



Nuclear data measurements and compilation at JAEA

ATSUSHI KIMURA

Contents



- Over View of nuclear data measurement in J-PARC
 - Experimental facility and detectors.
 - Example of Measurements
- Compilation at JAEA
- Request for EXFOR Editor

Nuclear Data Center in JAEA

JAEA

We have 6 sectors.

Sector of Fast Reactor and Advanced Reactor Research and Development

Sector of Fukushima Research and Development

Sector of Nuclear Science Research

J-PARC Center 5 centers in our sector.

Nuclear Science and Engineering Center

Nuclear Data Center (group)

Leader: Osamu Iwamoto

- Evaluation (5 staffs)
Making JENDL
- Experiment (3staffs + 1PhD.)
Experiments in J-PARC

<https://wwwndc.jaea.go.jp/>

Motivation for our team

The present status of experimental data for MAs and LLFPs is not sufficient both in quality and quantity.

This is because it is not easy to prepare enough amount of samples with high purity. Moreover, some MAs are highly radioactive.



Improvements in this study

1) Intense neutron source

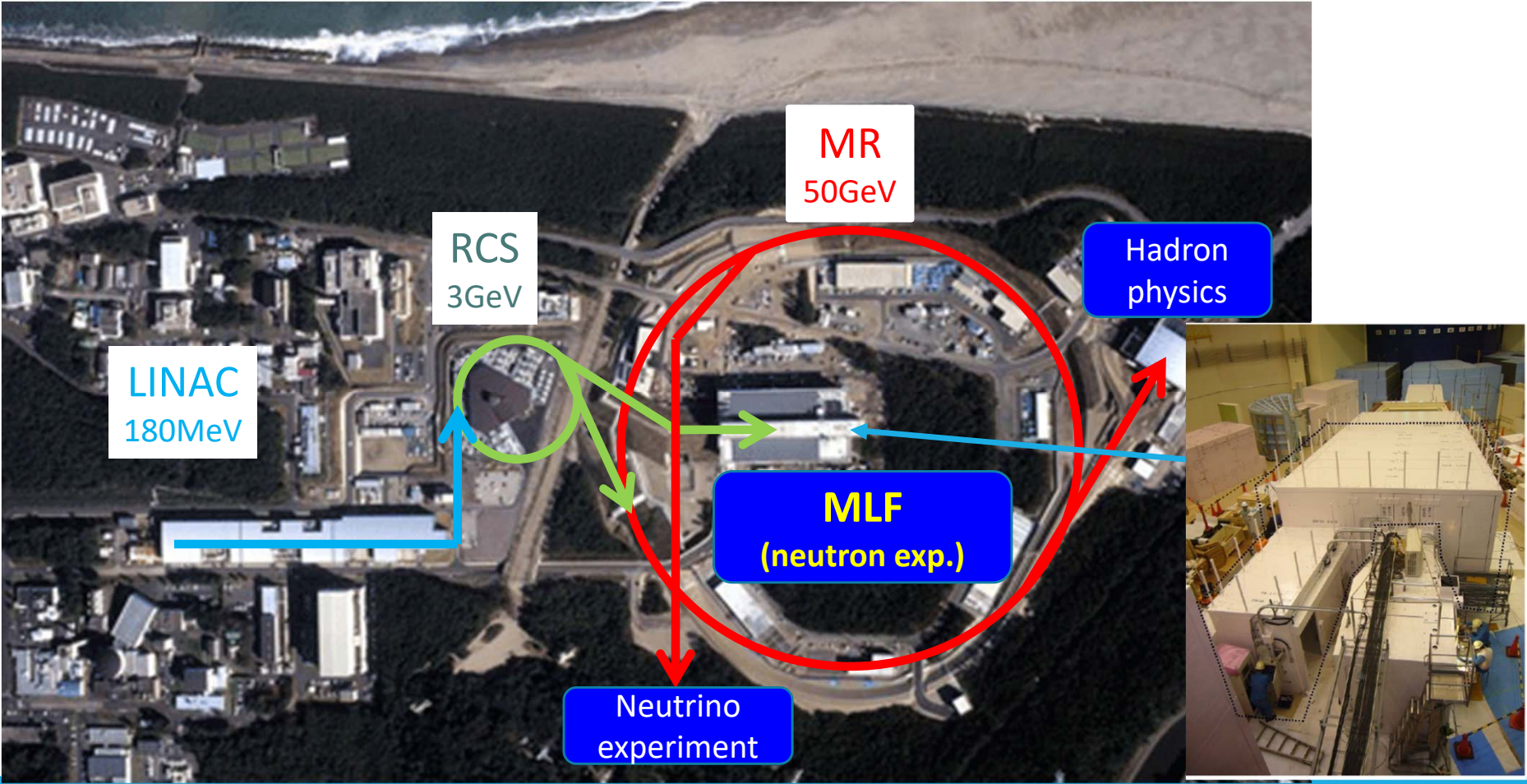
- A small amount of samples can be used for experiments
- Influence due to decay γ -rays can be reduced.

2) High energy resolution and high-efficiency γ -ray detector systems were applied to the TOF measurement (For LLFPs)

- Background due to impurities can be removed.

