Accuracy Improvement of Neutron Nuclear Data on Minor Actinides - AIMAC project -

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Present study includes the result of "Research and Development for accuracy improvement of neutron nuclear data on minor actinides" entrusted to the Japan Atomic Energy Agency by the Ministry of Education, Culture, Sports, Science and Technology of Japan (MEXT).

Contents

1) Accurate Nuclear Data Needs

2) Current Nuclear Data Status

3) Rule of State-of-Art facility

J-PARC/MLF/ANNRI

4) AIMAC project toward Accuracy

5) Related International Activity

OECD/NEA/WPEC /SG-41

Accurate Nuclear Data Needs

Data needs for advanced reactor systems such as FR, ADS, et al. were quantitatively deduced by M. Salvatores et al., OECD/NEA/WPEC/SG-26 Report (2008).

For example, FR and ADMAB need 0.3% accuracies on Multiplication factor k_{eff} in order to reduce margins, both for economic and safety reasons.

$$\frac{\delta k}{k} \Rightarrow \frac{\delta \boldsymbol{\sigma}}{\boldsymbol{\sigma}}$$

Examples of $\frac{\delta \boldsymbol{\sigma}}{\boldsymbol{\sigma}}$

Capture Cross Section of Am-241

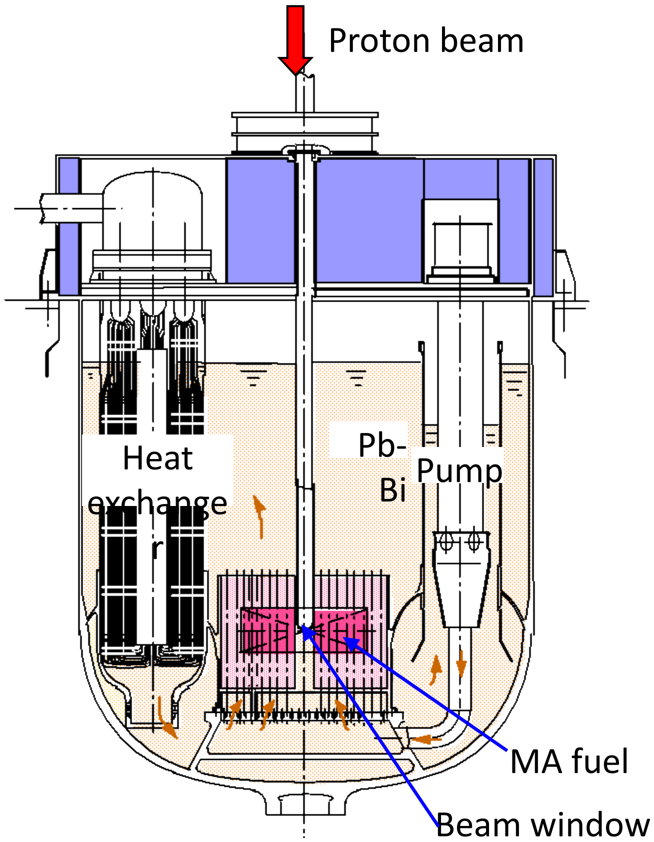
Advanced Reactor Type	Current Accuracy	Required Accuracy
GFR	8 %	3 %
ADMAB	8 %	2 %

Other MA	Current		Required
Am-242m	25%	→	12%
Am-243	10%	→	2%
Cm-244	20%	→	6%
Np-237	6%	→	3%

Current Accuracy 2-5 times larger than Required Accuracy

Ref. OECD/NEA Report NEA No. **6410** (2008)

The difference between calculated k_{eff} with JENDL-4.0 and 3.3 ?



Monte-Carlo Simulation

+

Evaluated Nuclear Data

(JENDL vs. ENDF vs. JEFF)

ADS proposed by JAEA

Courtesy of K. Tsujimoto (JAEA)

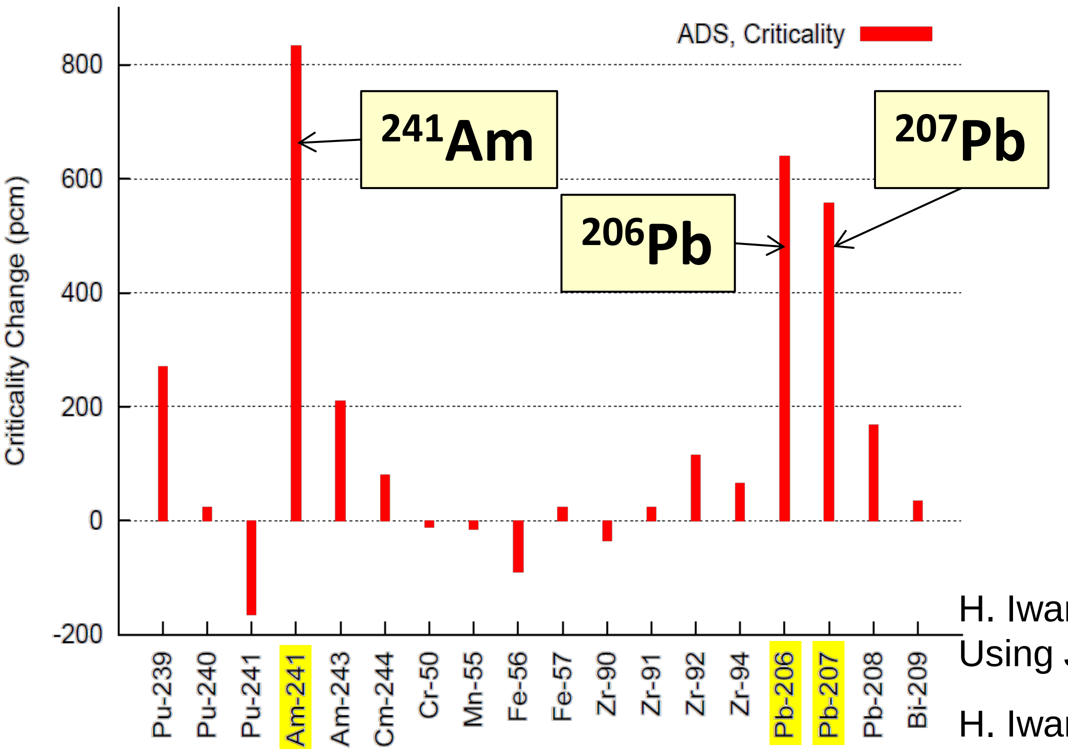
Ref. K. Tsujimoto at 2012 Symposium on Nuclear Data, November 15-16 2012.

Nuclide-wise contribution for the difference between calculated k_{eff} with JENDL-4.0 and 3.3

Criticality change 3% (>> 0.3%)

JENDL-3.3 → *JENDL-4.0*

0.971 +2,936 pcm **0.999**



Courtesy of K. Tsujimoto

Reaction-wise contributions of the criticality change

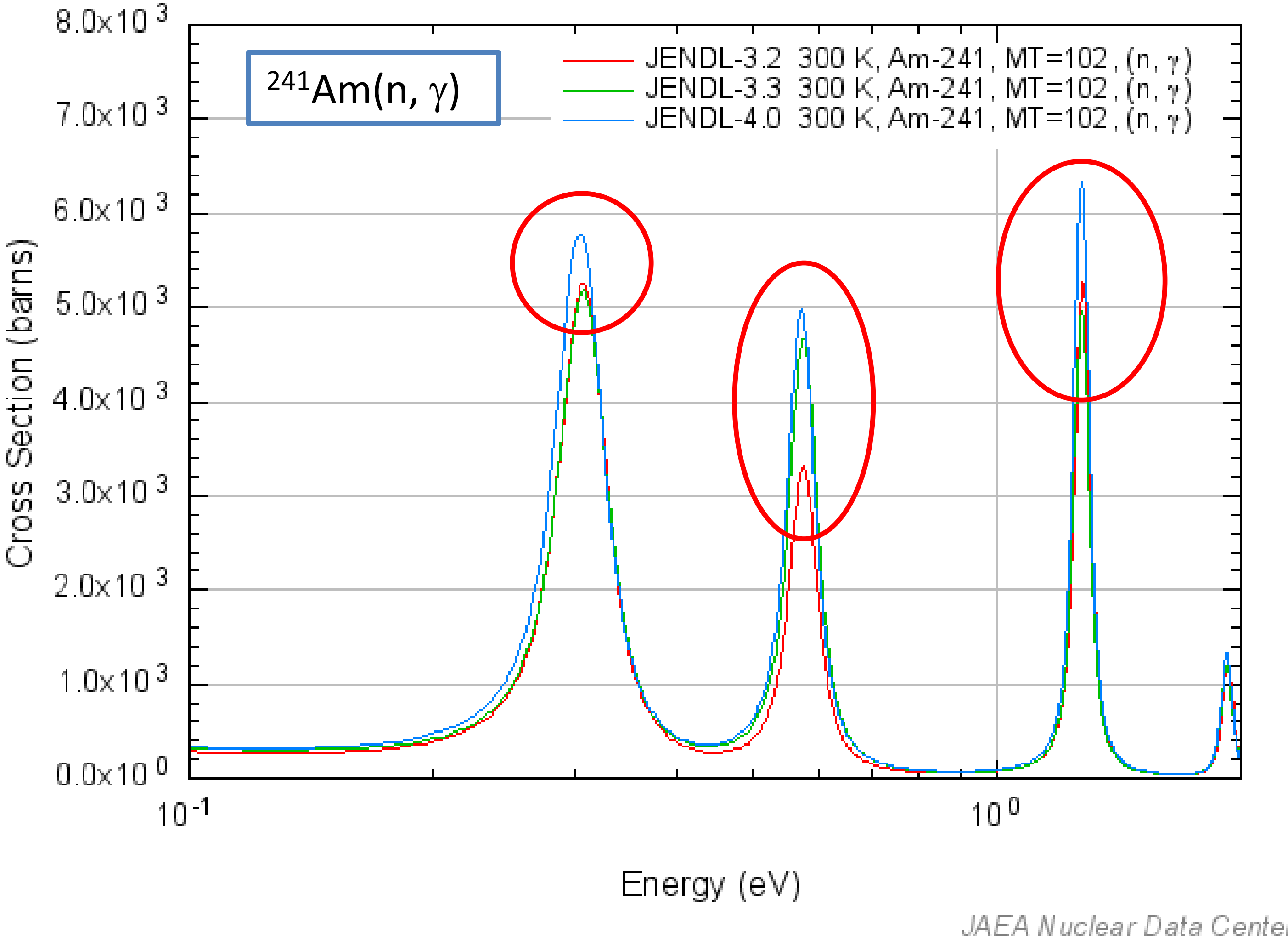
	fis	ν	cap	inl	el	μ
^{241}Am	76	208	255	293	1	1
^{206}Pb	—	—	-34	674	3	-5
^{207}Pb	—	—	-14	560	14	-4

