Neutron Capture Cross Section Measurement of Minor Actinides in Fast Neutron Energy Region for Study on Nuclear Transmutation System

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3) Kyoto University

Project Overview

Motivation

- Accurate nuclear reaction data of MAs are necessary for development of nuclear transmutation systems such as an accelerator-driven system (ADS).

Research Project on MA Neutron Capture Data

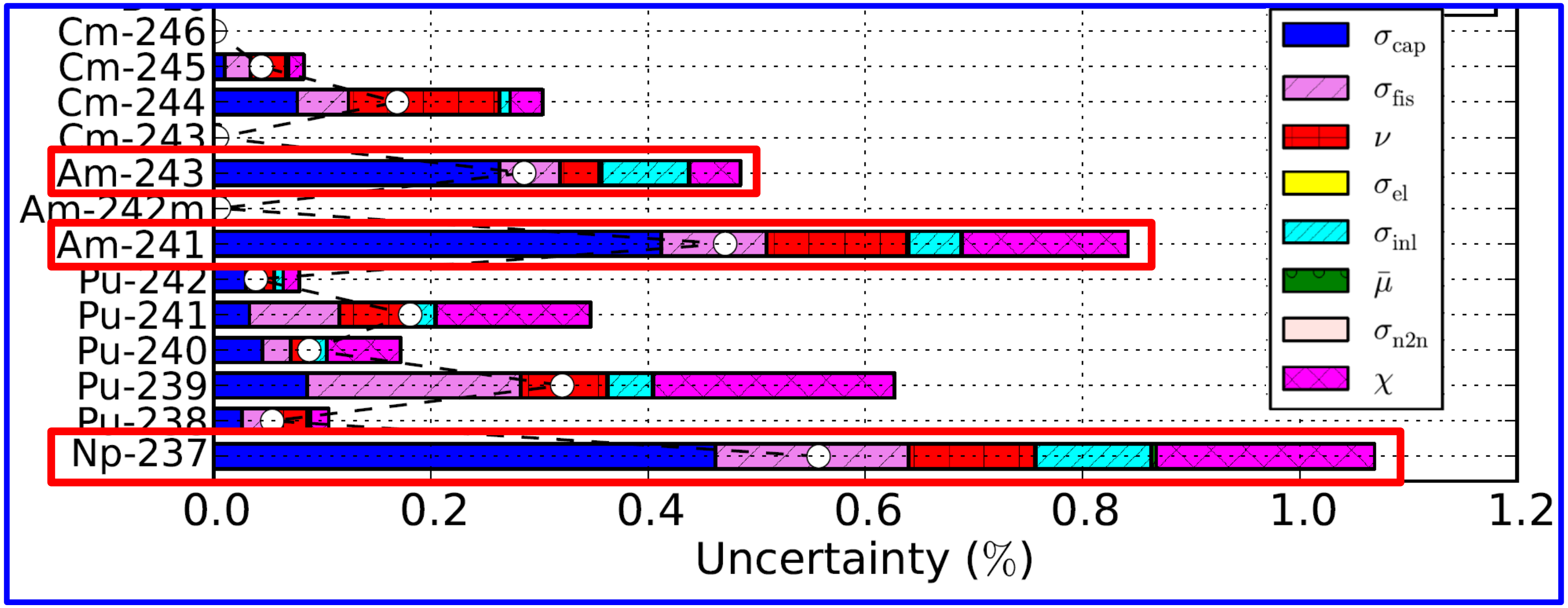
- Project "*Study on accuracy improvement of fast-neutron capture reaction data of long-lived MAs for development of nuclear transmutation systems*"
「核変換システム開発のための長寿命MA核種の高速中性子捕獲反応データの精度向上に関する研究」
- MEXT Innovative Nuclear Research and Development Program
原子力システム研究開発事業（放射性廃棄物減容・有害度低減技術研究開発 タイプB）
- Period: Oct. 2017 to Mar. 2021

Project Goal

- This project aims at improving accuracies of neutron capture cross section of long-lived minor actinides (^{237}Np, ^{241}Am, ^{243}Am) in the fast energy region (0.5 – 500 keV).

Research Background

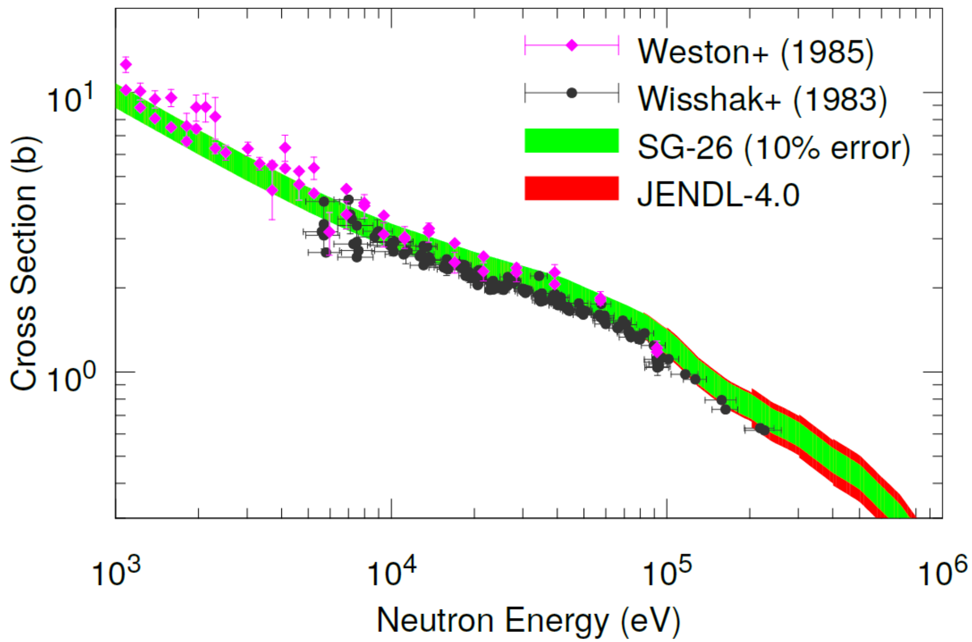
- The uncertainties of the present evaluated capture cross section data of MAs are not small enough to satisfy requirement for the design of nuclear transmutation systems.
- In particular, the uncertainties of neutron capture cross section data of ^{237}Np, ^{241}Am and ^{243}Am largely contribute to the uncertainty of criticality calculation of ADS.



Uncertainties by reaction and uncertainties by nuclide of criticality, H. Iwamoto et al., JAEA-Research 2014-033 (2015).

Present Status of MA Nuclear Data

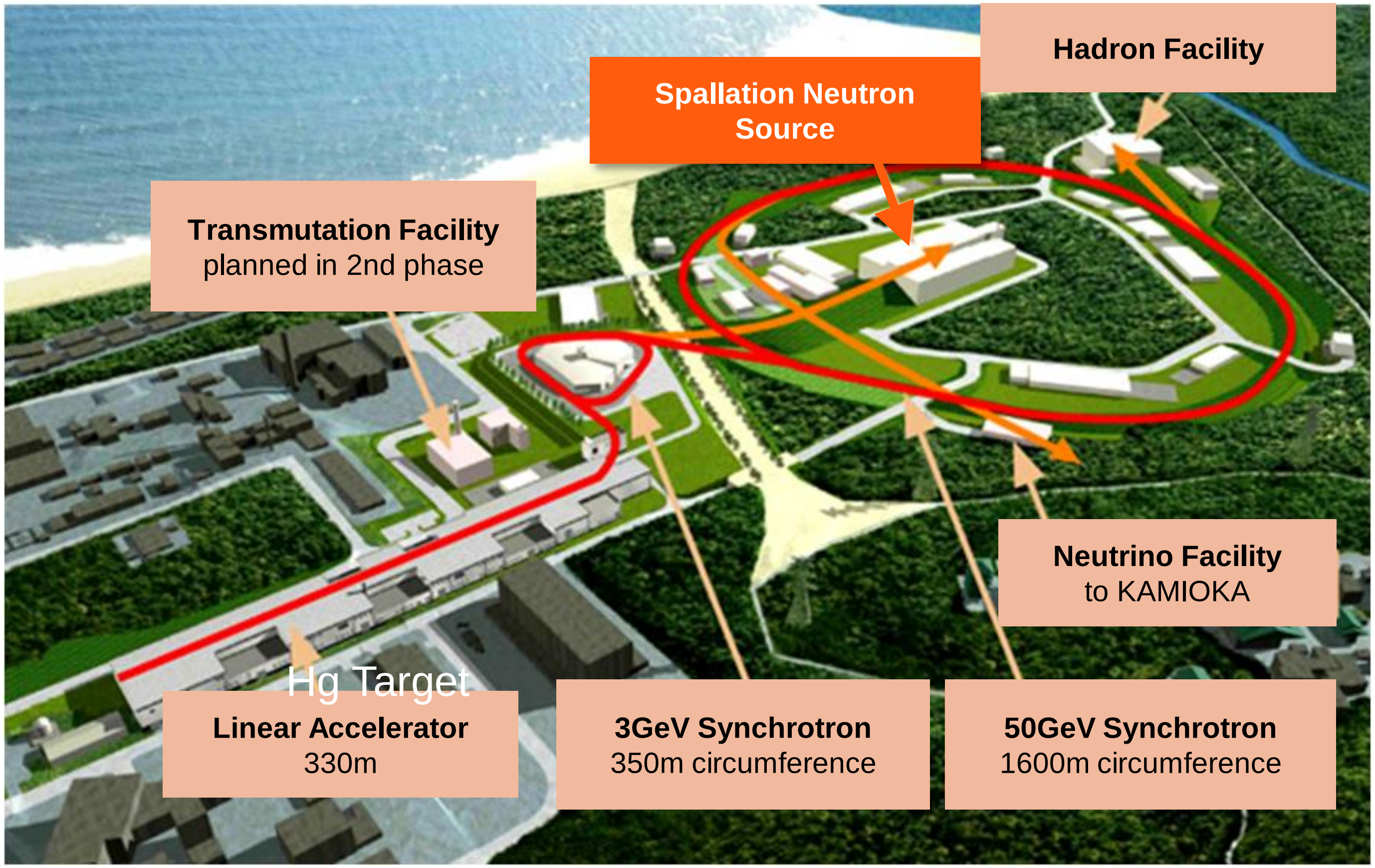
- **Uncertainties:**
  Present: >10%, Requirement: 2-3%
- **Previous works**
  - Less experimental data sets
  - Large disagreement between measured data
- **Large uncertainties of MAs are caused by:**
  - Background of decay γ-rays from radioactive samples
  - Difficult radioactive sample characterization such as total mass and impurities



