Activities of Nuclear Data Measurements using the ANNRI in J-PARC/MLF

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Content

- Introduction
- Our instrument, “ANNRI”
  - J-PARC and MLF
  - Detector system
- Example of Measurements
  - Experiment of Cm-244 and 246 (MAs)
  - Experiment of Zr-93 (LLFP)
  - Experiment of Sn-112 (stable isotope)
- Summary

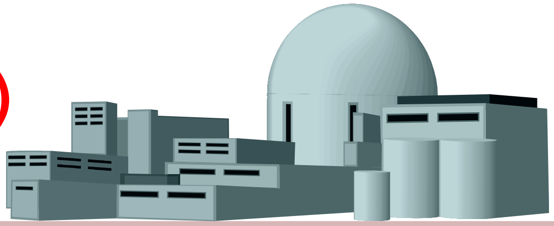
Introduction(1)
~Motivations of our study~

- To construct new types of reactors,
- To reduce High-Level radioactive Waste (Transmutation),
- To evaluate source terms from the Fukushima reactors,

Neutron Capture Cross Sections of

- **minor actinides (MAs) and**
- **long-lived fission products (LLFPs)**

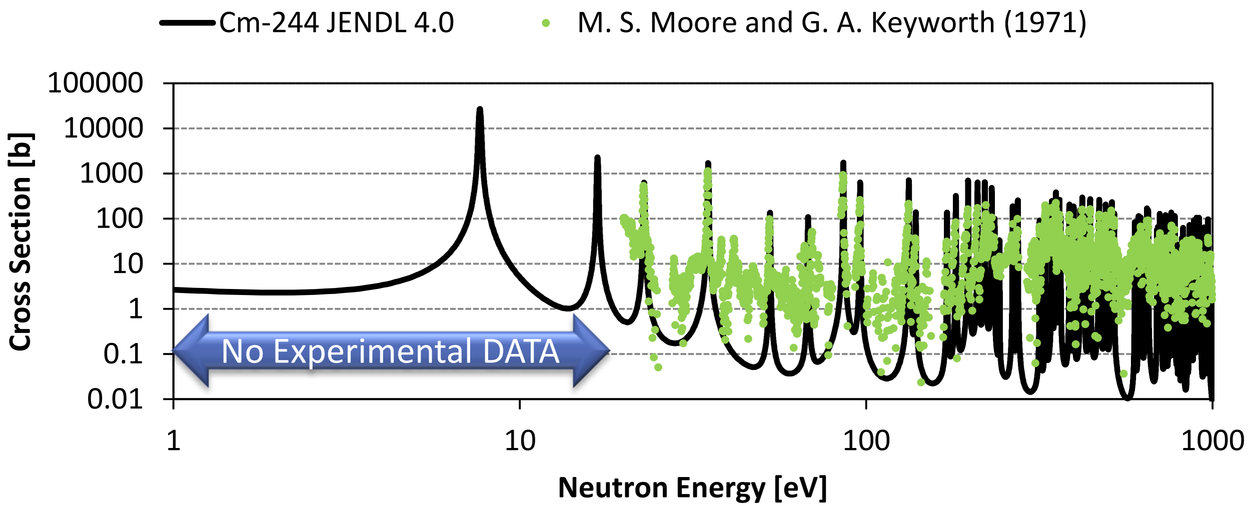
are very important.

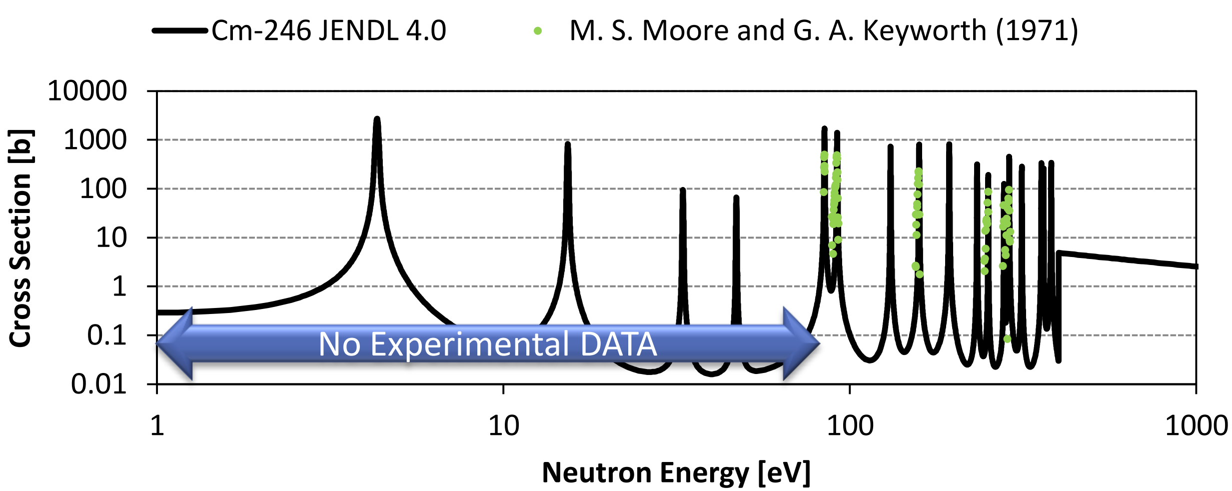


However, Experimental data of MAs and LLFPs is **not sufficient** both in quality and in quantity. For example…..

Introduction(2)

Present status of nuclear data (1)





Experimental results and evaluated neutron capture cross sections of ^{244}Cm (half-life: 18.1 years) and ^{246}Cm (4730 years).

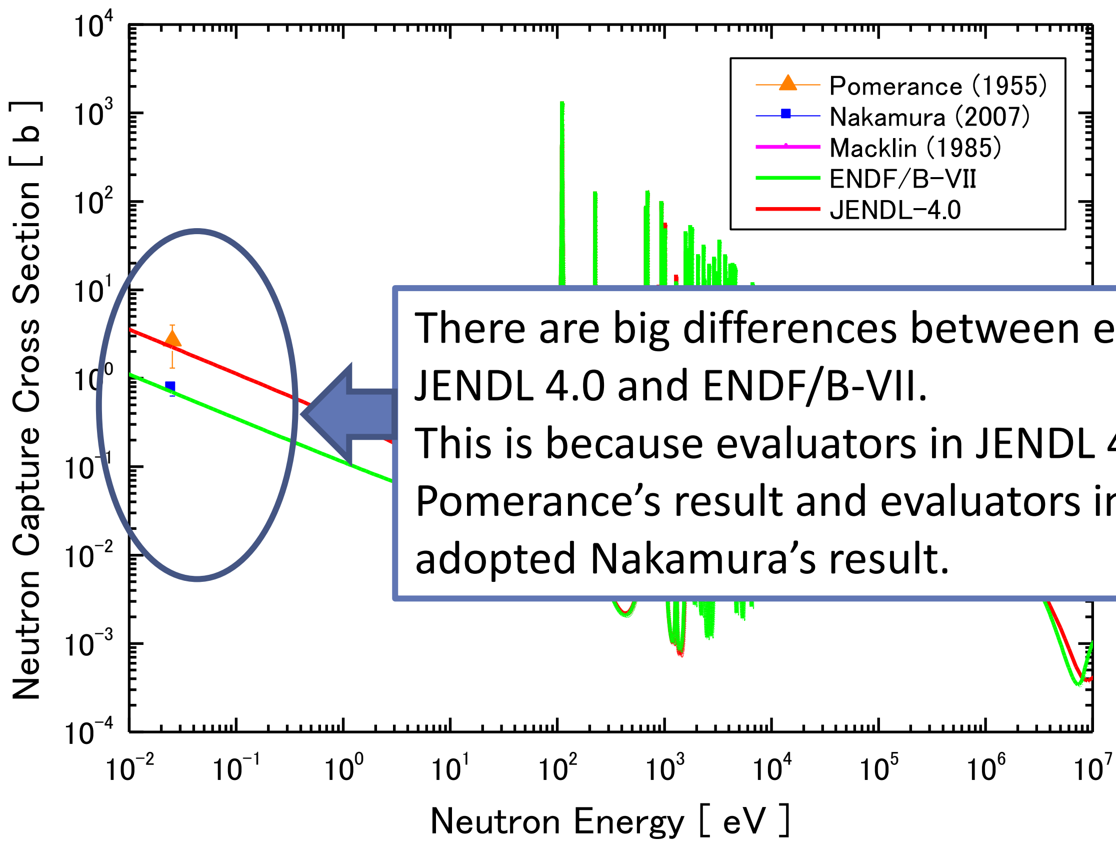
These are very important MAs.

However, there is only one experimental data set.

This previous measurement was performed using a nuclear explosion in 1969.

Introduction(3)

Present status of nuclear data (2)



There are big differences between evaluated data JENDL 4.0 and ENDF/B-VII.
This is because evaluators in JENDL 4.0 adopted Pomerance's result and evaluators in ENDF/B-VII adopted Nakamura's result.

Zr-93(LLFP)

σth
Num. of data : 2
・Pile oscillator
mpt gamma

region
n. of data : 1
F
ORELA (1985)
(a pair of C_6D_6)

Introduction(5)

Experimental data for MAs and LLFPs are not sufficient both in quality and in quantity.

This is because…

- It is not easy to prepare enough amount of MA and LLFP sample with a high purity.
- MAs and LLFPs are radio active nuclides.

To overcome these problems,

1) High Intense pulsed neutron source in J-PARC/MLF
   → Small amount of sample can be used
   → Influence due to decay γ-rays can be reduced.
2) High energy resolution γ-ray detector system
   → Background due to impurities can be removed.

were applied to the TOF measurement.

We have constructed new TOF instrument named "ANNRI".

Introduction(6)

~Measurement Plan~

In our project, we have been measuring these isotopes.

Red: Already Published. Blue: Already Measured. Black: Future Plan

Sealed RI Samples

MA Samples

^{237}Np, ^{241}Am, ^{244}Cm, ^{246}Cm

LLFP Samples

^{93}Zr, ^{99}Tc, ^{107}Pd, ^{129}I

Stable Samples

54, 56, ^{57}Fe

58, 60, 61, ^{62}Ni

74, 76, 77, 78, ^{80}Se

90, 91, 94, ^{96}Zr

104, 105, ^{108}Pd

112, 116, 117, 118, 119, 120, 122, ^{124}Sn

^{127}I

^{133}Cs

Introduction(7)

In this talk, as examples of our experiments, measurements for

- Cm-244, Cm-246, (MAs)
- Zr-93 (LLFP)
- Sn-112 (Stable)

will be demonstrated.

Cm-244,246: A. Kimura (JAEA)

A. Kimura, et. al. , J,NST,49,(7),708 (2012)

Zr-93(preliminary): J. Hori (KURRI)

J. Hori, et. Al., JKPS, 59, 1777, (2011)

Sn-112 (Very Preliminary results)

