Radioactivity Measurement of Reactor-produced Radionuclide ^{177}Lu using $4\pi\beta(\mathrm{LS}) - \gamma$ Coincidence System

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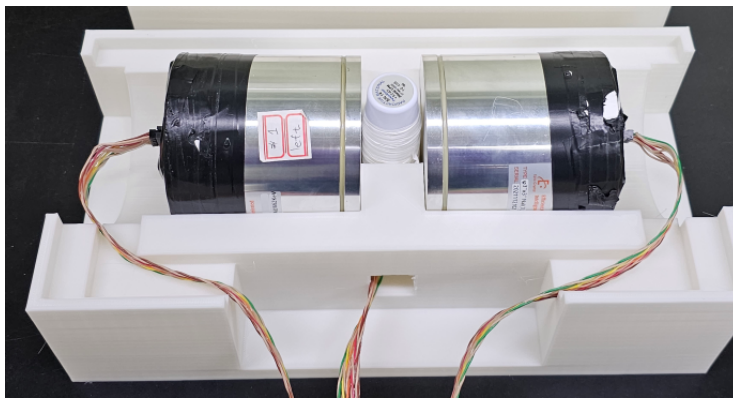
- HANARO

- Lutetium

- $4\pi\beta - \gamma$ coincidence counter



$4\pi\beta(\mathrm{LS}) - \gamma$ Coincidence Counting

- β & γ emissions from RI $\rightarrow$ coincidence counting
- Relation among observed counting rate & radioactivity

$$\frac{N_\beta N_\gamma}{N_c} = N_0[1 + k(\frac{1-\epsilon_\beta}{\epsilon_\beta})] = N_0[1 + k(\frac{N_\gamma}{N_c} - 1)] \quad (1)$$

$N_{\beta,\gamma,c}$: Observed counting rate of $\beta/\gamma/\beta$-γ coincidence events

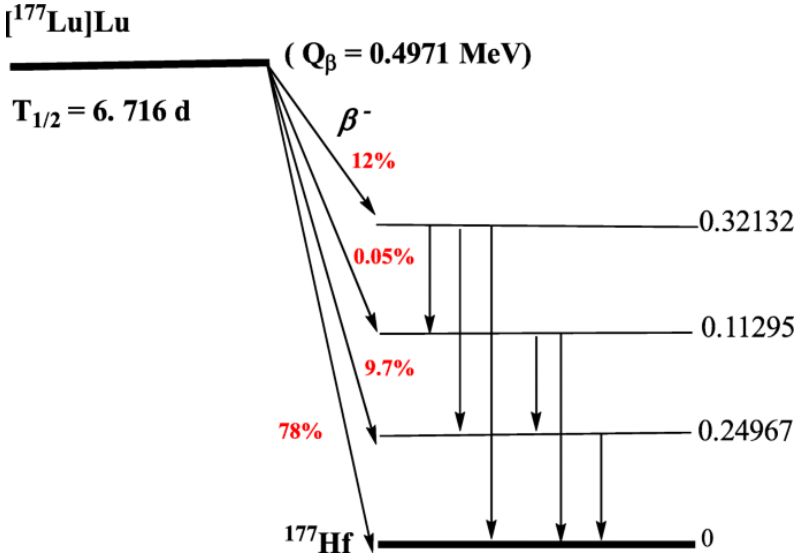
N_0: Radioactivity of RI

ϵ_β: β detection efficiency

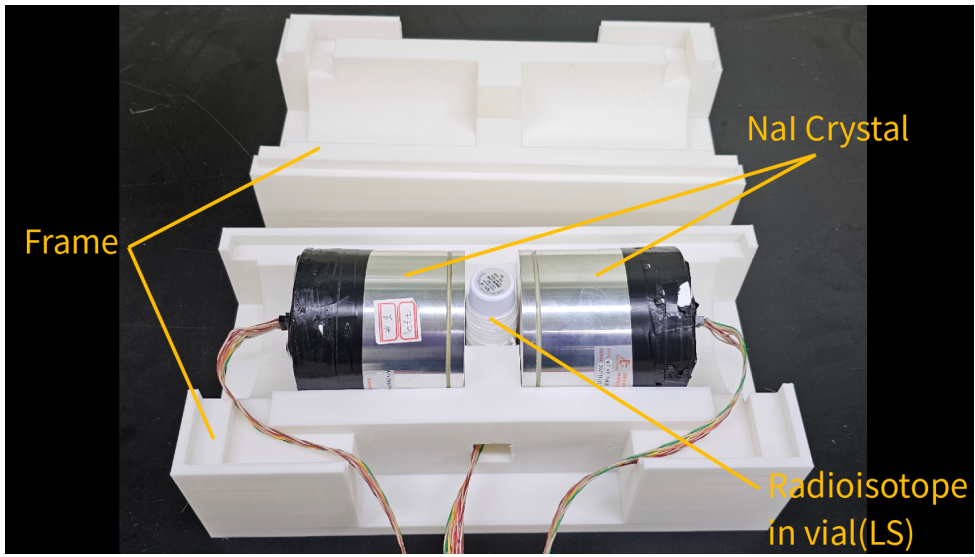
k: constant

- Efficiency-extrapolation ($\epsilon_\beta \rightarrow 1$): obtaining radioactivity of RI

- Decay scheme of radioisotope

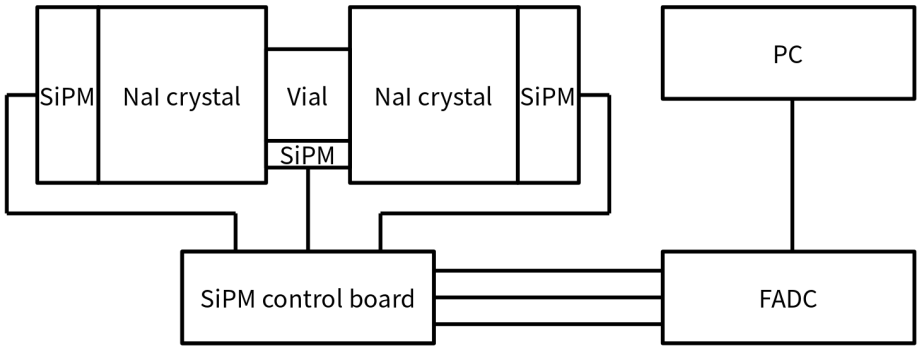


- $4\pi\beta - \gamma$ coincidence counter



$4\pi\beta(\mathrm{LS}) - \gamma$ Coincidence System

- Using SiPM instead of PMT → relatively small size → easy to move
- Liquid scintillator (LS) → uniform distribution of radionuclide to be measured
- Diagram

