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Activity determination of ^{88}Y by means of 4πβ(LS)-γ coincidence counting

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- Coincidence method – 4πβ(LS)-γ coincidence method
- Detection systems (*M1* and *M29*)
- Activity measurements
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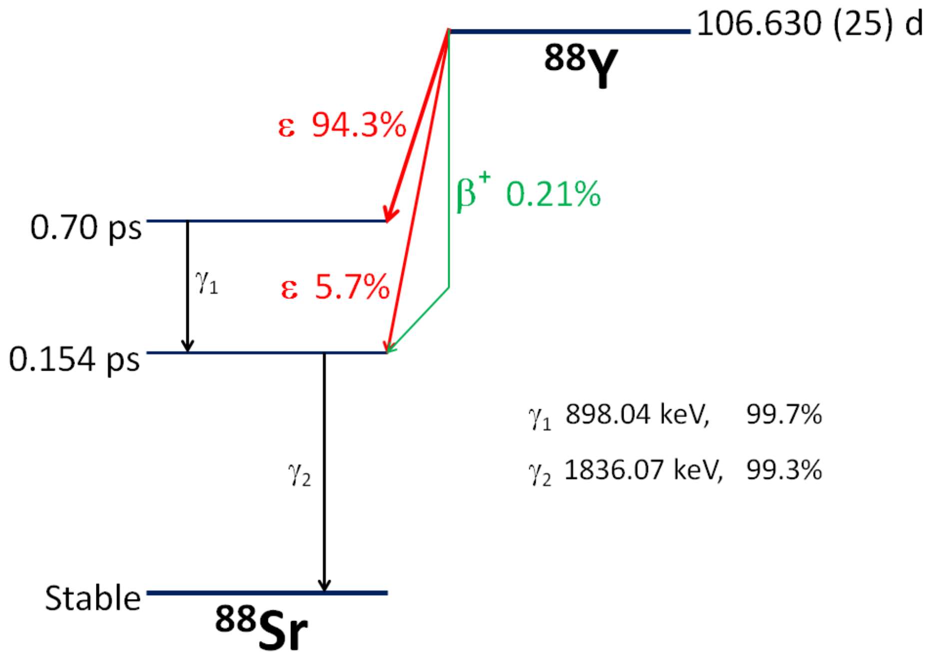
Introduction

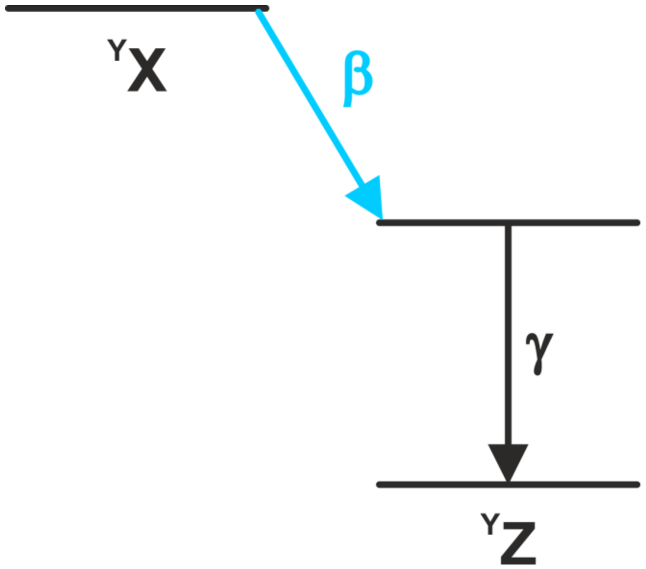


- ^{88}Y – electron capture nuclide (followed by a cascade of two γ-rays).
- ^{88}Y used as a radioactive source for the calibration of γ-ray spectrometers.
- A precise measurement of activity is not trivial due to low energy of X-ray and Auger electrons.
- Measurements were made by means of 4πβ-γ- coincidence counting with LS and PC detectors in β channels.

Coincidence method

Basic principle

- disintegrations with α-, β- or x-ray and Auger electrons emission plus γ-rays;
- β-, γ- and coincidence-channels in detection system;
- the activity follows from the counting rates;
- a priori knowledge of detection efficiency is not needed.



$$N_\beta = A \cdot \varepsilon_\beta$$

$$N_\gamma = A \cdot \varepsilon_\gamma$$

$$N_c = A \cdot \varepsilon_\beta \varepsilon_\gamma$$

$$A = \frac{N_\beta \cdot N_\gamma}{N_c}$$

A primary method !