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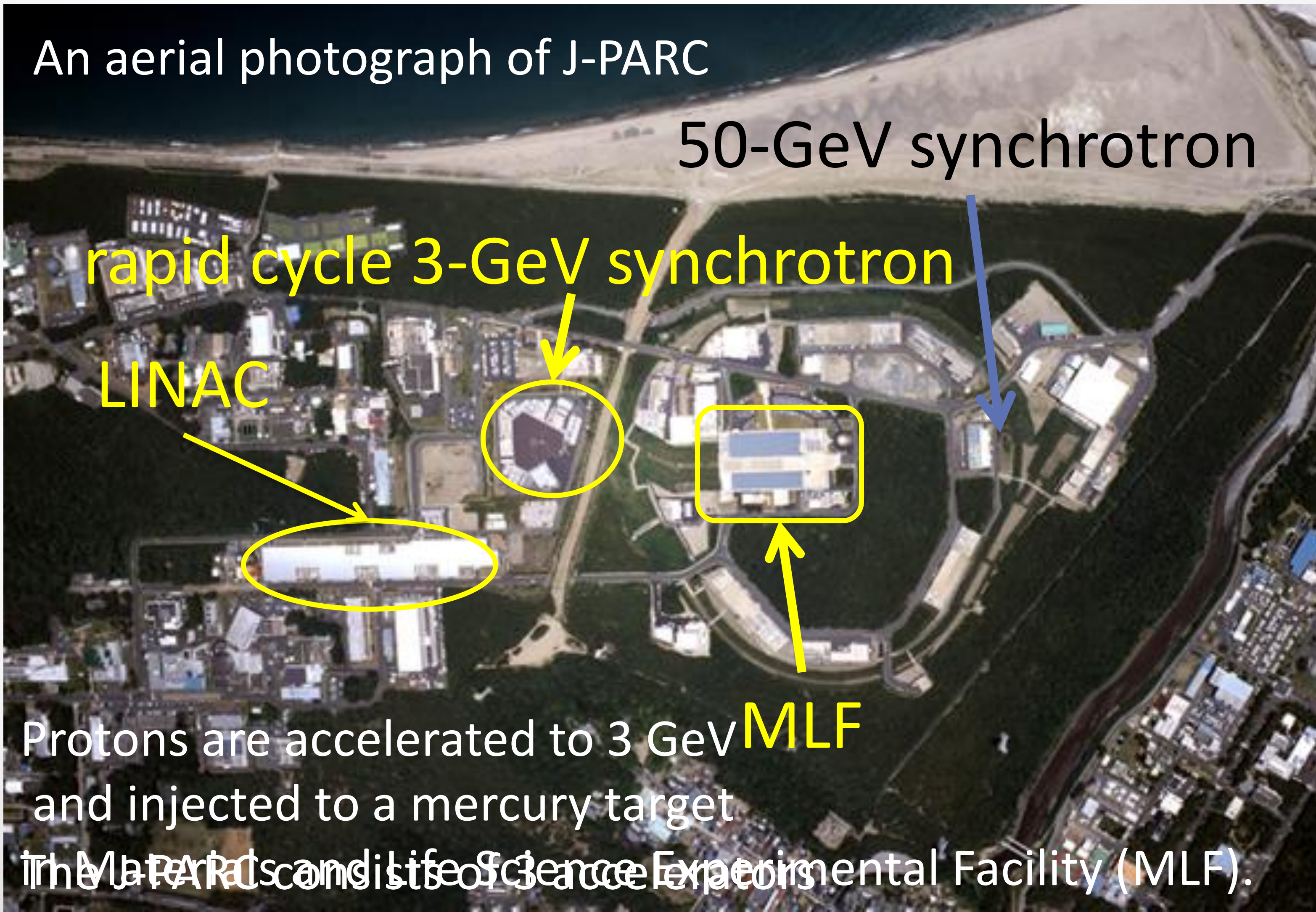
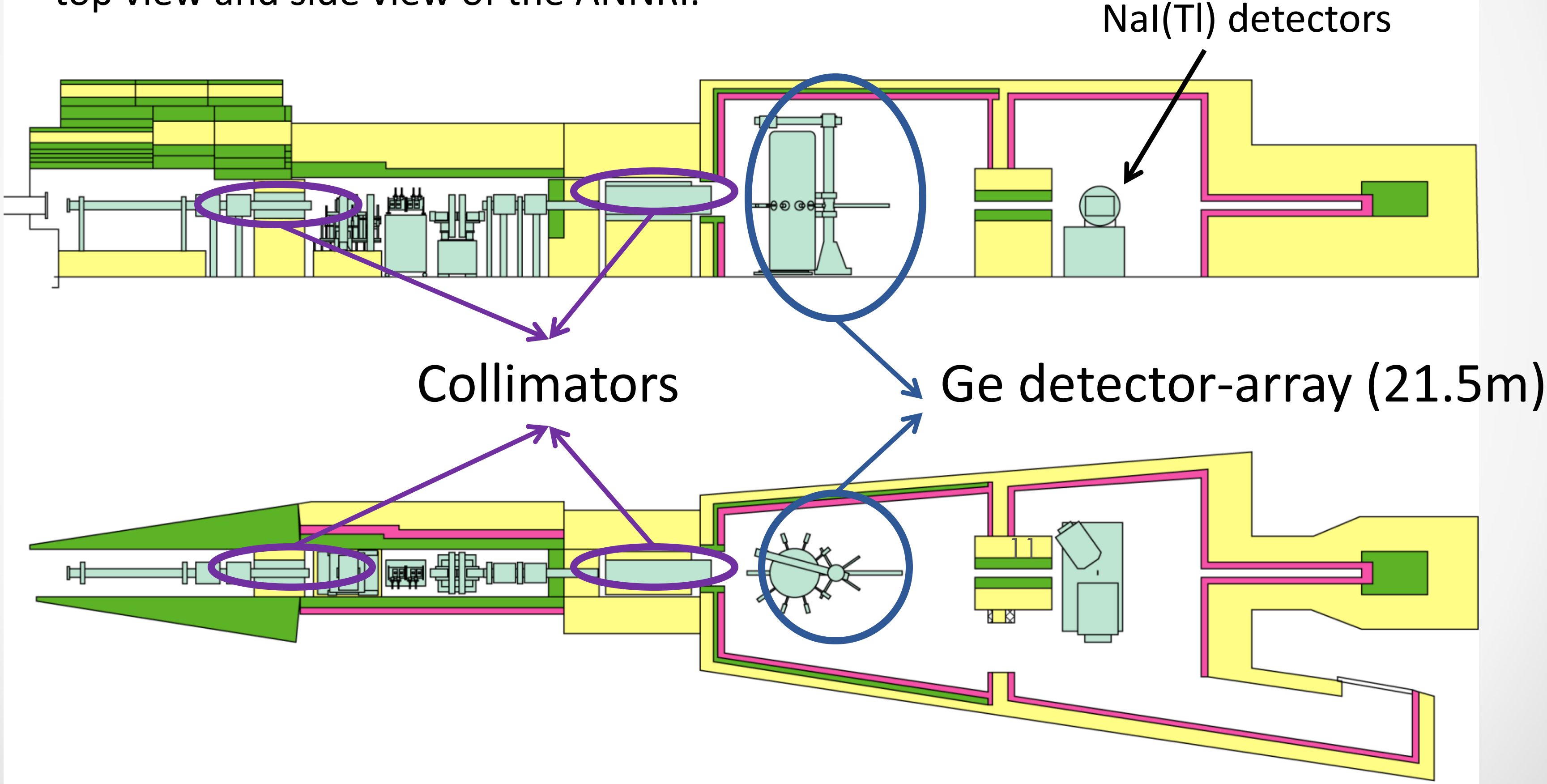
Our instrument, “ANNRI” ~ J-PARC~



Our instrument, “ANNRI”

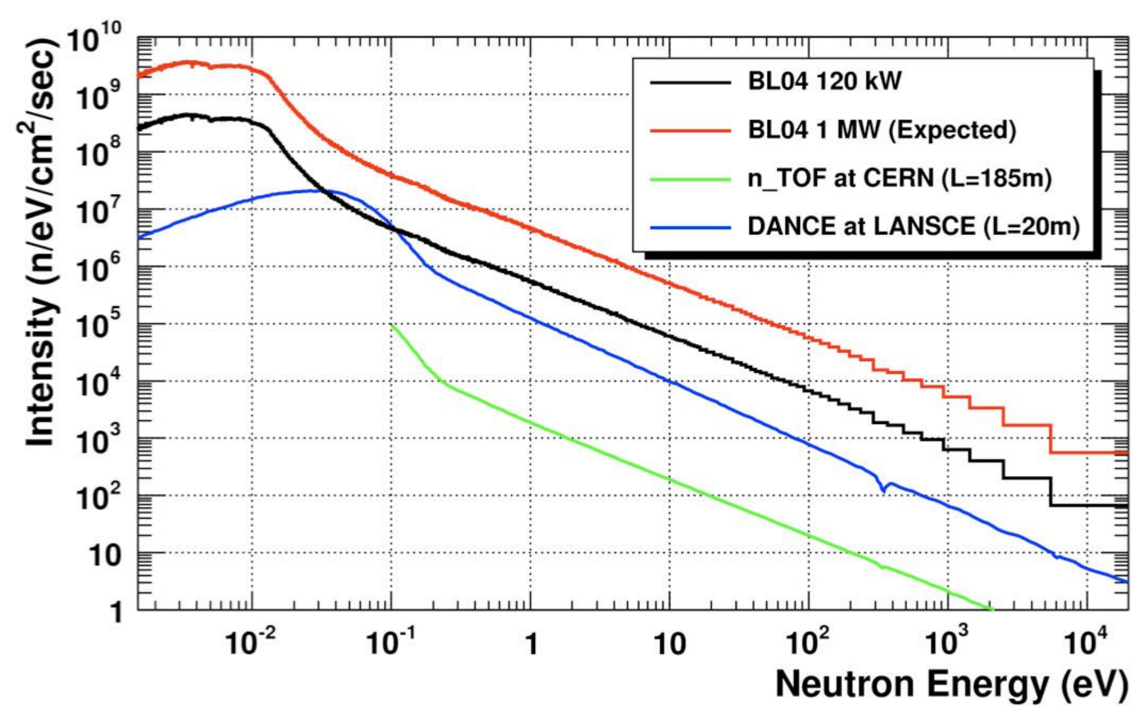
top view and side view of the ANNRI.



The neutrons were collimated to 7, 3, or 22 mm in diameter at the sample position.

Our instrument, “ANNRI” ~ Beam Intensity ~

By Dr. Kino



Neutron intensity of ANNRI under 120kW operation comparison to other facilities.

Only under 120kW operation, the neutron intensity is more than 7 times higher than the other instruments.
MLF is operated under beam power of 300kW.
(Furthermore, In future, the beam power will increase to 1MW.)

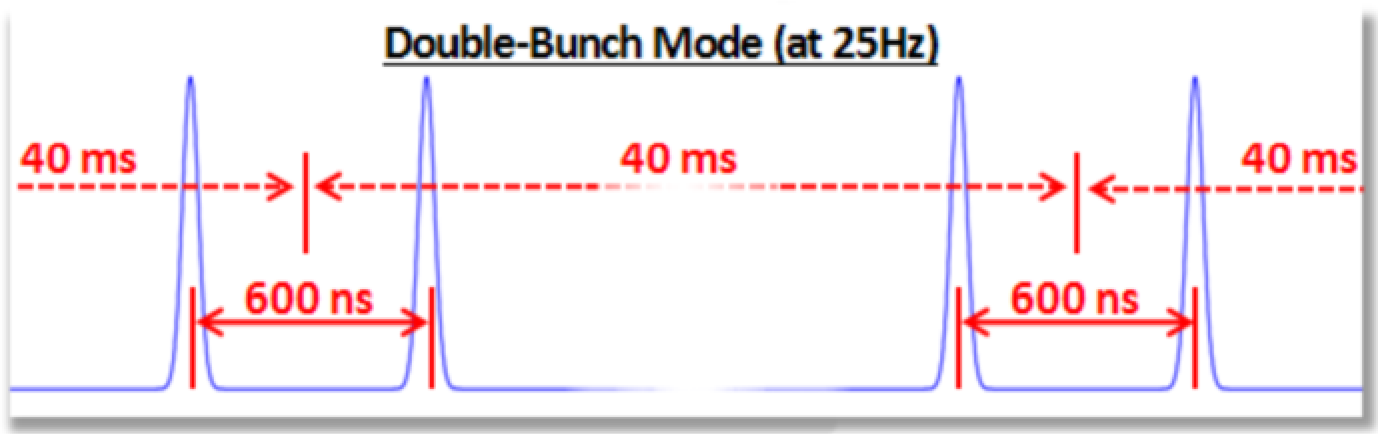
Very Strong pulsed neutron source!!

K. Kino, *et. Al.*, NIM-A, **626**, 58 -66 (2011).

energy-integrated intensities	1MW (Future)	120kW
1.5-25 meV	4.3×10^{7} n/s/cm^2	4.5×10^{6} n/s/cm^2
0.9-1.1 keV	6.3×10^{6} n/s/cm^2	6.6×10^{5} n/s/cm^2

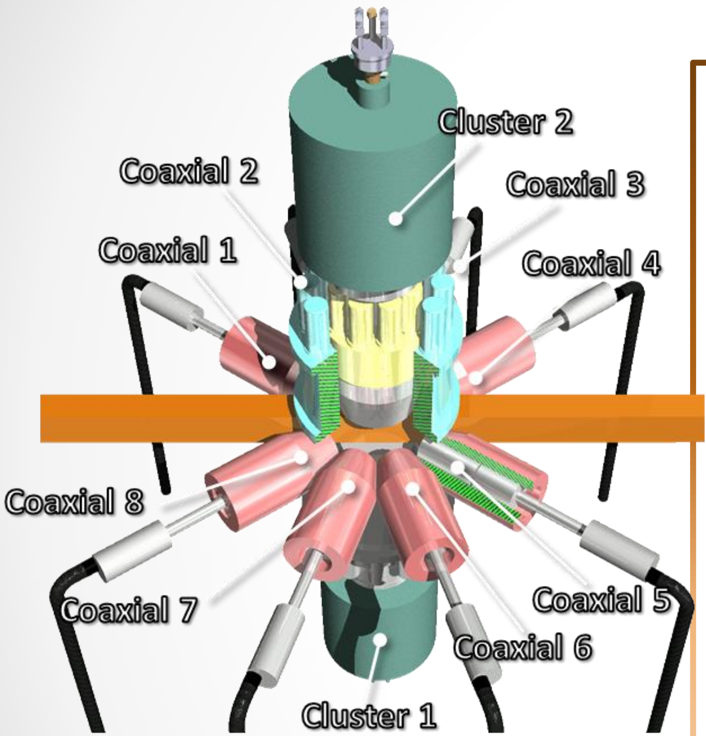
However, neutron energy resolution was not so good!!

- The moderator is large (14cm) and flight length is not so long (21.5m)
- The pulsed protons usually consist of two bunches, each with a width of 100 ns, at intervals of 600 ns (called “double-bunch mode”),



MLF (Materials and Life Science Experimental Facility) has 23 beam-lines, and most beam-lines are diffractometers, scattering spectrometers or reflectometers.
So, most users require only “neutron intensity”.

Our instrument, “ANNRI” ~Detector system~



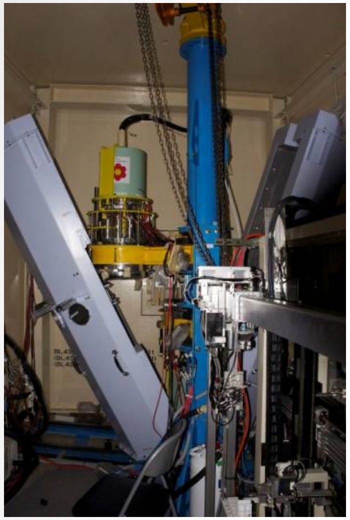
Our spectrometer has
- 2 cluster-Ge detectors
  (7 Ge crystals are installed in the detector)
- 8 coaxial-Ge detectors
  ⇒22 Ge Crystals.

Energy resolution for 1.33MeV γ-rays:
5.8keV (for 200kevents/s),
2.4keV (for 20kevents/s) [1]

Peak efficiency for 1.33MeV γ-rays:
3.64 ± 0.11 %

[1] T. Kin et. al., *the 2009 NSS-MIC Conf. Rec.* , N24-2, (2009).

The Great East Japan Earthquake



ANNRI was extensively damaged by the earthquake.

The γ-ray shields made of heavy metals for the large Ge spectrometer leaned to the walls. One of the shields was stopped by breaking an air-conditioner, and the other by a signal cable ruck installed along the walls. At the same time, several of the annular shaped BGO scintillators fell onto the floor, and their PMTs were damaged.

The Great East Japan Earthquake

3 March 2012

4 March 2012

The restoration of ANNRI was almost finished in this March.

Our project and user program in ANNRI has restarted.

- In this 2012 JFY, ANNRI has been used in user programs for more than 70 days.