H. Iwamoto, et.al, "Analysis of Transmutation Systems Using JENDL-4.0", JAEA-Research 2011-036.

H. Iwamoto, et.al, "Analysis of Transmutation Systems Using JENDL-4.0", ANS Annual Meeting, 2011 (2011).

Status of Evaluated Data

Resonance Region (^{241}Am case)



Recent Status of Evaluated Data

Thermal (^{241}Am case)

Evaluation	^{241}Am thermal neutron capture cross section
JEFF-3.1.2	647.2 b
ENDF/B IV → VII.0 → VII.1	581.5 → 619 → 684.3 b
JENDL-1 → 3.2 → 3.3 → 4.0	832 → 600 → 639.5 → 684.3 b

JENDL-4.0's evaluation 684.3 [b] is based on both of Activation and TOF

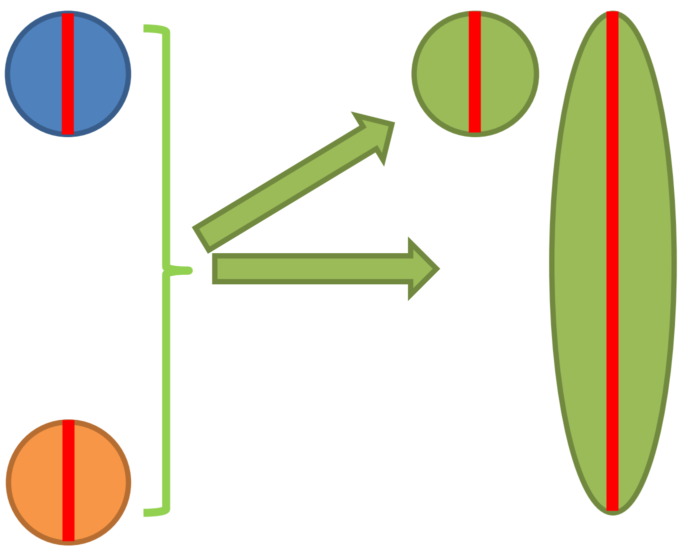
Current Nuclear Data Status

Sometimes, there are discrepancy between measurements

Evaluated nuclear data depends on experimental data

Experimental Uncertainty

- **Statistical Uncertainty**
- **Systematic Uncertainty**
- **<u>Unrecognized Bias effect</u>**

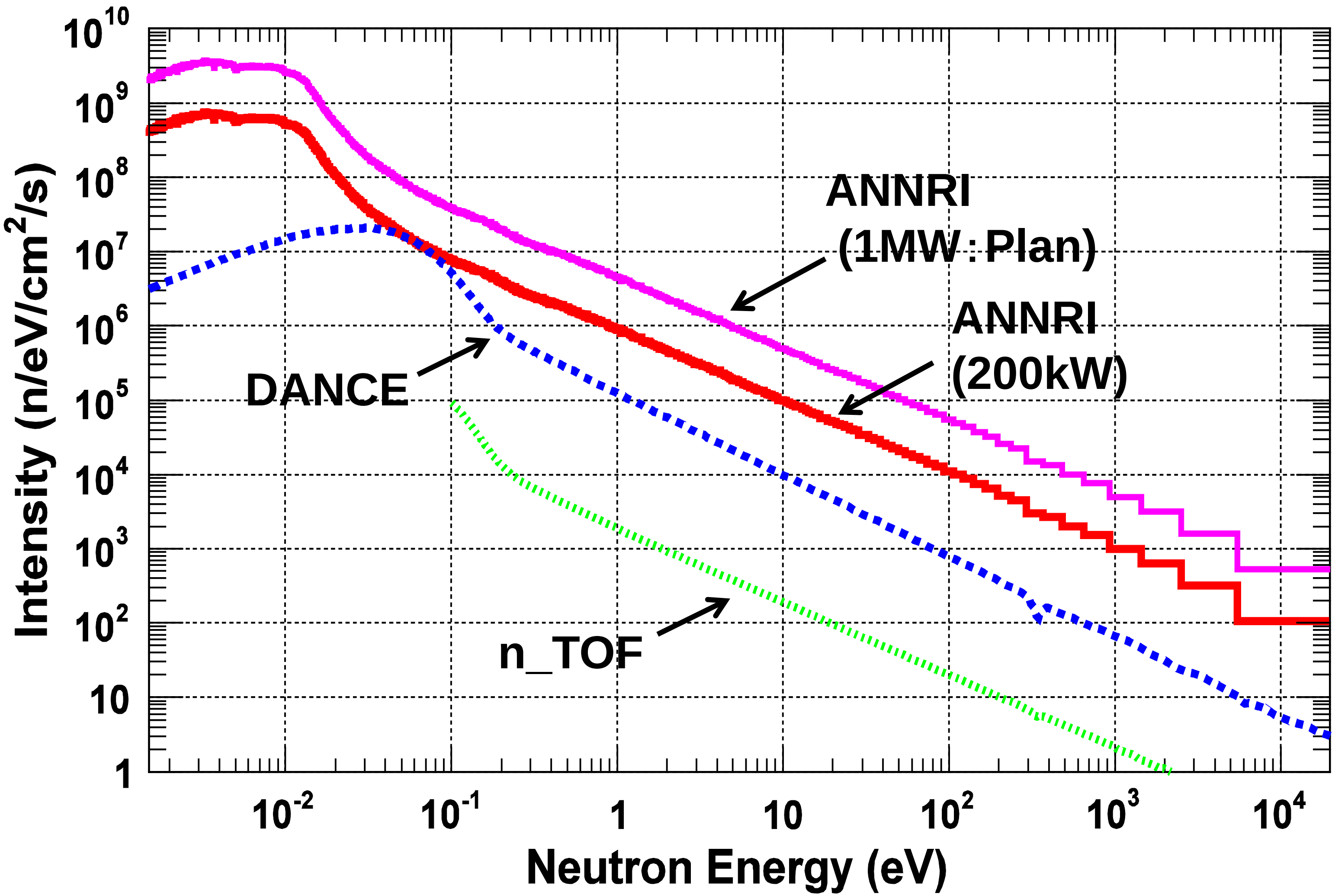


Important to solve Discrepancy for Accuracy

Rule of State-of-Art facility
J-PARC/MLF/ANNRI

State-of-Art TOF facility

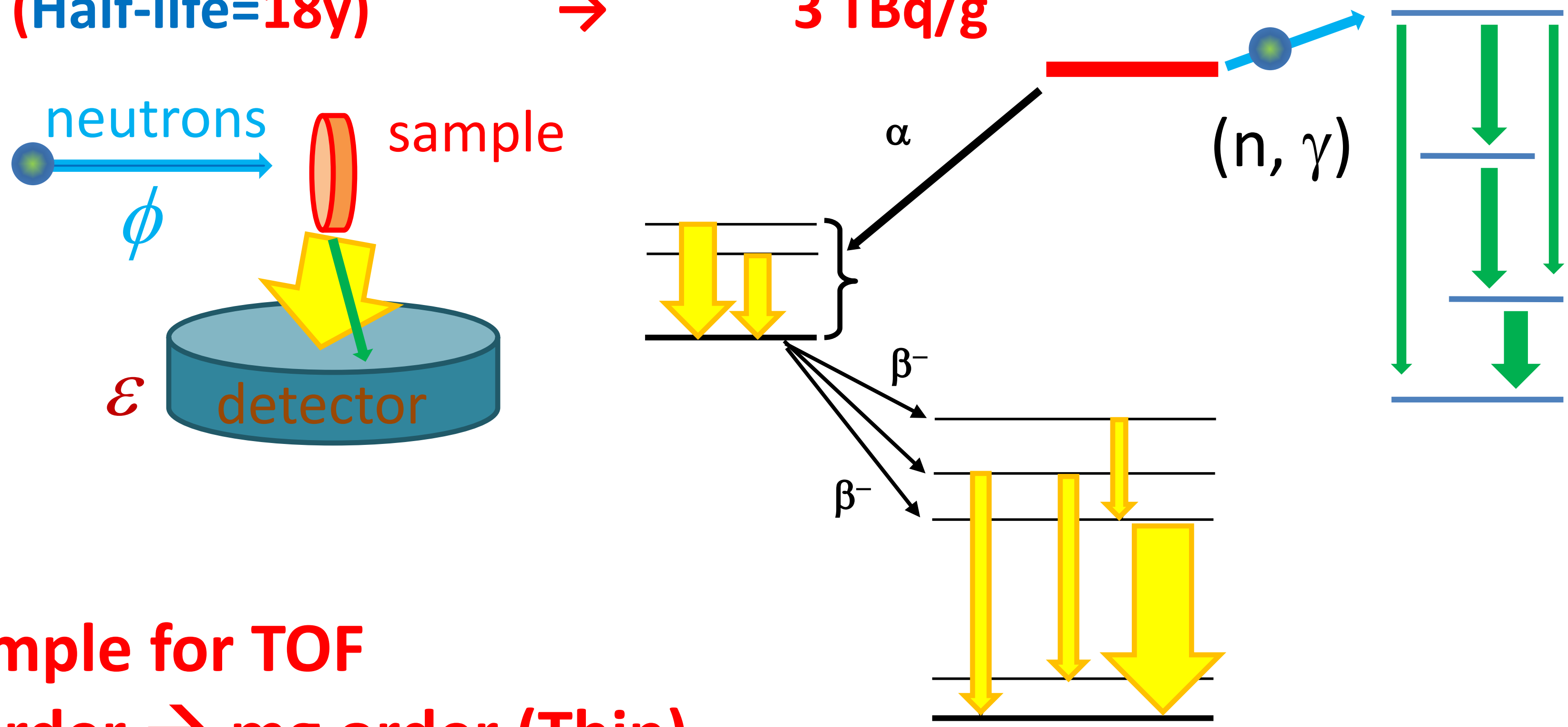
– High Intensity Pulsed Neutron Beam –



Possibility obtaining Capture data for highly radioactive nucleus by TOF

Example

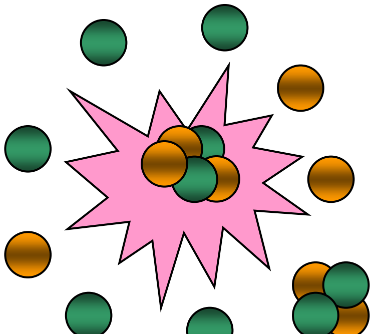
U235 (Half-life=704My) → 80 kBq/g
Am241(Half-life=432y) → 127 GBq/g
Cm244 (Half-life=18y) → 3 TBq/g