Coincidence method

The efficiency extrapolation method

- The β-channel detector – sensitive to the gamma-transition.

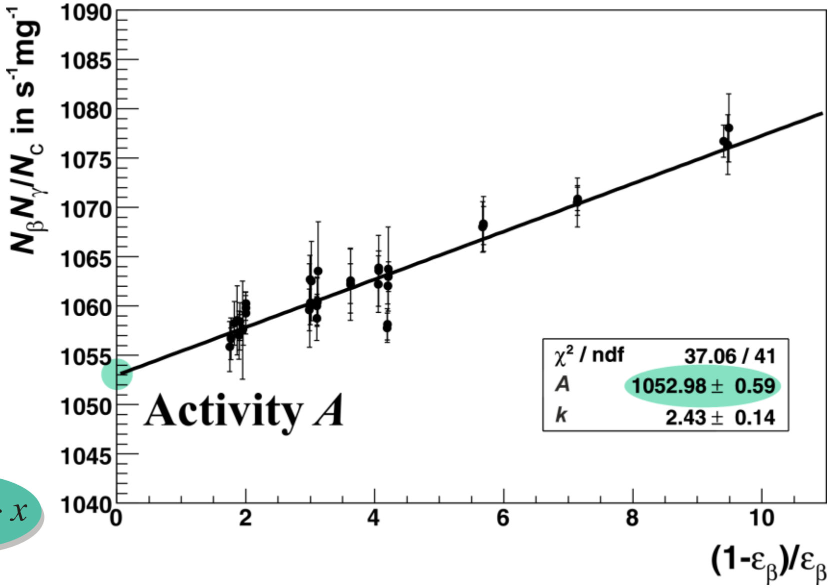
- The efficiency extrapolation method developed to compensate for:
  - the γ-sensitivity in the β-channel,
  - problems induced by conversion electrons,
  - additional coincidences from Compton scattering in the β-channel.

- The variation of the β-efficiency – the activity obtained from the extrapolation.

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

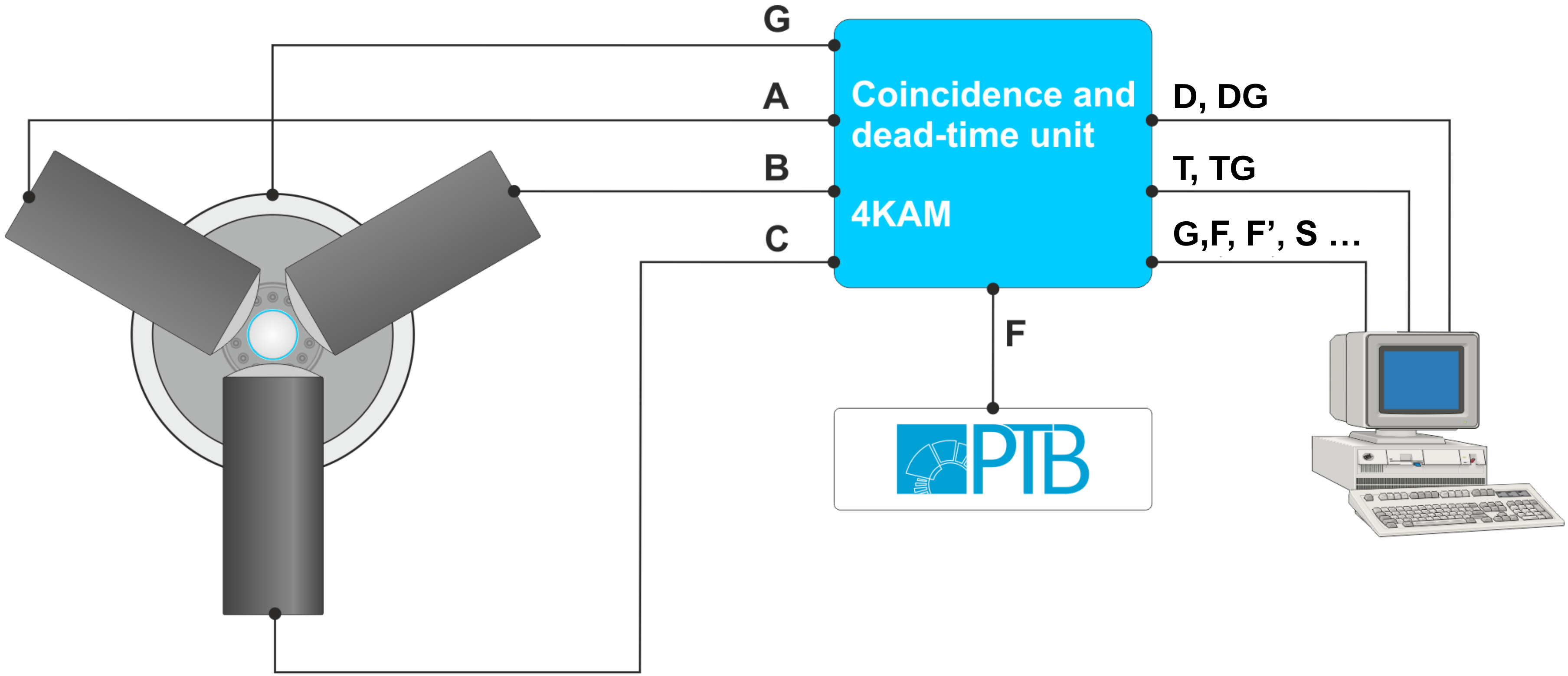
$$\left.\begin{aligned} N_\gamma &= A\cdot\frac{1}{1+\alpha}\cdot\varepsilon_\gamma \\ N_c &= A\cdot\frac{1}{1+\alpha}\cdot\varepsilon_\beta\varepsilon_\gamma \end{aligned}\right\} \rightarrow \frac{N_c}{N_\gamma} = \varepsilon_\beta$$