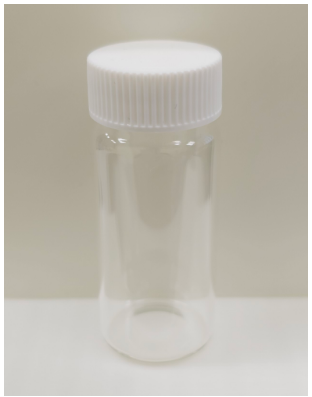


- $4\pi\beta - \gamma$ coincidence system

Components of $4\pi\beta(\mathrm{LS}) - \gamma$ Coincidence Counter

- Vial: containg LS & radioisotope, β detection
- NaI crystal: 3-inch, γ detection
- SiPM: scintillation light detection
- SiPM control board: power supply, thermometer
- Frame: 3-D printing, plastic

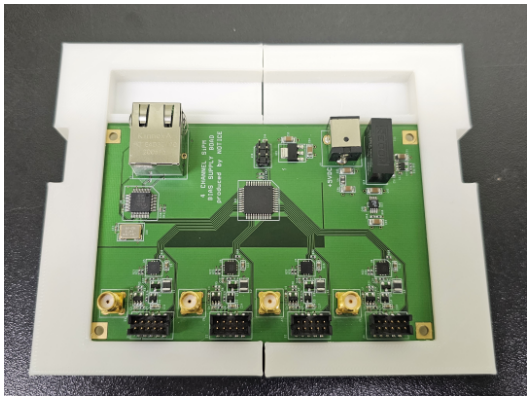
- Vial



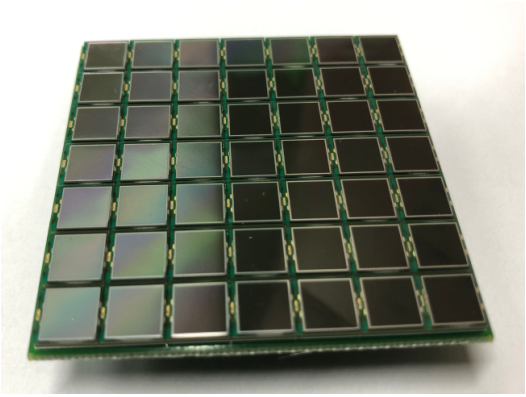
- LS

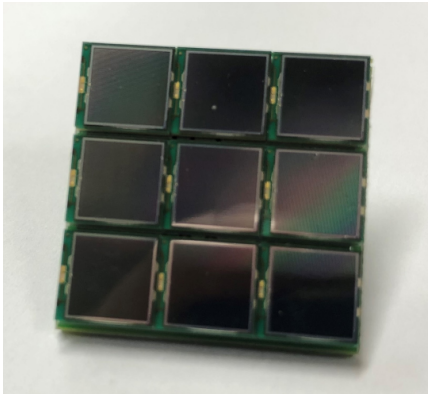
- NaI crystal

- SiPM control board

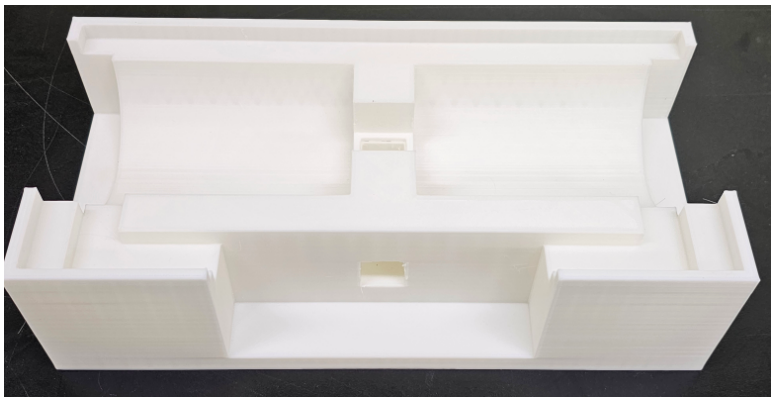


- SiPM





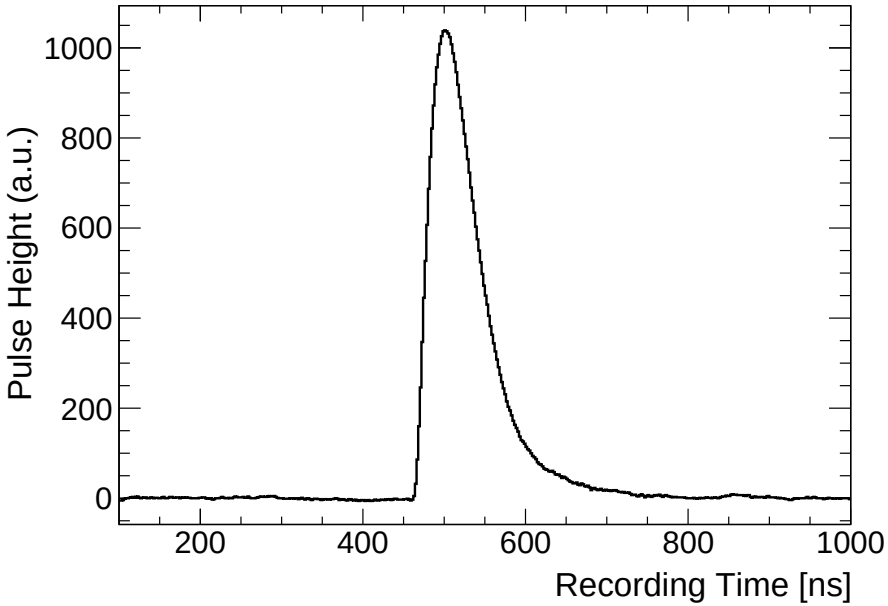
- Frame



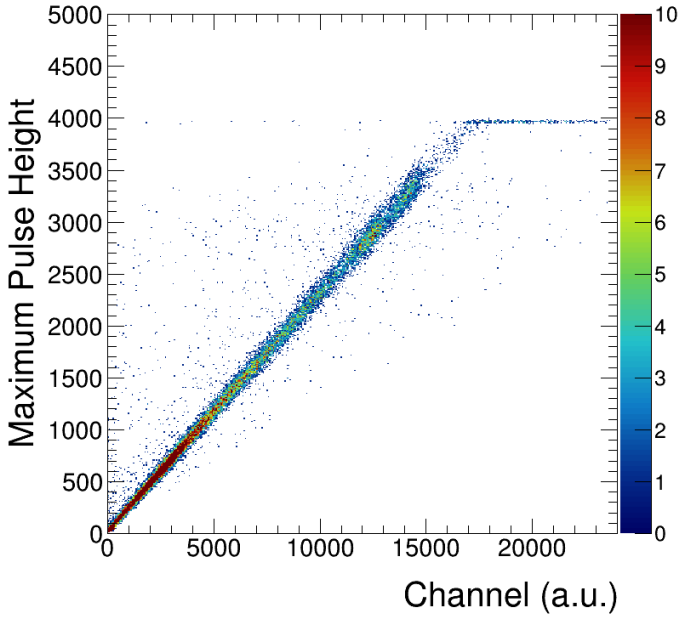
DAQ System

- FADC500 (Notice Korea)
- 4 channels
- 500 MHz sampling rate
- Dynamic range: 12 bit / 2.5 V
- Recording length: 0.1-32 μs
- Maximum trigger rate: $\sim$9.7 kHz for 4 channels