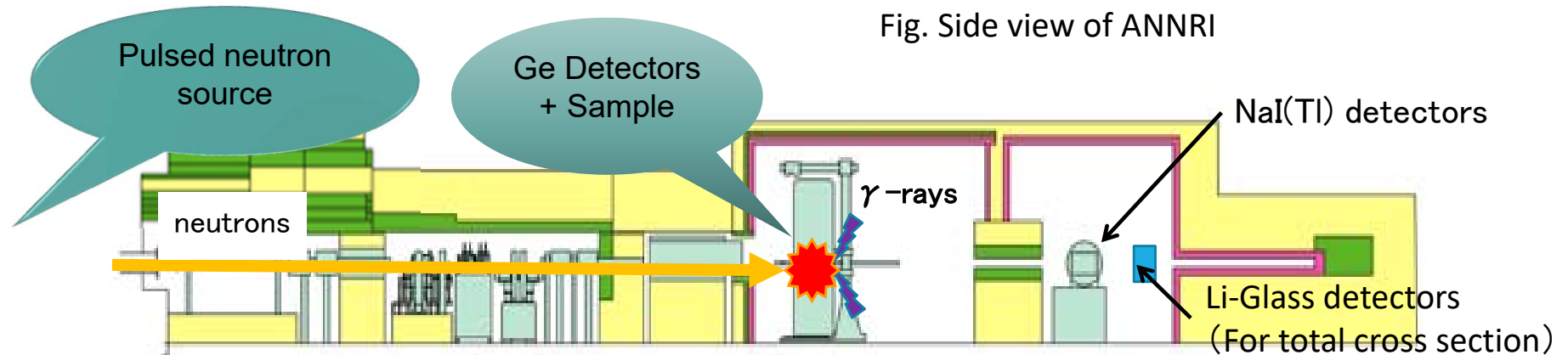
We constructed the Accurate Neutron-Nucleus Reaction measurement Instrument (ANNRI) in J-PARC.

Japan Proton Accelerator Research Complex (J-PARC)



ANNRI

Accurate Neutron-Nucleus Reaction measurement Instrument (ANNRI)



In ANNRI, there are three spectrometers.

- An array of large Ge detectors
 - NaI(Tl) detectors
 - Li-glass detectors for transmission experiments.
- } for capture cross section measurements

Neutron collimators, resonance filters, and chopper systems are installed

➡ ANNRI is used for nuclear data measurement and microanalysis.

~ Ge detector-array (For Capture) ~



Our spectrometer has

- 2 cluster-Ge detectors
(7 Ge crystals are incorporated in one cluster detector)
- 8 coaxial-Ge detectors
- Compton suppressing BGO detectors
⇒ 22 Ge Crystals.

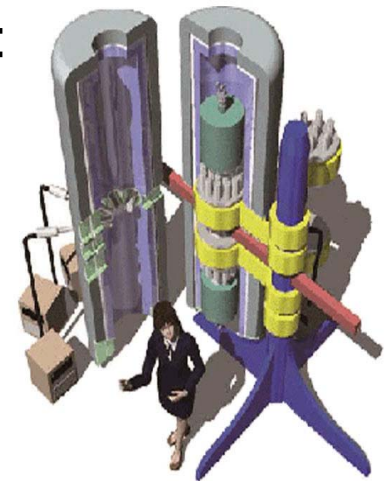
Energy resolution for 1.33MeV γ -rays:

5.8keV (for 200 kevents/s),

2.4keV (for 20 kevents/s) [1]

Peak efficiency for 1.33MeV γ -rays:

$3.64 \pm 0.11 \%$



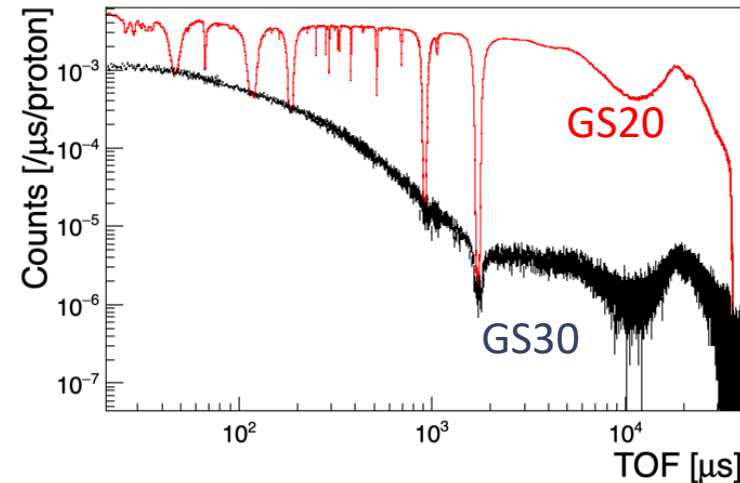
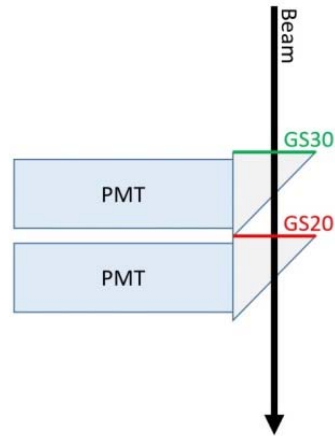
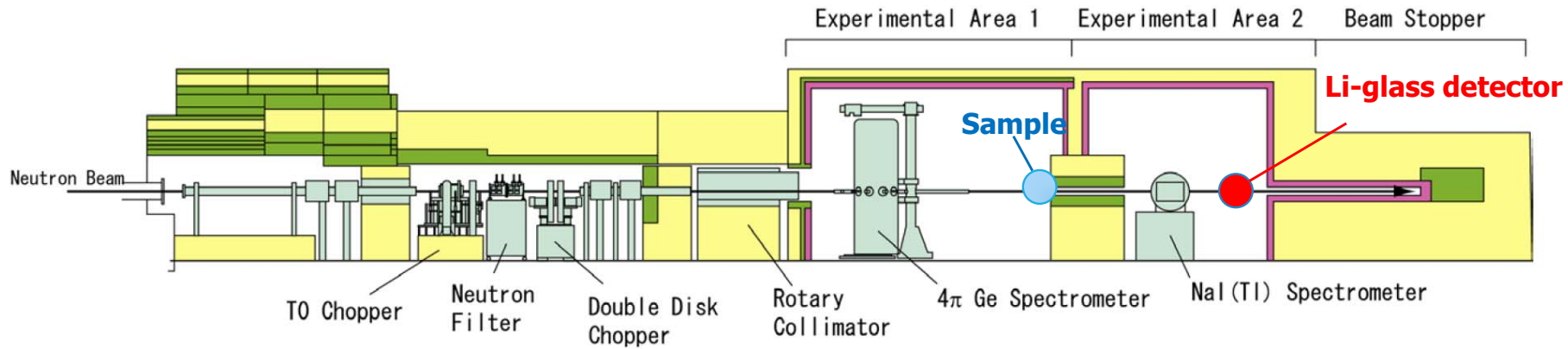
~NaI(Tl) detectors (For Capture)~



- **NaI(Tl) Detectors:**
 - 90° detector: 13" diam. x 8" long
 - 125° detector: 8" diam. x 8" long
- **Flight Length: 28m**
- **Shielding**
 - Borated Polyethylene, lead, ^6LiH , $^6\text{LiCO}_3$
- **Data acquisition**
 - Neutron TOF and detected γ -ray energy are recorded

Mainly used for High energy region.

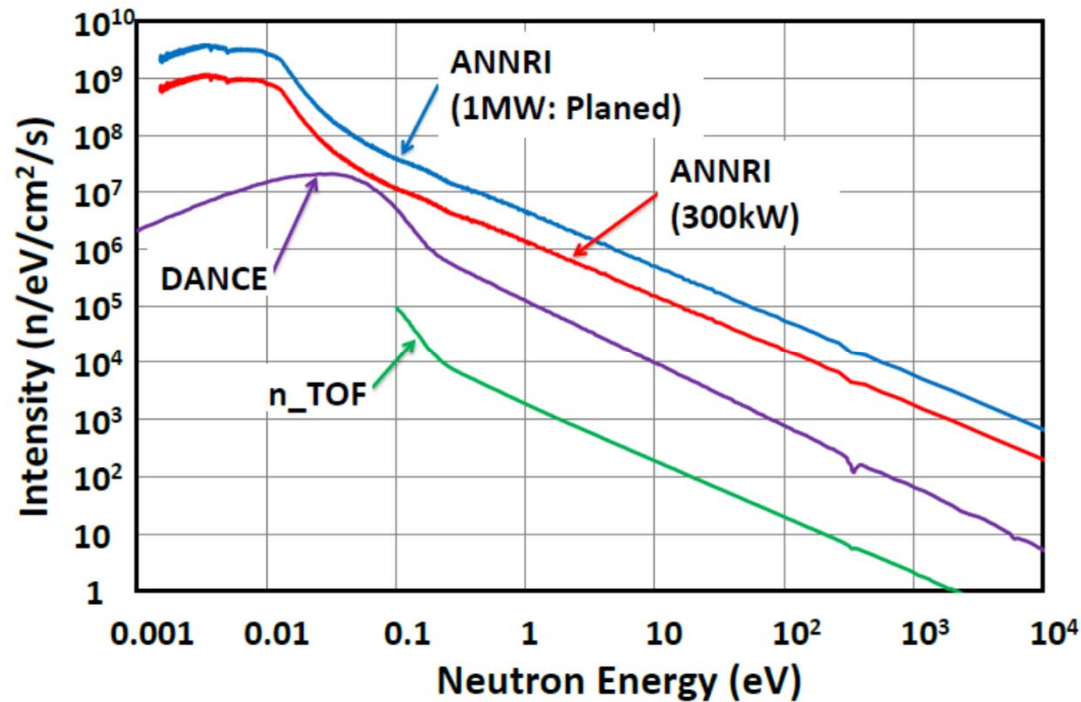
~ Li-glass Detectors (For total cross section) ~



A ^6Li -glass scintillation detector (GS-20) for neutron and a ^7Li -glass scintillation detector (GS-30) for B.G. were installed at the flight length of 28.7m

By taking a ratio of transmitted neutrons with/without a sample, total cross sections can be derived.