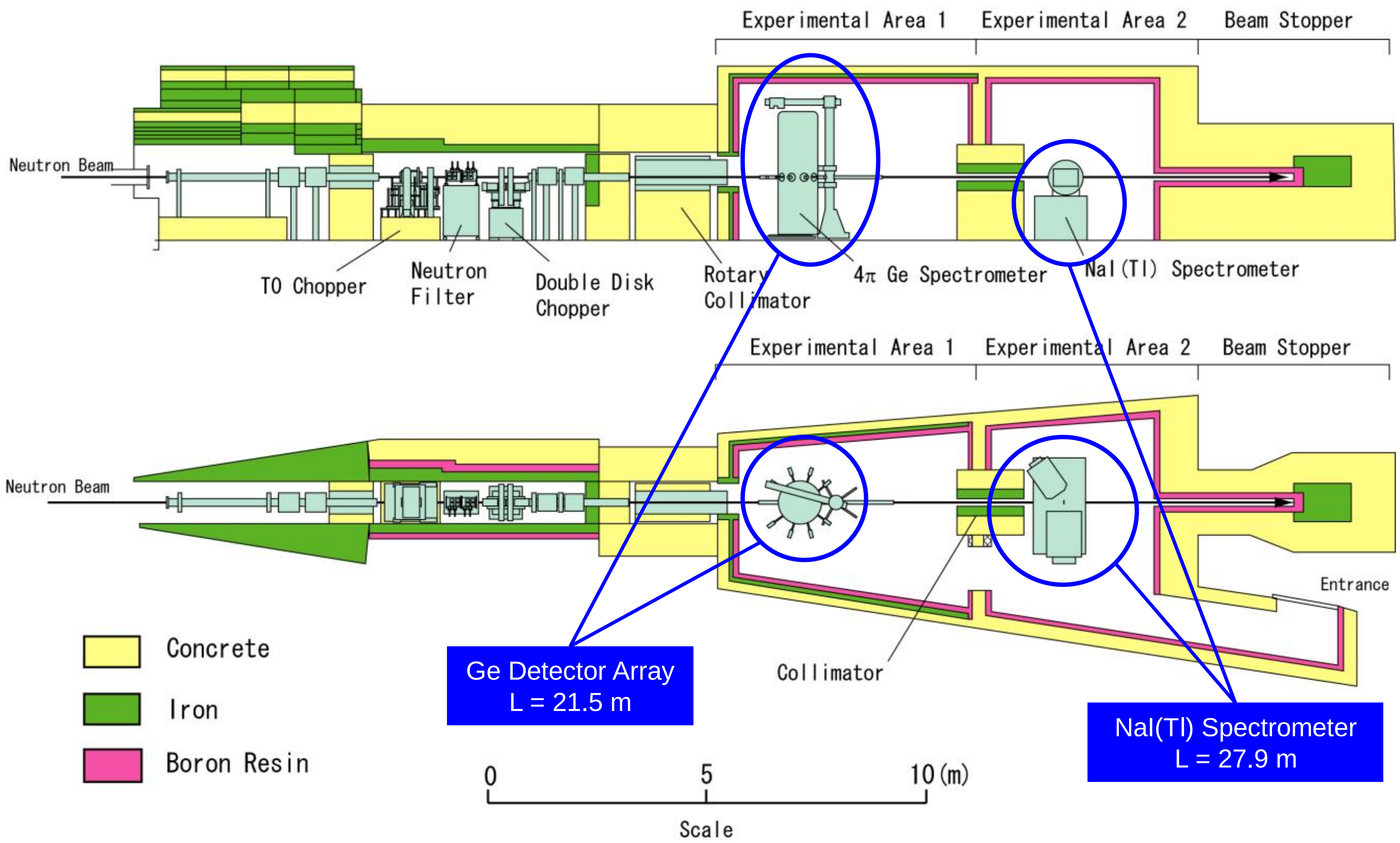
Neutron capture cross section of ^{243}Am

J-PARC

- Japan Proton Accelerator Research Complex (J-PARC)
- Spallation neutron source
  in Material and Life Science Experimental Facility (MLF)
- 3-GeV protons injected to a mercury target.



ANNRI



Experimental Area 1
Experimental Area 2
Beam Stopper
Neutron Beam
T0 Chopper
Neutron Filter
Double Disk Chopper
Rotary Collimator
4π Ge Spectrometer
NaI(Tl) Spectrometer
Experimental Area 1
Experimental Area 2
Beam Stopper
Neutron Beam
Entrance
Collimator
Concrete
Iron
Boron Resin
Ge Detector Array
L = 21.5 m
NaI(Tl) Spectrometer
L = 27.9 m
0
5
10 (m)
Scale

Issues and Tasks

- Issues

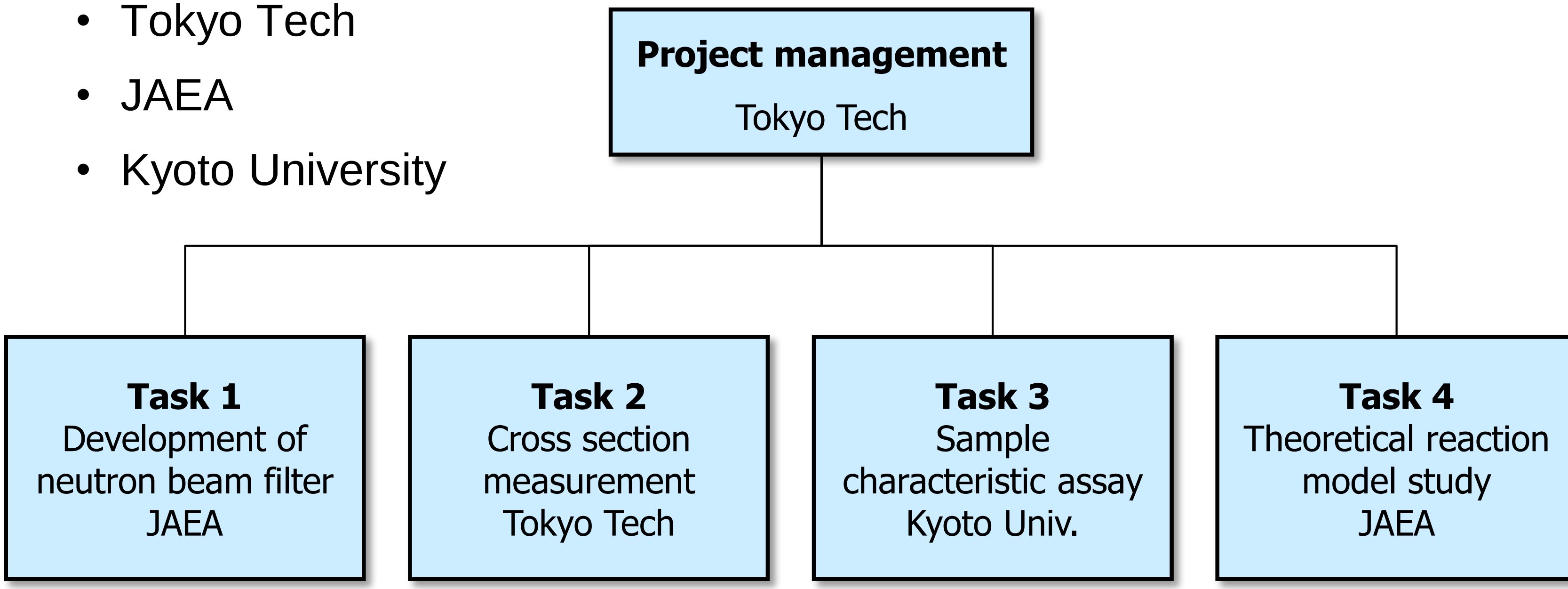
1. Double bunch beam structure of the proton beam pulse at J-PARC
2. Fast data acquisition for intense pulsed neutron beam at J-PARC
3. Impurity assay of MA samples