- Pulse



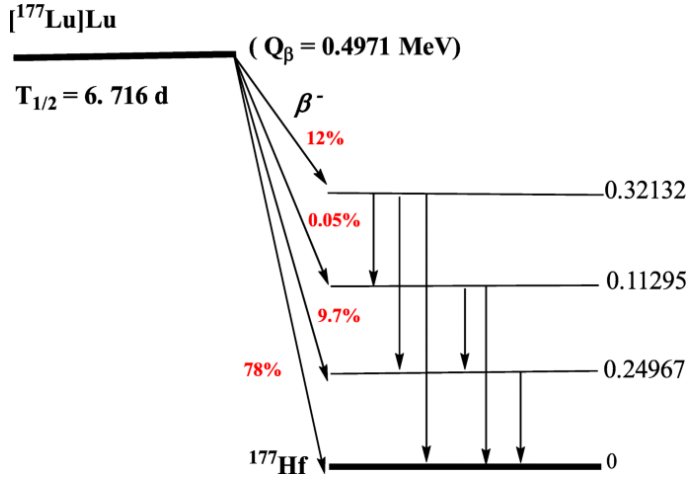
- Saturation



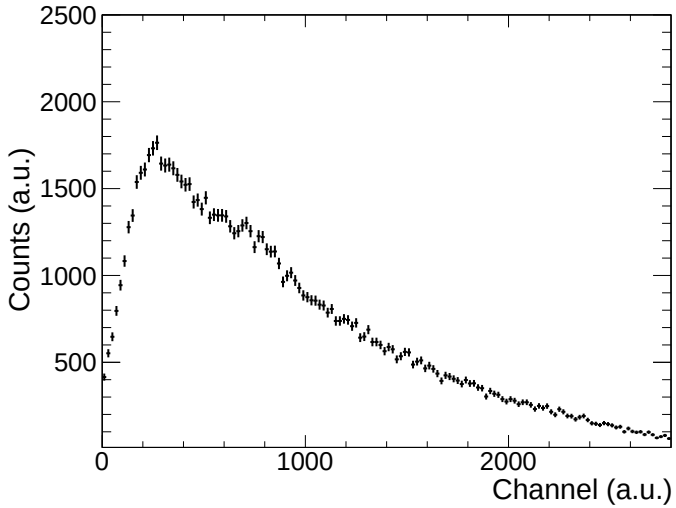
Experimental Setup

- Radioactivity measurement: ^{177}Lu
  - Experimental setup

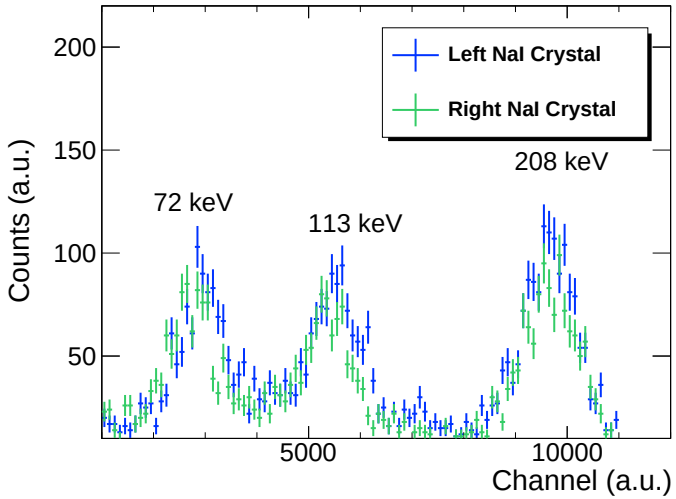
  - Decay scheme for ^{177}Lu



- β spectrum



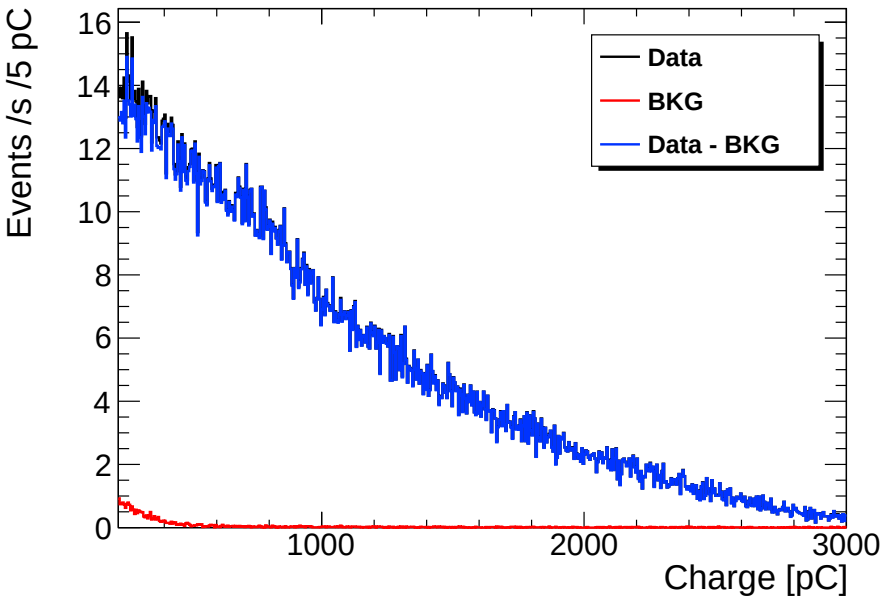
- γ spectra



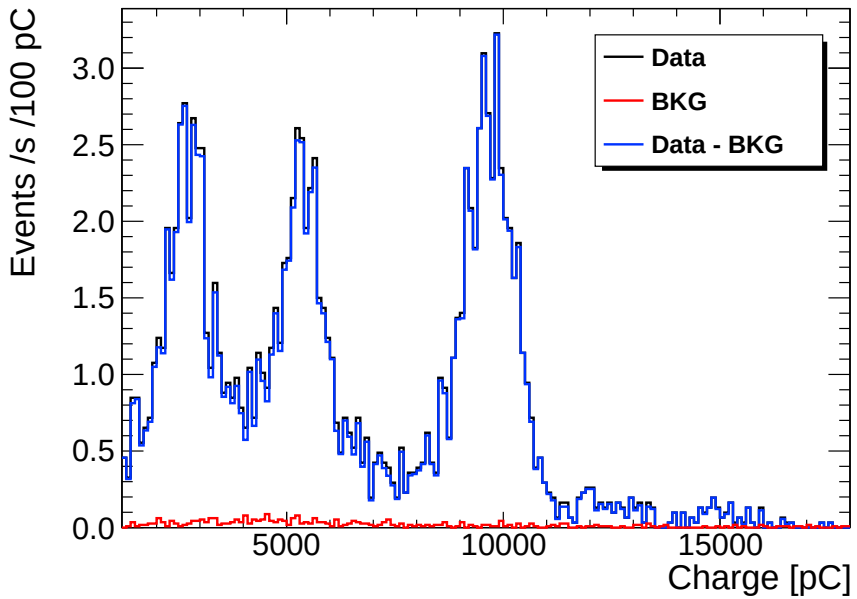
β/γ Spectra w/ Background

- Date: 2023-05-25
- Measuring background (BKG) before/after ^{177}Lu measurement → averaged