Beam Intensities Comparison to major facilities



U.S. LANSCE @LANL

Detector for Advanced Neutron Capture Experiments (DANCE)
BaF detectors
Flight length 20m

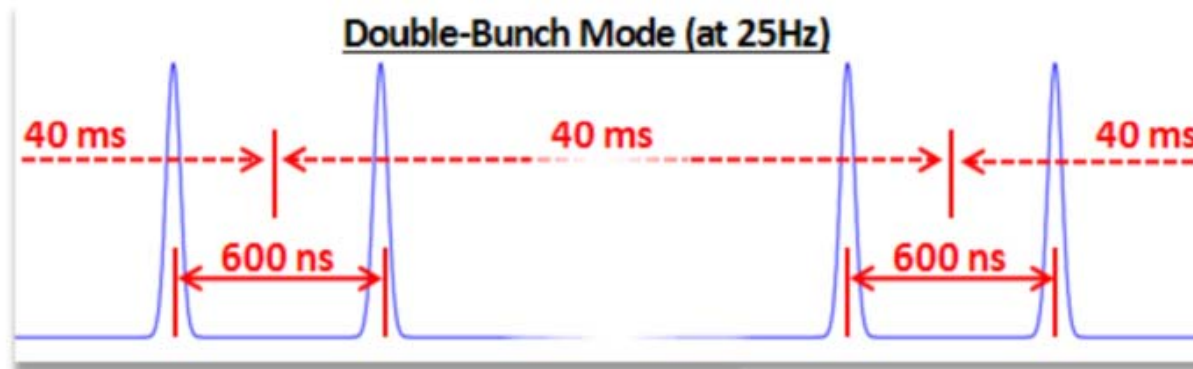
EU n_TOF@CERN

C6D6 detectors and BaF detectors
Experimental Room 1: 185m
Experimental Room 2: 20m
Beam Intensity per shot in Room2 is about 50% of ANNRI.
But beam frequency is 0.3~1.2Hz

In the epithermal energy region, the neutron intensity of ANNRI is more than 7 times as high as the values of the other instruments.

However, neutron energy resolution was not so good!!

- The pulsed protons usually consist of two bunches (called “double-bunch mode”), each with a width of 100 ns, at intervals of 600 ns



Most users in MLF are users of diffractometers, scattering spectrometers and reflectometers. They require “neutron intensity”.

Characteristics of ANNRI

- **High intensity pulsed neutron with High speed DAQ.**

✓ A small amount (less than 1 mg) sample can be used.

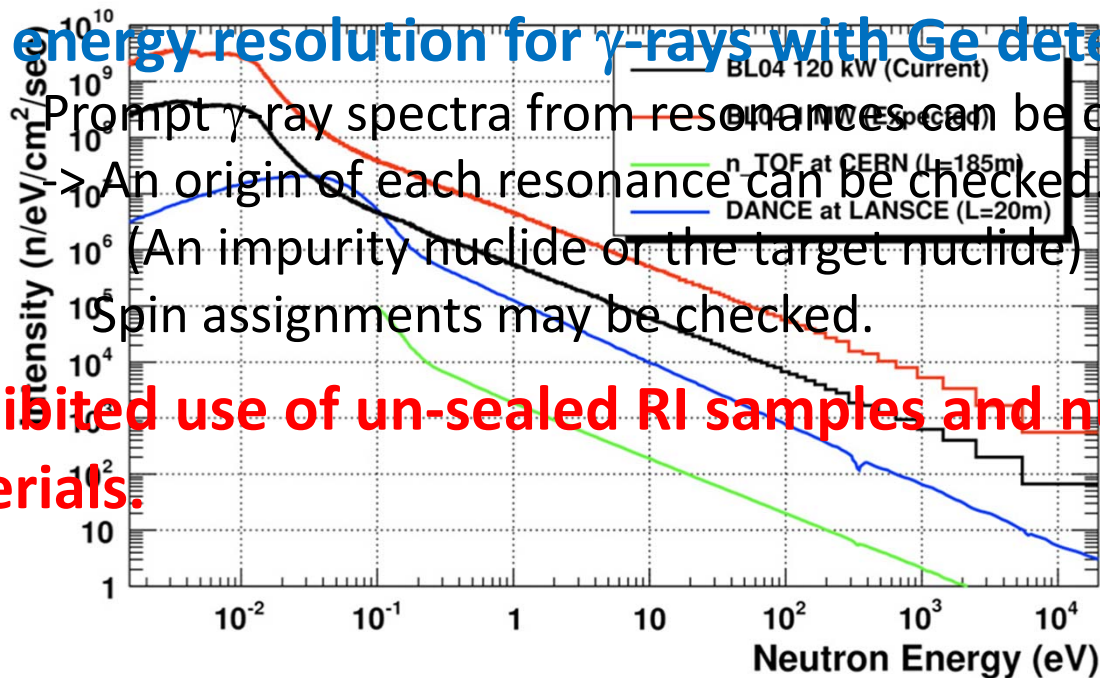
- **High energy resolution for γ -rays with Ge detectors.**

✓ Prompt γ -ray spectra from resonances can be obtained.