- Solutions and tasks

1. Development of neutron beam filter
   Solution of Issue 1
2. Cross section measurement of NaI(Tl)
   Solution of Issue 2
3. Sample characteristics assay
   Solution of Issue 3
4. Theoretical nuclear reaction model study
   Nuclear data evaluation for nuclear transmutation

Project Team Structure

- The project consists of four tasks:
  1. Development of neutron beam filter system in J-PARC
  2. Neutron capture cross section measurement at J-PARC
  3. Sample characteristic assay
  4. Theoretical reaction model study
- Institutes
  - Tokyo Tech
  - JAEA
  - Kyoto University



Plan

- Development and preparation were done from 2017 to 2018.
- Actual measurement will start from 2019.

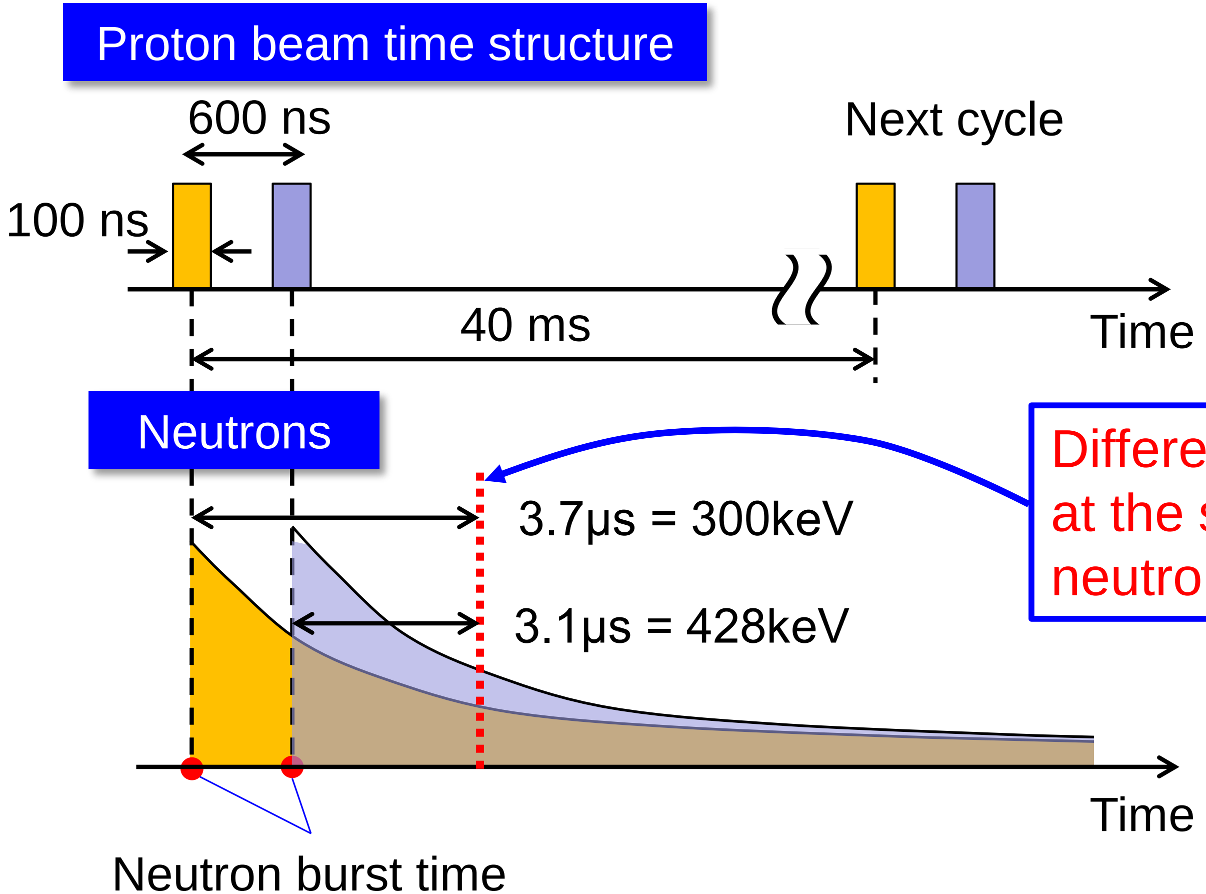
Tasks	2017 FY	2018 FY	2019 FY	2020 FY
1. Development of neutron filter	Design, fabrication, set up and test		Measurement of neutron spectrum	
2. Cross section measurement with NaI(Tl) detectors	Development and test		Cross section measurement at J-PARC	
		Sample rotation system		
3. Sample characteristic assay	Development		Sample preparation	Sample assay
4. Nuclear reaction model study	Development of methodology		Theoretical analysis	Evaluation

Development of Neutron Beam Filter

Double Bunch Beam at J-PARC

Double bunch operation

J-PARC is operated at a special mode that two proton beam pulses are injected into the spallation target every 40 ms.