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



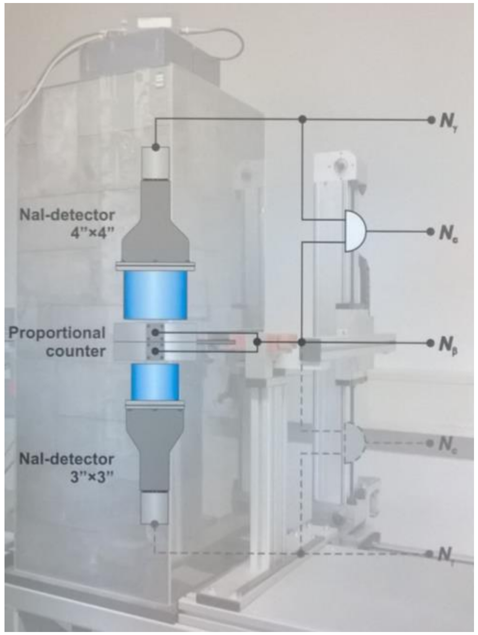
4πβ(LS)-γ coincidence method

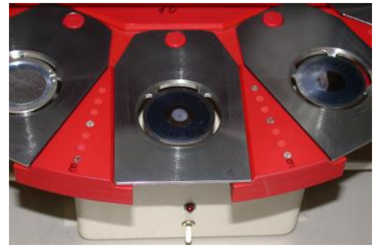
- the standard coincidence method ⟶ a proportional counter
- the "new" coincidence technique ⟶ a liquid scintillation detector



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

4πβ(PC)-γ coincidence system – *M1*





M1 system:

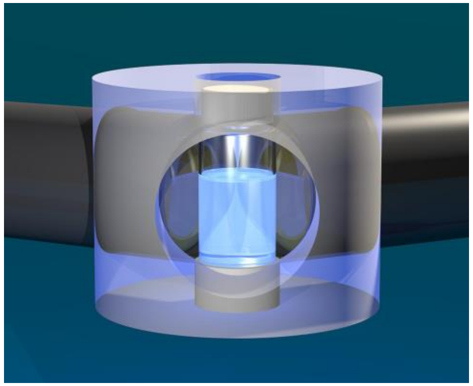
- a proportional counter (PC) in the β-channel;
- two scintillation detectors (NaI) in the γ-channel;
- samples are changed automatically.