→ An origin of each resonance can be checked.
(An impurity nuclide or the target nuclide)

Spin assignments may be checked.

- **Prohibited use of un-sealed RI samples and nuclear materials.**



~Measurement status@ANNRI~

Sealed RI Samples

MA Samples

237 ~~241~~ ~~243~~

244

LLFP

9

1

Stable Samples

54, 56, 57 Fe, 58, 60, 61, 62 Ni

74, 76, 77, 78, 80 Ce

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

- Cm-244, Ref: JNST,49,708 (2012)
 - Am-243 (MA) Ref: JNST, 56, 479 (2019)
 - Sn-112 (Stable) Ref: Nuclear Data Sheets, 119, 150 (2014)
- will be presented.

Red: Already Published. Green: Published with preliminary results.
Blue: Already Measured. Black: Future Plan in few years.

Experiments of ^{244}Cm

~Samples and Measurement conditions~



^{244}Cm sample (%)

^{244}Cm	89.57\pm1.68
^{245}Cm	2.66 \pm 0.34
^{246}Cm	7.08\pm0.33
^{247}Cm	Not Detect
^{248}Cm	Not Detect

- Sample :

^{244}Cm : **1.8GBq, 0.6mg**

Isotopic Ratio: 89.6%

Chemical form = CmO_2

Container = Al capsule

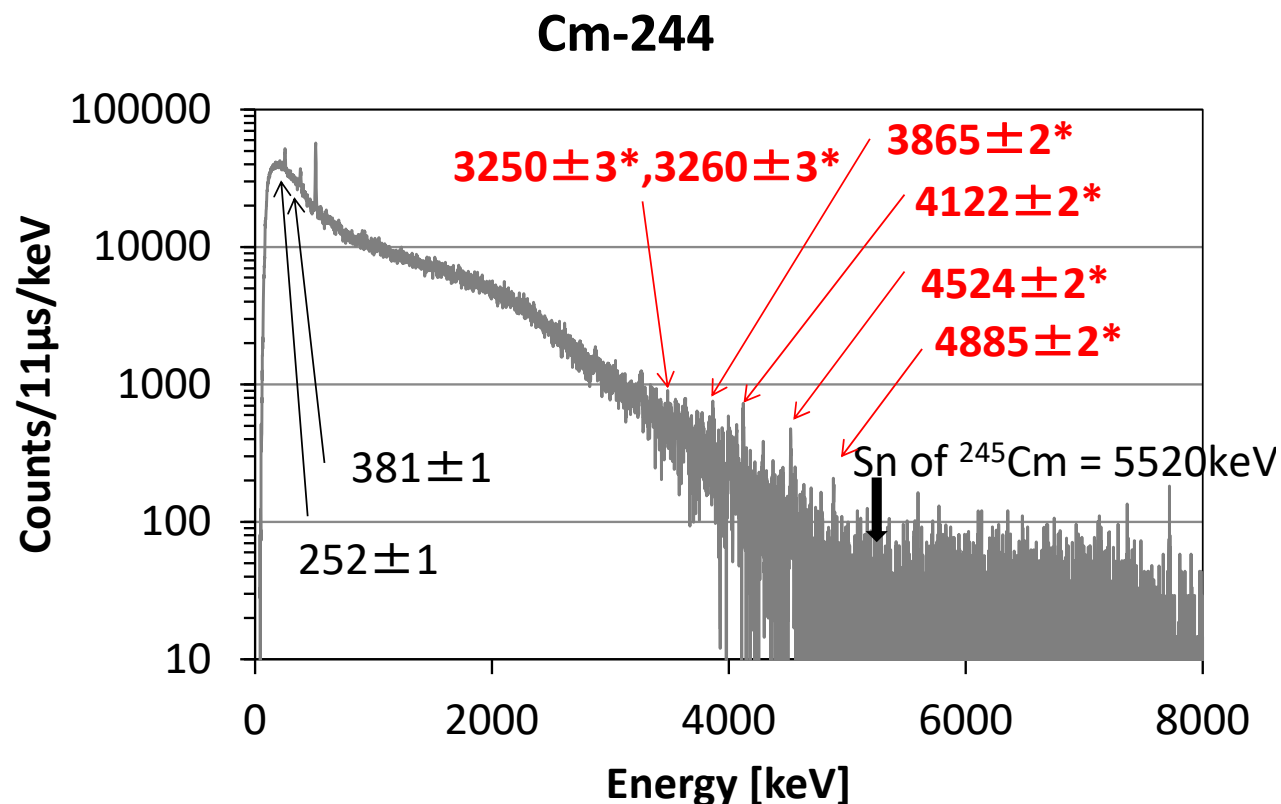
- Beam Operation : 120kW 25Hz

For the background estimation, a dummy case and a blank sample was measured 48 and 44 hours.

