determination of \(^{227}\text{Ac}\) and \(^{223}\text{Ra}\) by means of liquid scintillation counting and determination of nuclear decay data. Applied Radiation and Isotopes 95 (2015) 143

Recently completed: \(^{227}\text{Th}\) (not in equilibrium with progeny!)

Several NMIs/DIs also measure \(^{210}\text{Pb}\), \(^{222}\text{Rn}\) and other isotopes by means of LS method
Radionuclide metrology using LSC

$^{227}$Ac decay series

- Complex efficiency calculations (several $\alpha$-, $\beta$- and $\beta/\gamma$-transitions)
- Study of potential $^{219}$Rn losses
- Corrections for dead-time losses ($^{215}$Po)
- Decay data were determined

LSC measurements

Counters:
- Wallac 1414
- TriCarb 2800
- TDCR (M27)
- TDCR (M29)
$^{223}$Ra LSC measurements

- 2 solutions (citrate), one of them was diluted with a PTB carrier solution
- PE vials and glass vials; efficiency variation by means of chemical quenching
- Long-term measurements to determine the half-life and potential $^{227}$Ac impurity
- Gamma-ray spectrometry for determination of photon-emitting impurities and determination of photon emission probabilities.
Efficiency curves for TDCR

227Ac

223Ra

K. Kossert

Radionuclide metrology using LSC
Efficiency curves for CN

$^{227}$Ac

$^{223}$Ra

Radionuclide metrology using LSC
### Uncertainties

<table>
<thead>
<tr>
<th>Component</th>
<th>(^{227}\text{Ac}) u((a)/a) in %</th>
<th>(^{223}\text{Ra}) u((a)/a) in %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>TDCR</td>
<td>CIEMAT/NIST</td>
</tr>
<tr>
<td>Standard deviation of the mean (^{227}\text{Ac}: 6) samples with 9 or</td>
<td>0.12</td>
<td>0.02</td>
</tr>
<tr>
<td>more repetitions per sample and counter; (^{223}\text{Ra}: 10) samples</td>
<td></td>
<td></td>
</tr>
<tr>
<td>or more repetitions per sample and counter</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Weighing</td>
<td>0.06</td>
<td>0.06</td>
</tr>
<tr>
<td>Dead time (including corrections for (^{219}\text{Rn},^{215}\text{Po})</td>
<td>0.03</td>
<td>0.10</td>
</tr>
<tr>
<td>and (^{211}\text{Po}))</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Background</td>
<td>0.03</td>
<td>0.05</td>
</tr>
<tr>
<td>Duration of measurement</td>
<td>0.01</td>
<td>0.01</td>
</tr>
<tr>
<td>Adsorption</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Radionuclide impurities</td>
<td>0.03</td>
<td>0.03</td>
</tr>
<tr>
<td>TDCR value and fit</td>
<td>0.18</td>
<td>n.a.</td>
</tr>
<tr>
<td>(^{3}\text{H}) activity standard and fit of efficiency curve</td>
<td>n.a.</td>
<td>0.05</td>
</tr>
<tr>
<td>Model</td>
<td>0.8</td>
<td>0.05</td>
</tr>
<tr>
<td>Decay data (e.g. endpoint energies, beta shape factor functions, emission</td>
<td>0.9</td>
<td>0.9</td>
</tr>
<tr>
<td>probabilities)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ionization quenching</td>
<td>0.03</td>
<td>0.01</td>
</tr>
<tr>
<td>Quenching indicator ((S(QP(E),tSIE)))</td>
<td>n.a.</td>
<td>0.03</td>
</tr>
<tr>
<td>(\alpha) counting efficiency</td>
<td>0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Decay correction</td>
<td>&lt; 0.01</td>
<td>&lt; 0.01</td>
</tr>
<tr>
<td>Potential (^{219}\text{Rn} loss</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>PMT asymmetry</td>
<td>0.02</td>
<td>0.02</td>
</tr>
<tr>
<td>Square root of the sum of quadratic components</td>
<td>1.24</td>
<td>0.93</td>
</tr>
</tbody>
</table>
Motivation:
Patients exhale significant amounts of actinon. Hence, there are radiation protection difficulties for health personal.