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Experiments of $^{244,246}Cm$

~Samples and Measurement conditions~

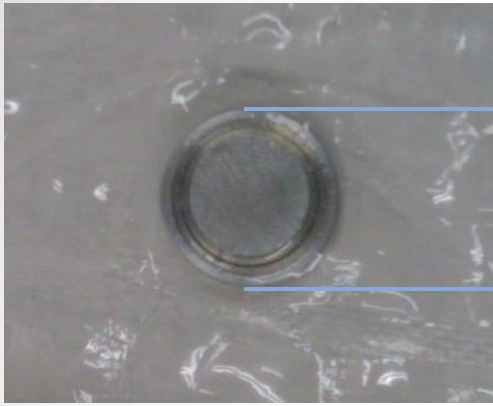


Table 1 The isotopic composition of the ^{244}Cm sample or the ^{246}Cm sample.[1]

	^{244}Cm sample	^{246}Cm sample
	TIMS (mole%)	TIMS (mole%)
^{244}Cm	90.1±1.7	27.5±0.5
^{245}Cm	2.71±0.34	1.06±0.28
^{246}Cm	7.22±0.34	59.4±1.3
^{247}Cm	N.D.	2.9±0.4
^{248}Cm	N.D.	9.10±0.24

Samples:

Cm-244 ($T_{1/2}$=18.1y: MA)

Net weight = 0.6 mg

Activity = 1.8 GBq

Measurement Periods: 64 h

Cm-246 ($T_{1/2}$=4753y: MA)

Net weight = 2.1 mg

Activity = 12.1 MBq

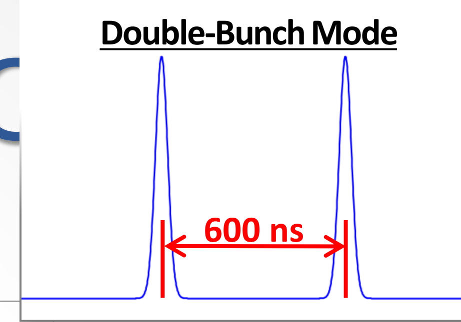
(^{244}Cm: 1.7GBq)

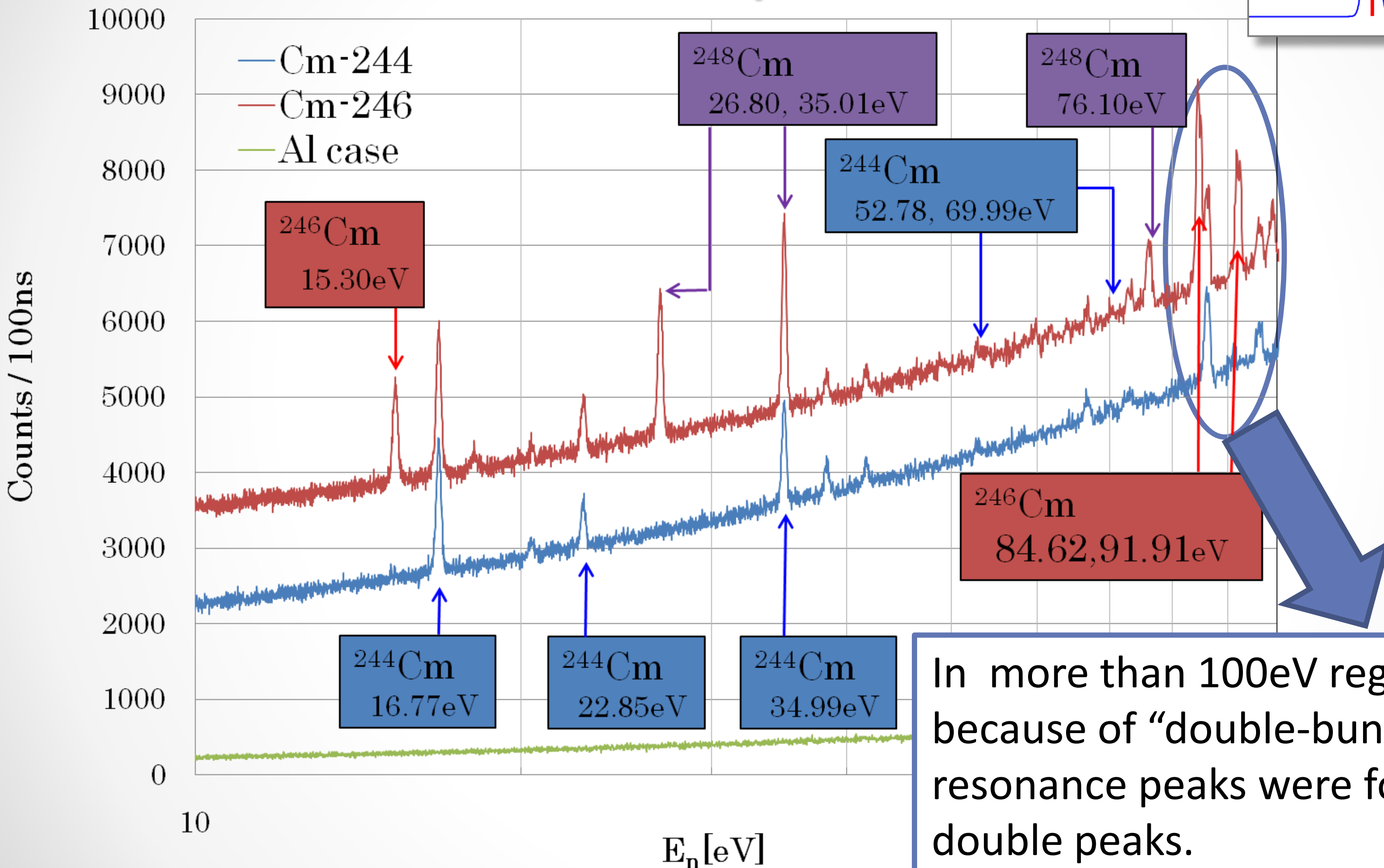
Measurement Periods: 94 h

Because of reguration of MLF, the samples are sealed in Al capsuel (278mg).

Experiments of ^{244,246}C

~TOF Spectrum ~



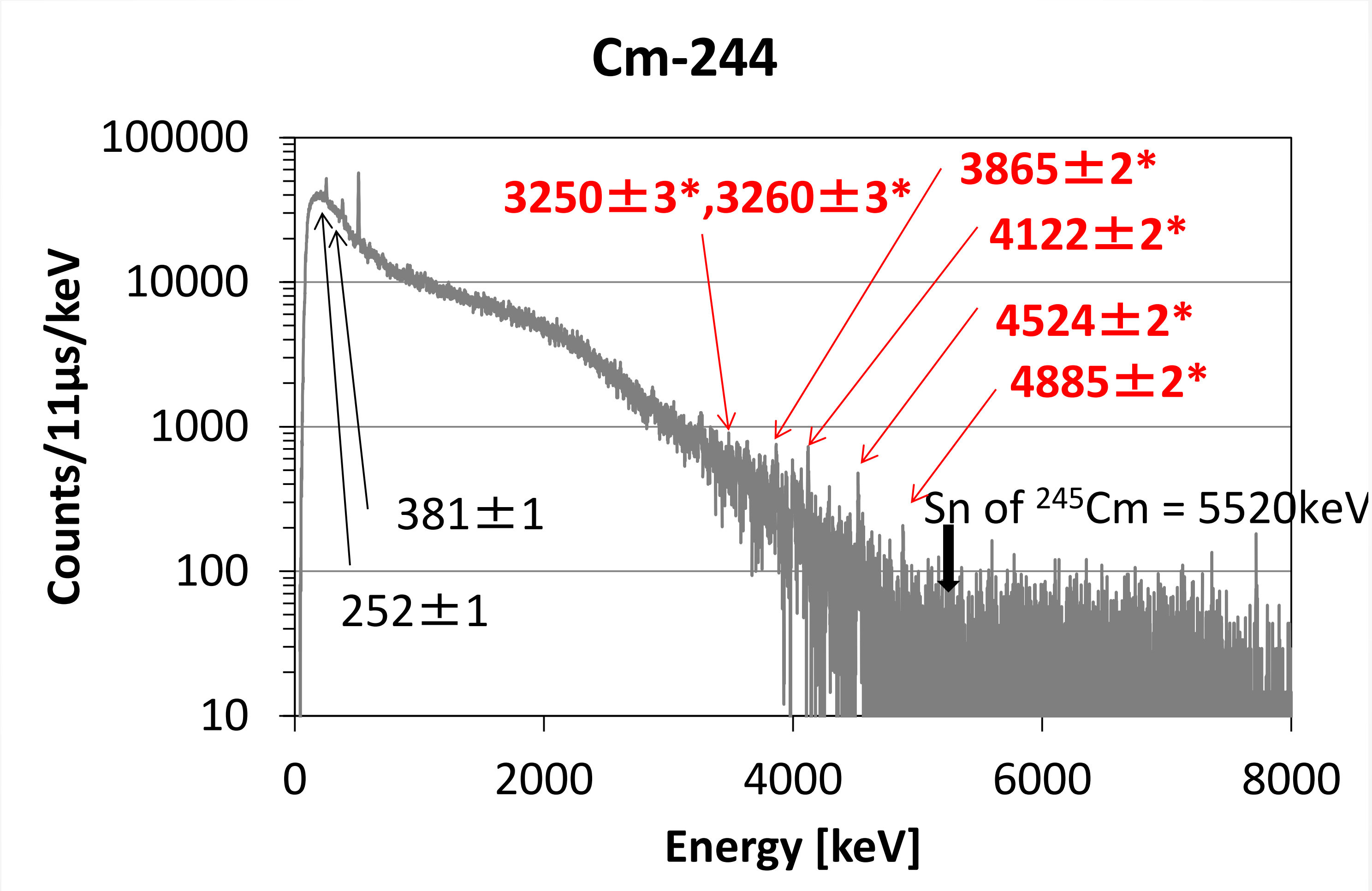


In more than 100eV region, because of "double-bunch mode", resonance peaks were formed double peaks.

This graph shows TOF spectra of the ^{244}Cm, the ^{246}Cm sample, and the dummy case. Many resonance peaks were observed.

Experiments of $^{244,246}Cm$

~ γ-ray Spectrum at the 1st resonance of ^{244}Cm ~

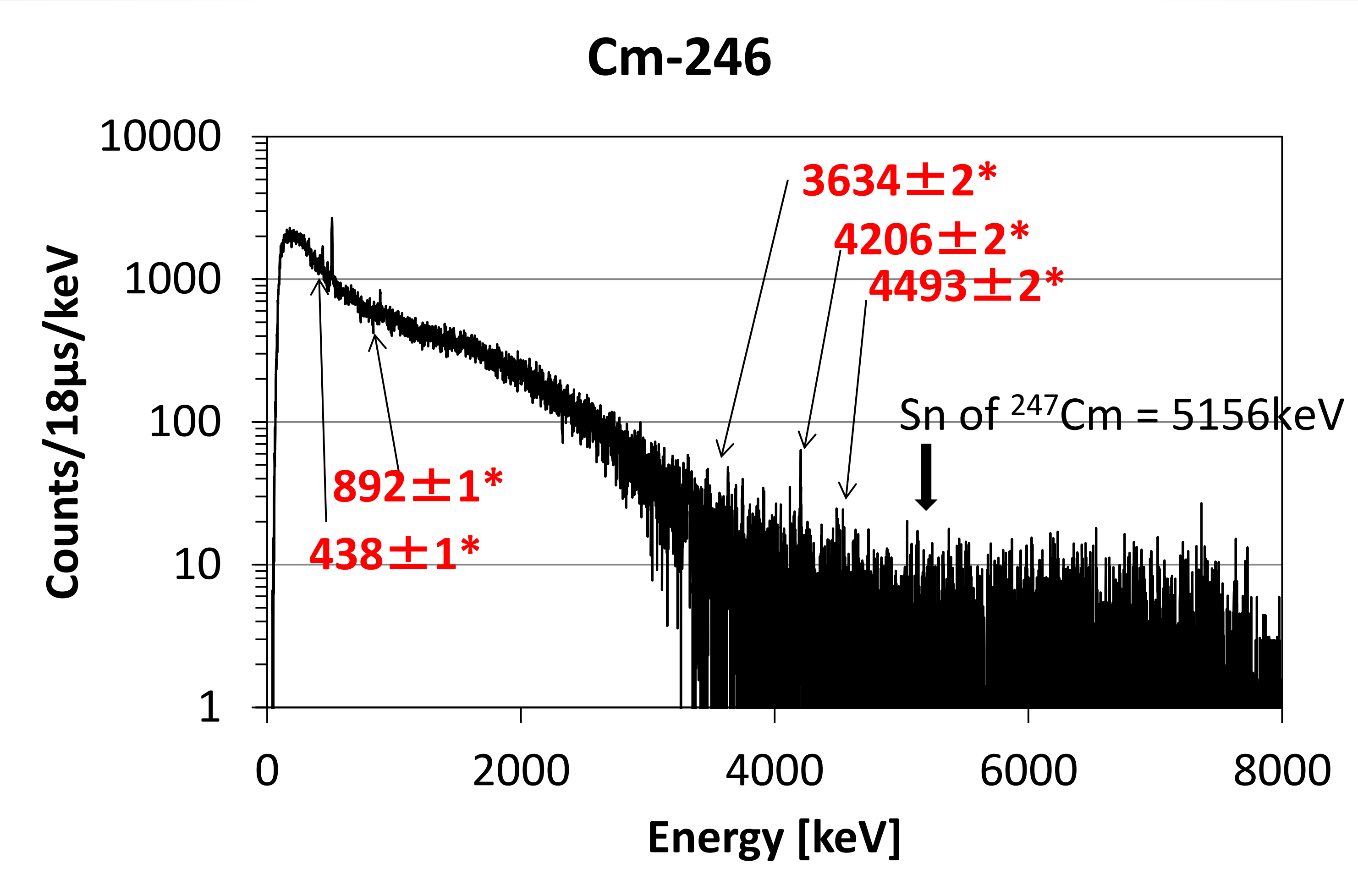


The252.4- and 380.8-keV γ-rays have already been studied in α decay of ^{249}Cf, electron capture decay of ^{245}Bk, and β-decay of ^{245}Am.

The other γ-rays were previously unknown γ rays.

Experiments of $^{244,246}Cm$

~γ-ray Spectrum at the 1st resonance of ^{246}Cm ~



The all observed γ-rays were previously unknown γ rays.

Experiments of $^{244,246}Cm$ ~Analysis~

The data were analyzed with this procedure.