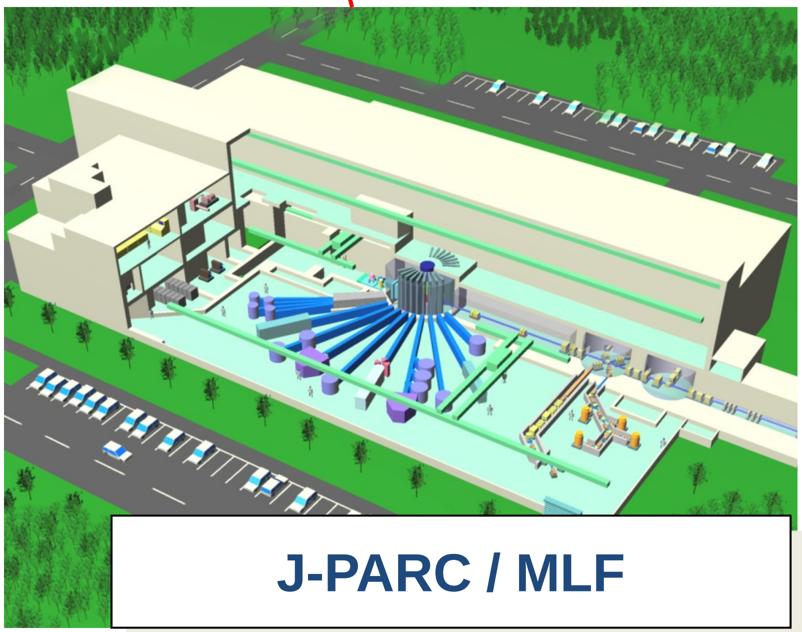


Sample for TOF
g order → mg order (Thin)

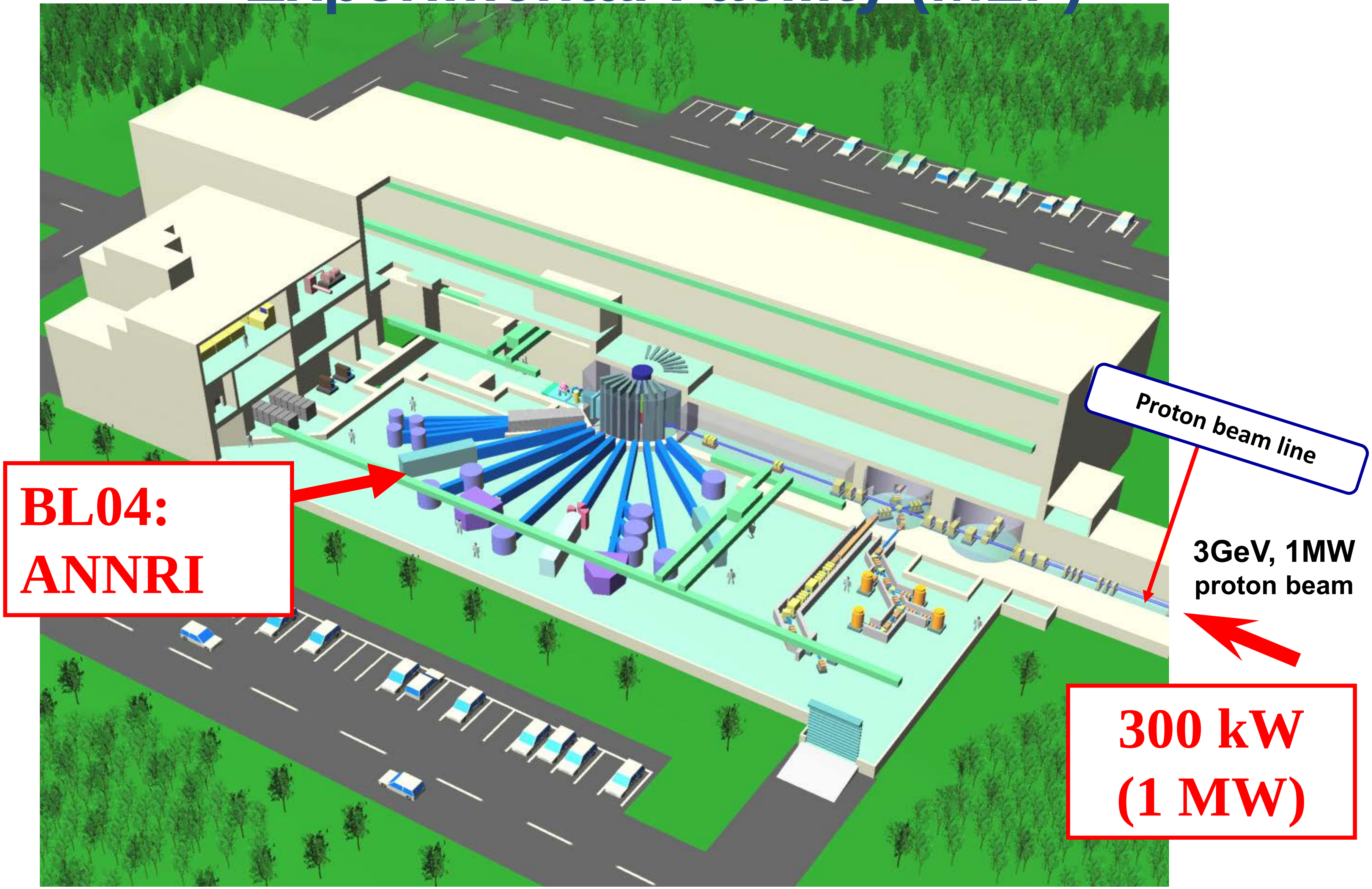
3 GeV
proton beam

Spallation
pulsed
neutron



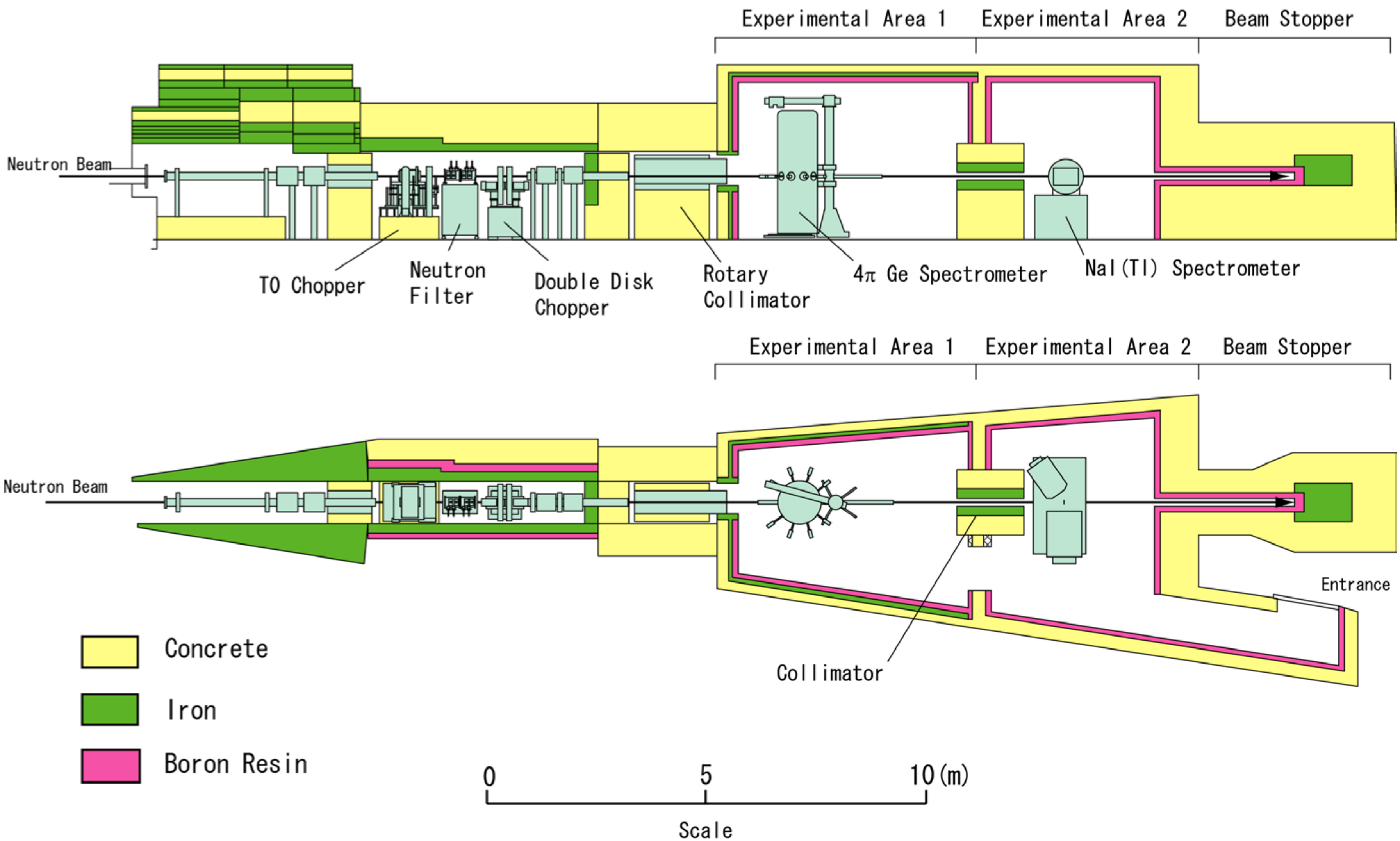


J-PARC Materials and Life Science Experimental Facility (MLF)