The longest-lived progeny is $^{211}\text{Pb}$, but the evaluated half-life in DDEP was based on only two measurements.
A $^{219}$Rn trap

Glass beaker

Large area source with dried drops of a $^{223}$Ra solution (approx. 2 MBq)

Petri dish filled with Mineral Oil scintillator

K. Kossert

Radionuclide metrology using LSC
An \(^{219}\text{Rn}\) trap

- TDCR values are stable when using PE-Vials
- Cherenkov sources were also measured

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Coincidence channel</th>
<th>(T_{1/2}) in min</th>
<th>Type of source</th>
<th>Pre-treatment of PE vials with a carrier solution</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>T</td>
<td>36.159</td>
<td>LS</td>
<td>No</td>
</tr>
<tr>
<td>1</td>
<td>D</td>
<td>36.151</td>
<td>LS</td>
<td>No</td>
</tr>
<tr>
<td>2</td>
<td>T</td>
<td>36.161</td>
<td>LS</td>
<td>No</td>
</tr>
<tr>
<td>2</td>
<td>D</td>
<td>36.164</td>
<td>LS</td>
<td>No</td>
</tr>
<tr>
<td>3</td>
<td>T</td>
<td>36.175</td>
<td>LS</td>
<td>No</td>
</tr>
<tr>
<td>3</td>
<td>D</td>
<td>36.174</td>
<td>LS</td>
<td>No</td>
</tr>
<tr>
<td>4</td>
<td>T</td>
<td>36.161</td>
<td>LS</td>
<td>Yes</td>
</tr>
<tr>
<td>4</td>
<td>D</td>
<td>36.157</td>
<td>LS</td>
<td>Yes</td>
</tr>
<tr>
<td>5</td>
<td>T</td>
<td>36.188</td>
<td>LS</td>
<td>Yes</td>
</tr>
<tr>
<td>5</td>
<td>D</td>
<td>36.187</td>
<td>LS</td>
<td>Yes</td>
</tr>
<tr>
<td>6</td>
<td>T</td>
<td>36.160</td>
<td>LS</td>
<td>Yes</td>
</tr>
<tr>
<td>6</td>
<td>D</td>
<td>36.159</td>
<td>LS</td>
<td>Yes</td>
</tr>
<tr>
<td>7</td>
<td>T</td>
<td>36.179</td>
<td>Cherenkov</td>
<td>Yes</td>
</tr>
<tr>
<td>7</td>
<td>D</td>
<td>36.170</td>
<td>Cherenkov</td>
<td>Yes</td>
</tr>
<tr>
<td>8</td>
<td>T</td>
<td>36.145</td>
<td>Cherenkov</td>
<td>Yes</td>
</tr>
<tr>
<td>8</td>
<td>D</td>
<td>36.156</td>
<td>Cherenkov</td>
<td>Yes</td>
</tr>
</tbody>
</table>
A $^{220}$Rn trap

- Similar experiment were carried out using $^{228}$Th sources
- In this case, $^{224}$Ra recoil atoms must be stopped, e.g. using a thin foil as a barrier.
- The half-life of $^{212}$Pb is important since $^{212}$Pb/$^{212}$Bi is of increasing interest in cancer therapy (e.g. breast cancer, ovarian cancer, melanoma)
A $^{220}$Rn trap

$^{212}$Pb half-life determination

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>Coincidence channel</th>
<th>$T_{1/2}$ in h</th>
<th>Pre-treatment of PE vials with a carrier solution</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>T</td>
<td>10.623</td>
<td>Yes</td>
</tr>
<tr>
<td>1</td>
<td>D</td>
<td>10.621</td>
<td>Yes</td>
</tr>
<tr>
<td>2</td>
<td>T</td>
<td>10.621</td>
<td>Yes</td>
</tr>
<tr>
<td>2</td>
<td>D</td>
<td>10.620</td>
<td>Yes</td>
</tr>
<tr>
<td>3</td>
<td>T</td>
<td>10.623</td>
<td>No</td>
</tr>
<tr>
<td>3</td>
<td>D</td>
<td>10.623</td>
<td>No</td>
</tr>
</tbody>
</table>
Radionuclide metrology using LSC

A $^{220}$Rn trap

<table>
<thead>
<tr>
<th>Reference</th>
<th>Half-life in h</th>
</tr>
</thead>
<tbody>
<tr>
<td>Buttlar (1952)</td>
<td>10.67(5)</td>
</tr>
<tr>
<td>Marin et al. (1953)</td>
<td>10.64(3)</td>
</tr>
<tr>
<td>Tobailem and Robert (1955)</td>
<td>10.643(12)</td>
</tr>
<tr>
<td>DDEP (2011), data evaluation</td>
<td>10.64(1)</td>
</tr>
<tr>
<td>Kossert (2017)</td>
<td>10.622(7)</td>
</tr>
</tbody>
</table>

$T_{1/2}$ in h

Do decay rates depend on the Earth-Sun distance?