- β spectrum



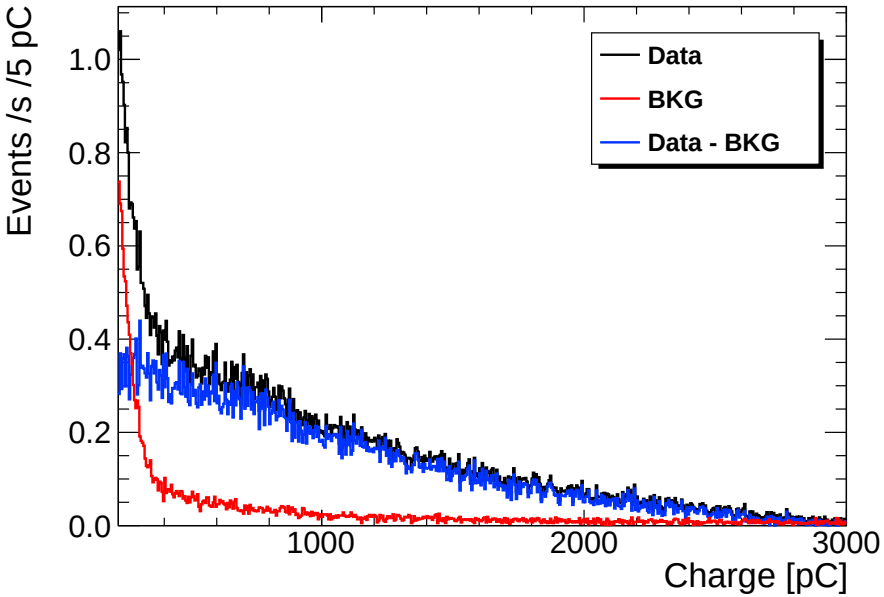
- γ spectrum



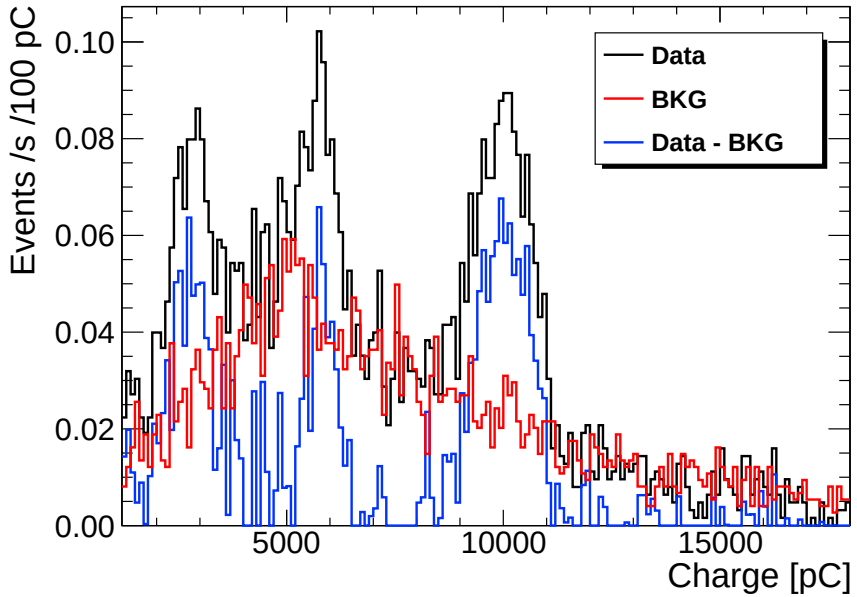
β/γ Spectra w/ Background

- Date: 2023-06-29
- Measuring background (BKG) before/after ^{177}Lu measurement → averaged