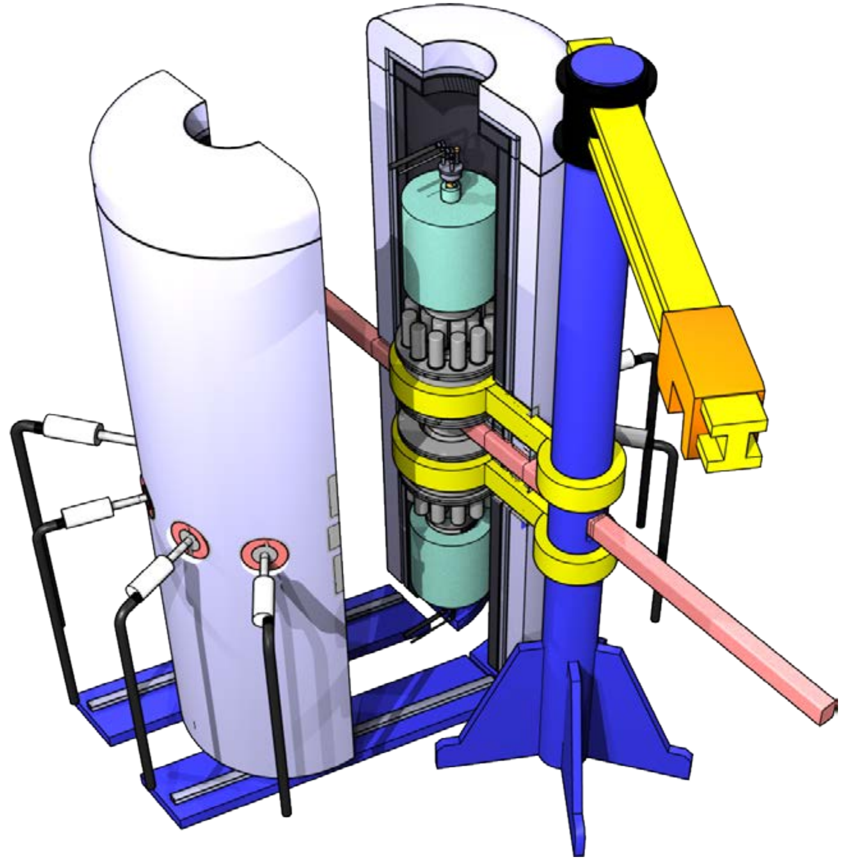
ANNRI

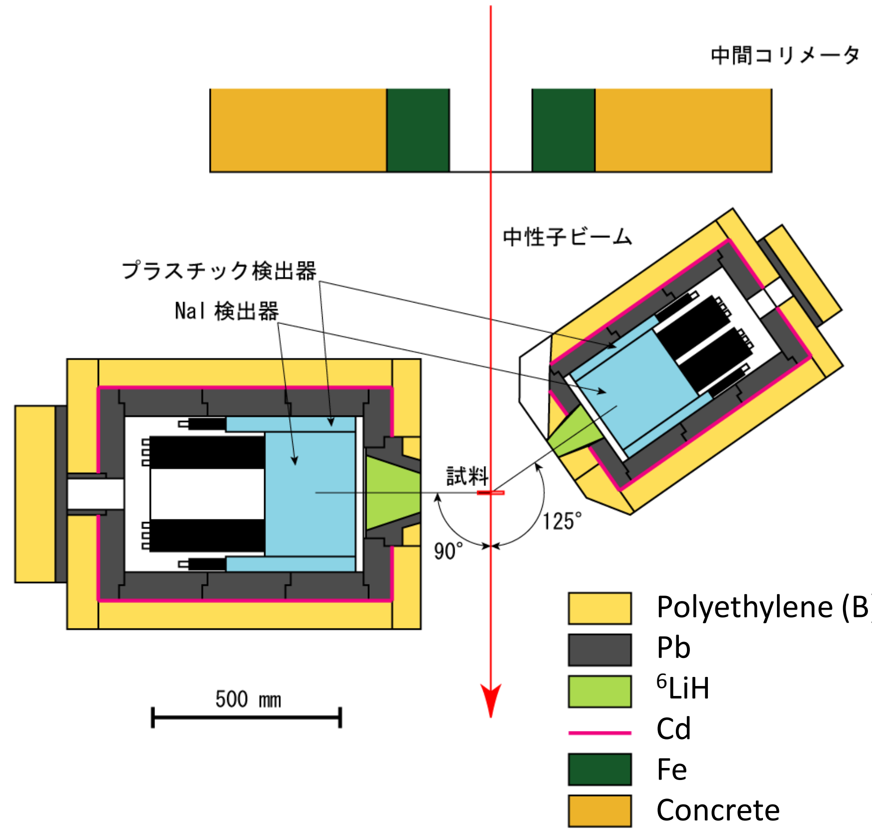
Accurate Neutron-Nucleus Reaction Measurement Instrument



ANNRI is open to users, Call for Proposals: Twice a year

Two kinds of gamma ray spectrometers at ANNRI





High efficiency Ge spectrometer at L = 21.5 m

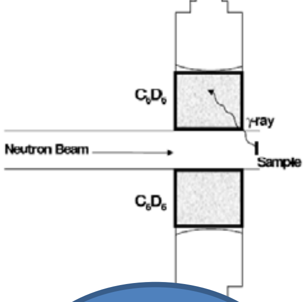
NaI(Tl) spectrometers at L = 27.9 m

T. Kin, *et al.*, J. Korean Phys. Soc., 59, 1769-1772 (2011).

From Tokyo Tech

Energy resolution of Gamma-ray detector

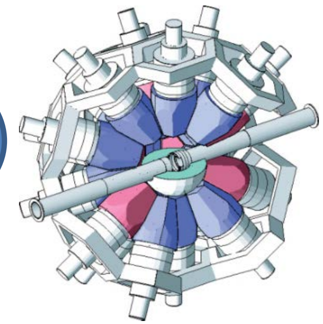
Ref: PRC **73**, 034604 (2006)



BaF_2

DANCE

n_TOF

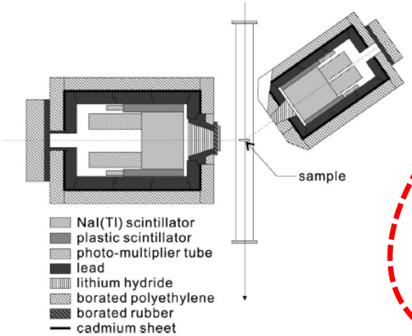


Ref: PRC **85**, 044616 (2012)

Detection Efficiency

C_6D_6

n_TOF



Ref: JNST **50**, 188-200 (2013)

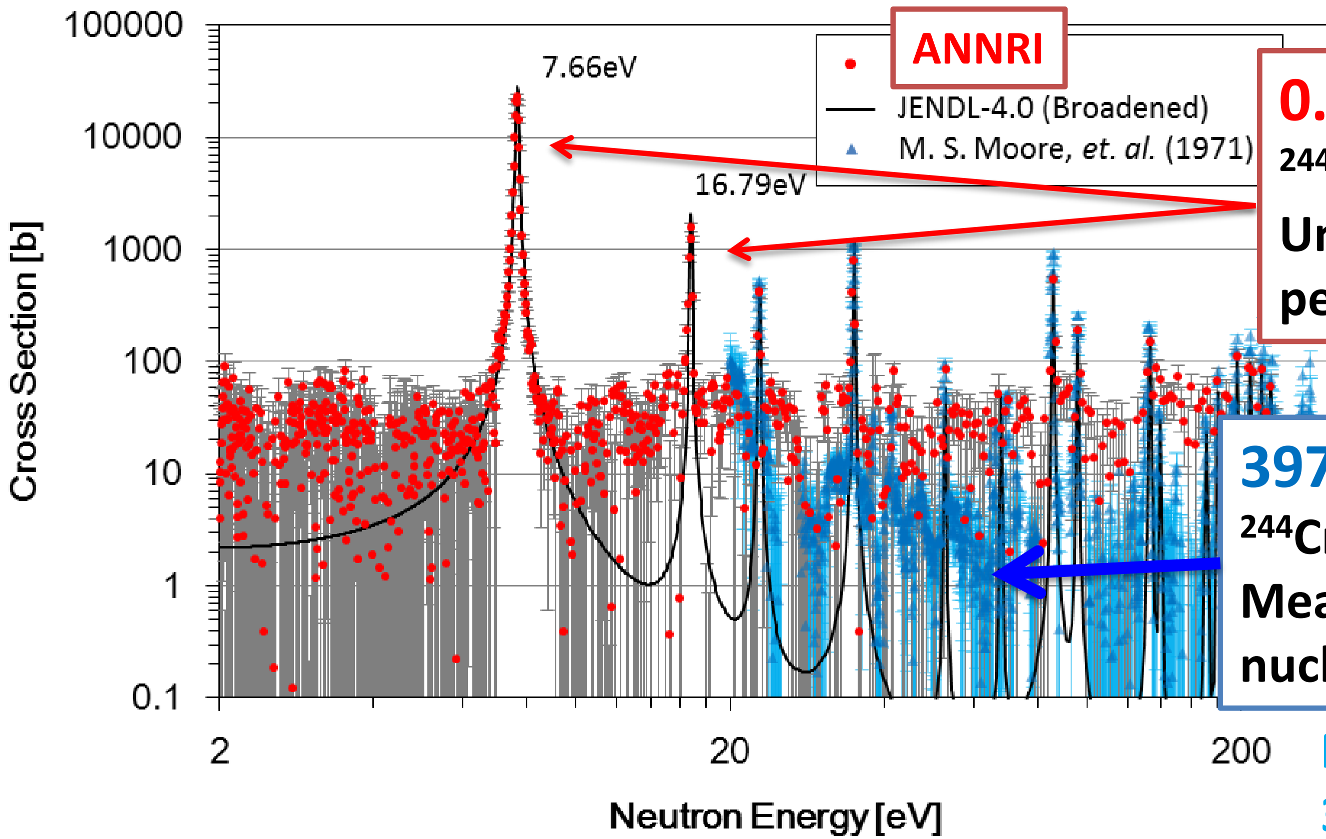
Ge

ANNRI

NaI

Energy Resolving Power = E/dE

Measurement of ^{244}Cm(n, γ) cross section Using Ge spectrometer at ANNRI



0.6 mg Sample

^{244}Cm 90% and ^{246}Cm 7%

Uncertainties < 6 % for peak cross sections

397 mg Sample

^{244}Cm 79% and ^{246}Cm 16%

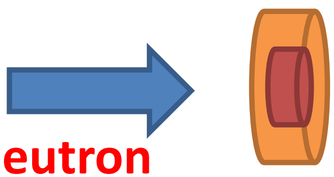
Measurement using the nuclear explosion neutron