Normalized $^{32}\text{Si}/^{36}\text{Cl}$ (BNL) Ratio With Earth-Sun Distance

from Jenkins et al., Astroparticle Physics 32 (2009) 42–46

Radionuclide metrology using LSC
Do decay rates depend on the Earth-Sun distance?

No evidence from LSC

Where are the oscillations?

Do decay rates depend on the Earth-Sun distance?

Also no evidence for $^{90}\text{Sr}/^{90}\text{Y}$

Where are the oscillations?

from Kossert and Nähle, Astroparticle Physics 69 (2015) 18-23
Do decay rates depend on the Earth-Sun distance?

Facts:

Authors (Fischbach, Jenkins, Sturrock et al.) mix detector response (counting rate) and decay rate (activity) and misinterpret data.

The efficiency can vary with time (in particular for gas detectors).

Observed variations vary from detector to detector and from institute to institute. Thus, the Sun-Earth distance can be excluded as common reason for variations.

Effects triggered by solar neutrinos are possible but too small to be significant.
No evidence for influence of Earth-Sun distance

Kossert, Nähle: Long-term measurements of $^{36}$Cl to investigate potential solar influence on the decay rate. Astroparticle Physics 55 (2014) 33


Kossert, Nähle: Disproof of solar influence on the decay rates of $^{90}$Sr/$^{90}$Y. Astroparticle Physics 69 (2015) 18


Pommé et al.: On decay constants and orbital distance to the Sun – Part I: alpha decay. Metrologia 54 (2017) 1

Pommé et al.: On decay constants and orbital distance to the Sun – Part II: beta minus decay. Metrologia 54 (2017) 19

Research related to LS methods
The importance of beta spectra

Facts
- Beta spectra are required for the TDCR method as well as for CIEMAT/NIST efficiency tracing.
- The results for the efficiency depend on the shape of beta spectra which are used as input data.

Problems
- Calculation methods in LS programs are simplified; effects as Screening and Atomic exchange are not taken into account.
- Calculation methods for certain forbidden transitions are inaccurate.
- Experimentally determined spectra with high precision are missing.
The importance of beta spectra

EMPIR-Project MetroBeta “Radionuclide beta spectra metrology”

Partner:

<table>
<thead>
<tr>
<th></th>
<th>Partner</th>
<th>Address</th>
<th>Country</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>CEA</td>
<td>Commissariat à l'énergie atomique et aux énergies alternatives</td>
<td>France</td>
</tr>
<tr>
<td>2</td>
<td>CMI</td>
<td>Cesky Metrologicky Institut Brno</td>
<td>Czech Republic</td>
</tr>
<tr>
<td>3</td>
<td>PTB</td>
<td>Physikalisch-Technische Bundesanstalt</td>
<td>Germany</td>
</tr>
<tr>
<td>4</td>
<td>Gonitec</td>
<td>Gonitec BV</td>
<td>Netherlands</td>
</tr>
<tr>
<td>5</td>
<td>UHEI</td>
<td>Ruprecht-Karls-Universitaet Heidelberg</td>
<td>Germany</td>
</tr>
<tr>
<td>6</td>
<td>UMCS</td>
<td>Uniwersytet Marii Curie-Sklodowskiej</td>
<td>Poland</td>
</tr>
<tr>
<td>7</td>
<td>CHUV</td>
<td>University Hospital of Lausanne</td>
<td>Switzerland</td>
</tr>
</tbody>
</table>

Website: [http://metrobeta-empir.eu/](http://metrobeta-empir.eu/)
Coordinator: Mark Kellett (LNHB)
## The importance of beta spectra

### Work packages:

<table>
<thead>
<tr>
<th>Work Package Title</th>
<th>Active Partners</th>
<th>Months</th>
</tr>
</thead>
<tbody>
<tr>
<td>WP1 Theoretical calculations of beta spectra</td>
<td>CEA; UMCS</td>
<td>21.5</td>
</tr>
<tr>
<td>WP2 High-resolution beta spectrometry based on Metallic Magnetic Calorimeters (MMCs)</td>
<td>PTB; CEA; UHEI</td>
<td>60.9</td>
</tr>
<tr>
<td>WP3 Measurements of beta spectra with other methods</td>
<td>CHUV; CMI; Gonitec</td>
<td>28.5</td>
</tr>
<tr>
<td>WP4 Comparison and validation of measurements</td>
<td>PTB; CEA; CHUV</td>
<td>16.0</td>
</tr>
<tr>
<td>WP5 Creating impact</td>
<td>CMI; all partners</td>
<td>11.5</td>
</tr>
<tr>
<td>WP6 Management and coordination</td>
<td>CEA; all partners</td>
<td>10.9</td>
</tr>
</tbody>
</table>