- β spectrum



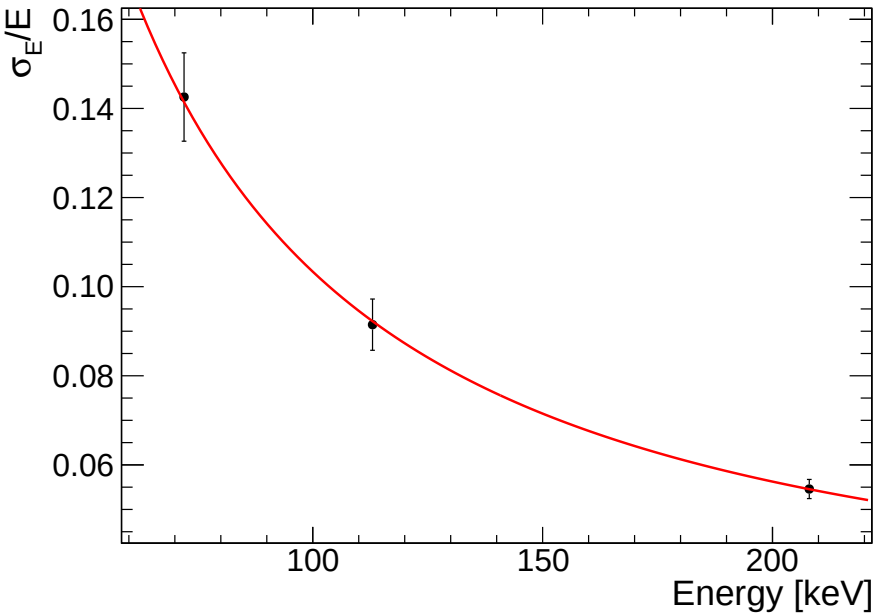
- γ spectrum



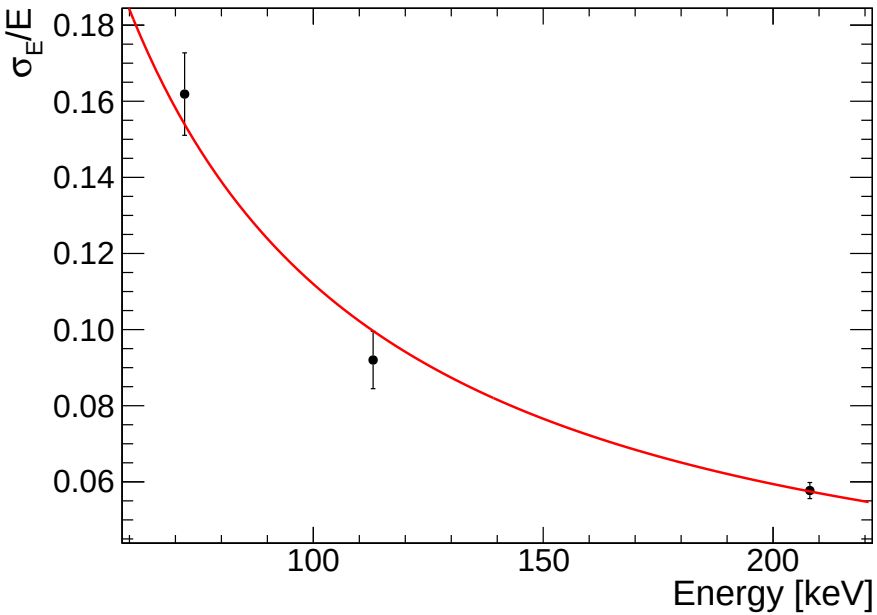
Energy Resolution

- Evaluating energy resolution of γ detectors using γ spectra for ^{177}Lu
- Fitting function: $\frac{\sigma_E}{E} = \sqrt{a^2 + \frac{b^2}{E} + \frac{c^2}{E^2}}$

- Left γ detector



- Right γ detector



γ detector	72 keV	FWHM	113 keV	FWHM	208 keV	FWHM	662 keV	FWHM
Left	14.1%	33.3%	9.22%	21.7%	5.45%	12.8%	2.98%	7.01%
Right	15.4%	36.2%	9.97%	23.5%	5.75%	13.5%	2.83%	6.67%

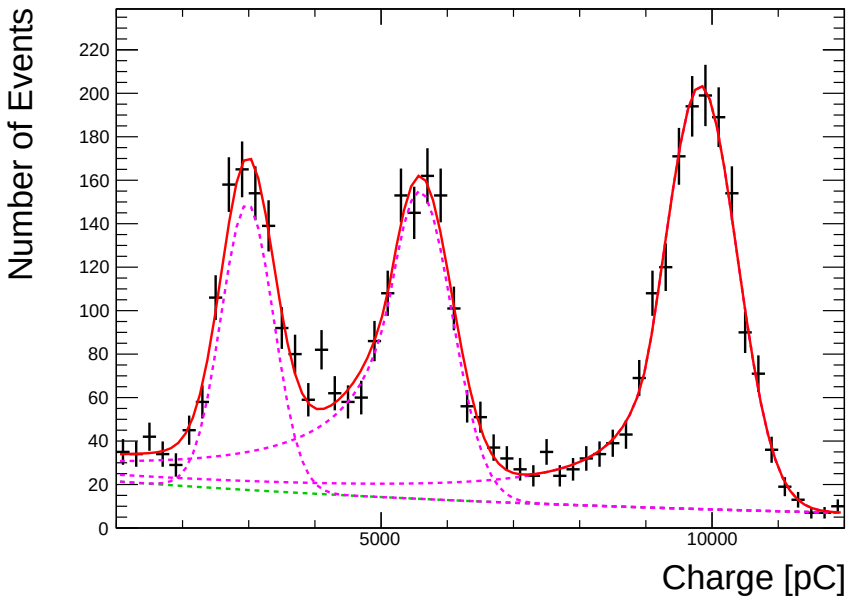
SiPM Gain Change with Temperature Change

- Averaged temperature of SiPMs

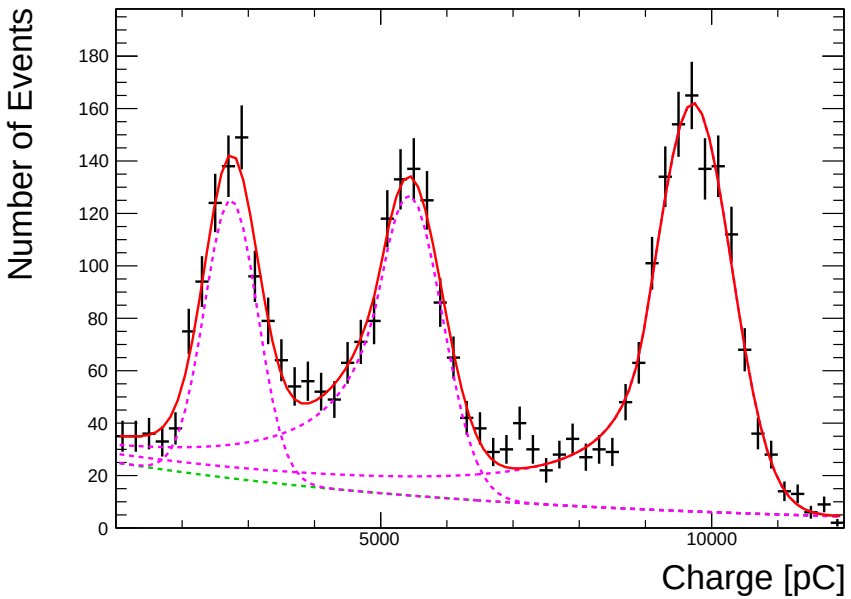
Date	2023-05-25	2023-06-19	2023-06-23
Temperature [°C]	21.5	30.0	26.5

- Fitting function: Crystal ball + Exponential

- γ spectrum for left γ detector (2023-05-25)



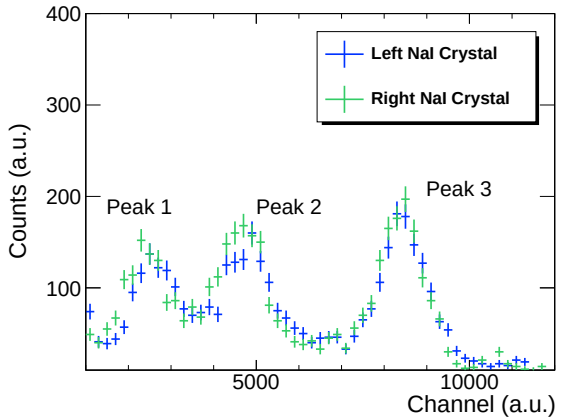
- γ spectrum for right γ detector (2023-05-25)