M. S. Moore *et.al.* , PRC, 3, 1656 (1971).

A. Kimura et al., J. Nucl. Sci. Technol., [7] 708 (2012).

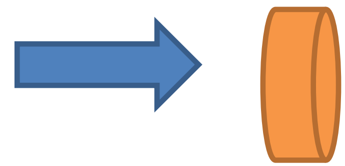
Importance using thin sample

Example: ^{241}Am capture at ANNRI

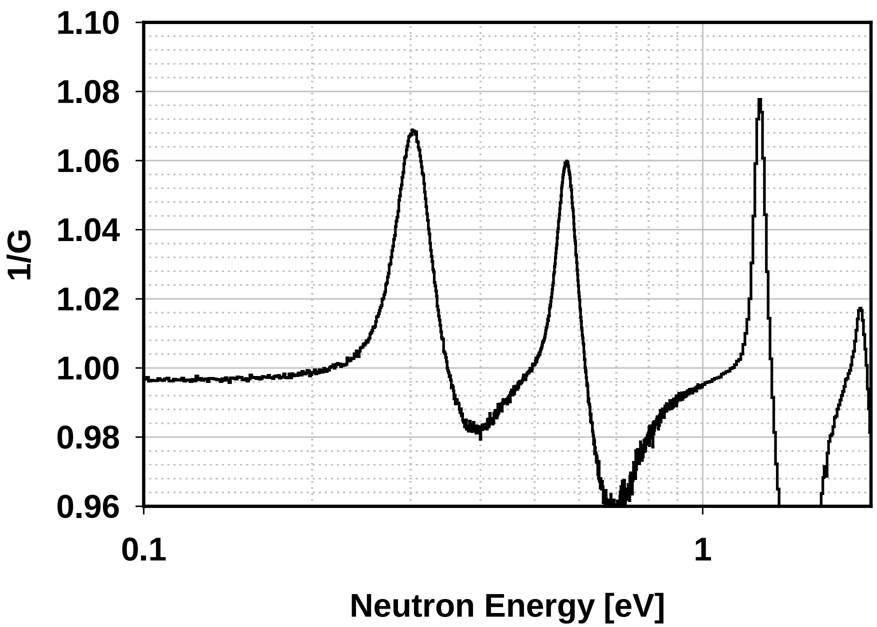


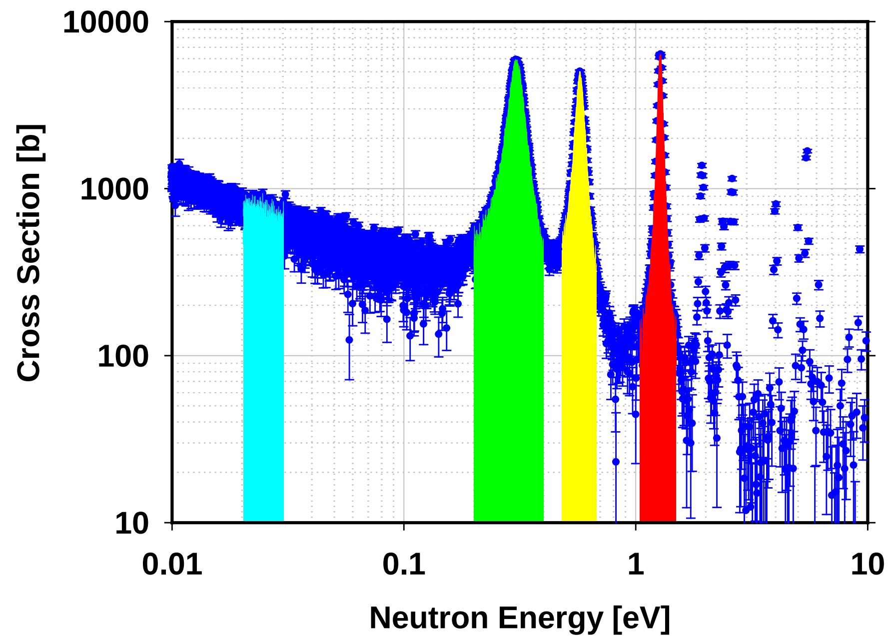
Neutron Beam

^{241}Am
(7.5 mg, 10mmϕ)
in Al case



Al case for Background





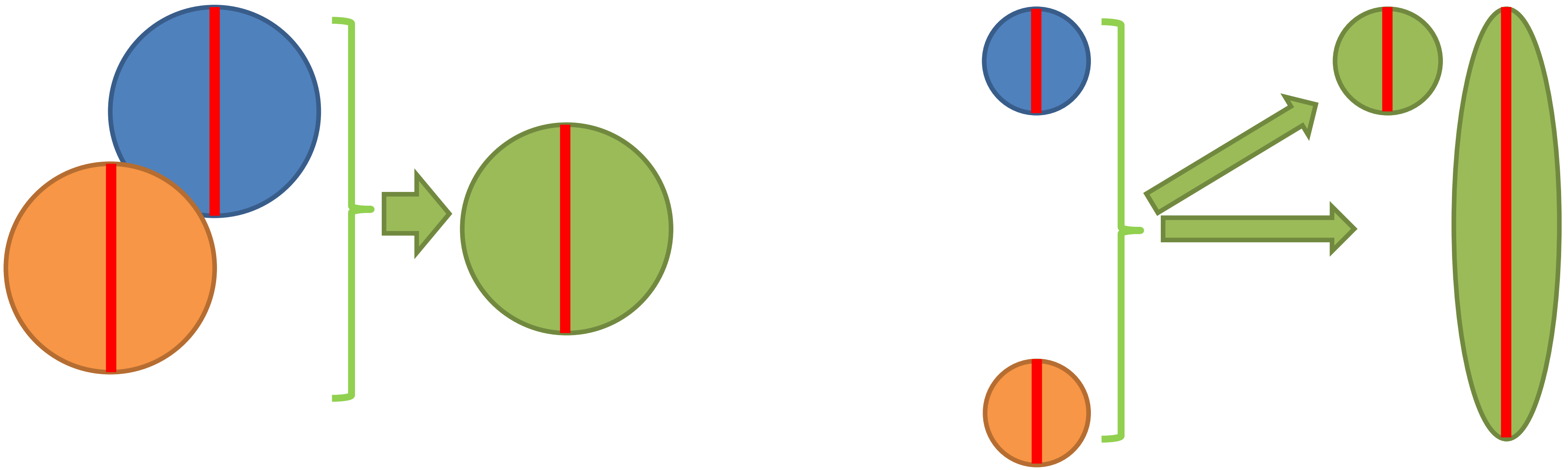
Small Self-shielding and multiple scattering correction factor

Statistics is high !

H. Harada et al., NDS, 119 (2014) 61.

Precision to Accuracy

Although high quality energy dependent capture data can be measured by ANNRI, accurate normalization is challenging issue. It is also important to identify unrecognized bias effect (and to know physical reason on discrepancy).

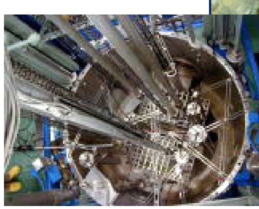


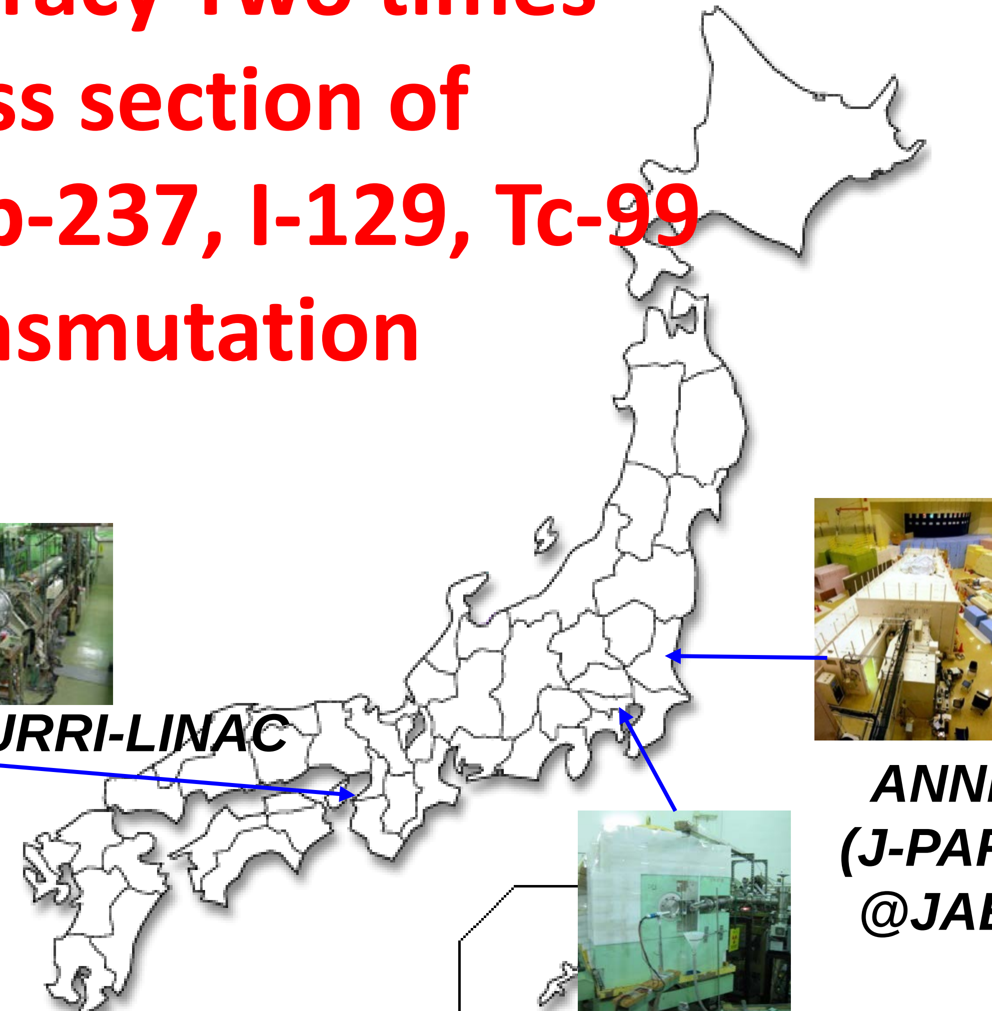
AIMAC project toward Accuracy

October 2013 – March 2017

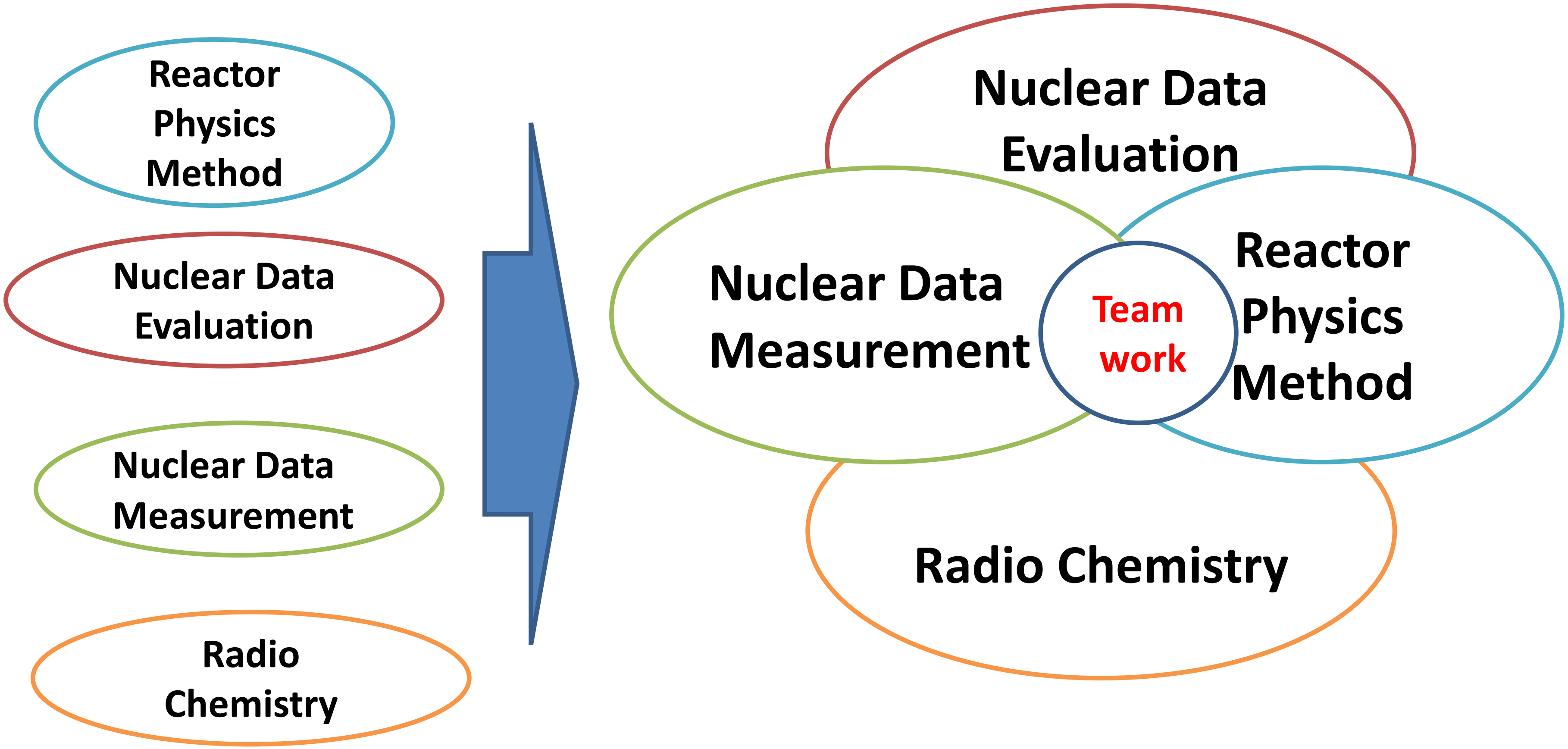
AIMAC collaboration

Improving Accuracy Two times on capture cross section of Am-241,243, Np-237, I-129, Tc-99 For nuclear transmutation