Sample	Measurement Time[h]
Cm-244	74
Dummy Case	48
Blank(Holder+Film)	44

Experiments of ^{244}Cm

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



The 252.4- 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}Cm ~Analysis~

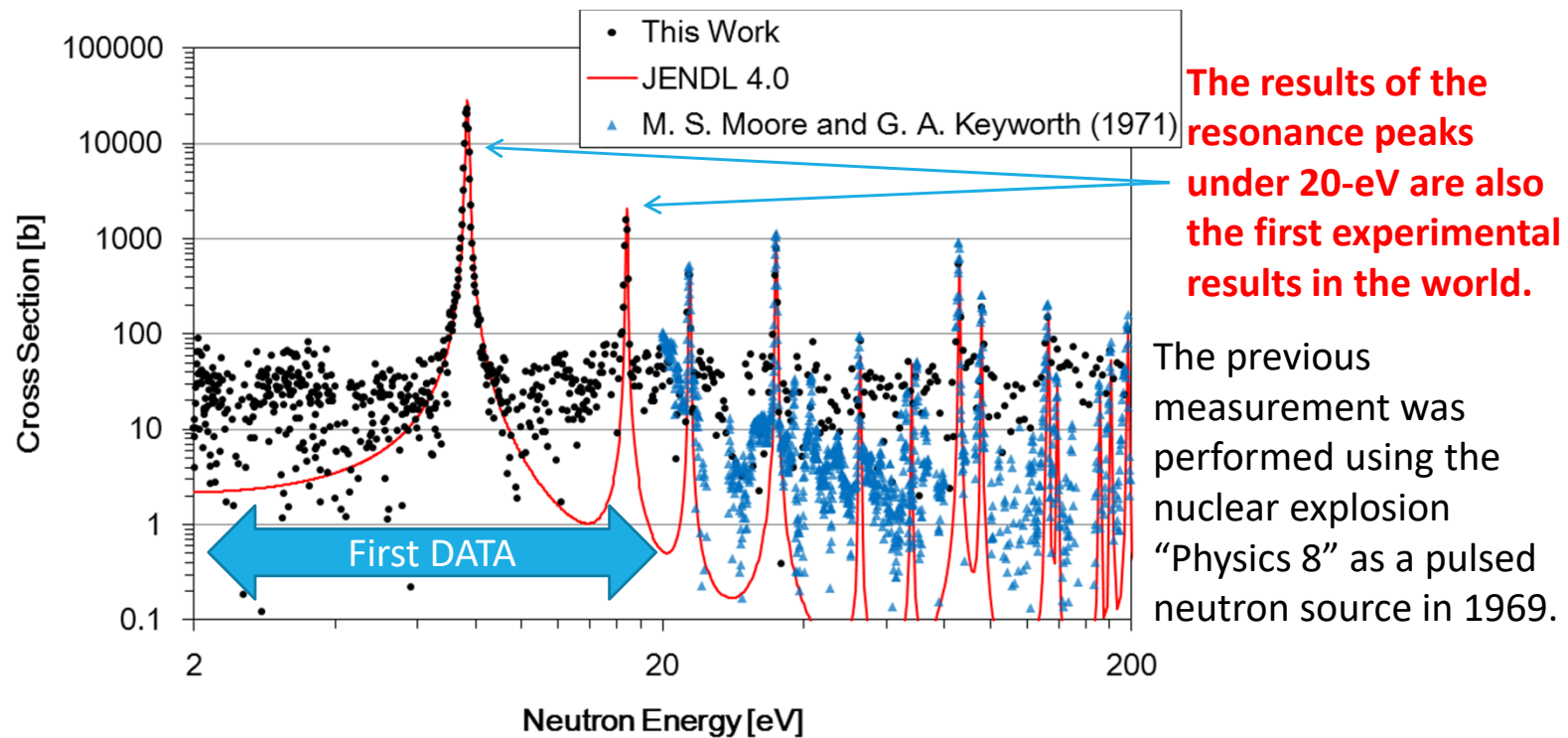
The data were analyzed with the procedure.