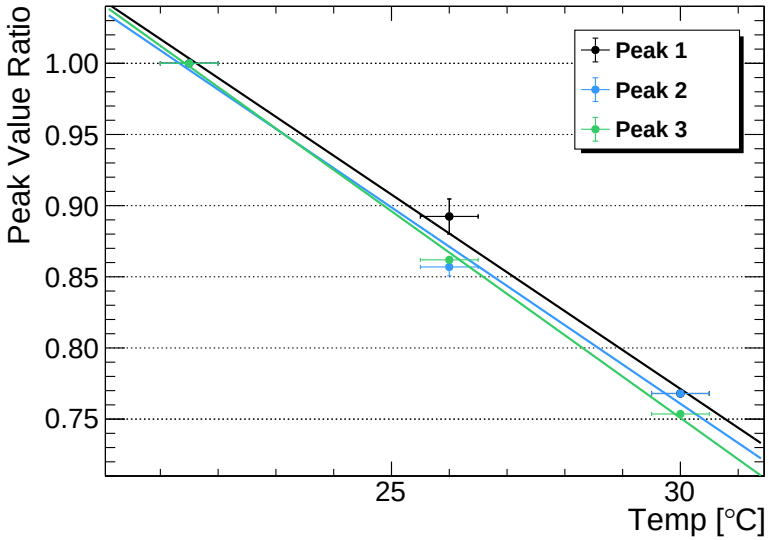
SiPM Gain Change with Temperature Change

- Comparison of peak values for 3 different temperatures
- Comparisons for the same channel are drawn in the same plot
- Fitting function: 1st order polynomial
- Difference among peaks
  - left: 0.3-2.7%
  - NaI right: 0.3-1.0%

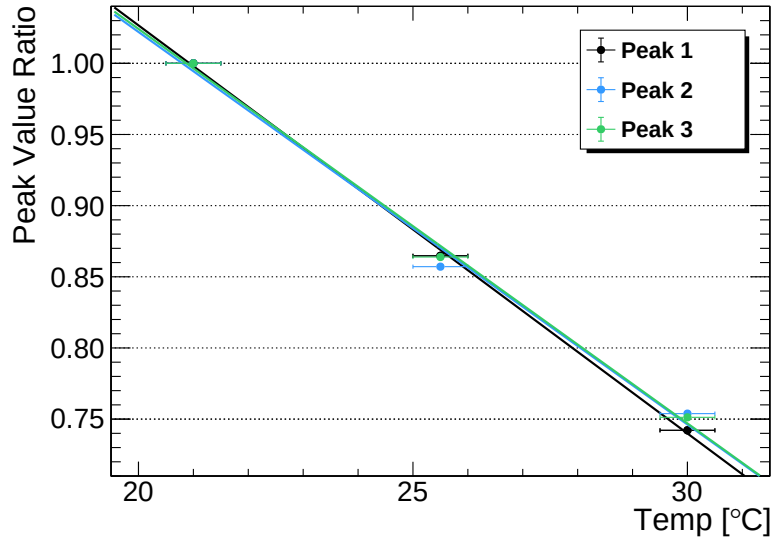
- Peaks in γ spectrum



- γ spectrum for left γ detector



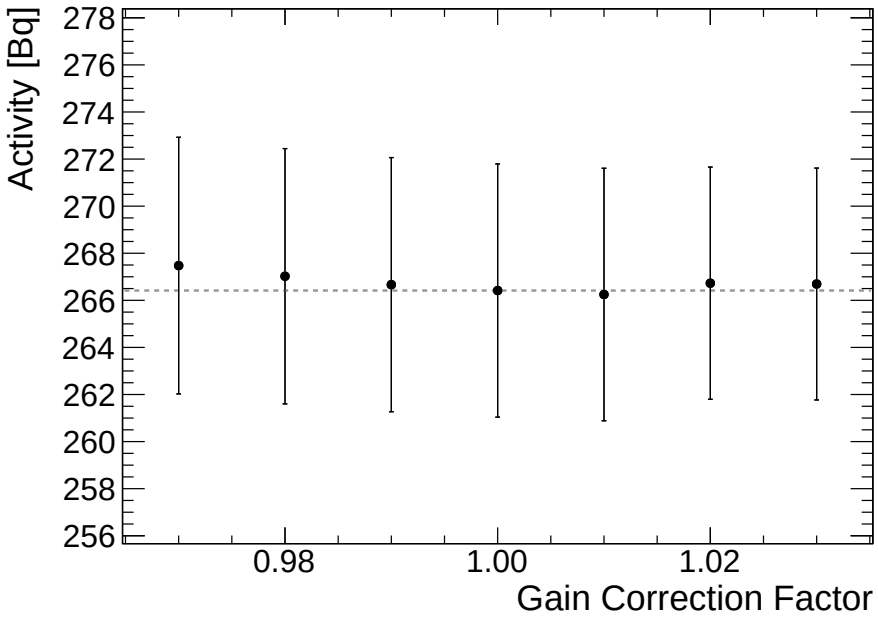
- γ spectrum for right γ detector



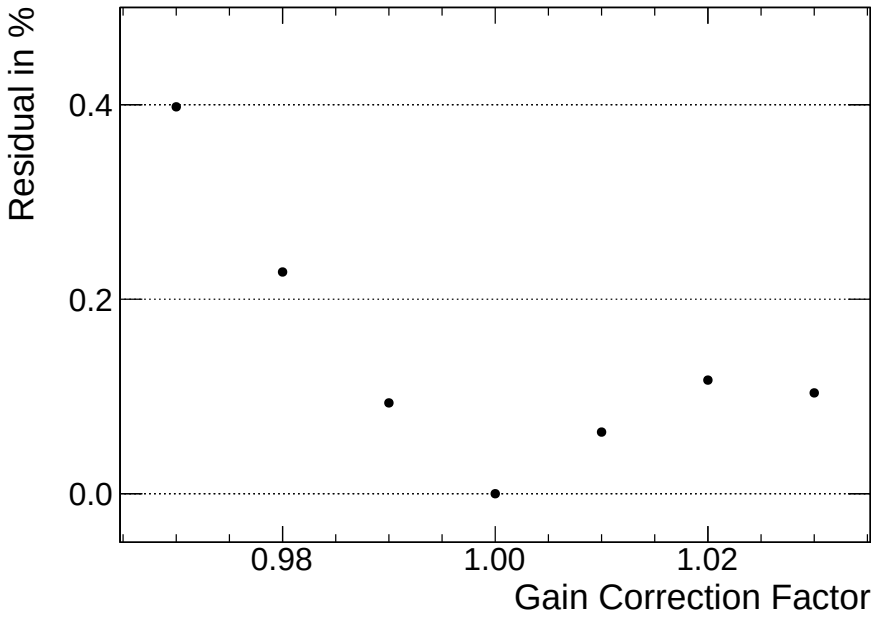
The Effect of Gain Change on Radioactivity Measurement

- Changing gain correction factor: 1 → 0.97, 0.98, ..., 1.02, 1.03 (assuming $\pm 1^{\circ}\mathrm{C}$ temp. change, 7 steps)
- Testing data obtained for 3 difference periods