AIMAC approach



Collaboration by researchers from 4 different research fields

AIMAC Project

① Accurate measurements of
thermal neutron capture cross-sections

② High-precision quantification of
sample amount used for TOF measurement

③ Resonance parameter determination
by combining total and capture cross sections

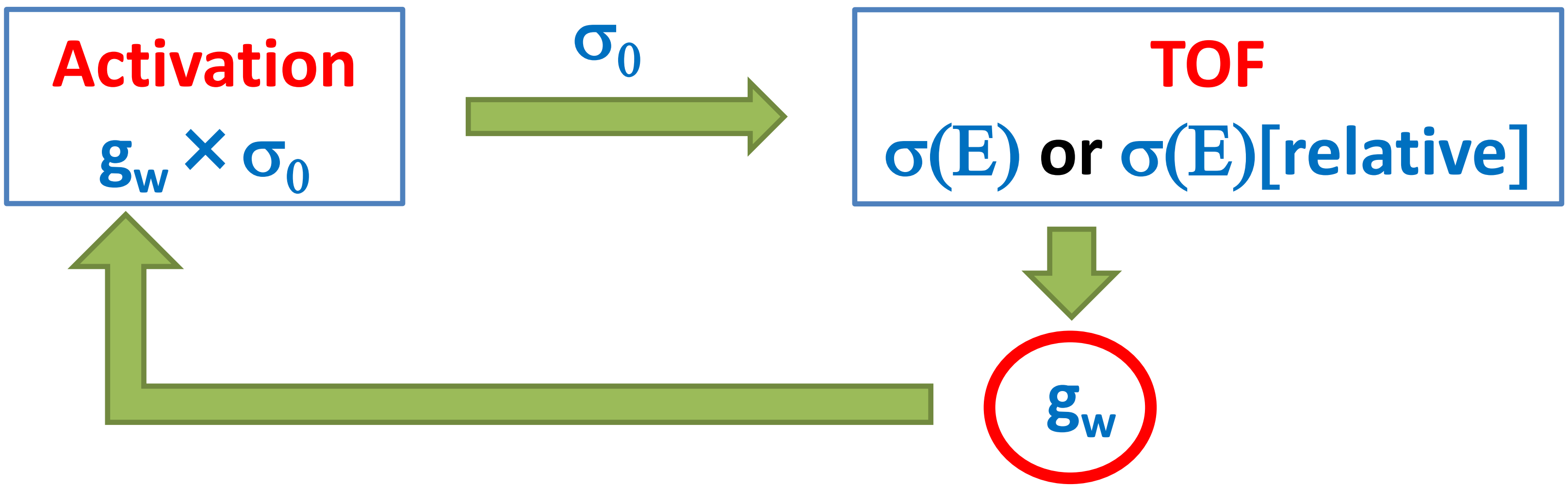
④ Extension of capture cross sections
to high energy neutrons

⑤ High quality evaluation based on
iterative communication with experimenters

Importance of Thermal for normalization

Ratio between σ(E) weighted by Maxwellian and σ_0

$$g_w = \frac{1}{\sigma_0}\int_0^{\infty}\frac{2}{\sqrt{\pi}}\frac{E}{{E_0}^2}e^{-\left(\frac{E}{E_0}\right)}\sigma(E)dE, \qquad (in\ case\ T = T_0)$$

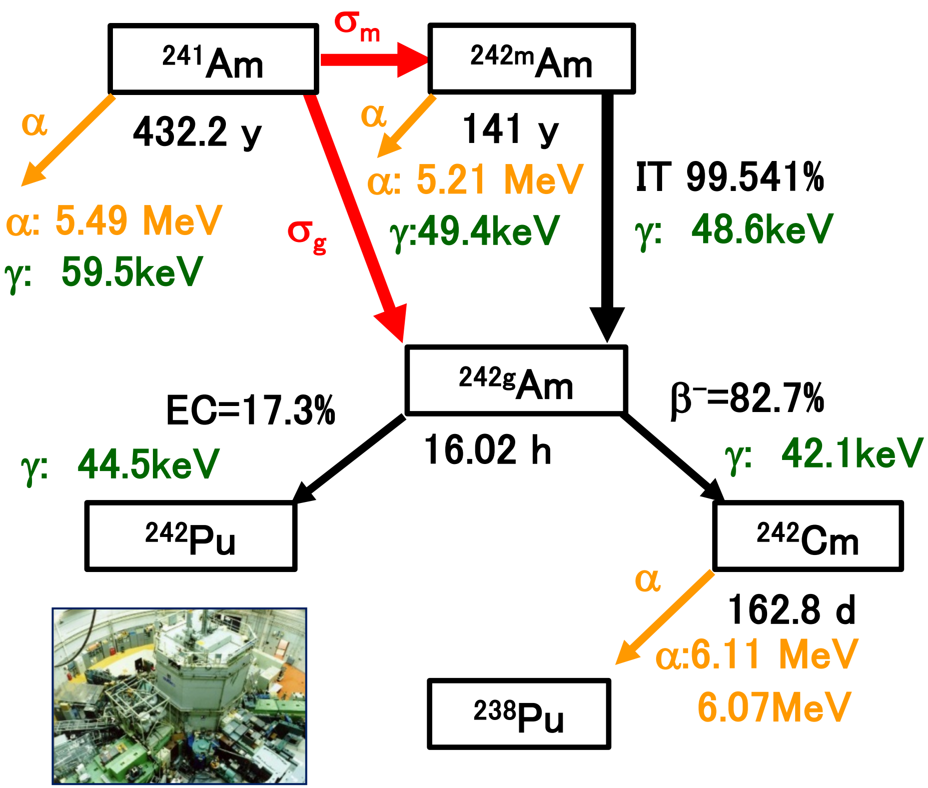


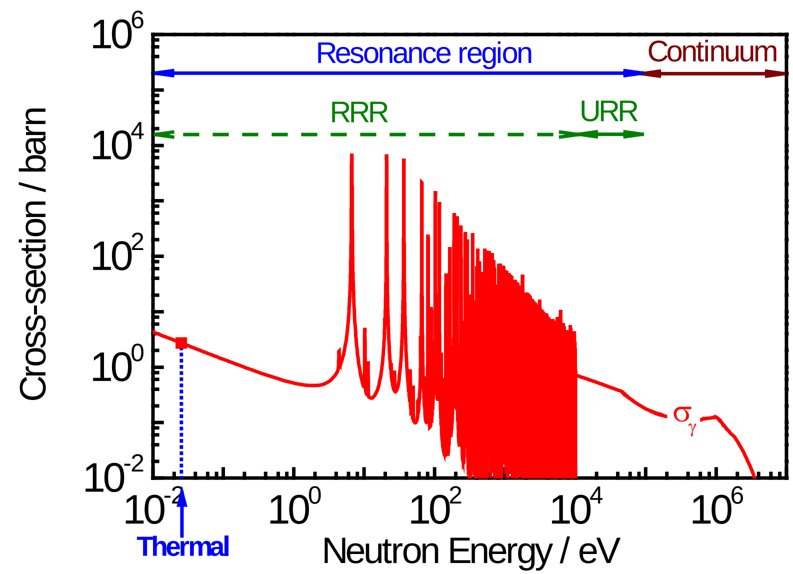
	JENDL-4.0	Mughabghab
Westcott factor	1.010	1.051