**Total months**: 149.3
MMC @ PTB

Principle of an MMCs (metallic magnetic calorimeter)

Measurement of the change of a magnetic field via SQUID (superconducting quantum interference device)

Working temperature: a few mK
Resolution: approx. < 3 eV bei 60 keV

Source f. both figures:
Fleischmann, Uni Heidelberg

Radionuclide metrology using LSC
Planned MMC system at PTB

- $^3\text{He}/^4\text{He}$ dilution refrigerator
- MMC technology from Uni. Heidelberg
- SQUID technology from PTB Berlin
- Expertise from LNHB
- Source preparation: electrolytic deposition etc.

Source: Bluefors
Radionuclides which shall be measured within MetroBeta (LNHB+PTB):

- $^{14}$C
- $^{151}$Sm
- $^{99}$Tc
- $^{36}$Cl

Source: Bluefors
MMC @ PTB

K. Kossert

Radionuclide metrology using LSC
Absorber preparation

Requirements:
- Full energy must be deposited within absorber (including secondary bremsstrahlung)
- Falsification due to chemical effects shall be avoided

Ideas:
- Galvanic deposition of radionuclide on an Au foil
- Cutting Au foil and placing between 2 Au plates
- Combination using diffusion welding

Range of electrons in gold
Validation and importance

Measurement of beta spectra using MMCs at LNHB


K. Kossert
Validation and importance

Example: $^{63}\text{Ni}$

From Kossert & Mougeot
Validation and importance

Example: $^{63}\text{Ni}$

From Kossert & Mougeot

$k_B = 0.0075 \text{ cm/MeV}$
Validation and importance

Example: $^{60}\text{Co}$

Kossert et al., ICRM 2017

$^{60}\text{Co}$

- $\beta^-$ 99.88%
- $\beta^-$ 0.12%
- 5.2711 (8) a
- 0.30 ps
- 1173.228 keV
- 99.85%
- 0.713 ps
- 1332.492 keV
- 99.98%
- Stable
- $^{60}\text{Ni}$

Advantage when analysing $^{60}\text{Co}$:
Possibility to compare with coincidence counting (no model dependence)
Validation and importance

Example: $^{60}$Co

When using the “classical“ spectrum, the results from LSC methods are too low by a few per mille !!!

When using the “new“ spectrum, the deviation between LSC methods and the reference value from $4\pi\beta(\text{PC})-\gamma$ and $4\pi\beta(\text{LS})-\gamma$ is lower than 0.1%.
Conclusions concerning beta spectra:
- It is recommended to apply TDCR and CNET.
- Discrepancies are considerably reduced when using screening corrections and when taking into account the atomic exchange effect.
- Improved calculation methods are also needed for forbidden transition.
- MMC measurements can be used to measure beta spectra with distinguished energy resolution.
- The EMPIR project MetroBeta will help to improve calculation procedures as well as to get experimentally determined beta spectra.
Analysis of light transport

K. Kossert

Radionuclide metrology using LSC
Radionuclide metrology using LSC

Reduction due to total reflection

Glass + Scotch tape

Glass

PE
Detector developments

Several metrology institutes developed their own LS counters. Examples: (trans)portable system:

- **PTB**
- **LNHB**
- **NPL**
- **ENEA**
- **PTB**
Detector developments

Examples: Automated systems:
Detector developments

Examples: Electronics

Several institutes developed coincidence modules

Often FPGA ICs are used.

K. Kossert  Radionuclide metrology using LSC
LSC in Radionuclide metrology

Other ongoing R&D topics:

- Investigation of the role of (reverse) micelles

Examples:
- NIST: micelle sizes and size distributions
- IRA: new MC-based model

\[ l=3 \]
\[ l=2 \]
\[ l=1 \]

(inverse) micelle

4 nm
LSC in Radionuclide metrology

Other ongoing R&D topics:

- (Full) digitized data acquisition (high sampling rates >250 MHz)
  Examples:
  ANSTO: Modules from National Instruments (after CFD)
  ENEA: CAEN modules
  IRA: Modules from National Instruments
  ...

Future (possibilities):
- Analysis of one data set with various conditions after the measurement
- Easy variation of coincidence resolving time, deadtime, discriminator threshold, etc. (without need to repeat the measurement)
- Ideal for short lived isotopes and/or isotopes with delayed states (e.g. $^{67}$Ga)
Current limitations and challenges
Limitations

Problem 1: Commercial counters

Reliable counters with automated sample changers and external standard sources are desirable.