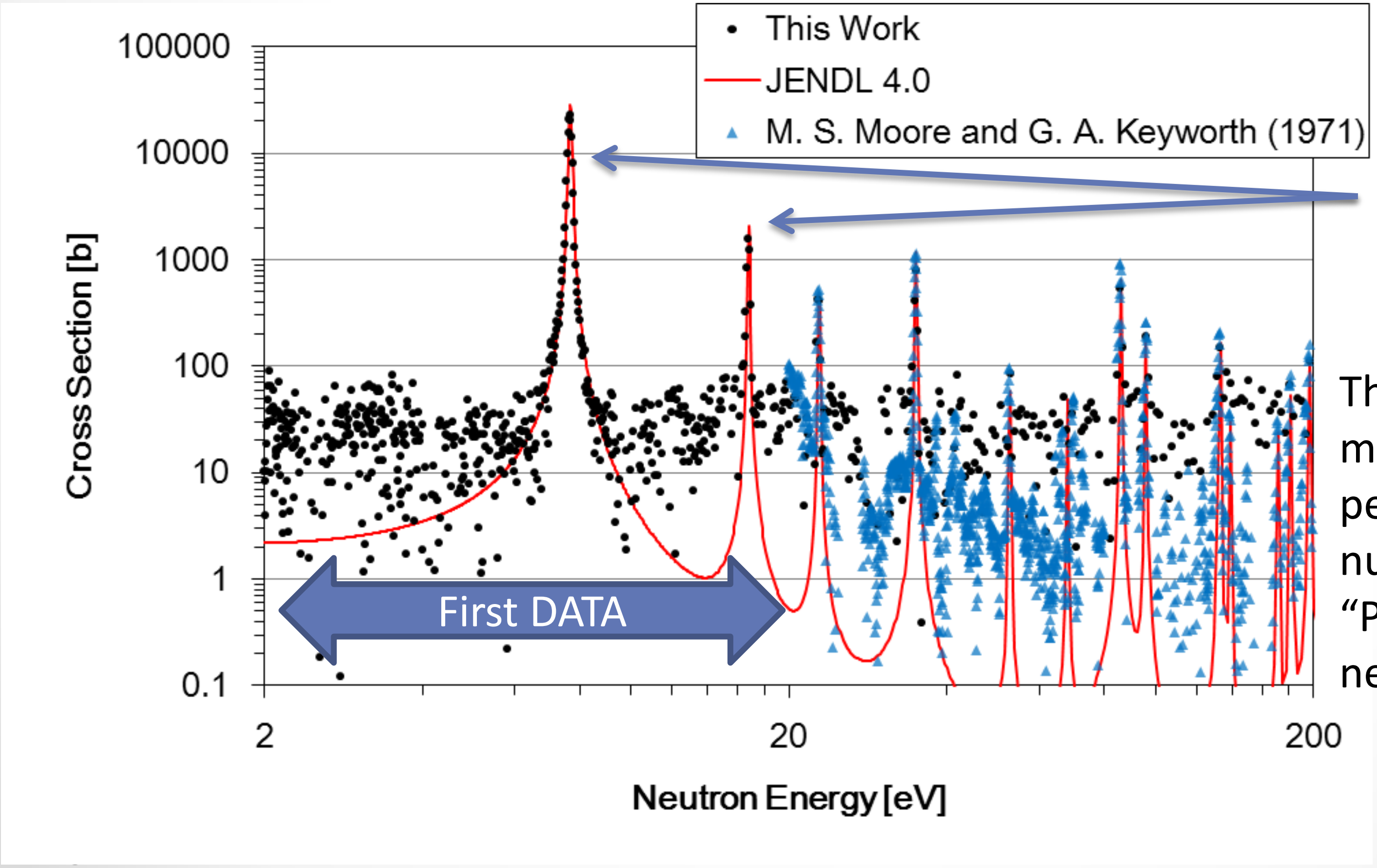
- **Dead-Time Correction**
- **Background Estimation and Subtraction**
- **Self-Shielding and Multiple-Scattering Correction**
- **Normalization (Using the 1st resonance of ^{240}Pu)**
- **Evaluation and Subtraction of Influence of Fission Events**
- **Evaluation and Subtraction of Influence of Impurities**

The obtained neutron-capture cross sections are....

Experiments of $^{244,246}Cm$

~Cross Section of ^{244}Cm~

Only one neutron-capture cross-section data of ^{244}Cm (n,g) was reported in 1969[1].



The results of the resonance peaks under 20-eV are the first experimental results in the world.

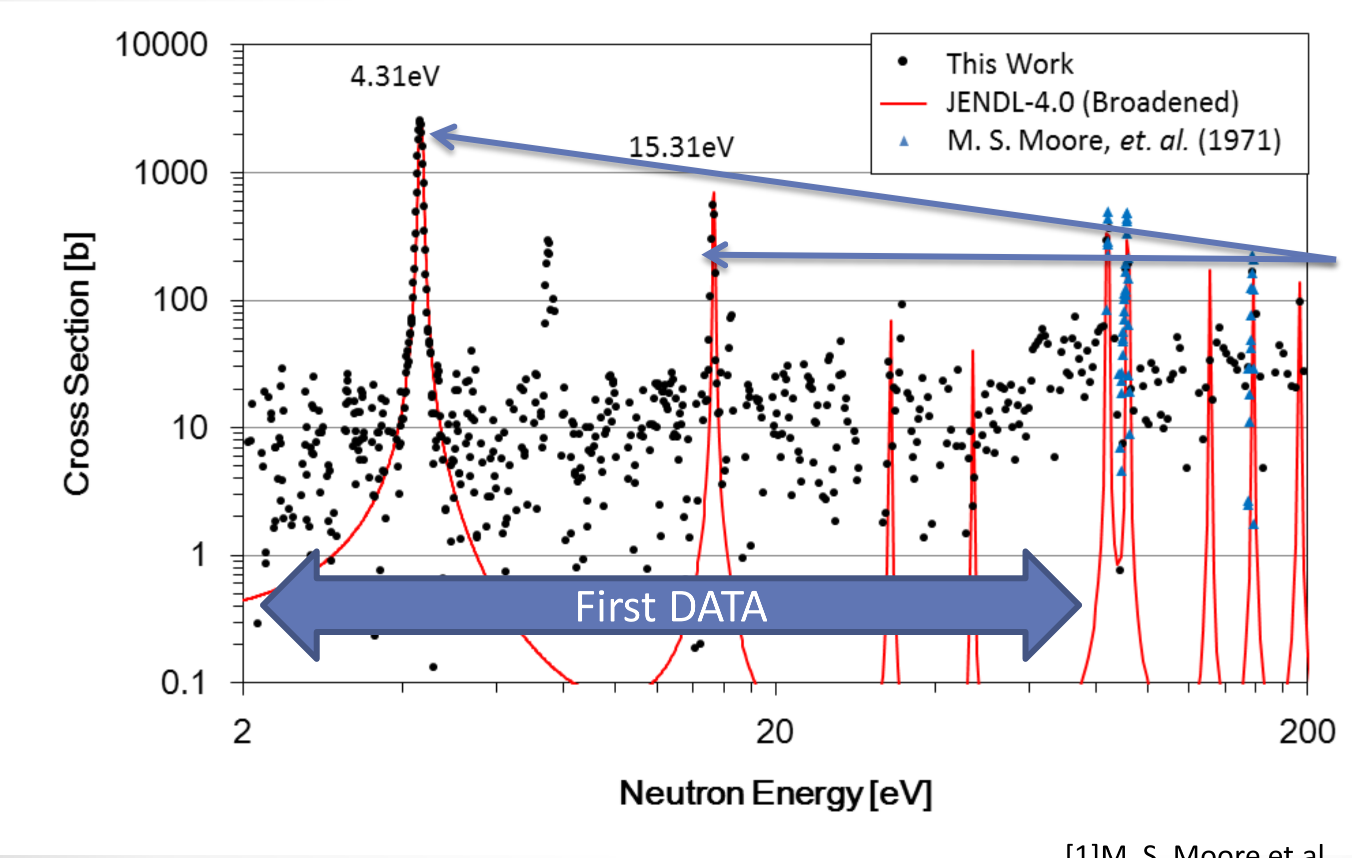
The previous measurement was performed using the nuclear explosion “Physics 8” as a pulsed neutron source in 1969.

[1]M. S. Moore et.al. , Physical Review C, 3, 1656 (1971).

Experiments of $^{244,246}Cm$

~Cross Section of ^{246}Cm~

Only one neutron-capture cross-section data of ^{246}Cm (n,g) was made in 1971[1].



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[1]M. S. Moore et.al. , Physical Review C, 3, 1656 (1971).

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Experiments of ^{93}Zr

~ Sample and Measurement conditions ~

By Dr. Hori

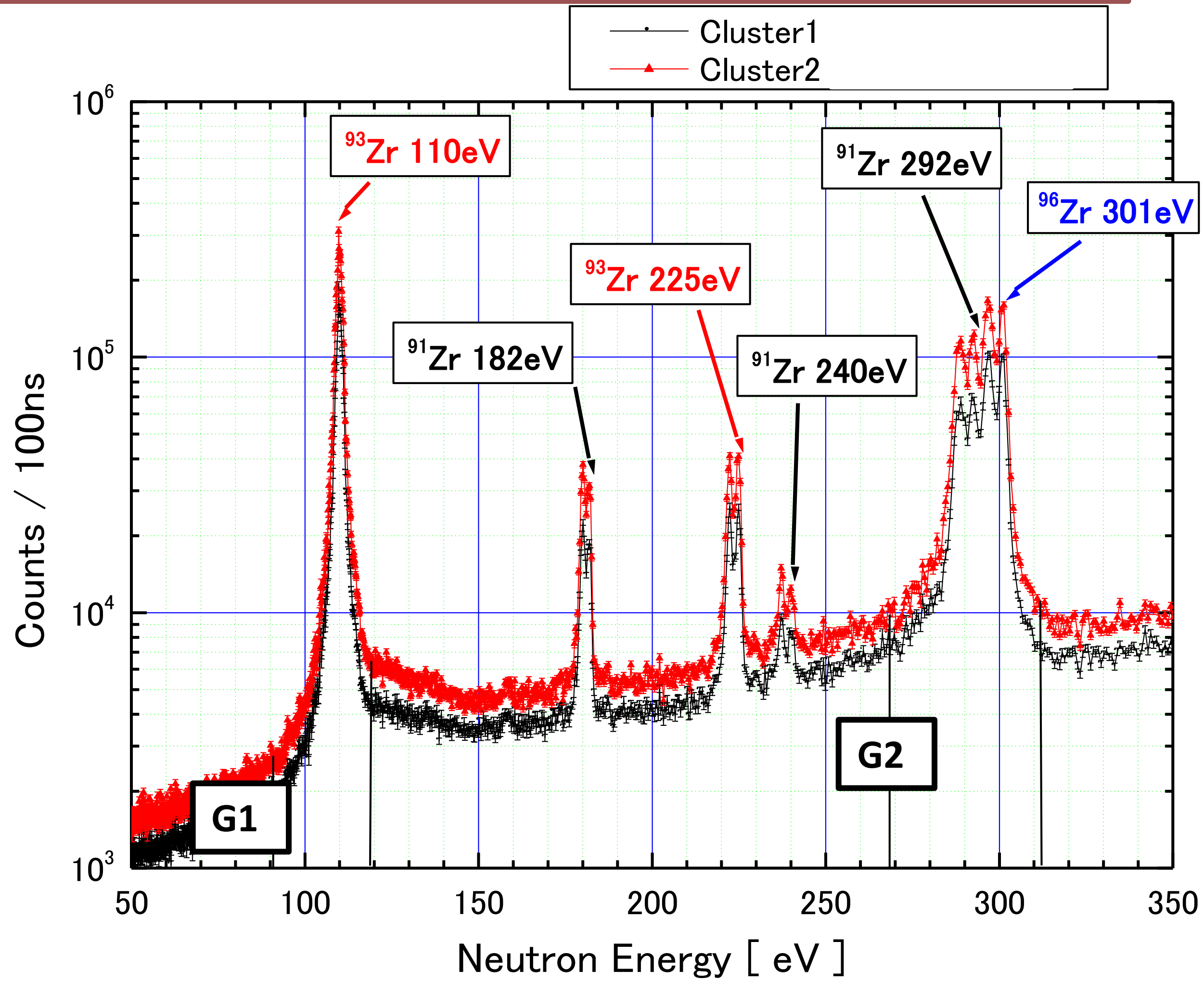
LLFP	Activity (Bq)	Net weight (Total weight)	Measurement Periods	Size (mm)	Manufacture
Zr-93	**47 M**	**470mg (3.31g)**	**52H**	**20ϕ × 5.3**	**USA ORNL**

Zr Isotopes	purity[%]
Zr-90	5.04±0.06
Zr-91	12.36±0.04
Zr-92	15.53±0.03
Zr-93	**18.86±0.04**
Zr-94	22.53±0.02
Zr-96	25.67±0.09

Isotopic purities were determined with a Thermal Ionization Mass Spectrometer (TIMS) at KURRI.
Purity is not enough.
Contamination with impurities are very large.