Discrepancy
4 %

① Accurate measurements of thermal neutron capture cross-sections

1) Combination of α and γ spectroscopy





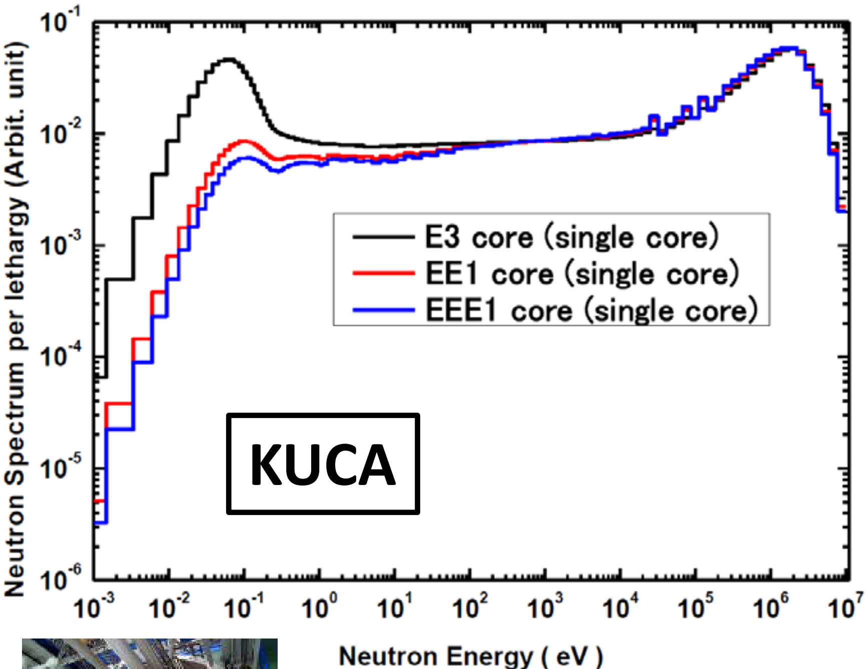
Normalization at thermal

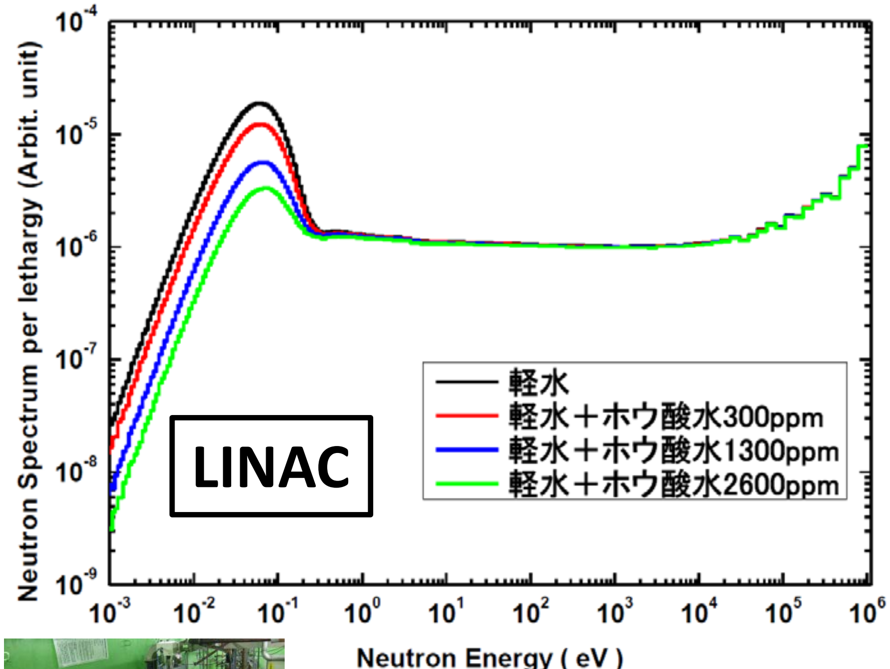
Capture cross section by α and γ spectroscopy, and Comparison

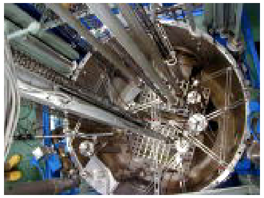
To know unrecognized systematic error

① Accurate measurements of thermal neutron capture cross-sections

2) Test of evaluated data using Variable neutron flux



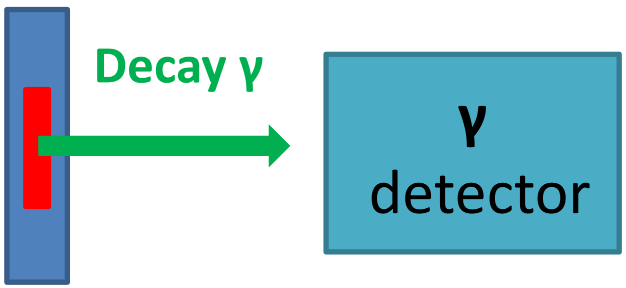




Measure $\int \phi(E)\sigma_r(E)dE$ ➡ **Comparison with JENDL**

② High-precision quantification of sample amount used for TOF measurement (1/2)

① Determination of decay γ-ray intensity



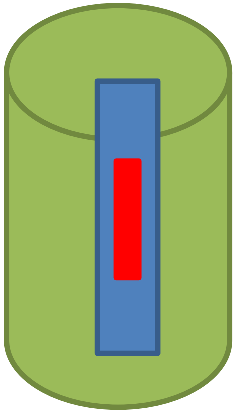
Example : **Am-243**

Uncertainty of γ-ray intensity for 118-keV

: ±15%

By combining **α and γ spectroscopy,**
γ-ray intensity is determined

② Utilization of Micro Calorimetry



Ex: Am-243
Uncertainty of Q_α value is 0.02%
and
Uncertainty of half-life $T_{1/2}$ 0.2%

⇩

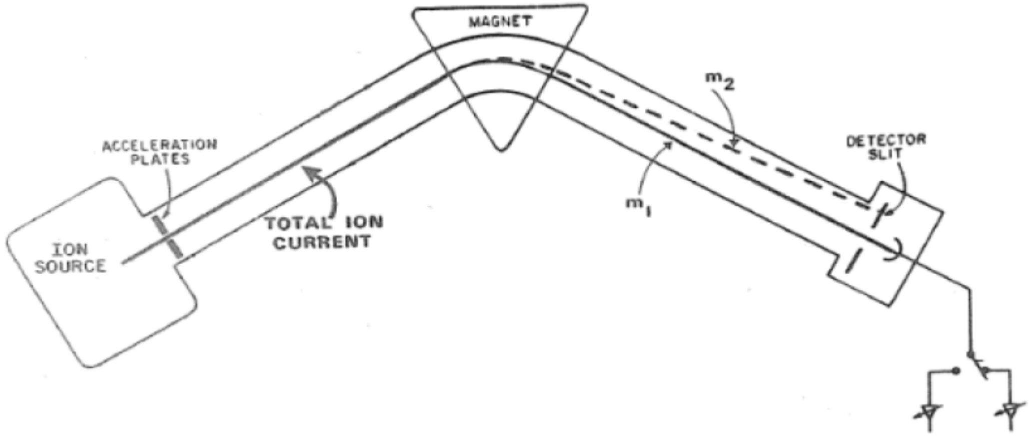
Absolute Amount

② High-precision quantification of sample amount used for TOF measurement (2/2)

3) Preparation of MA sample

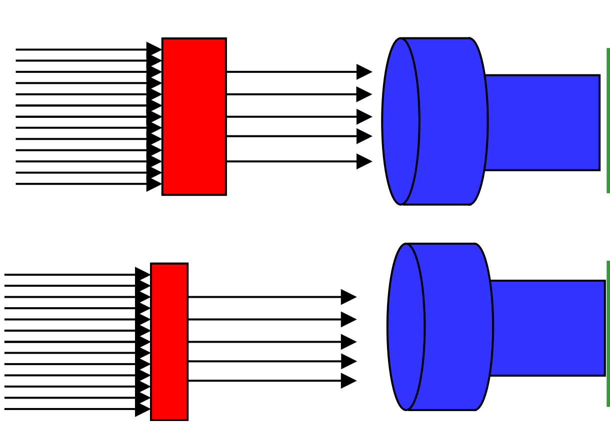
Different Thickness Samples for Identification of unrecognized origin of error

Analyses of materials used for the sample by Mass (TIMS), α-ray, and γ-ray spectroscopy



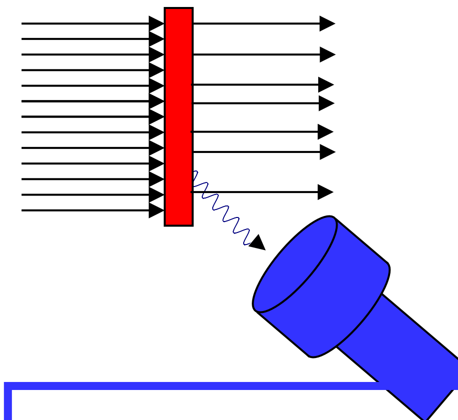
TIMS: Thermal Ionization Mass Spectrometer

③ Resonance parameter determination by combining total and capture cross sections (1/2)



$$A_{t,thick} \propto \sqrt{n\,g\,\Gamma_n\,\Gamma}$$

$$A_{t,thin} \propto n\,g\,\Gamma_n$$



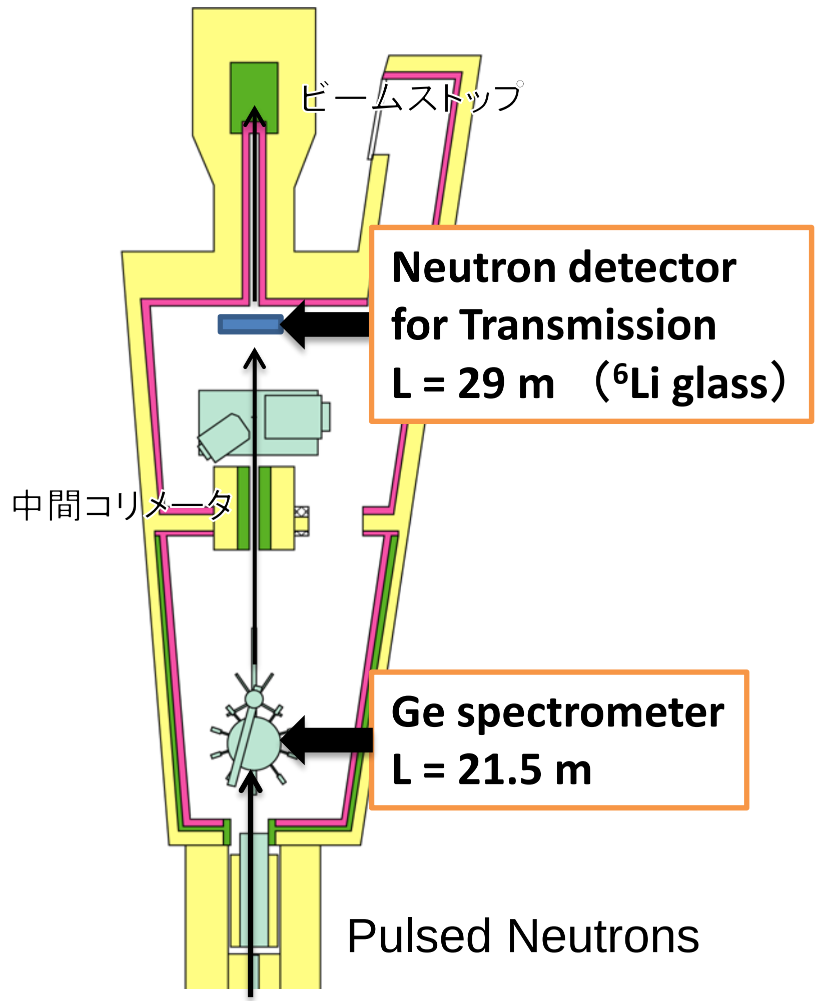
$$A_{r,thin} \propto n\,g\,\frac{\Gamma_n \Gamma_r}{\Gamma}$$