M1 samples consist of:

- VYNS (polyvinylchloride-polyvinylacetate co-polymer) foils;
- coated with a thin Au-Pd layer;
- mounted on stainless steel rings;
- pre-treated by electrospraying with a colloidal SiO_2 suspension;
- the radioactive solution is deposited on the center of the sources and then slowly dried;
- β-efficiency variation by absorption foils.

Detection systems

4πβ(LS)-γ coincidence system – *M29*



M29 system:

- a liquid scintillation (LS) counter in the β-channel;
- an automated sample changer;
- a scintillation detector (NaI) in the γ-channel;
- β-γ coincidence and TDCR measurements are possible simultaneously.

M29 samples:

- mixing the radioactive solution directly into the scintillation cocktail (Ultima Gold™);
- low-potassium borosilicate glass or polyethylene vials;
- β-efficiency variation by chemical quenching (nitromethane-CH_3NO_2) or/and by the neutral density grey-foil filters.

The activity measurement

4πβ(LS)-γ coincidence counting

Two sets of samples:

set (1) – 3 samples, the efficiency variation was made by chemical quenching (nitromethane from 0 μL to 60 μL) combined with ND filters, with light transmission from 69.3% to 6.6%, foils were placed around and under the vial;

set (2) – 8 samples, the efficiency variation was made by pure chemical quenching (nitro-methane from 0 μL to 210 μL).

The γ energy window was adjusted to use three different settings:

1) a lower threshold to cover the whole energy spectrum,
2) a window to cover the full-energy peak of one of the most intensive line – 898.042 keV
3) a window to cover the full-energy peak of the 898.042 keV line together with the Compton continuum.

The coincidence resolving times: 77.5 ns and 76.5 ns for the β channel and for the β-γ coincidences, respectively.

The common extendable dead time – 30 μs.

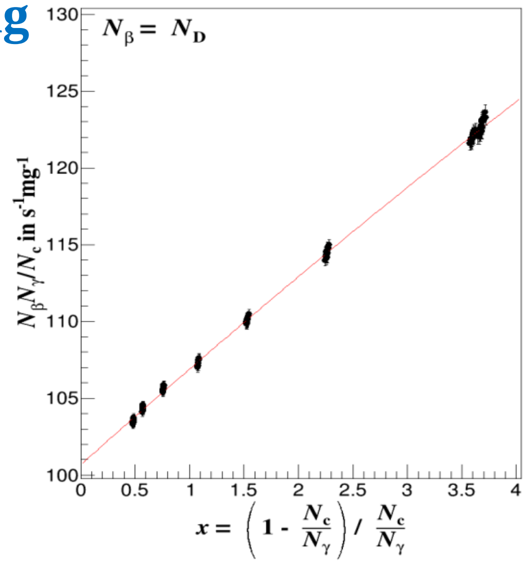
A delay time of 2.212 μs was used for the β signals to compensate for the slower signal processing of the γ channel.

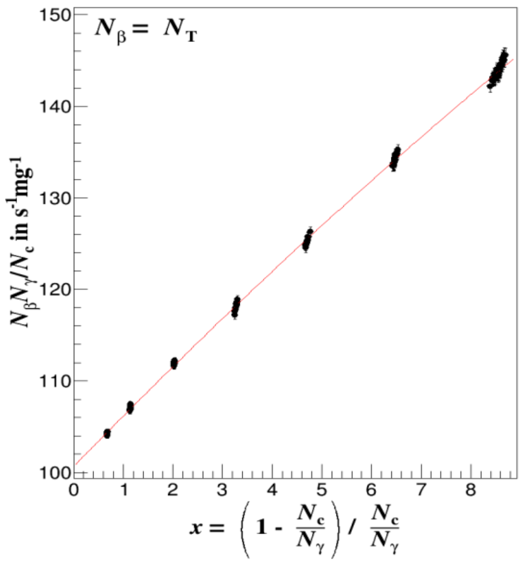
The activity measurement

4πβ(LS)-γ coincidence counting

Set (1) of samples:
a = (100.69 ± 0.69) kBq/g
Set (2) of samples
a = (100.79 ± 0.58) kBq/g

Final result
***a* = (100.75 ± 0.58) kBq/g**





The counting rates of β, γ and β-γ coincidence events, were used to plot the ratio $N_\beta N_\gamma / N_c$ as a function of $x = (1-N_c/N_\gamma)/(N_c/N_\gamma)$.