- **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}Cm ~Cross Section~

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



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

Experiments of ^{243}Am

~Capture Cross sections~

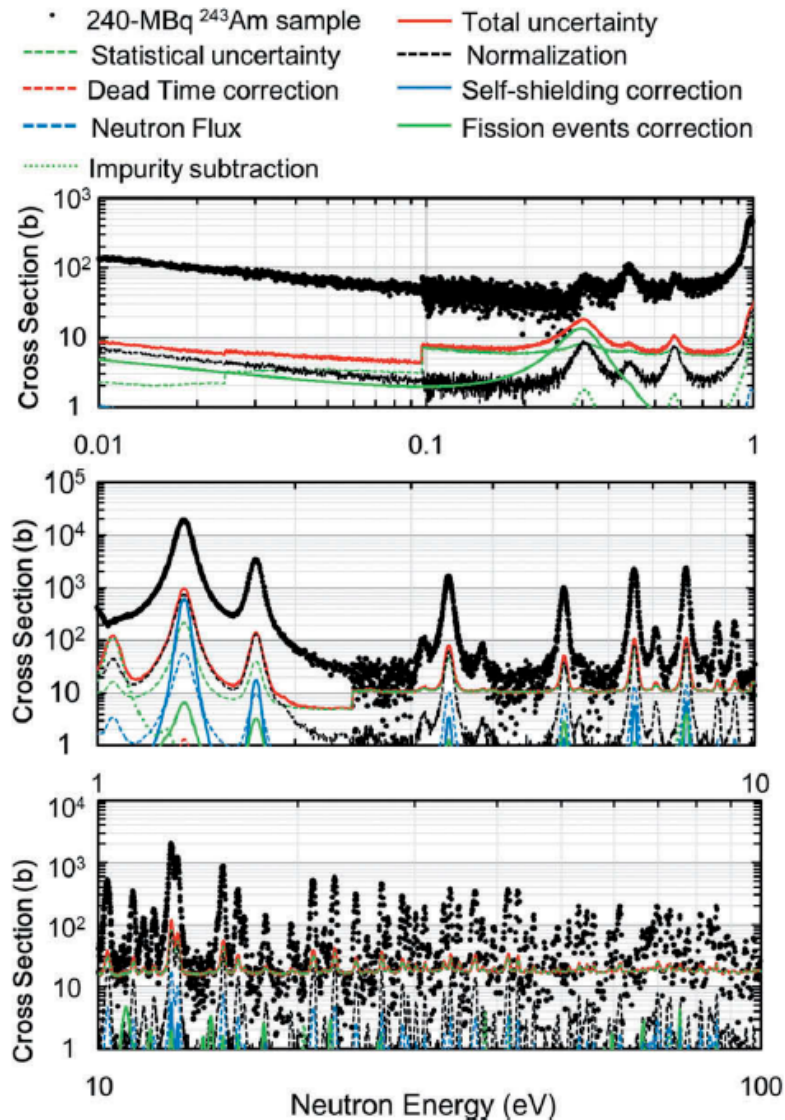
Sample: Am-243 240MBq
(Isotopic ratio: 97.67%, 30mg)

Measurement Time: 13hours

- Capture cross-section of Am-243 was deduced from 10 meV to 100 eV.
- $87.7 \pm 5.4\text{b}$ @thermal energy.

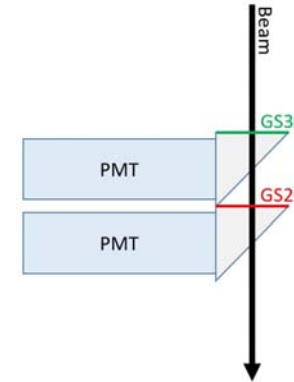
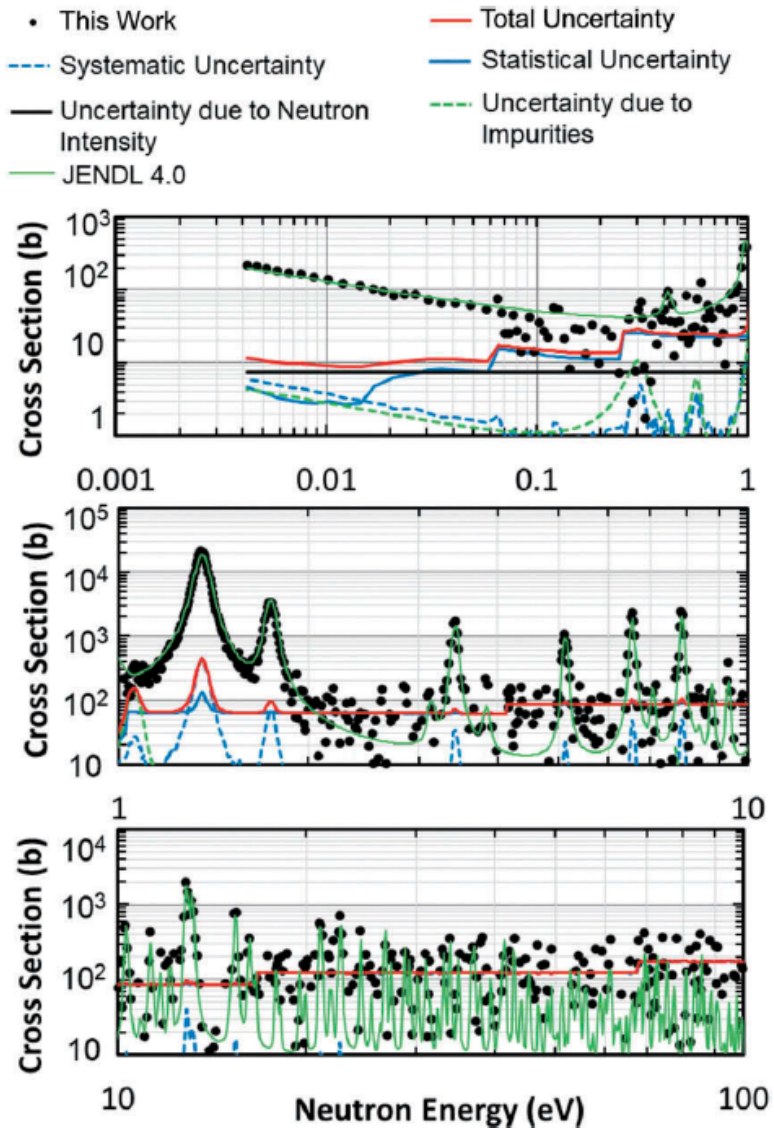
Table 2. Thermal capture cross sections σ_0 provided by different integral measurements and evaluations of ^{243}Am .