TOF spectrum of Zr-93

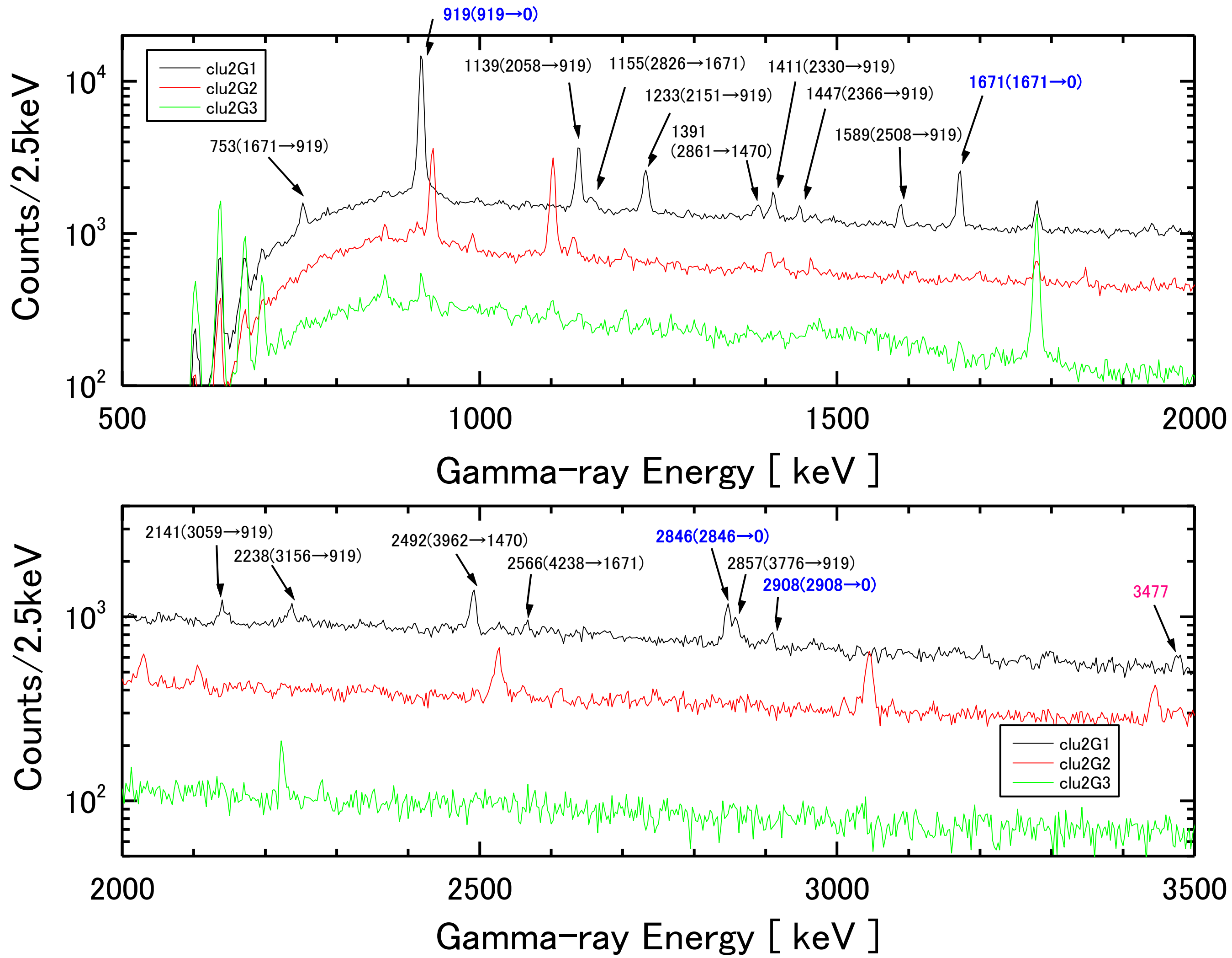
Resonances of ^{93}Zr and stable Zr isotopes were observed.

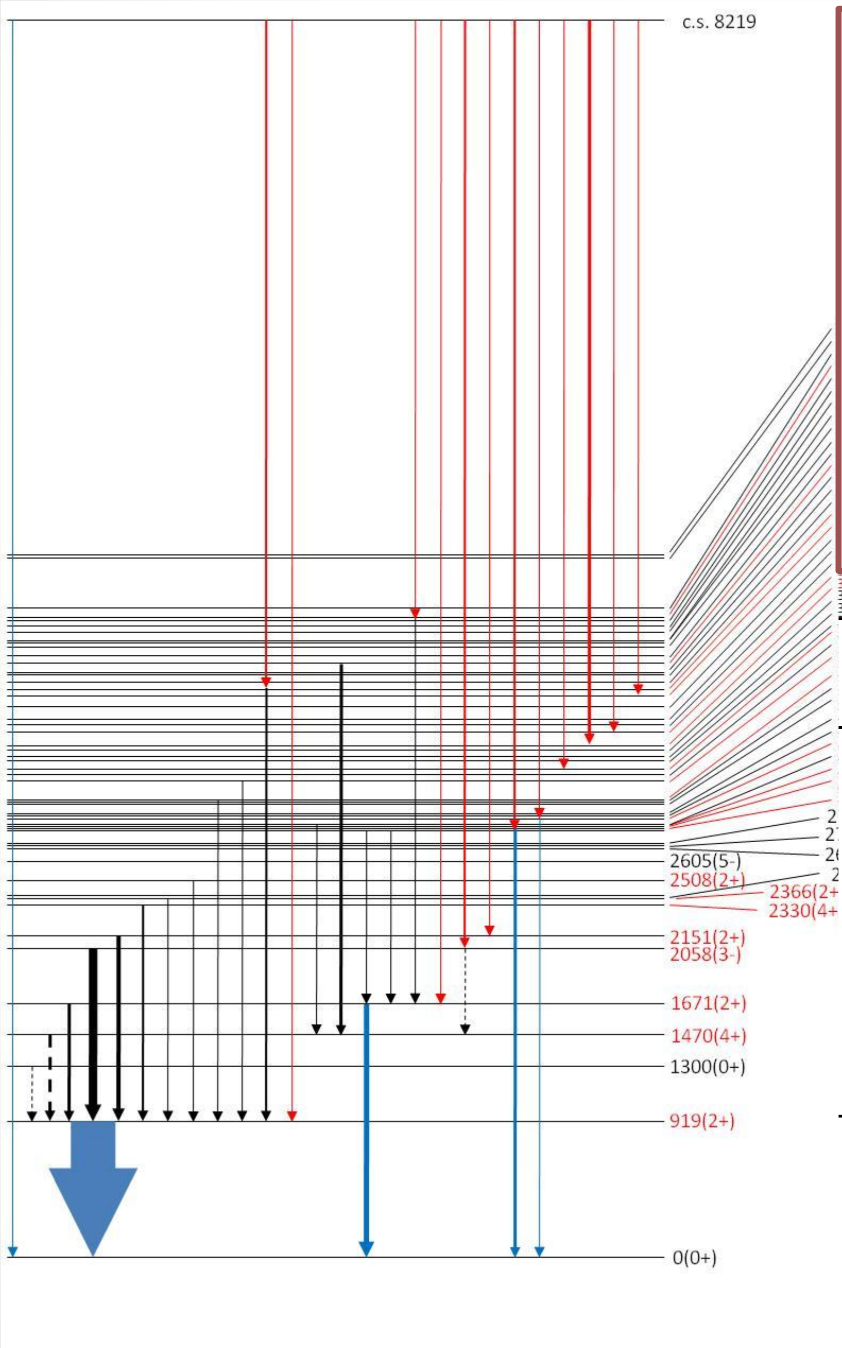


G1(Zr−93):1491−1550ch.(60ch.)
G2(Zr−91,96):936−990ch.(55ch.)
G3(off resonance):1391−1490ch.(100ch.)

P. H. Spectra gated by TOF regions

Black : Zr-93, Red : Zr-91 and Zr-96 resonances, Green : Off resonance





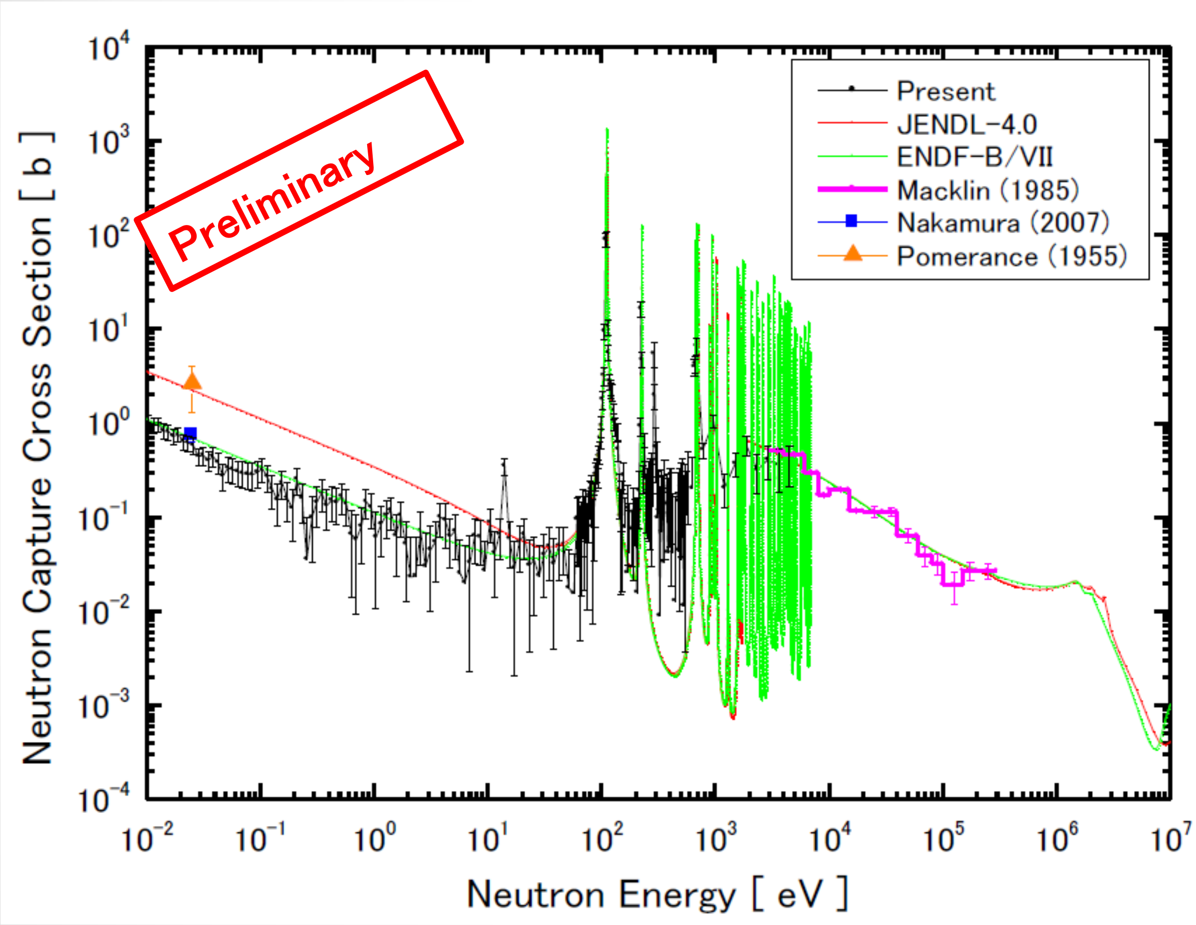
We have observed 5 ground-state, 12 primary and 13 cascade transitions for ^{93}Zr.

Neutron capture cross sections were derived with gating on these 5 ground-state transitions.

Initial state (keV)	Final state (keV)	Energy (keV)	Relative intensities
919	0	919	82.9±4.1
1671	0	1671	9.1±0.5
2846	0	2846	6.0±1.2
2908	0	2908	1.0±0.2
8219	0	8219	1.0±0.2
Total			100

Experiment of ^{93}Zr

~^{93}Zr ~



- We have obtained absolute neutron cross sections of ^{93}Zr by TOF and ground-state transition methods in the energy range from 0.01 eV to 5 keV.
- At thermal neutron energy, our results supported Nakamura's data and evaluated value of ENDF-B/VII. (There is an obvious disagreement with JENDL-4.0.)
- We have found a new resonance at 14 eV.

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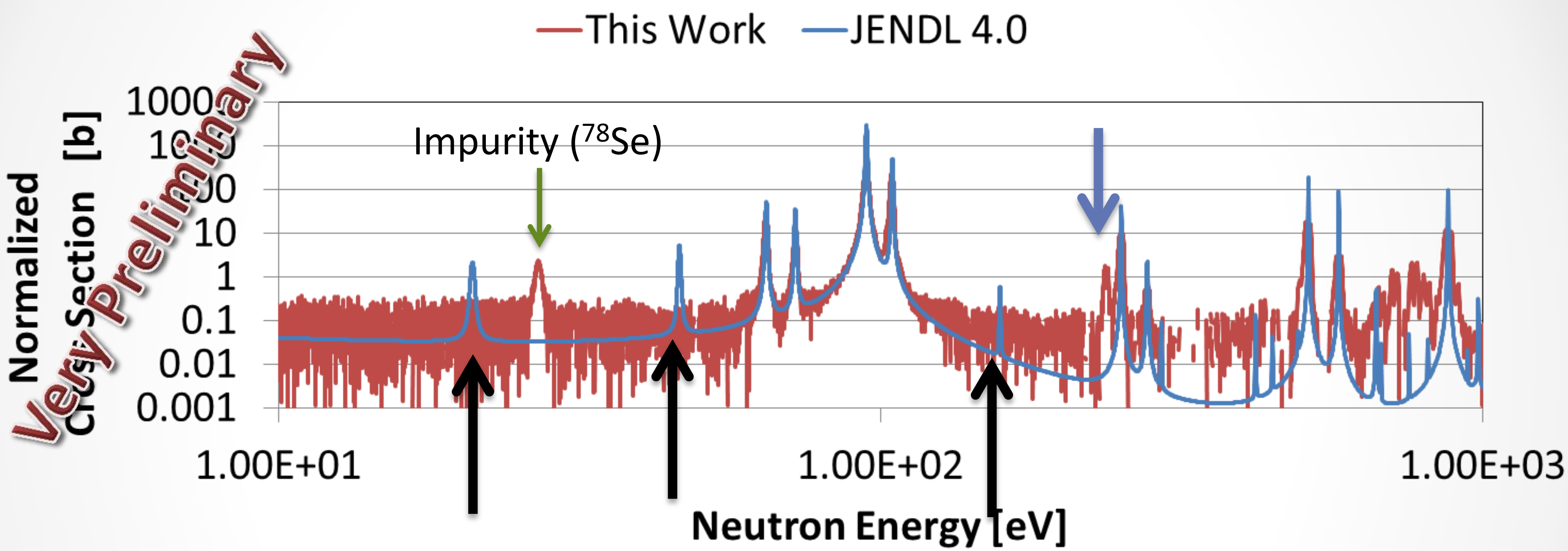
Experiment of ^{112}Sn