The counting rate determination includes: the background subtraction, the decay correction to the reference date, and the correction for decay during the measurement.

2nd deg. polynomial functions were fitted to the experimental data and extrapolated to ε_β = 100%.

Results were determined for double and triple coincidences in the β channel and for three different γ settings.

The activity concentrations were determined separately for each set of samples.

The activity measurement

4πβ(PC)-γ coincidence counting

To validate the result from the new PTB custom-built 4πβ(LS)-γ coincidence counting system.

Three samples were prepared – 10 mg, 15 mg and 22 mg.

The β counting efficiency was varied by successive addition of VYNS absorption foils.

Both the β and γ channels included dead-time units with the same non-extendable dead time of about 8 μs.

To compensate for the delay between the γ and β events, a delay unit was used.

The coincidence resolving time: about 1 μs.

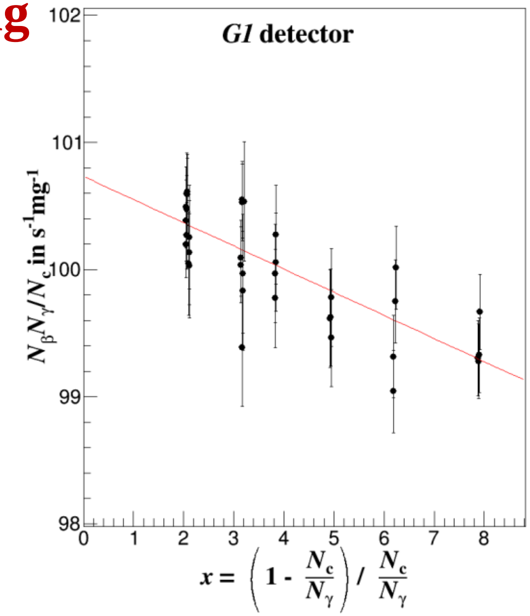
The γ energy window was adjusted with a lower threshold to cover the whole energy spectrum.

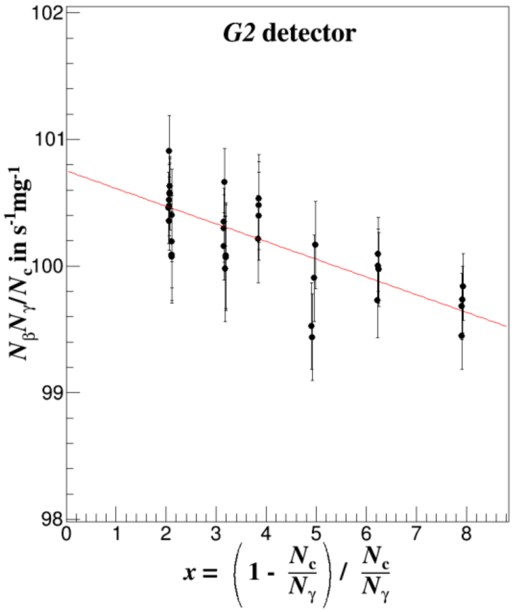
The activity measurement

4πβ(PC)-γ coincidence counting

With *G1* detector:
a = (100.73 ± 0.29) kBq/g
With *G2* detector
a = (100.75 ± 0.27) kBq/g

Final result
a = (100.74 ± 0.29) kBq/g





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The counting rates were corrected for background, decay to the reference date, decay during the measurement and analyzed based on the expression given in Smith D. (Nucl. Instrum. Meth. 152:505-519, 1978) for the coincidence counting rate.

1st deg. polynomial functions were fitted to the experimental data and extrapolated to ε_β = 100%.

Comparison

4πβ(LS)-γ coincidence counting

***a* = (100.75 ± 0.58) kBq/g**

4πβ(PC)-γ coincidence counting

***a* = (100.74 ± 0.29) kBq/g**

0.01 % relative deviation

The good agreement between results from both systems confirms the correct operation of the 4πβ(LS)-γ coincidence counting system.