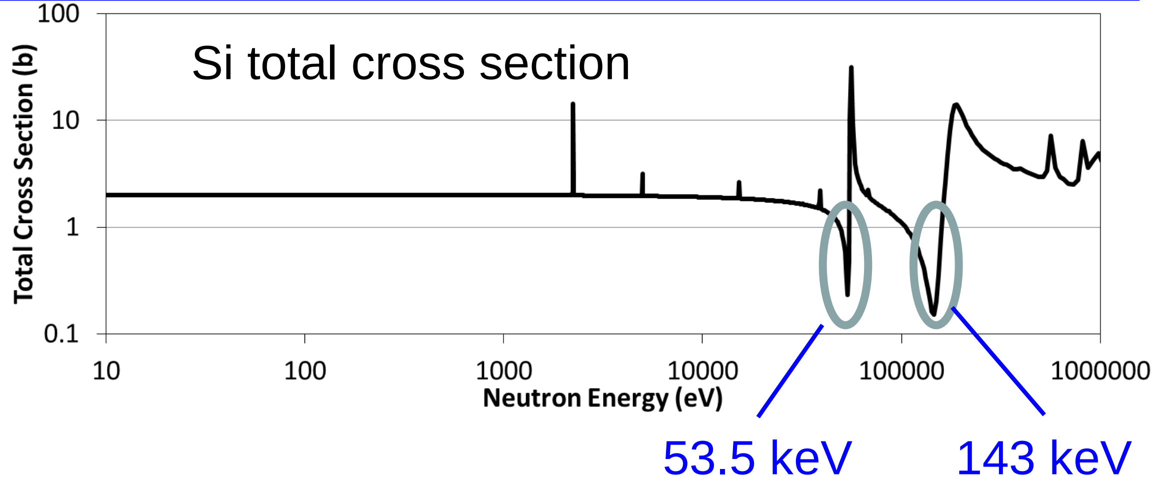


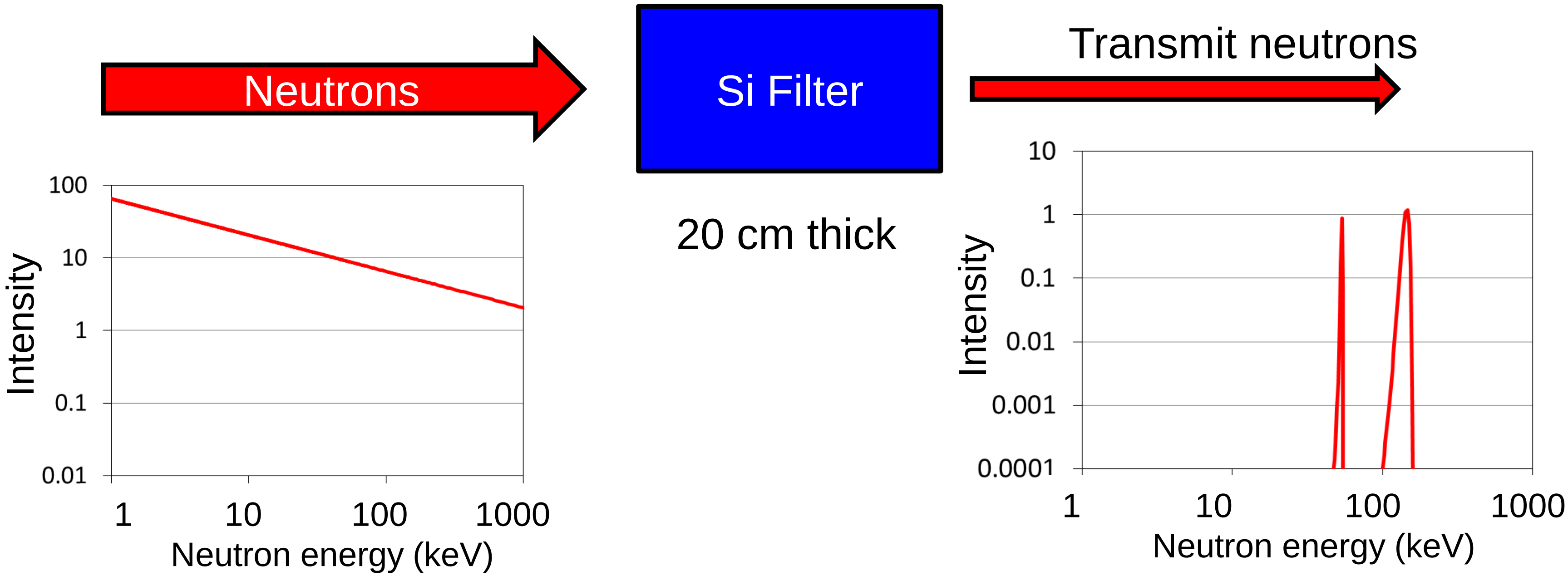
Pulse width 100 ns
Repetition rate 25 Hz
Cycle 40 ms

Different energy neutrons exist at the same time due to two neutron burst times.

Neutron Beam Filter Technique

A neutron beam filter allows neutrons to pass only at energies for cross section minimums

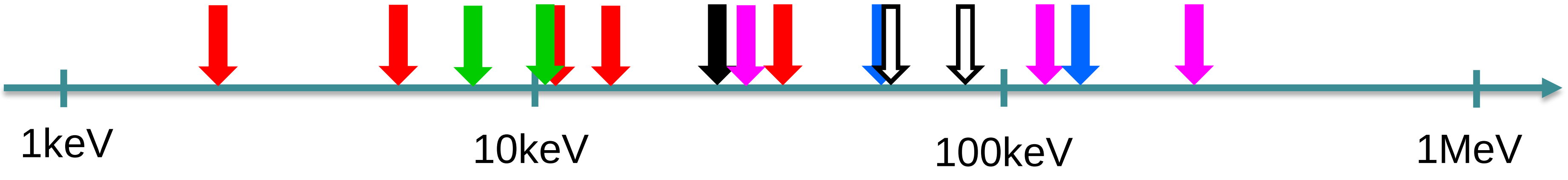




Selection of Filter Materials

Materials selected for neutron beam filters

	Material	En (keV)	Thickness (cm)
Fe filter	Fe	24	30
Bi filter	Bi	2.2, 5.1, 11.5, 15.3, 32.9	30
Al filter	Al	27, 125, 265	60
Si filter	Si	54, 146	40
Cr filter	Cr	56, 82	20
Sc filter	Sc_2O_3	7.5, 10.5	$100 g/cm^2$

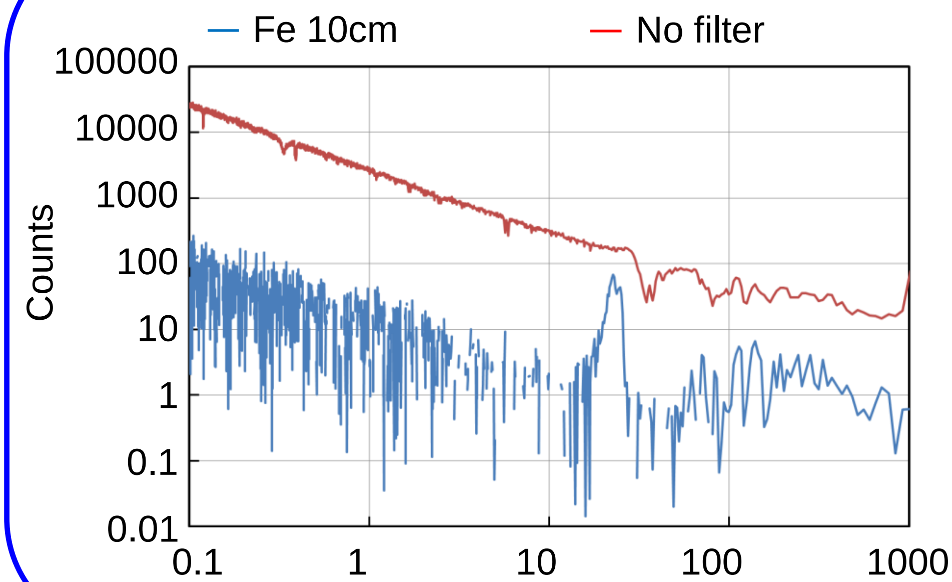


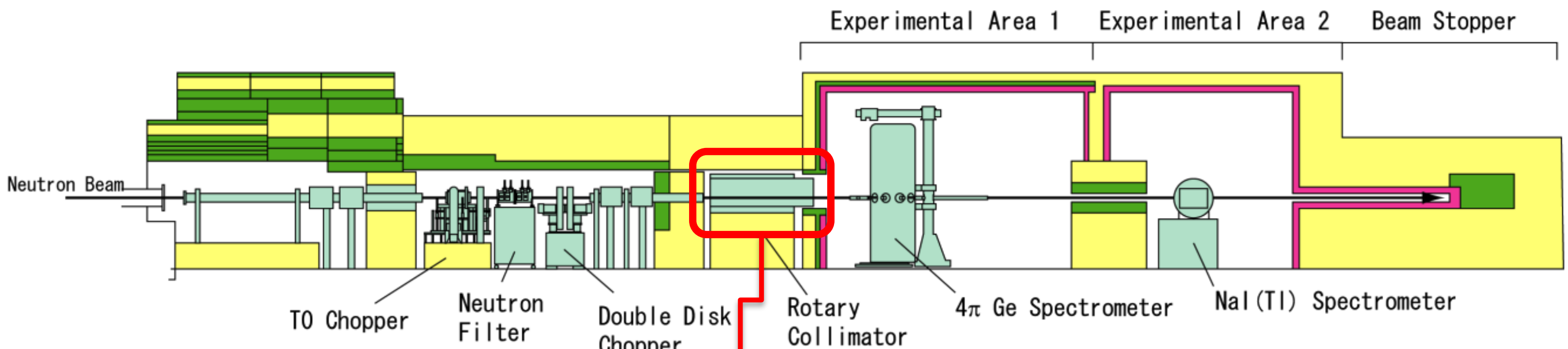
Test Experiments

Filtered materials were tested at
- J-PARC ANNRI (right)
- $^{7}Li(p,n)^{7}Be$ neutron source at Tokyo Tech (bottom)

Examples for 10-cm Fe filter are shown in figures.

J-PARC ANNRI



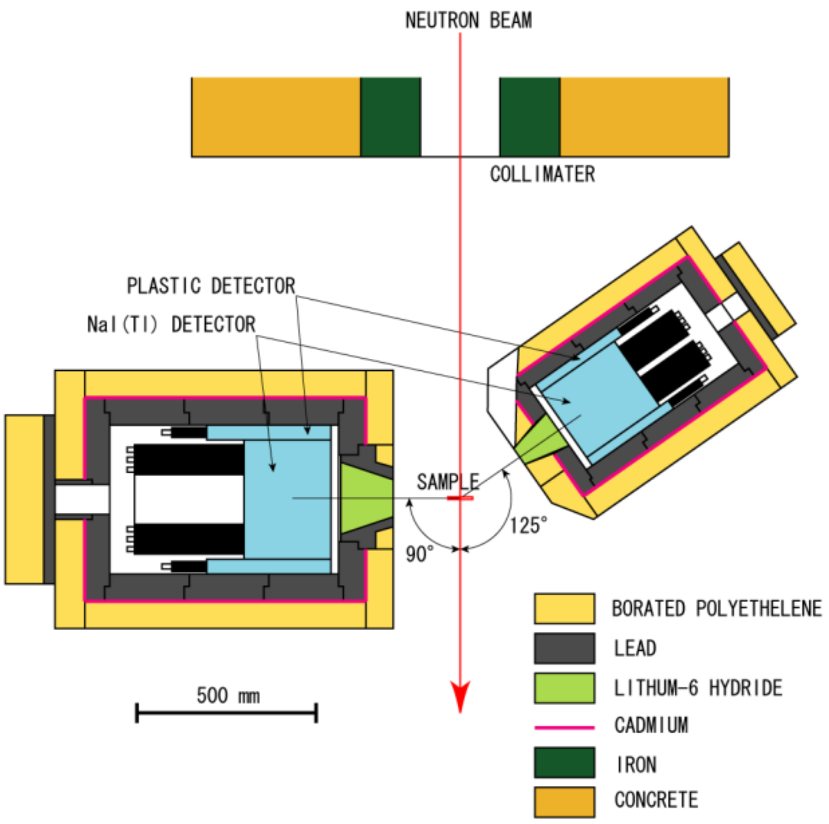


The neutron beam filter was inserted into one of the rotary collimator holes.