~Very Preliminary Cross Section ~

Sample: ^{112}Sn(99.6%) 83.8mg, Beam Power: 210kW, Measurement Periods: 36 hours



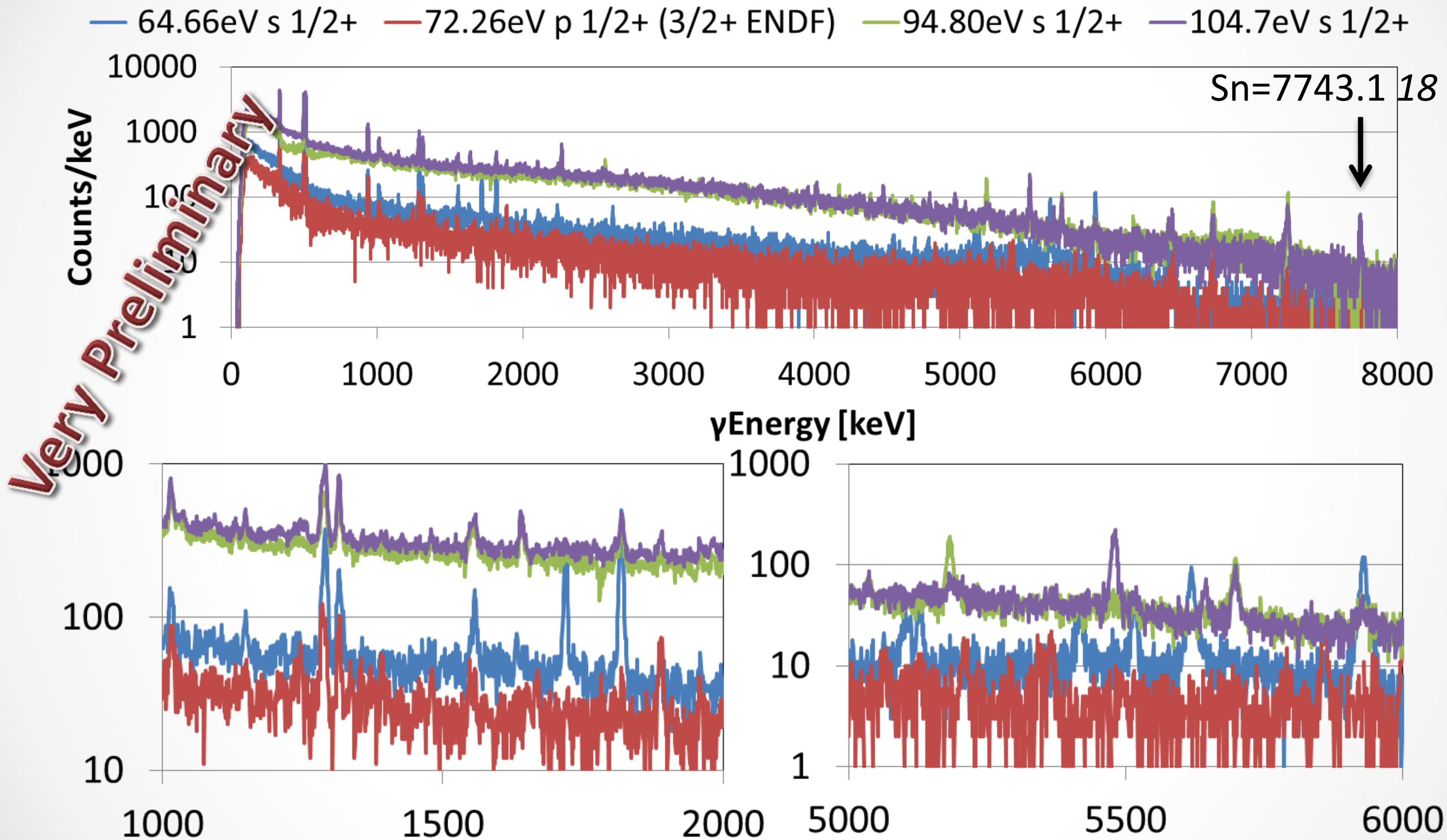
Resonances at 21, 46, and 151 eV were not observed, (although they are listed in both JENDL 4.0 and ENDF B-VII).

Resonance at 240 eV was observed.
This is listed in ENDF B-VII but not listed in JENDL 4.0.

Resonance parameters in JEFF 3.1 are the same as those in JENDL 4.0, except for negative resonance.

Experiment of ^{112}Sn

~Very Preliminary γ-ray spectrum~

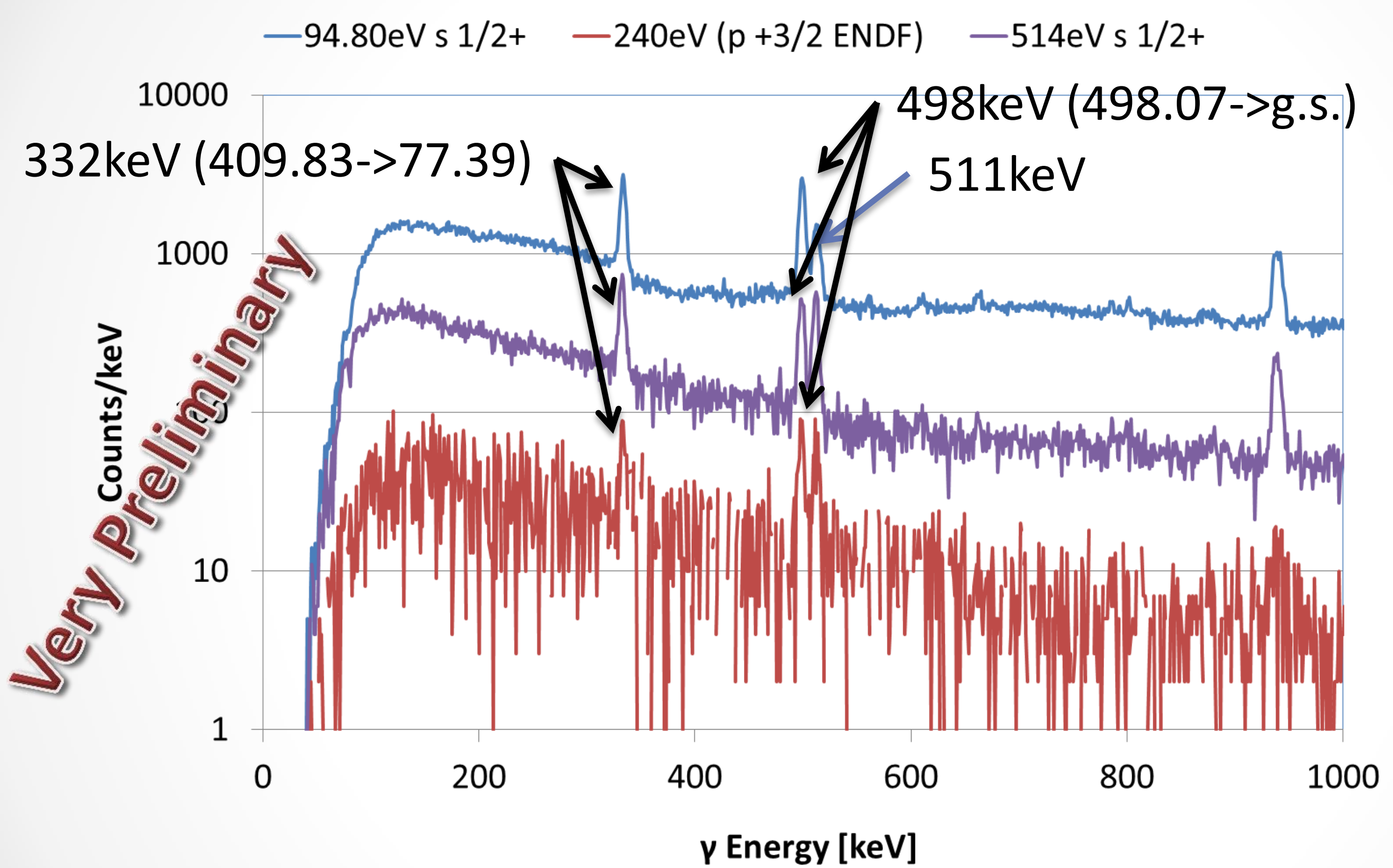


Prompt γ-ray spectra from ^{112}Sn resonances are obtained.
There are many differences between the resonances.

Experiment of ^{112}Sn

~Check the origin of the 240-eV resonance~

Sn-112 0+
Sn-113 ½+



This 240-eV resonance is listed in ENDF B-VII but not listed in JENDL 4.0.

The 332- and 498-keV γ-rays are clearly observed in all resonances.
They have already been studied in other reactions. (^{110}Cd(α,nγ), ^{114}Sn(p,d)...)

This result indicates that
"the 240-eV resonance is clearly one of the ^{112}Sn resonances".

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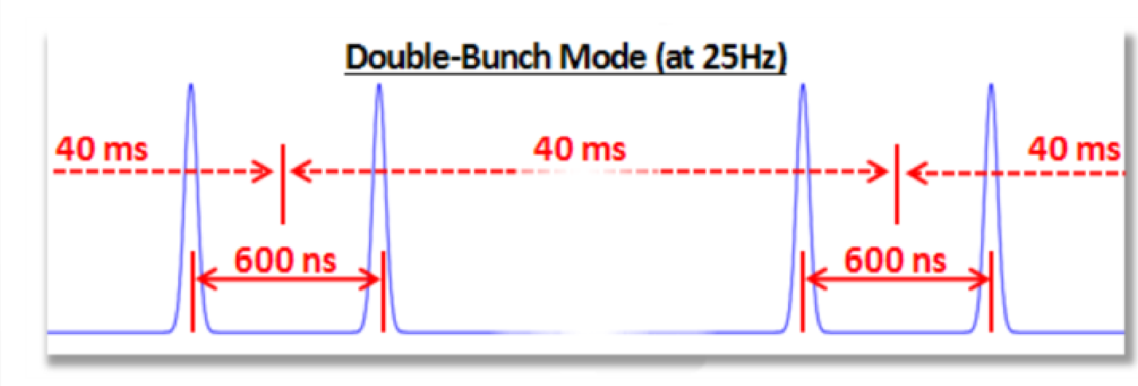
SUMMARY

~Disadvantages of ANNRI~

- Neutron energy resolution is not good!!
  - ✓ The moderator is large (14cm) and flight length is not so long (21.5m).
  - ✓ MLF usually operated with “double-bunch mode”. (with a width of 100 ns, at intervals of 600 ns)

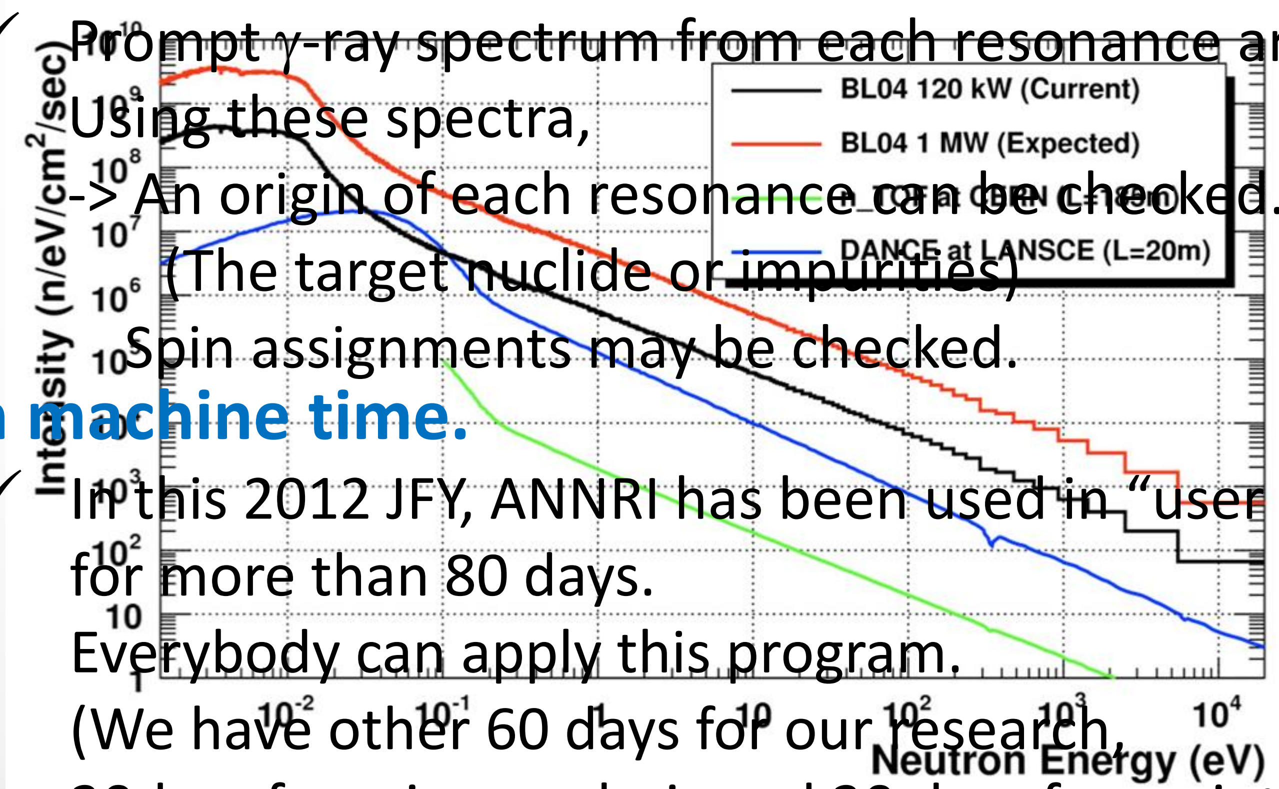


- Non-sealed RI and nuclear fuel materials (U, Pu, and Th) cannot be used, because of regulation of MLF.

SUMMARY

~Advantages of ANNRI~

- **Strong pulsed neutron with High speed DAQ**.
  - ✓ A small amount (less than 1 mg) sample can be used.
- **High γ-ray energy resolution with Ge detectors.**



  - ✓ Prompt γ-ray spectrum from each resonance are obtained.
    Using these spectra,
    -> An origin of each resonance can be checked.
    (The target nuclide or impurities)
    Spin assignments may be checked.
- **Rich machine time.**
  - ✓ In this 2012 JFY, ANNRI has been used in "user program" for more than 80 days.
    Everybody can apply this program.
    (We have other 60 days for our research, 30days for microanalysis and 30 days for maintenance.)

SUMMARY

We have built a new experimental instrument (ANNRI) , and obtained preliminary neutron-capture cross sections of MAs and LLFPs.