- Gain vs. activity



- Residual

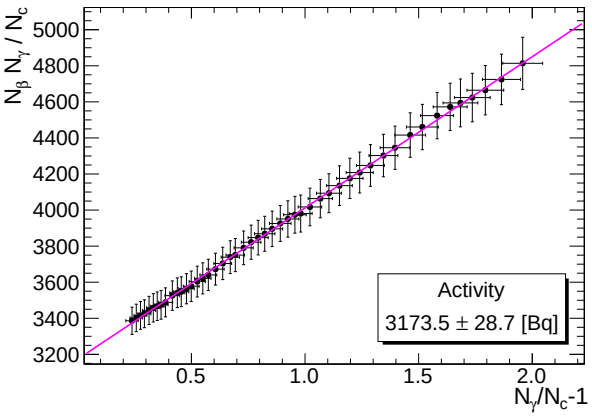


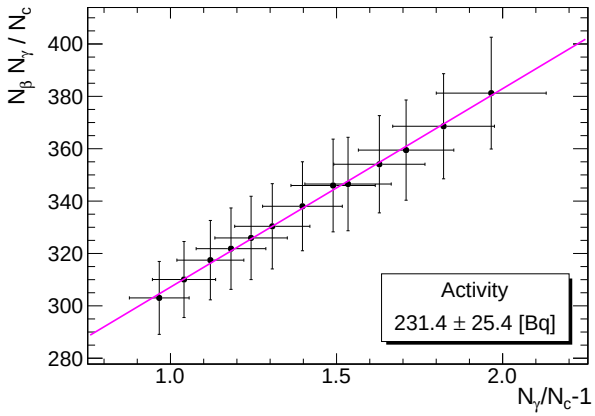
- Temperature change → gain change → radioactivity change
- $\pm 1\ ^{\circ}\mathrm{C}$ change → radioactivity change $< 0.4\%$ (smaller than statistical uncertainty $> \sim 1\%$)

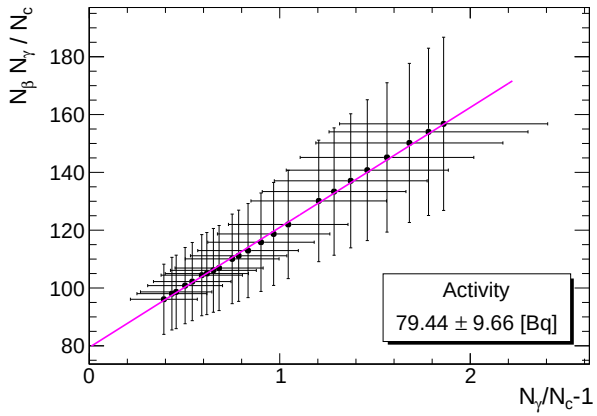
Radioactivity Measurement w/ BKG Subtraction

- $N_\beta N_\gamma / N_c$ for various ϵ_β values → efficiency-extrapolation → radioactivity of RI
- Changing threshold for β → various $N_\gamma / N_c - 1$ and $N_\beta N_\gamma / N_c$ values
- Fitting function: $N_\beta N_\gamma / N_c = N_0[1 + C(N_\gamma / N_c - 1)]$ (N_0: radioactivity, C: constant)
- Error bar: statistical uncertainty only

- Efficiency-extrapolation







- Results of radioactivity measurements

Date	2023-05-25	2023-06-19	2023-06-29
Radioactivity [Bq]	3174±29	231±25	79.4±9.7

Uncertainties and Total Error

- Calculating total error including uncertainties

$$\sigma_{tot} = \sqrt{\sum_{i=1}^{n} \sigma_i^2} \tag{2}$$

σ_{tot}: total error
σ_i: ith uncertainty
n: the number of uncertainties

- Uncertainties for radioactivity measurement

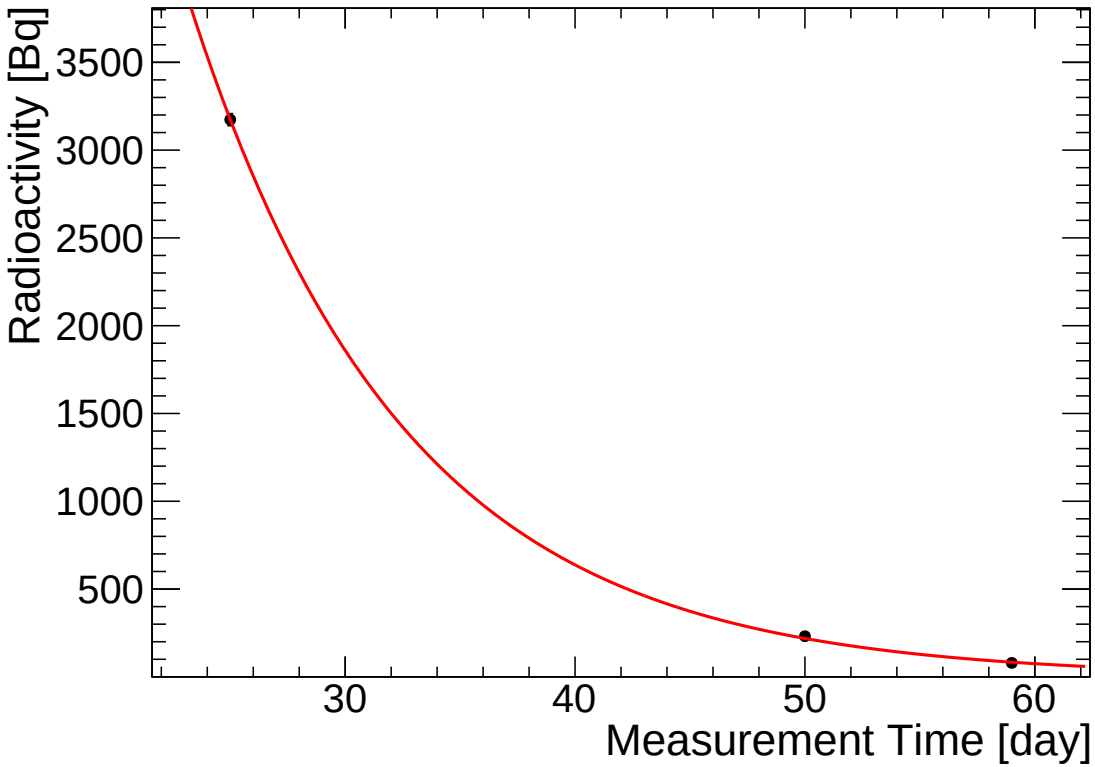
Statistical	1.02-10.0%
Temperature dependence	0.4%
Background difference	1-15% for background rate
Volume of the sample	0.964%