Requirements:
- Threshold adjustment of PMT discriminators (below SEP)
- Deadtime / Lifetime (should be known, variable, extendable); linearity, reference clock
- Time (start time) needs to be known with hours, minutes and seconds
- Features as PSA, PAC, guards (vetos), other background reduction algorithms etc. should be detachable

See also contribution #203 from Philippe Cassette

K. Kossert
Limitations

Problem 2: Electron-capture (EC) isotopes

After an EC process a complex atomic relaxation takes place.

Requirements:
- Knowledge of fractional EC probabilities
- Knowledge of probabilities and energies of Auger electrons and x-rays

But:
- Existing models suffer from inaccurate input data and make many simplifications.
- Electron shake-off and shake-up effects are not taken into account
- Atom is considered as single ionized configuration
- ...
Limitations

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

Problem 3: The ionization quenching function

The Birks function $Q(E)$ is semi-empirical. The $kB$ value is one of the most frequently discussed parameters. Electron stopping powers $dE/dx$ are not well known at low energies. The sample composition is not always well known.

Potential way out:

Measure $Q(E)$ with a TDCR-γ Compton spectrometer
Limitations

\[ Q(E) = \frac{1}{E} \int_{0}^{E} \frac{dE}{1 + kB \frac{dE}{dx}} \]

**Problem 3:** The ionization quenching function

The Birks function \( Q(E) \) is semi-empirical. The \( kB \) value is one of the most frequently discussed parameters. Electron stopping powers \( dE/dx \) are not well known at low energies. The sample composition is not always well known.

**Potential way out:**

Measure \( Q(E) \) with a TDCR-\( \gamma \) Compton spectrometer.

**Very challenging!**
Literature

Recommendation:
Comprehensive text book written by Agustín Grau Malonda
Radionuclide metrology using liquid scintillation counting

Ryszard Broda¹, Philippe Cassette²,⁴ and Karsten Kossert³

¹ Institute of Atomic Energy, Radioisotope Centre POLATOM, 05-400 Otwock-Świerk, Poland
² CEA Saclay DRT/DETECS/LNHB, 91191 Gif Sur Yvette Cedex, France
³ Physikalisch-Technische Bundesanstalt, Department 6.1, D-38116 Braunschweig, Germany

Received 9 February 2007
Published 2 August 2007
Online at stacks.iop.org/Met/44/S36

Abstract
Liquid scintillation counting (LSC) techniques can be used for radionuclide standardization when the calculation of detection efficiency is possible. This is done using a model of the physicochemical processes involved in light emission and also of the statistics of photon emission: the free parameter model. This model can then be applied in two ways: by deducing the free parameter from the measurement of a tracer (the CIEMAT/NIST method) or by calculating this free parameter from coincidence ratio in a specific LS counter (the TDCR method). The purpose of this paper is to describe both these models and some practical issues that need to be addressed if LSC is to be effectively used in radionuclide metrology.
Literature

Uncertainty determination for activity measurements by means of the TDCR method and the CIEMAT/NIST efficiency tracing technique

Karsten Kossert¹, Ryszard Broda², Philippe Cassette³, Guy Ratel⁴ and Brian Zimmerman⁵

¹ Physikalisch-Technische Bundesanstalt (PTB), Bundesallee 100, 38116 Braunschweig, Germany
² National Centre for Nuclear Research, Radioisotope Centre POLATOM 05-400 Otwock, Poland
³ Laboratoire National Henri Becquerel, CEA-LNHB, CE-Saclay, 91191 Gif sur Yvette Cedex, France
⁴ Bureau International des Poids et Mesures (BIPM), Pavillon de Breteuil, F-92312 Sèvres Cedex, France
⁵ Physical Measurement Laboratory, National Institute of Standards and Technology (NIST), Gaithersburg, MD20899-8462, USA

More about uncertainties in contribution #143 from Brian Zimmerman

K. Kossert

Radionuclide metrology using LSC
Messages

- Liquid scintillation counting is a wonderful tool for radionuclide metrology.

- Significant progress has been achieved in the past decade.

- More research is needed to solve existing problems (e.g. determination of beta spectra, investigation of $Q(E)$, counter development including full digital data acquisition).

- Good commercial counters are desirable.
ICRM 2017
21st International Conference on Radionuclide Metrology and its Applications

15th to 19th of May, 2017
Buenos Aires - Argentina

Radionuclide metrology using LSC
Thank you for your attention