Cross Section Measurement with NaI(Tl) Detectors

NaI(Tl) Spectrometer

- **Use for:**
  - Complementary use to Ge array
  - Faster response: can be used in the high energy range
- **NaI(Tl) detectors**
  - 90° detector: 13” diam. × 8” long
  - 125° detector: 8” diam. × 8” long
- **Shielding**
  - Borated polyethylene, Pb, ^{6}LiH, Cd
- **Data analysis**
  - The pulse-height weighting technique is established.



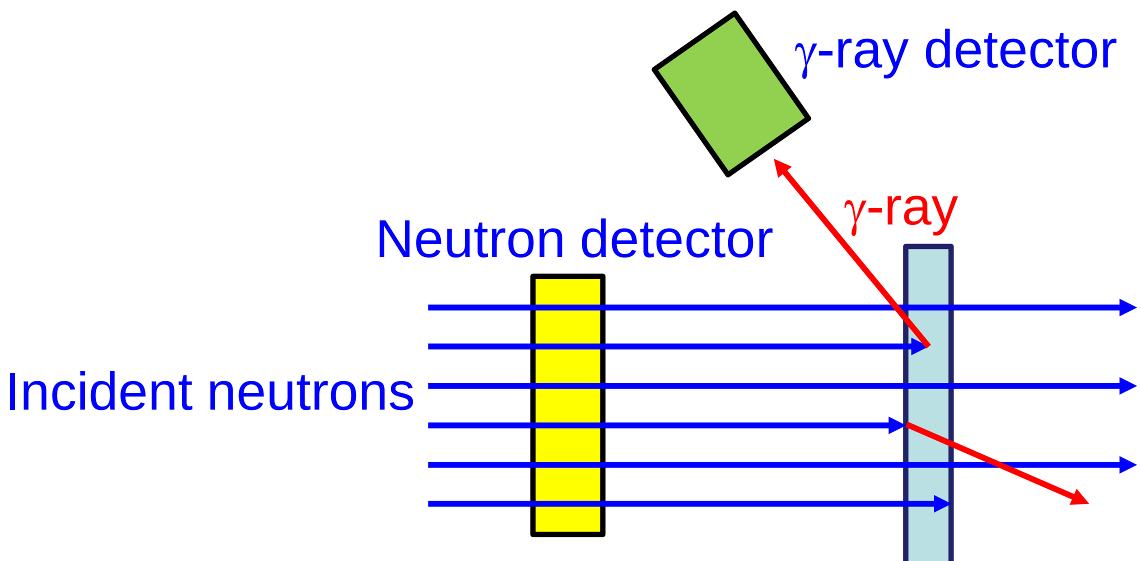
Neutron capture yield and cross section

Cross section σ (cm^2) $\sigma = \dfrac{Y}{\phi\xi}$

Neutron capture yield

Number of incident neutrons

Area density of sample atoms (atoms/cm^2)



γ-ray counts → Y

$$N_\gamma \rightarrow Y = \frac{N_\gamma}{\varepsilon_\gamma}$$

Neutron counts → ϕ

$$N_n \rightarrow \phi = \frac{N_n}{\varepsilon_n}$$

Sample

Sample mass and area → ξ

Origin of uncertainties

Cross section σ (cm²) $\sigma = \dfrac{Y}{\phi\xi}$

Neutron capture yield

Number of incident neutrons

Area density of sample atoms (atoms/cm²)

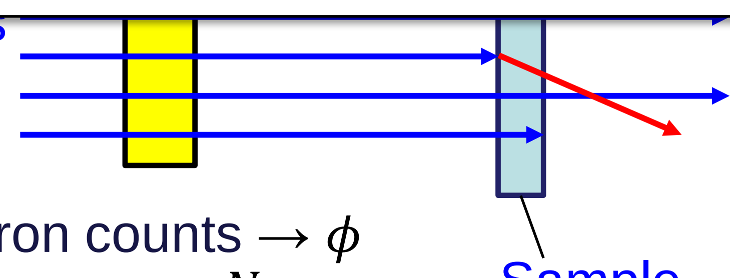
Statistical uncertainties of N_γ, N_n

Systematic uncertainty:

- Detection efficiency of γ-ray detector ε_γ
- Detection efficiency of neutron detector ε_n
- Sample mass, area and area density

γ-ray counts → Y

$$N_\gamma \rightarrow Y = \frac{N_\gamma}{\varepsilon_\gamma}$$



Neutron counts → ϕ

$$N_n \rightarrow \phi = \frac{N_n}{\varepsilon_n}$$

Sample mass and area → ξ

Relative measurement to well-known cross section data

$$\sigma = \frac{Y}{\phi\xi} \times \frac{\phi_{st}\xi_{st}}{Y_{st}} \times \sigma_{st}$$

Standard cross section

$$= \frac{\varepsilon_\gamma N_\gamma}{\varepsilon_\gamma N_{\gamma.st}} \times \frac{\varepsilon_n N_{n.st}}{\varepsilon_n N_n} \times \frac{\xi_{st}}{\xi} \times \sigma_{st}$$

$$= \frac{N_\gamma}{N_{\gamma.st}} \times \frac{N_{n.st}}{N_n} \times \frac{\xi_{st}}{\xi} \times \sigma_{st}$$

- Standard cross section: *eg*. ^{197}Au(n,γ)^{198}Au
- First resonance peak
- Thermal cross section

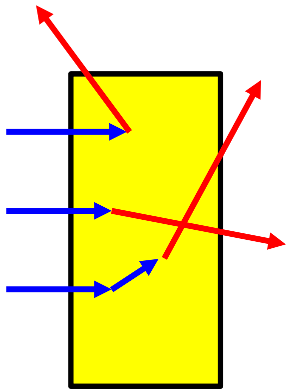
Pros: easy and can be used for many nuclides

Cons: uncertainties of nuclear data introduced.

Absolute measurement: Saturated resonance method

Capture yield with neutron self shielding

$$Y = \phi \frac{\sigma_c}{\sigma_t}(1 - e^{-\xi\sigma_t})$$



When the sample is very thick,

$$Y = \phi \frac{\sigma_c}{\sigma_t}$$

In addition, scattered neutrons are captured in the multiple scattering process. Then,

$$Y = \phi$$

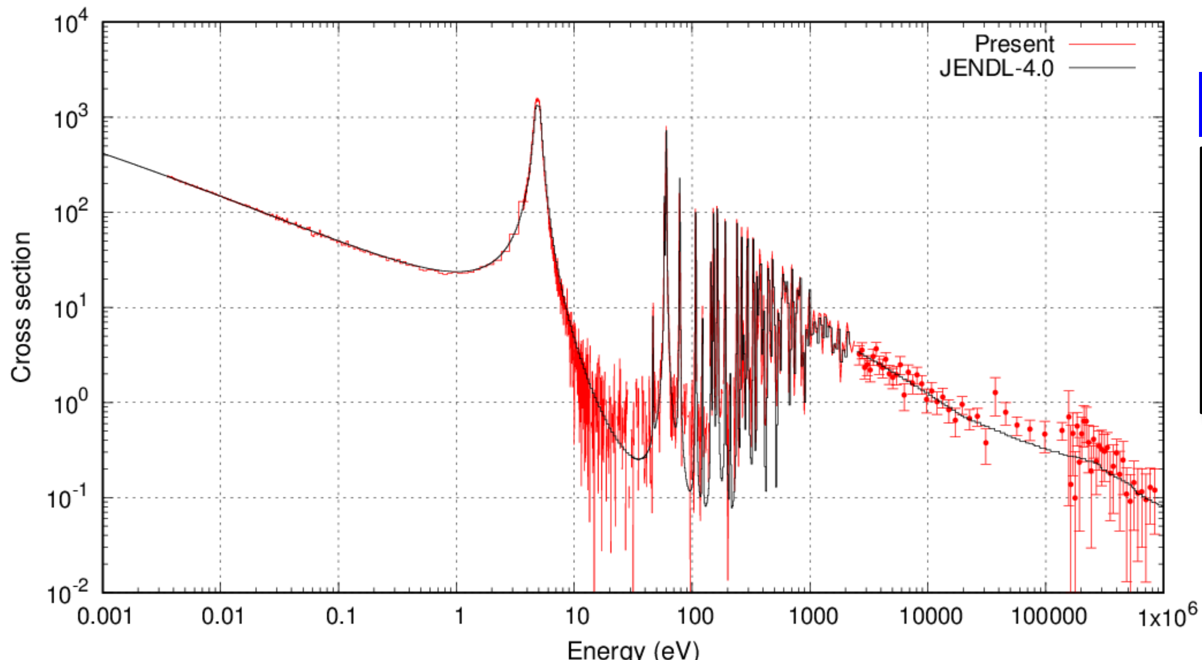
This constraint can be used to determine the absolute value of neutron capture cross section.