- The results of radioactivity measurements and total errors

Measurement data	Radioactivity [Bq]
2023-05-25	3174±35 (1.09% error)
2023-06-19	231±27 (11.5% error)
2023-06-29	79.4±10.8 (13.6% error)

Radioactivity Measurement

- Testing radioactivity measurement results
- Assuming total 5% uncertainty
- Fitting function: $N(t) = N_0 e^{-t\lambda} = N_0 e^{-t\ln(2)/t_{1/2}} = N_0 2^{-t/t_{1/2}}$ (λ: mean lifetime)
- $t_{1/2} = 6.478 \pm 0.183$ [day] (2.54% difference from the reference 6.647 [day])



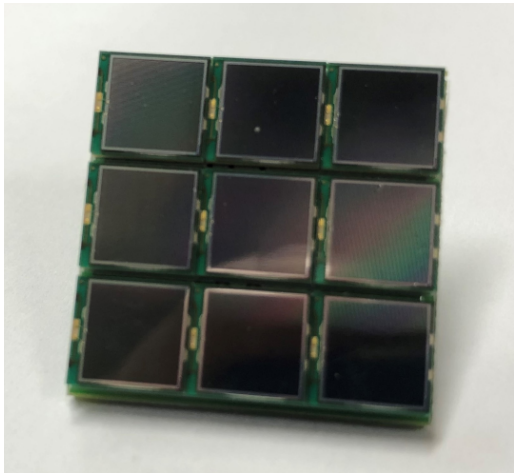
Summary

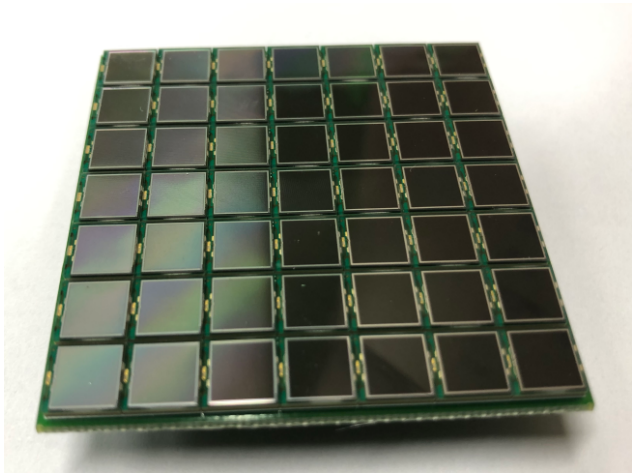
- Development of a radioactivity measurement system
  - radioactivity measurement for ^{177}Lu produced in HANARO
- Responses of the radioactivity measurement system
  - energy resolution of γ detector: $\sim$3% (FWHM: $\sim$7%) at 662 keV
  - temperature dependence of SiPM gain / radioactivity: $\pm$3% / $<$ 0.4% for $\pm$1 $^{\circ}$C
  - differences between backgrounds: 1-15%
- Radioactivity measurement for ^{177}Lu
  - evaluation of uncertainties included in total error
  - $\sim$1.1% total error for $\sim$3 kBq radioactivity
  - evaluation of half-life: 2.54% difference from the reference
- Operation of radioactivity measurement system
  - improvement for analysis method, experimental environment, structure of the system, etc.
  - measurements for other radioactive nuclides

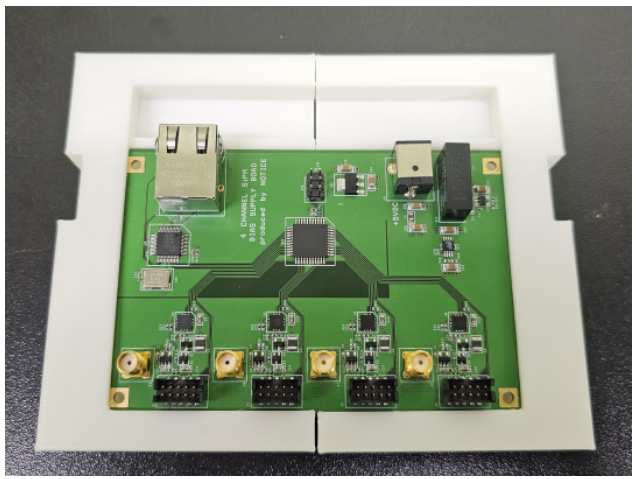
Backup slides

SiPM & Control Board

- SiPM: Hamamatsu S13360-1350PE
  - Operating voltage: 52-60 V
  - Operating temperature: -20 to 60 °C
  - Gain: 1-5×10^6 @25 °C
  - Spectral response range: 320-900 nm
  - Photon detection efficiency: 40% @450 nm
- SiPM array & control board
  - Production: Notice Korea
  - 3×3 array: for vial(LS),
  - 7×7 array: for NaI,
  - Control board: 4 channels,
  - Connection: TCP/IP



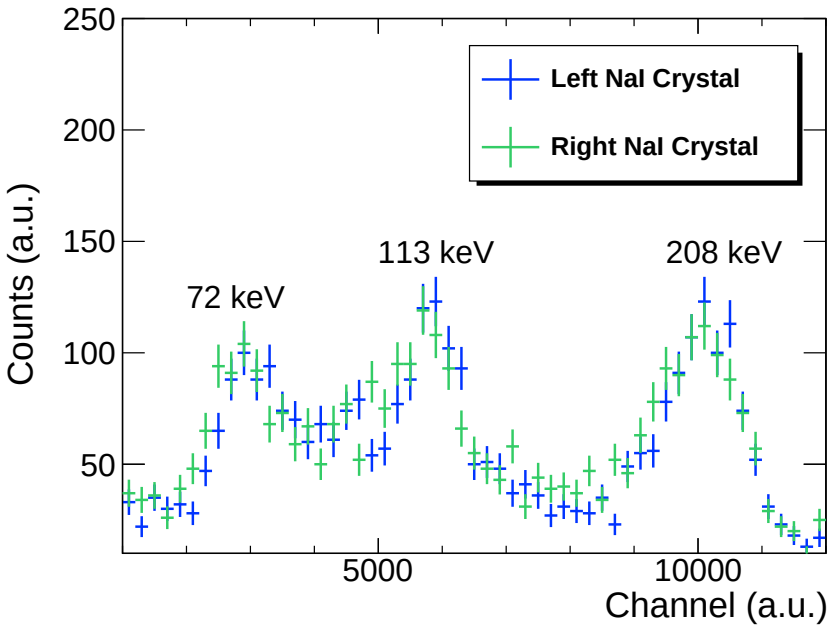




γ Spectra for ^{177}Lu + ^{109}Cd

- ^{177}Lu: 72, 113, 208 keV γs
- ^{109}Cd: 88 keV γ

- γ spectra for ^{177}Lu



- γ spectra for ^{177}Lu + ^{109}Cd