Uncertainties

Component	$u(a)/a$ in %	
	4πβ(PC)-γ CC	4πβ(LS)-γ CC
Counting statistics	0.18*	0.02*
Weighing	0.08	0.02
Dead time	negligible	0.20
Background	0.04	0.03
Resolving time	negligible	0.20
Counting time	negligible	negligible
Adsorption	0.05	0.05
Decay correction	<0.01	<0.01
Extrapolation of efficiency curve	0.05	0.04
Impurities (no radioactive impurity detected)	<0.03	<0.03
Fitting uncertainty	0.19	0.49
Square root of the sum of quadratic components	0.29	0.57

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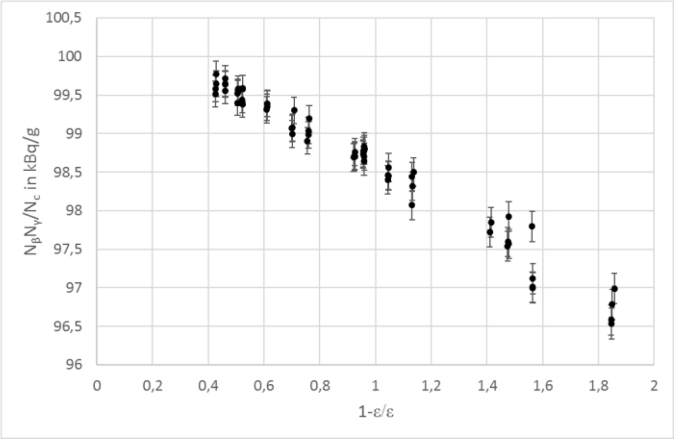
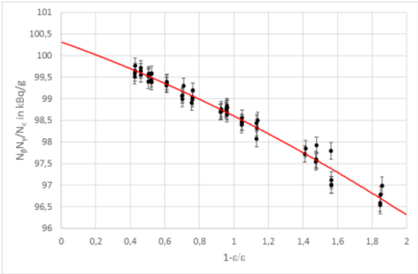
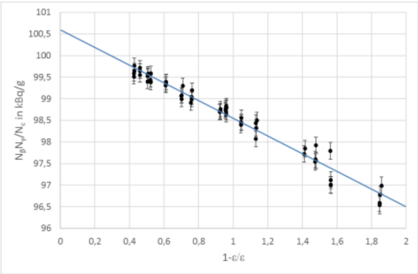
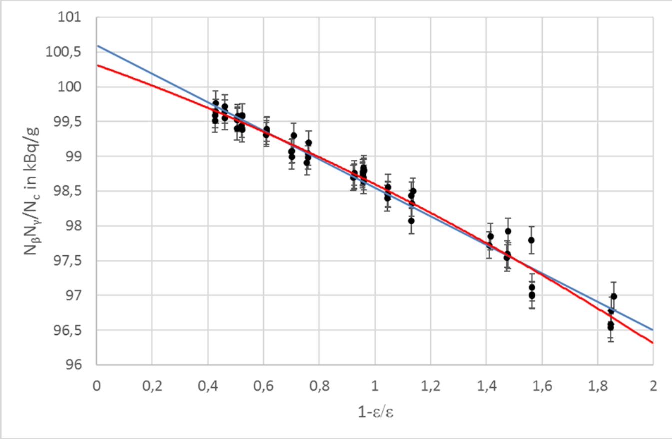
This uncertainty might be reduced by using a Monte-Carlo-simulation to provide a proper fitting function.









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In the 4KAM module, the determination of coincidences for the β channels (double and triple) is realized by a delay line with increments of 2 ns, while the determination of β-γ coincidences is set by the FPGA clock resulting in a jitter and increments of about 5.882 ns. Insufficient knowledge of these differences was the reason to estimate rather large uncertainties for the uncertainty components due to dead time and resolving time.

A significant decrease of these uncertainties could be achieved by a more comprehensive analysis of the 4KAM module.

Summary

- The 4πβ(LS)-γ coincidence counting is a good primary activity standardization technique, and sample preparation is much easier than in classical coincidence counting.

- The activity measurement of ^{88}Y is important for the calibration of γ-ray spectrometers.

- The good agreement between results from the 4πβ(LS)-γ coincidence counting and the 4πβ(PC)-γ coincidence counting confirms the correct operation of the 4πβ(LS)-γ coincidence counting system.

Thank you
for your attention