- **Results of these experiments show that neutron-capture cross sections are deduced with a small amount (less than 1 mg) and high radioactive sample.**
- **From JFY 2011, ANNRI has been started “user program”. Everyone can be used ANNRI using this program.**

→ http://is.j-parc.jp/uo/index_e.html J-PARC User office

OR Dr. Harada harada.hideo@jaea.go.jp

Acknowledgment

Thank you
for your kind attention!!

LLFP

Nuclei	$T_{1/2}$ [y]	Y[%]	ALI[MBq]	Hazard Index
Se-79	6.50×10^{4}	4.53×10^{-2}	21～160	0.04～0.3
Zr-93	1.53×10^{6}	6.39	54～110	0.35～0.71
Tc-99	2.11×10^{5}	6.11	140	1.9
Pd-107	6.50×10^{6}	0.14	1000～1300	(1.5～1.9)E-4
Sn-126	1.00×10^{5}	5.49×10^{-2}	10	0.5
I-129	1.57×10^{7}	0.72	0.2～0.67	0.63～2.1
Cs-135	2.30×10^{6}	6.53	260	1

$T_{1/2}$: Half-life
Y: Cumulative fission yield
ALI: Annual limit on intake
Hazard Index : $Y/T_{1/2}/ALI$

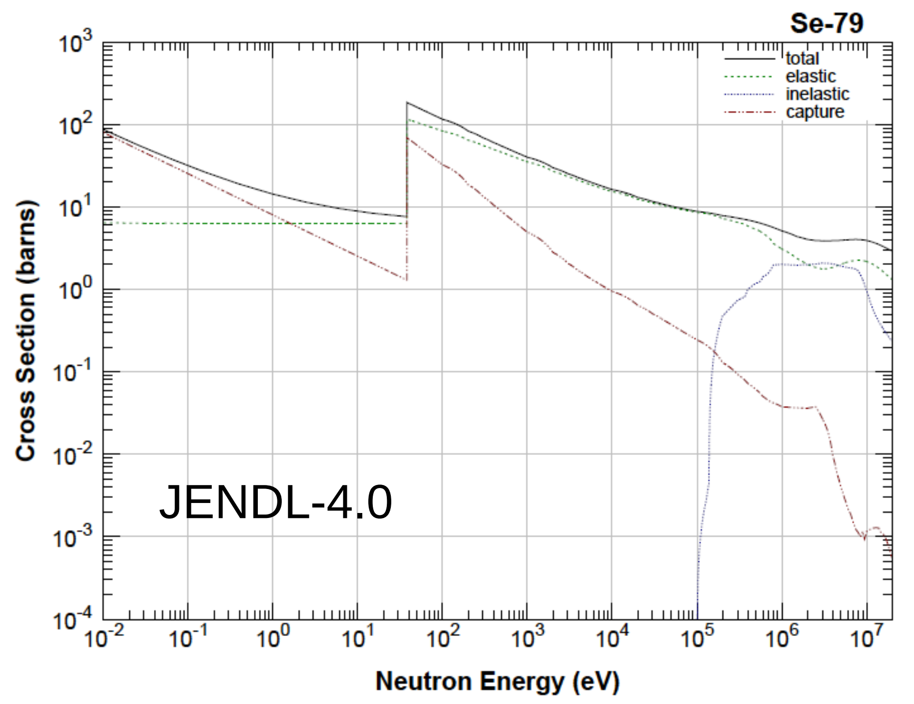
Seven nuclei were selected as candidate nuclei for transmutation.

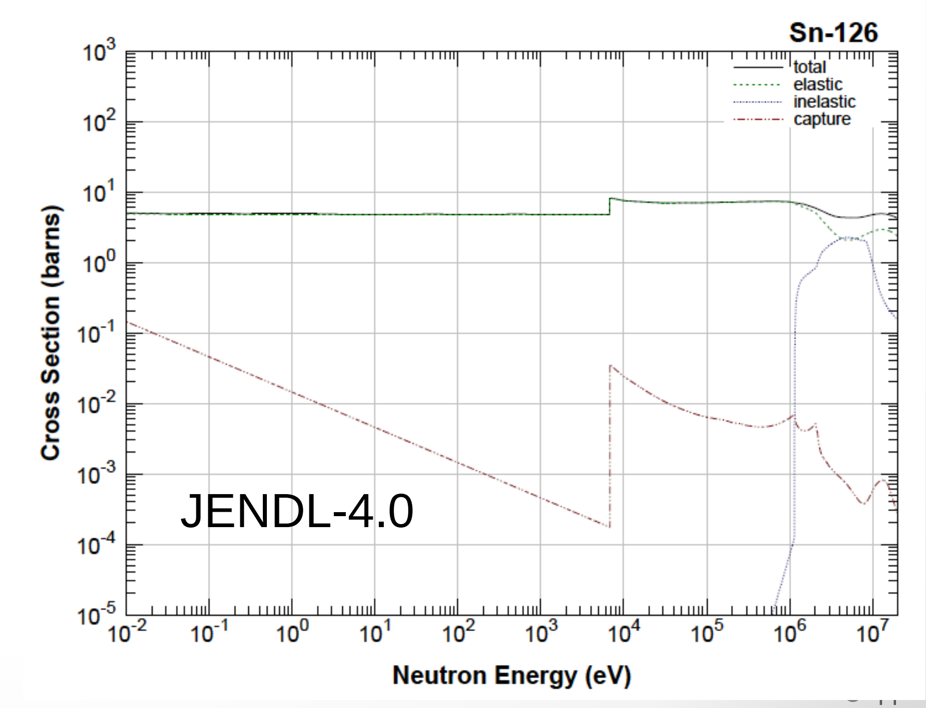
First priority nuclei are Tc-99 and I-129.
Other LLFPs are also important in future.

Introduction(4)

Present status of nuclear data (3)

- Se-79, Sn-126
  No experimental data

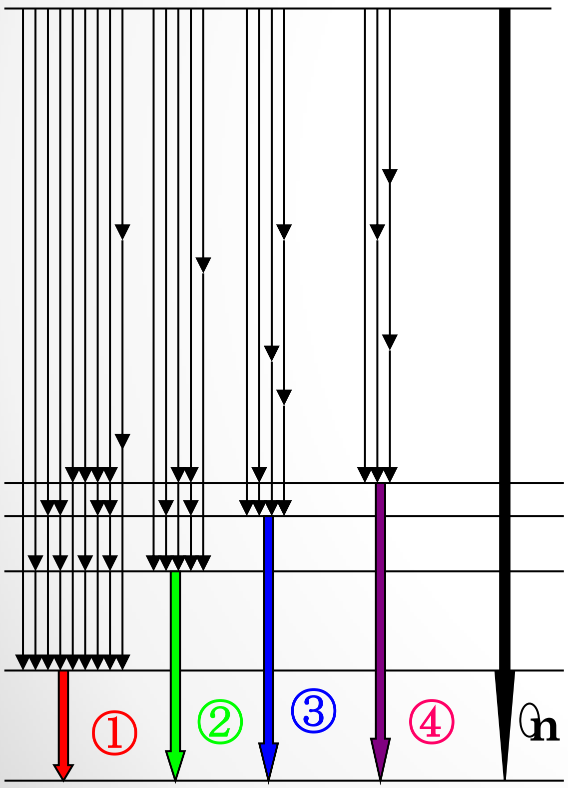




Experiments of LLFPs

~ Ground-state transition method ~

Ground-state transition method was applied for low purity samples.



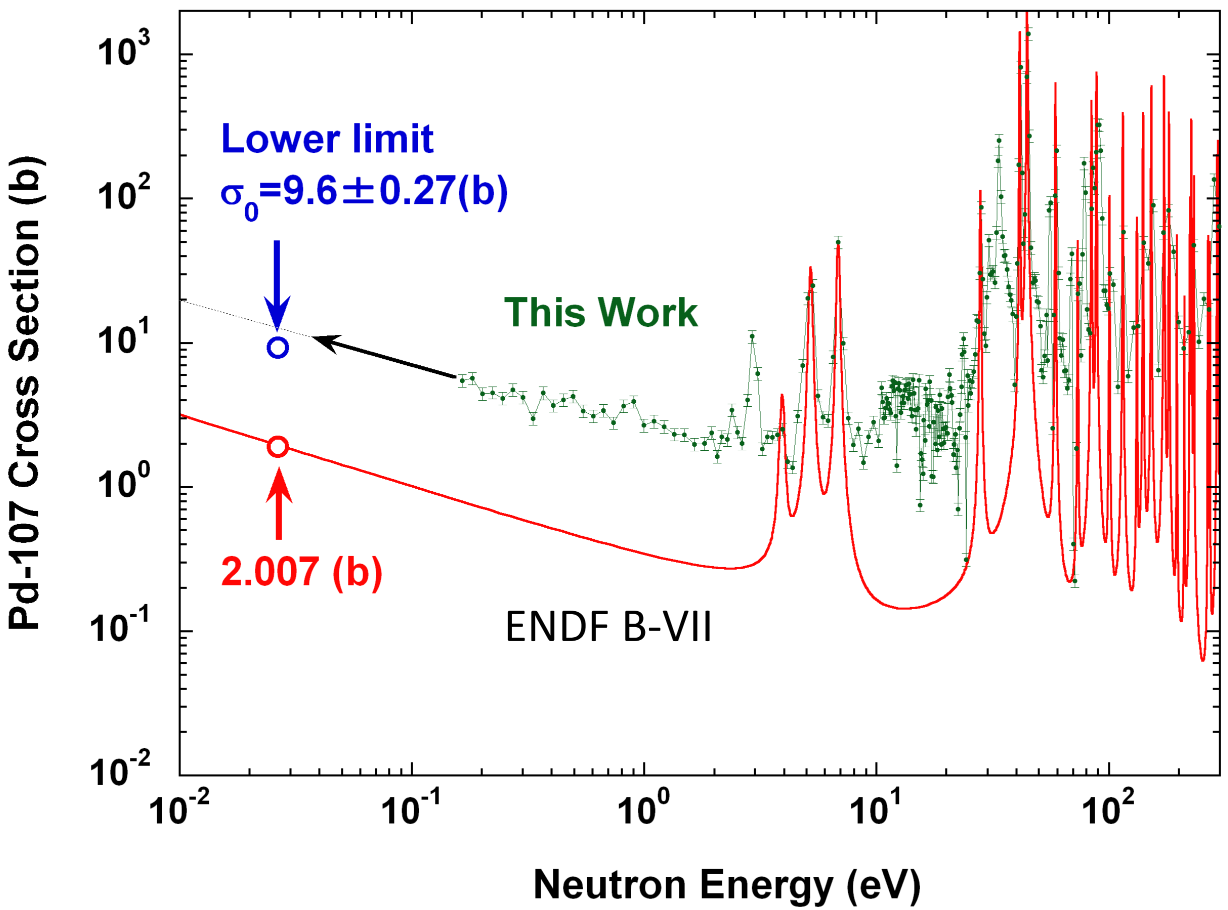
Ground-state transition method :
Cross section was obtained by summing up partial cross section for each ground-state transition.

Advantage

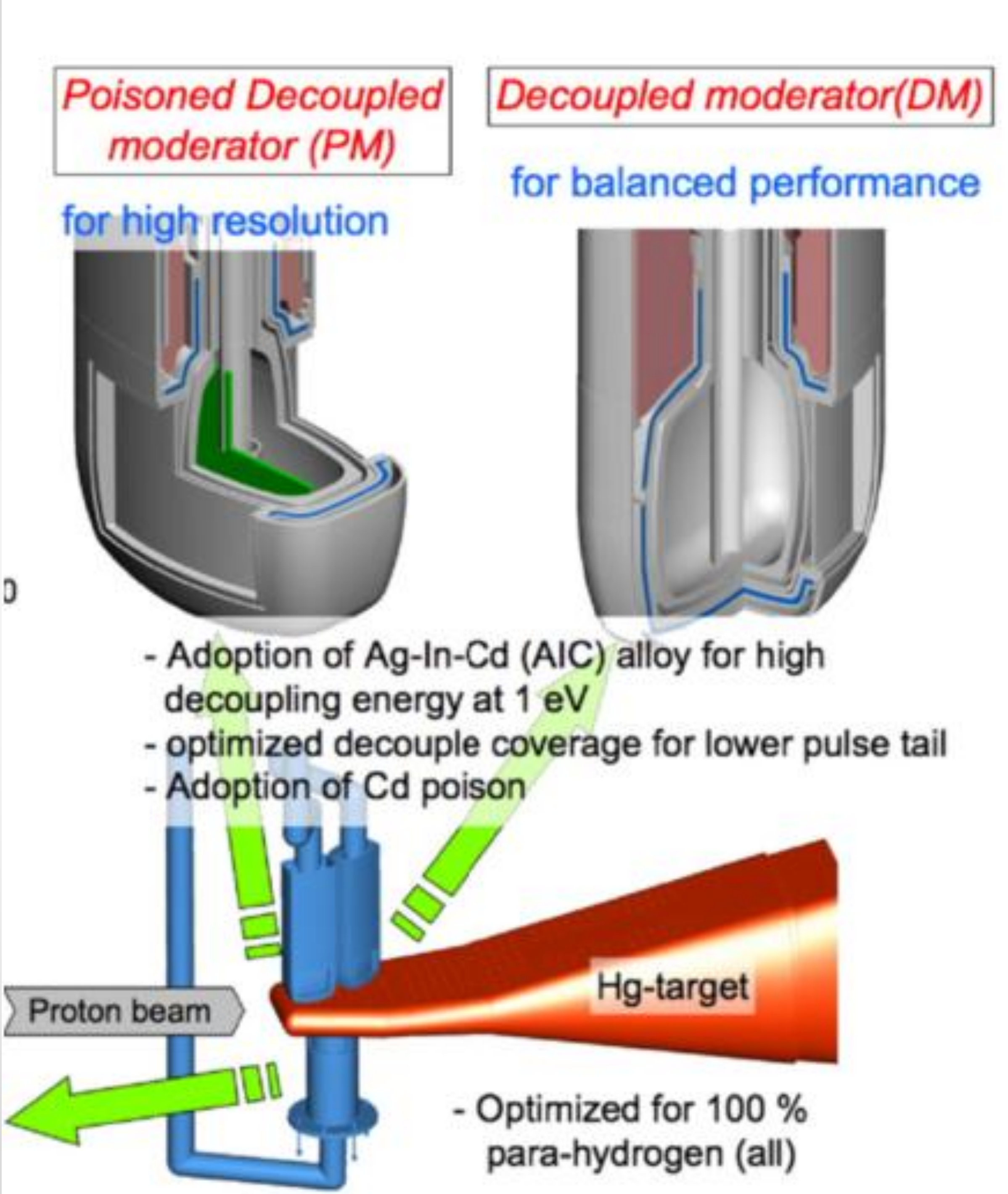
- **The true capture events can be separated from the background due to impurities by using unique γ rays from target.**
- **Lower limit of capture cross sections or absolute yields of the γ rays can be obtained.**
  (Capture cross sections were obtained by normalizing sum of the yields using relative intensities of the unique γ rays)