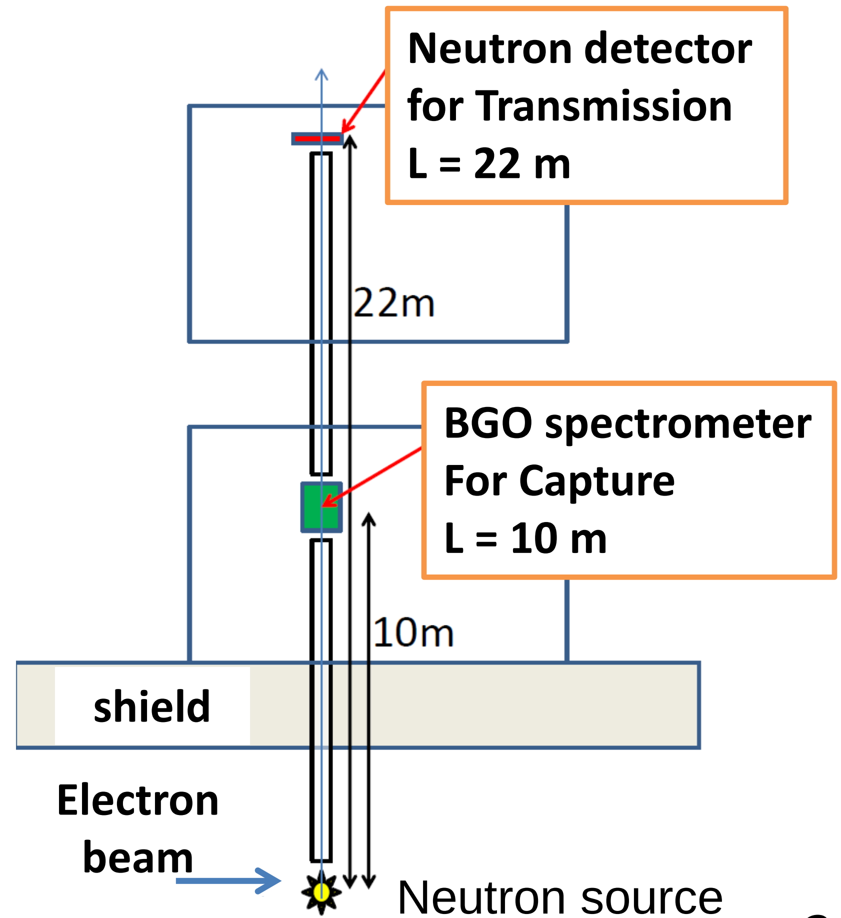
σ_0, Γ_γ, Γ ($= \Gamma_n + \Gamma_\gamma$)

Determination of resonance parameter
→ Normalization at the first resonance

③ Resonance parameter determination by combining total and capture cross sections (2/2)

J-PARC/ANNRI High Intensity Double pulse（100ns width）	KUR LINAC Normal Intensity High time resolution（2～100 ns width）

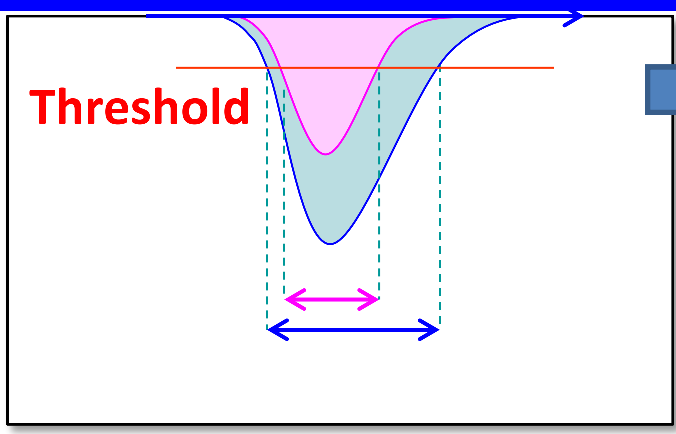




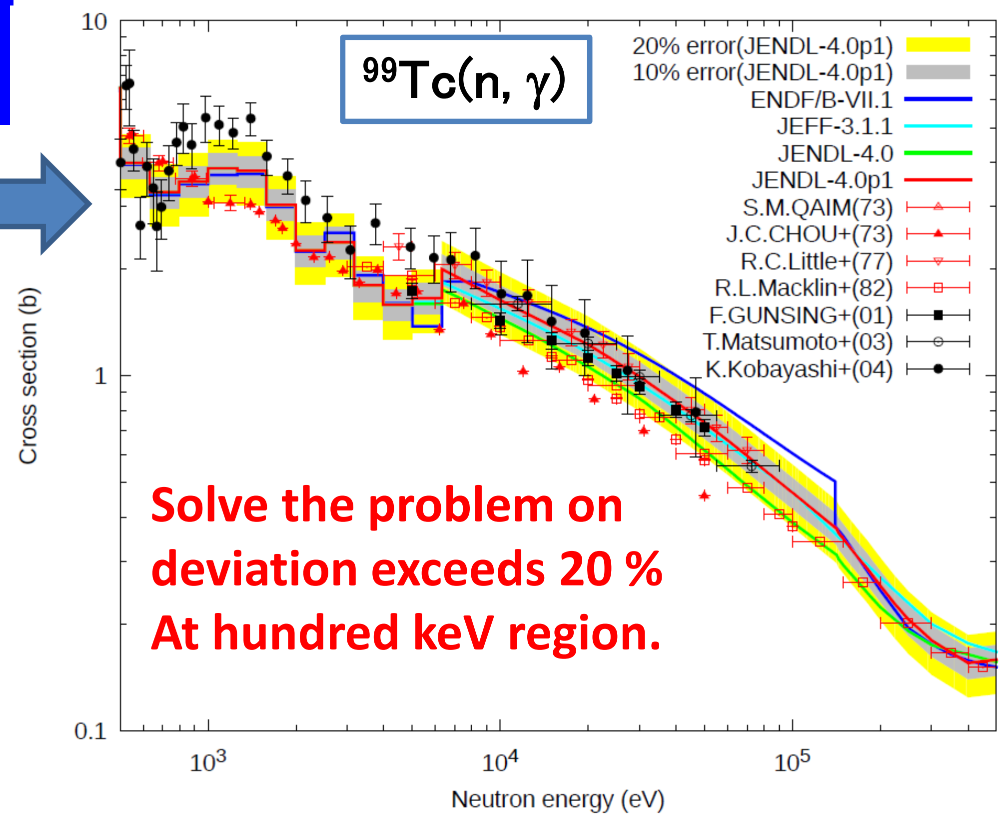
④ Extension of capture cross sections to high energy neutrons (1/2)

Extend energy region for capture cross section by ANNRI-NaI(Tl) from 100 keV to a few hundred keV.

Pulse width measurement methods, to be published in NIM by Katabuchi et al.



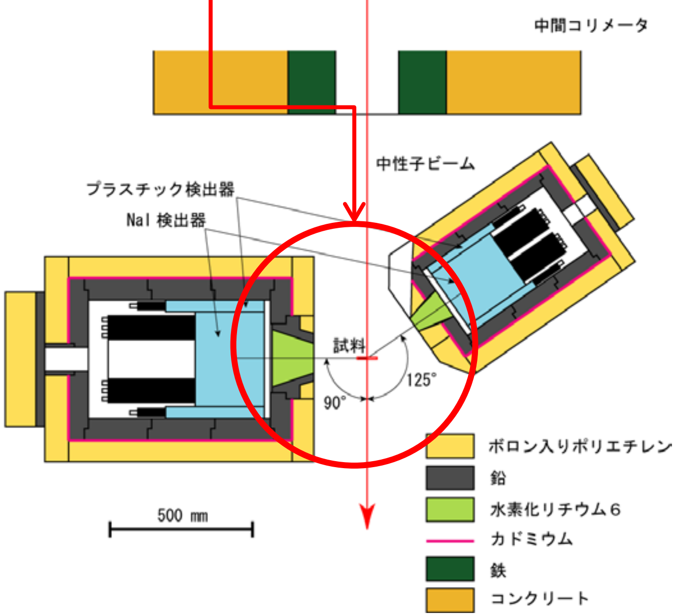
Achieve accuracy Under 5 % for neutron energy exceeding 100 keV

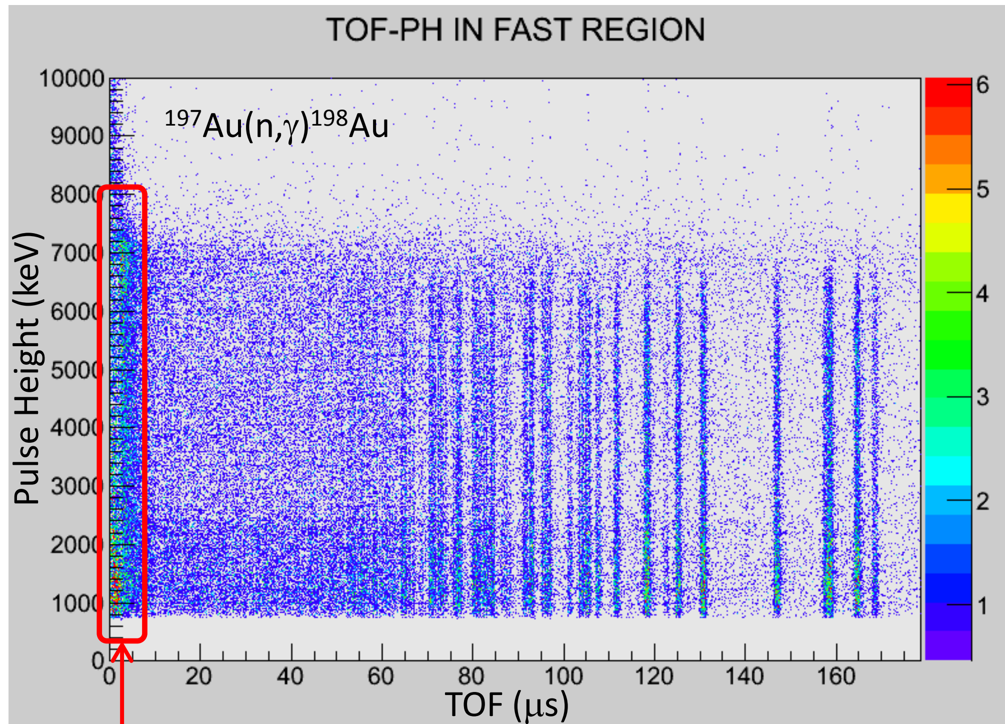


④ Extension of capture cross sections to high energy neutrons(2/2)

High neutron background for high energy region（TOF<5μs、E_n>150keV） is planned to be reduced by adding neutron shields

Insert neutron shield





Neutron Background At high energy region

⑤ High quality evaluation based on iterative communication with experimenters

Usual Approach

- **Deviation of measured values**
- **There were no intense communication between Evaluators and Experimentalists**

→ Main origin of uncertainty in evaluated data

AIMAC Approach

- **Evaluators and Experimentalists communicate intensively including detailed discussions.**

→ reliable evaluated values for MA (parts of LLFP)

Related International Activity

Next OECD/NEA/WPEC subgroup 41 (INDA) will start soon: *Improving nuclear data accuracy of ^{241}Am and ^{237}Np capture cross sections*

Expert's knowledge on evaluations, energy dependent methods , spectrum averaged methods , nuclear structure data related to capture cross sections are integrated internationally for accuracy improvement of nuclear data.

https://www.oecd-nea.org/science/wpec/meeting2014/SG_INDA_draft.pdf

Thank you very much for your attention!