$$\sigma = \frac{Y}{\phi\xi} \rightarrow \frac{1}{\xi}$$

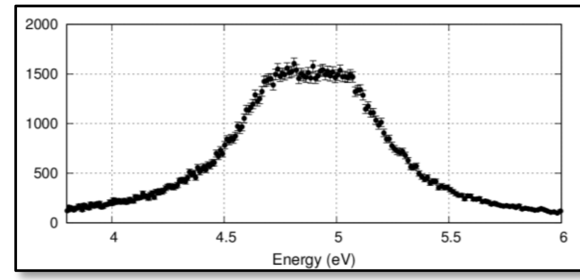
or

$$\frac{Y}{\phi} = \frac{N_\gamma}{\varepsilon_\gamma}\frac{\varepsilon_n}{N_n} = 1 \ \therefore \frac{\varepsilon_n}{\varepsilon_\gamma} = \frac{N_n}{N_\gamma}$$

Example of saturated resonance method : $^{197}Au(n,\gamma)^{198}Au$



First resonance @ 4.9 eV



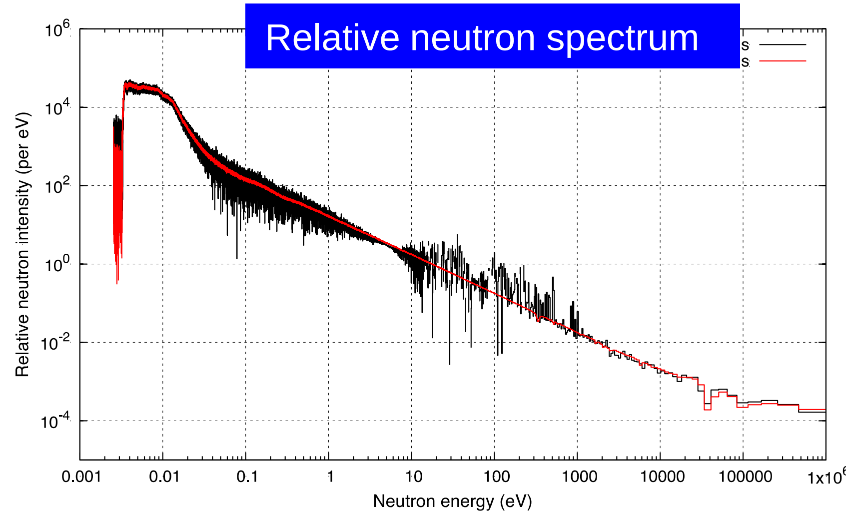
$$\frac{\varepsilon_n(4.9\ \mathrm{eV})}{\varepsilon_\gamma} = \frac{N_n}{N_\gamma}$$

$$\varepsilon_n(E_n) \propto f(E_n)$$

$$\varepsilon_n(E_n) = \frac{\varepsilon_n(4.9\ \mathrm{eV})}{\varepsilon_\gamma} f(E_n)$$

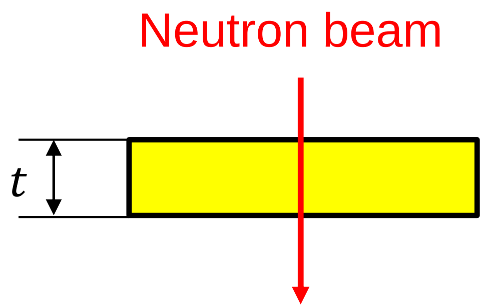
Pros: No nuclear data is needed.

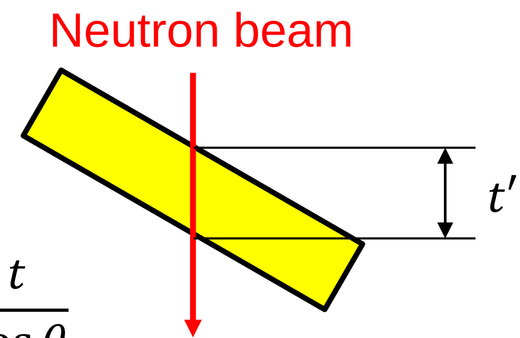
Cons: Thick sample is needed.



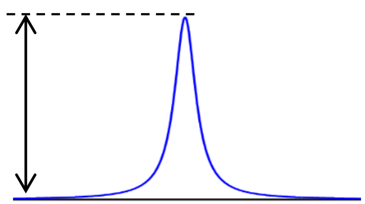
New method: sample rotation method

Self-shielding ratio can be determined by changing the effective thickness of sample by rotation



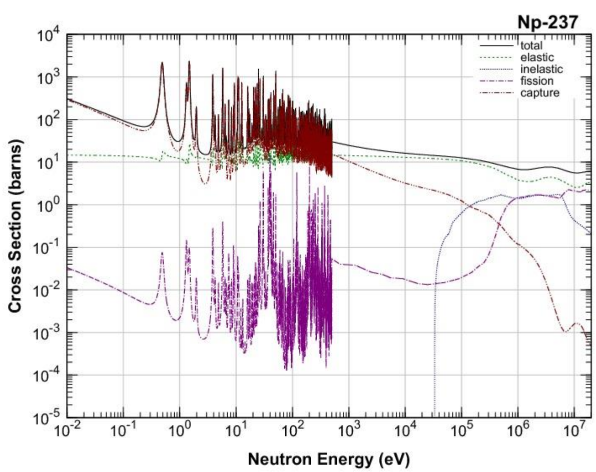


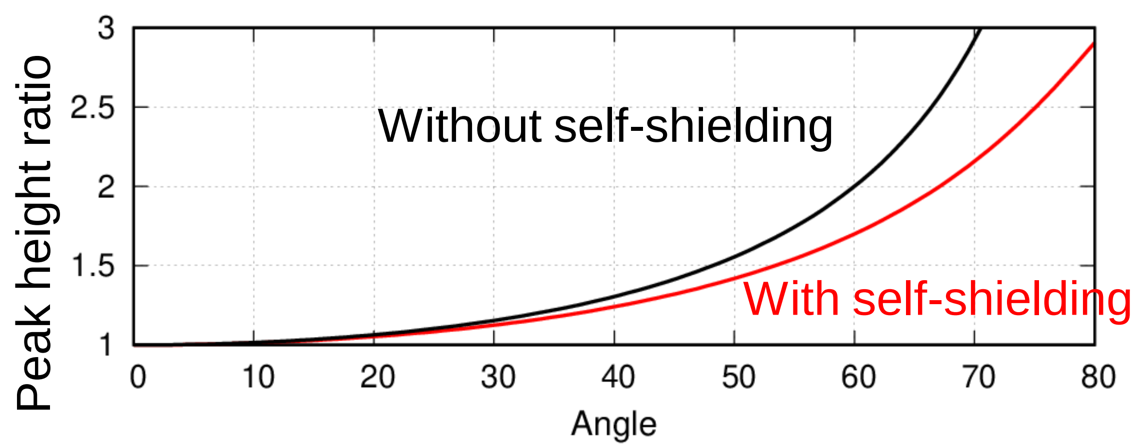
$$t' = \frac{t}{\cos\theta}$$



First resonance peak
^{237}Np: 0.49 eV, ^{241}Am: 0.30 eV
^{243}Am: 1.35 eV

$\frac{Y}{\phi}(< 1)$ can be determined.





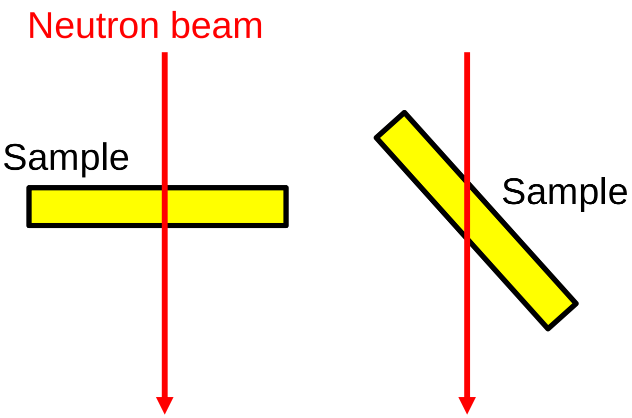
System upgraded

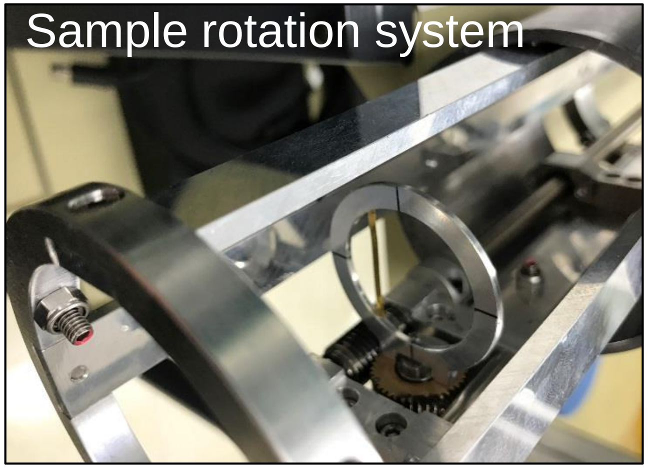
Fast data acquisition

- ✓ A waveform digitizer board (CAEN V1720) is used for fast data acquisition. The time-of-flight and the pulse height of each event are calculated from offline analysis.
- ✓ Count loss at a high counting rate is minimized.