Experiments of LLFPs
~^{107}Pd ~

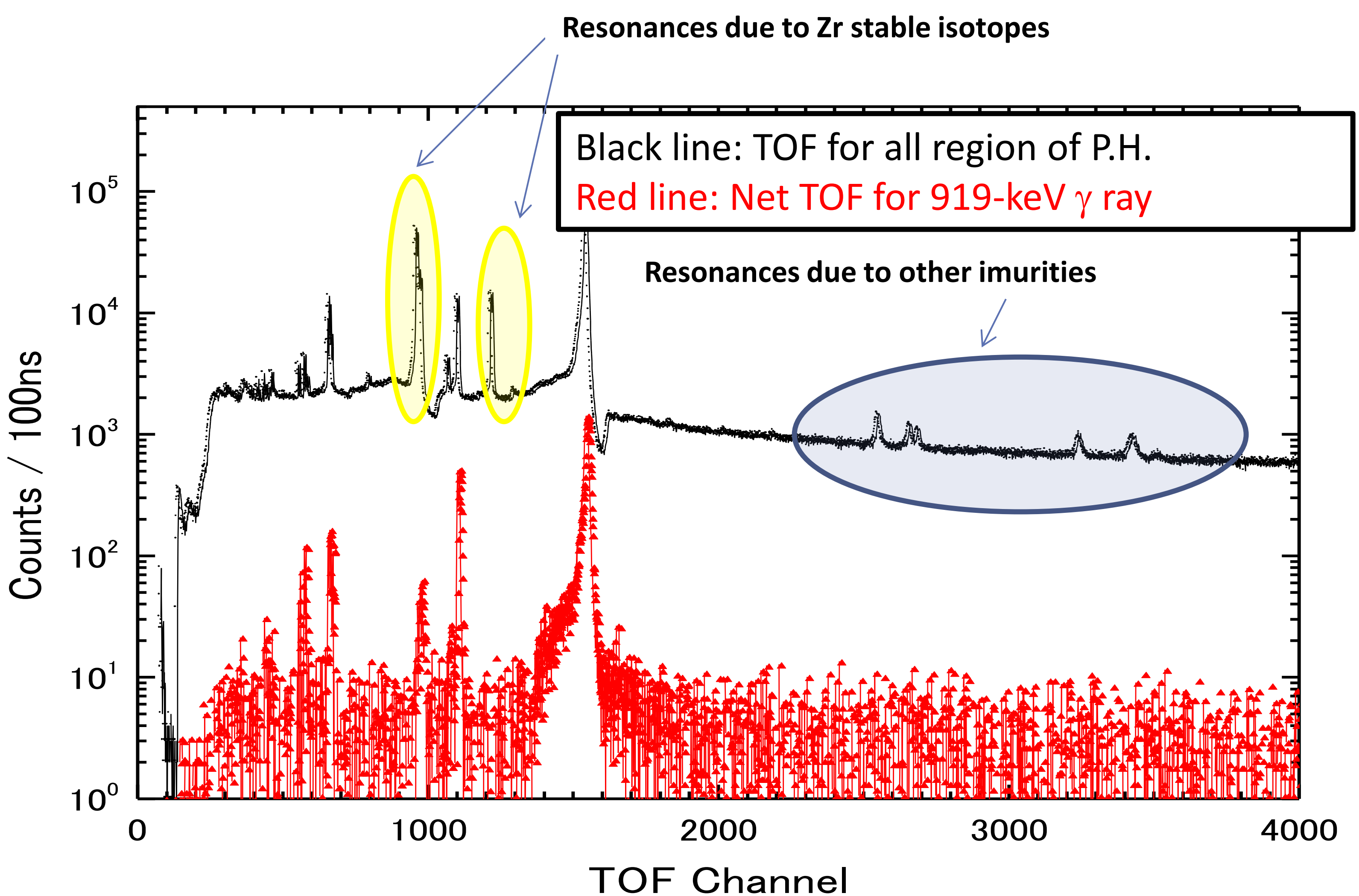
By Dr. Nakamura



- We have obtained neutron cross sections of ^{107}Pd by TOF and ground-state transition methods in the energy range from 0.1 eV to 300 eV.
- The present results supported Nakamura’s data at thermal neutron capture cross section and JENDL 4.0.

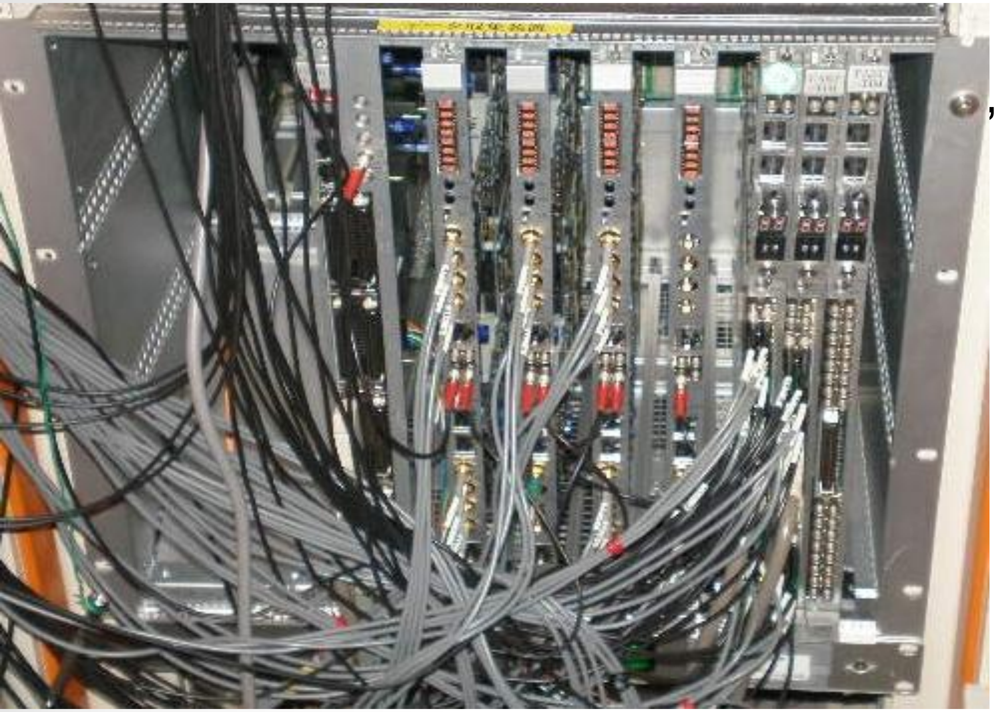

Poisoned Decoupled moderator (PM)
Decoupled moderator(DM)
for balanced performance
for high resolution
- Adoption of Ag-In-Cd (AIC) alloy for high decoupling energy at 1 eV
- optimized decouple coverage for lower pulse tail
- Adoption of Cd poison
Hg-target
Proton beam
- Optimized for 100 % para-hydrogen (all)

TOF spectrum gated at 919keV gamma-ray peak



Extraction of background components due to impurities was done successfully.

Experimental set up
~ Data acquisition system~



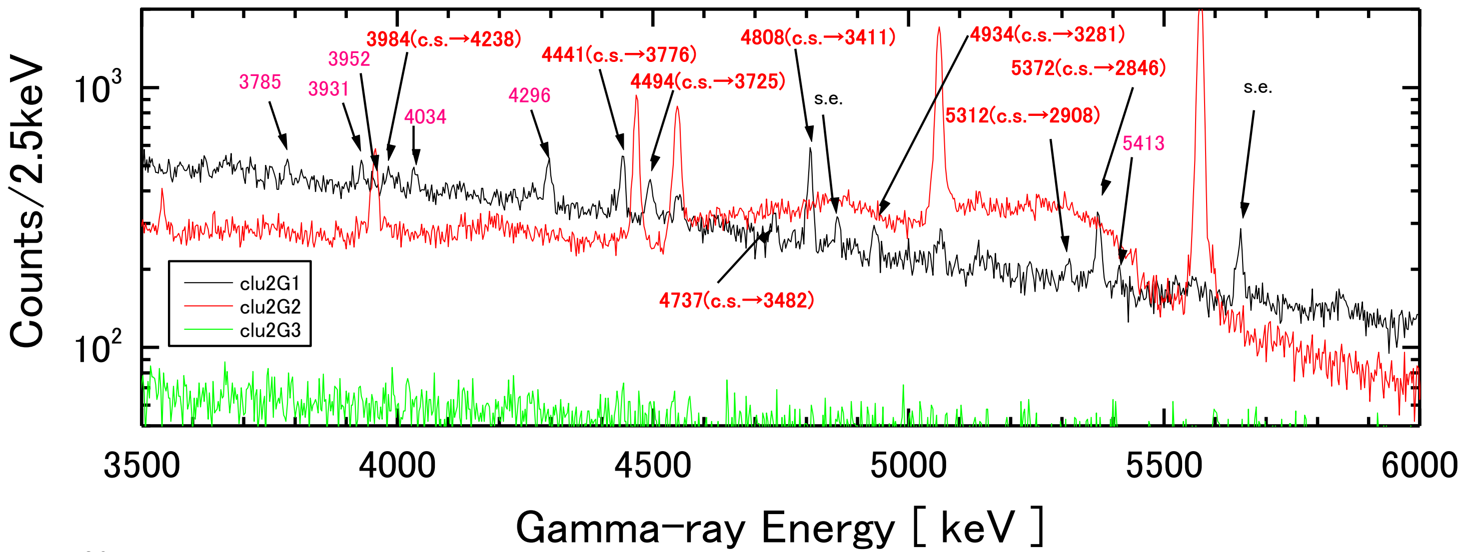
The DAQ were stored
- γ-ray pulse height
- TOF

Time resolution (For TOF):
10 ns (Max 16.7ms)

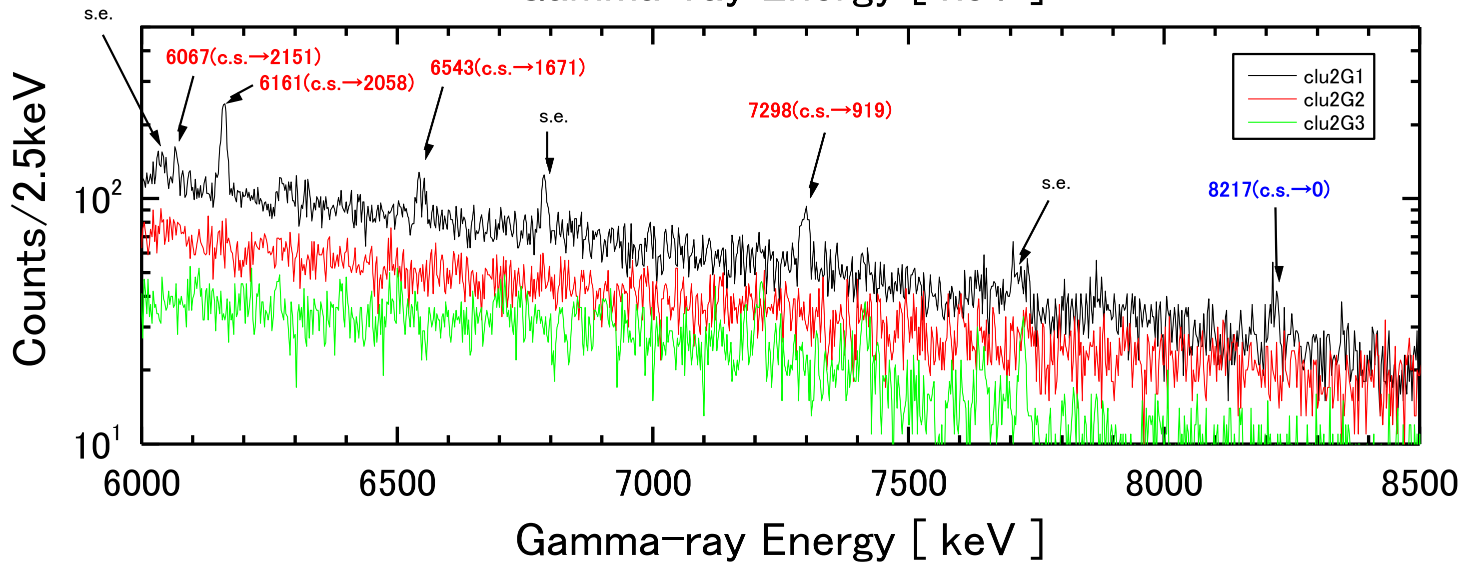
Maximum event rate:
More than 200k events/s.

For dead-time correction, pulses from the random-pulse Generator were input and measured with the DAQ.

This DAQ is a VME-based.
Each channel has
- analog circuits for pulse shaping,
- a high-speed ADC, and
- a high-power Digital Signal Processor



3785
3931
3952
3984(c.s.→4238)
4034
4296
4441(c.s.→3776)
4494(c.s.→3725)
4737(c.s.→3482)
4808(c.s.→3411)
s.e.
4934(c.s.→3281)
5312(c.s.→2908)
5372(c.s.→2846)
5413
s.e.
clu2G1
clu2G2
clu2G3
Counts/2.5keV
Gamma-ray Energy [keV]



s.e.
6067(c.s.→2151)
6161(c.s.→2058)
6543(c.s.→1671)
s.e.
7298(c.s.→919)
s.e.
8217(c.s.→0)
clu2G1
clu2G2
clu2G3
Counts/2.5keV
Gamma-ray Energy [keV]