Sample rotation system

- ✓ A sample rotation system was designed and built to change the sample thickness by rotating the sample.
- ✓ Different sample thickness measurements can be achieved without purchasing multiple samples.





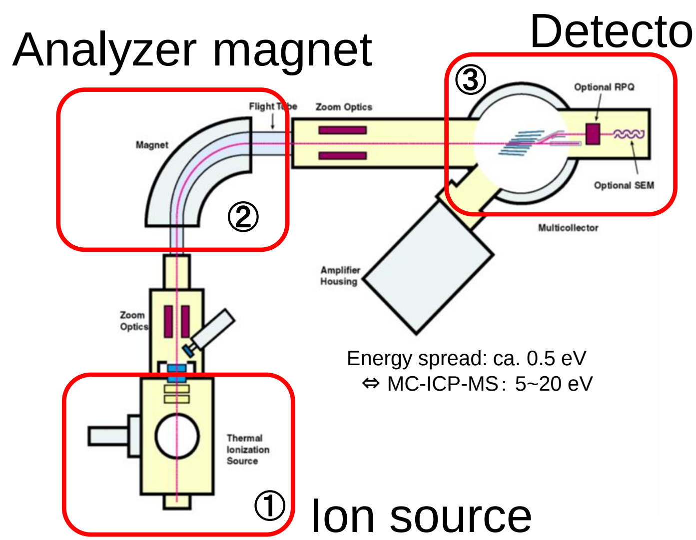
CAEN V1720

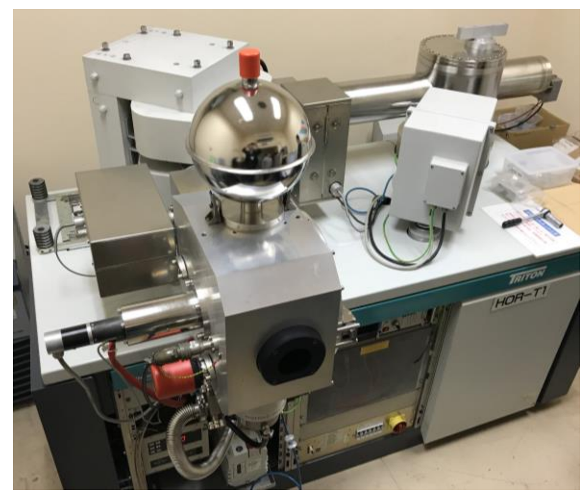
Sample Characteristic Assay

Sample Characteristic Assay

Thermal ionization mass spectrometer（TIMS）

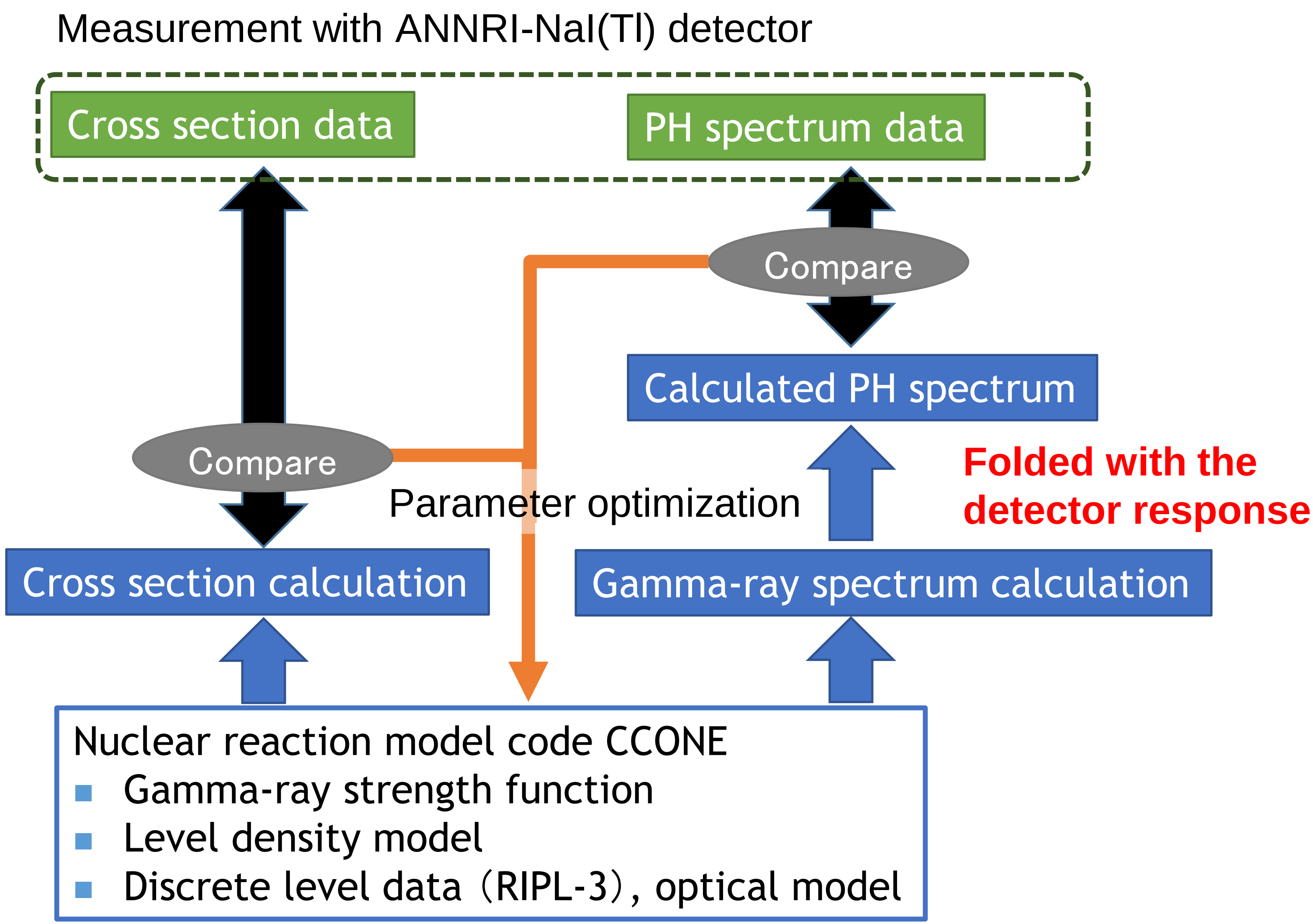
- We use a TIMS of Kyoto University to analyze the isotope compositions and impurities of samples.
- Unsealed MA solution taken from the same batch as the sealed MA samples were prepared.
- In the past two years, the TIMS was upgraded to improve its vacuum system and stability control of the beam intensity.
- In the previous project, Pu impurities were found in Am samples. To analyze the Pu impurities precisely, standard Pu solution is planned to purchase in 2019.
- The target precision is less than 1%.





Nuclear Reaction Model Study

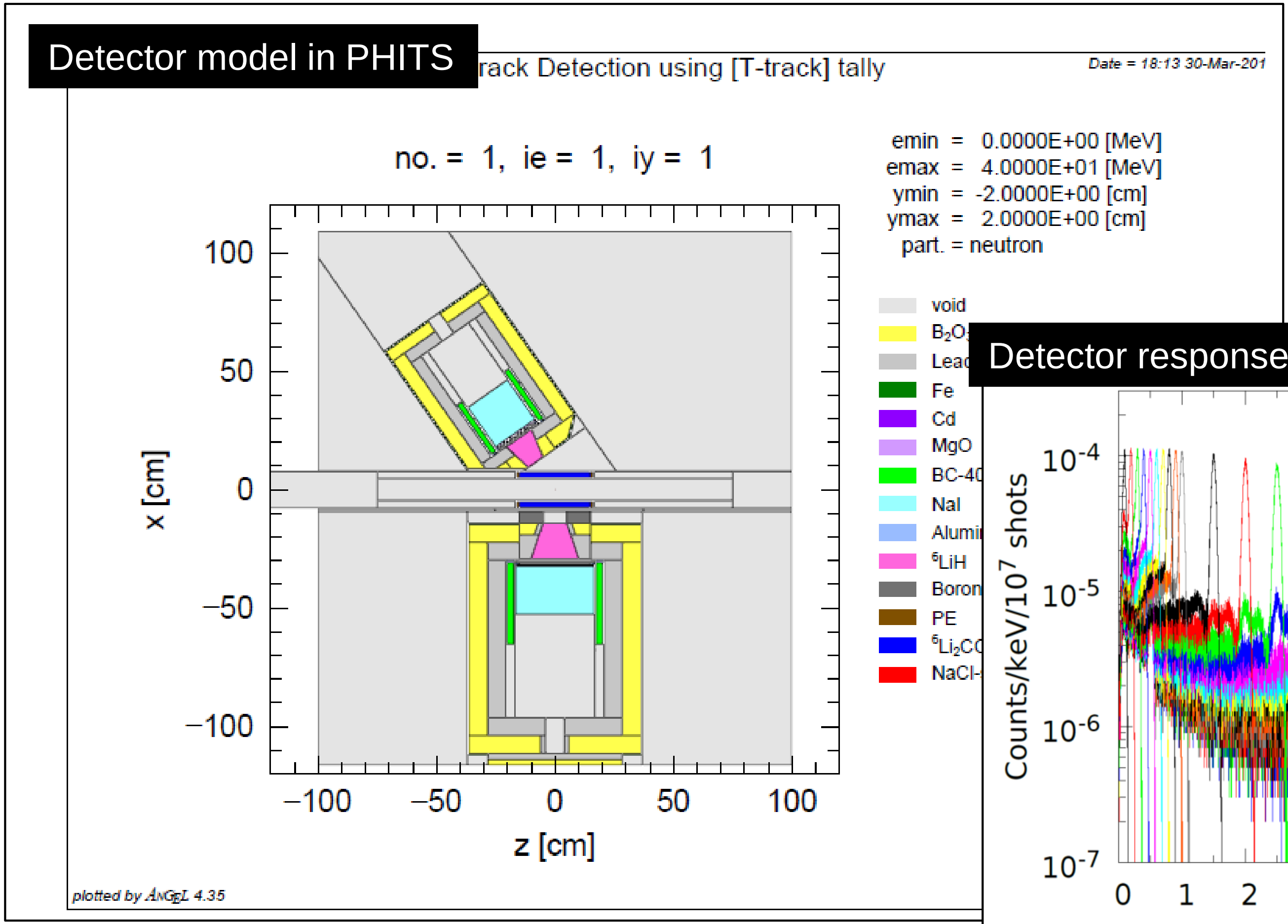
Evaluation Flow



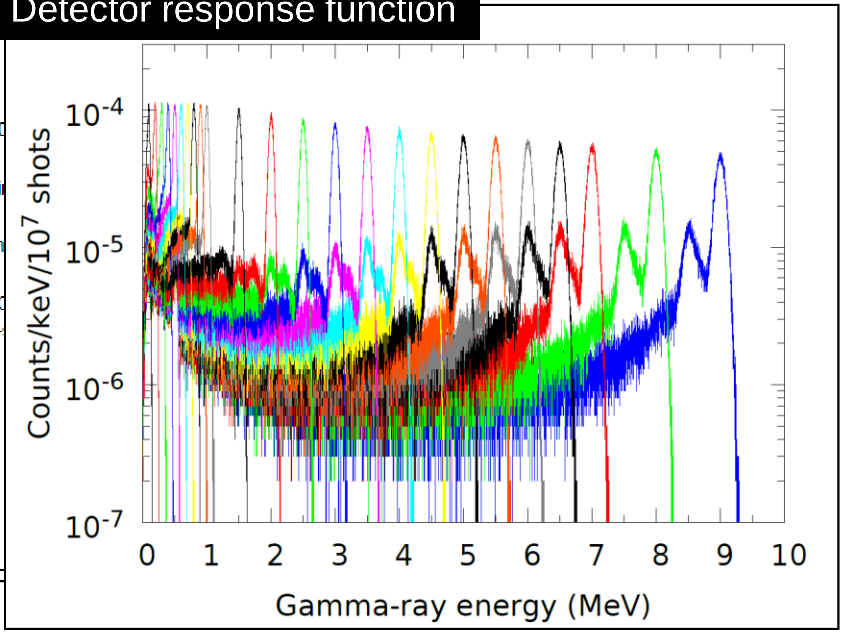
Calculation of Detector Response Function

Monte Carlo simulation code PHITS was used to calculate detector response functions.

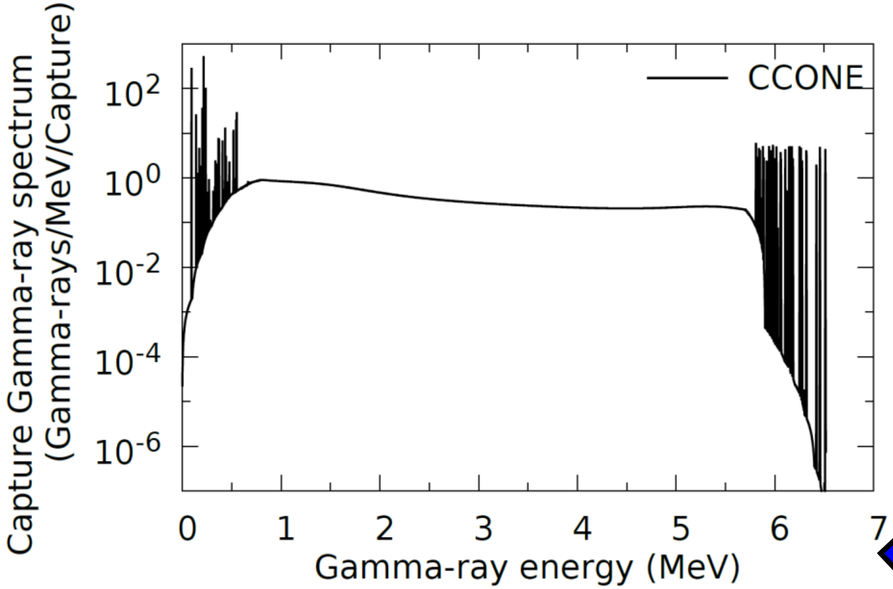
Detector model in PHITS



Detector response function

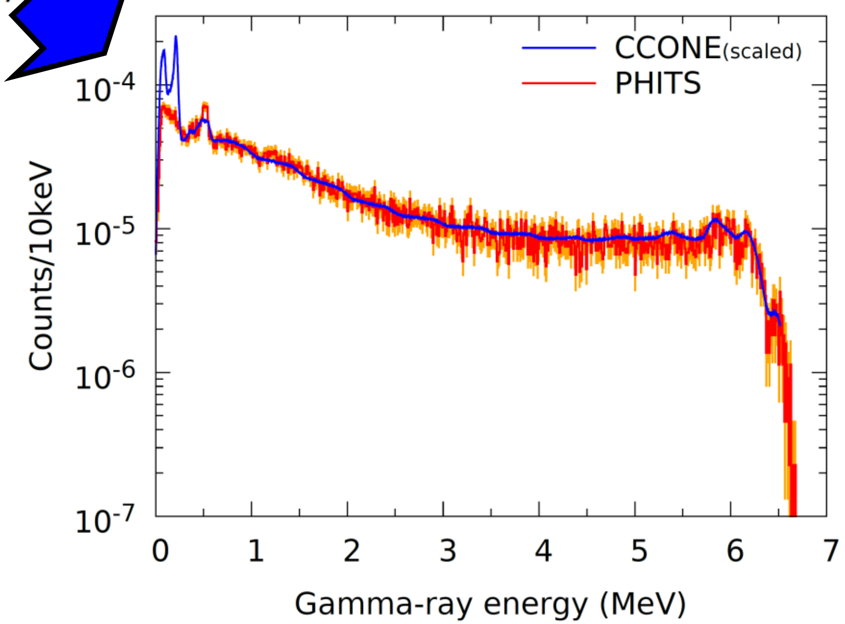


Folding with Detector Response Functions



Gamma-ray **spectrum** calculate with CCONE

Folding



Gamma-ray spectrum folded with detector response functions

Summary

- A project "*Study on accuracy improvement of fast-neutron capture reaction data of long-lived MAs for development of nuclear transmutation systems*" is ongoing as a joint project of the three institutes, Tokyo Tech, JAEA and Kyoto University.
- The project focuses on neutron capture reaction of Mas, especially ^{237}Np, ^{241}Am and ^{243}Am, in the fast neutron energy region. An intense neutron beam from a spallation source of J-PARC will be employed to improve the neutron capture data of MA.
- The past two years were spent for development and preparation. Actual measurement will start in 2019.

Acknowledgements

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