Reference	Year	$\sigma_0(\text{b})$
This Work	2019	87.7 ± 5.4
Hori et al. [6]	2009	76.6
Marie et al. [14]	2006	81.8 ± 3.6
Hatsukawa et al. [15]	1997	84.4
Gavrilov et al. [16]	1977	83 ± 6
Eberle et al. [17]	1971	77 ± 2
Folger et al. [18]	1968	78
Bak et al. [19]	1967	73 ± 6
Ice et al. [20]	1966	66–84
Butler et al. [21]	1957	73.6 ± 1.8
Harvey et al. [22]	1954	140 ± 50
Stevens et al. [23]	1954	115
ENDF/B-VII.1 [24]	2011	80.4
JENDL-4.0 [25]	2011	79.3



Experiments of ^{243}Am

~Total Cross section~

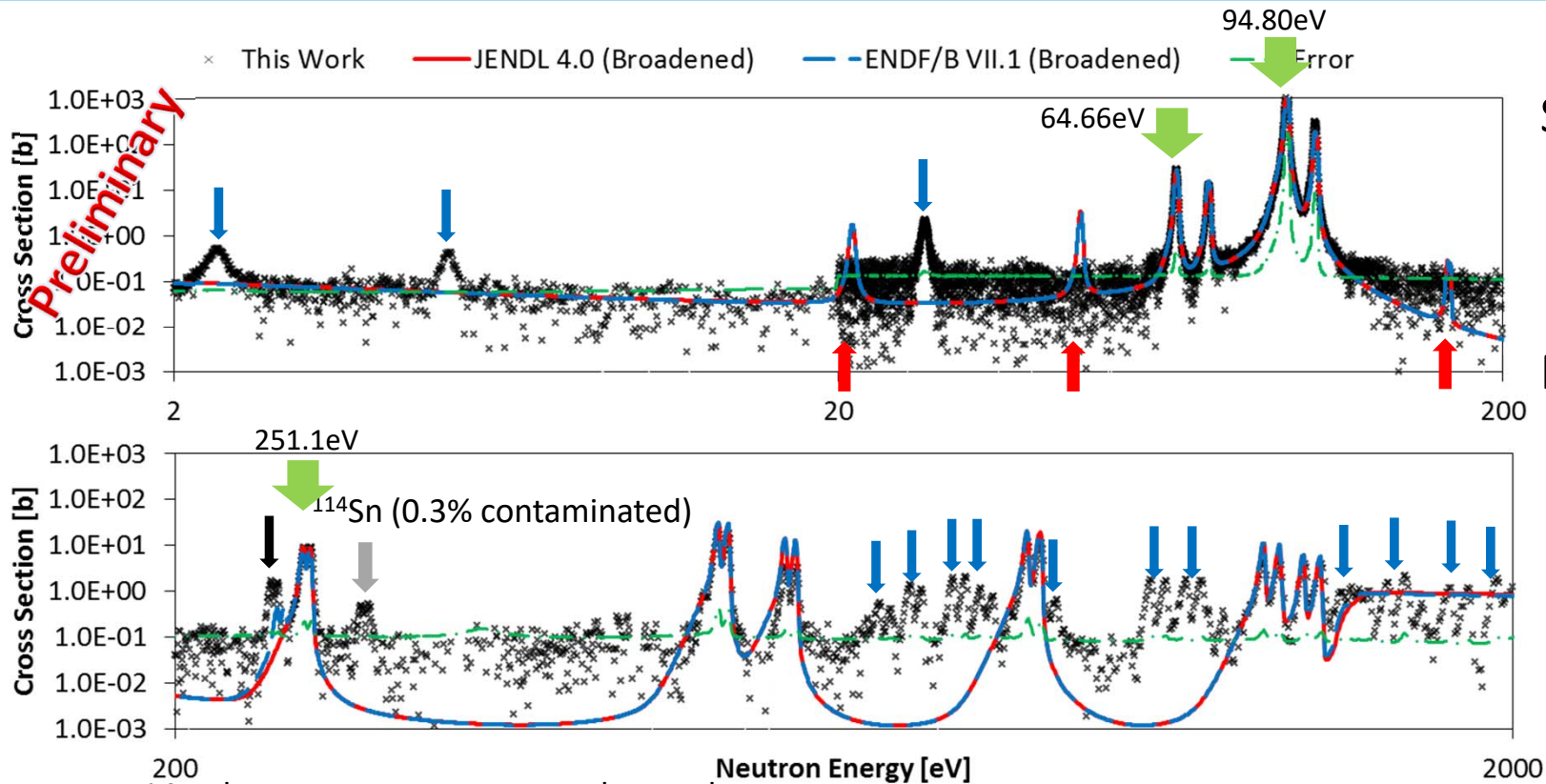


- A ^6Li enriched Li-glass detector (GS-20) and a ^7Li enriched detector were used (GS-30)
- Measurement time: 15時間
- Total cross-section was deduced from 4meV to 100 eV.
- 96 ± 11 b @thermal energy

• Total cross section can be measured in ANNRI.

Experiments of ^{112}Sn

~Tof spectrum~

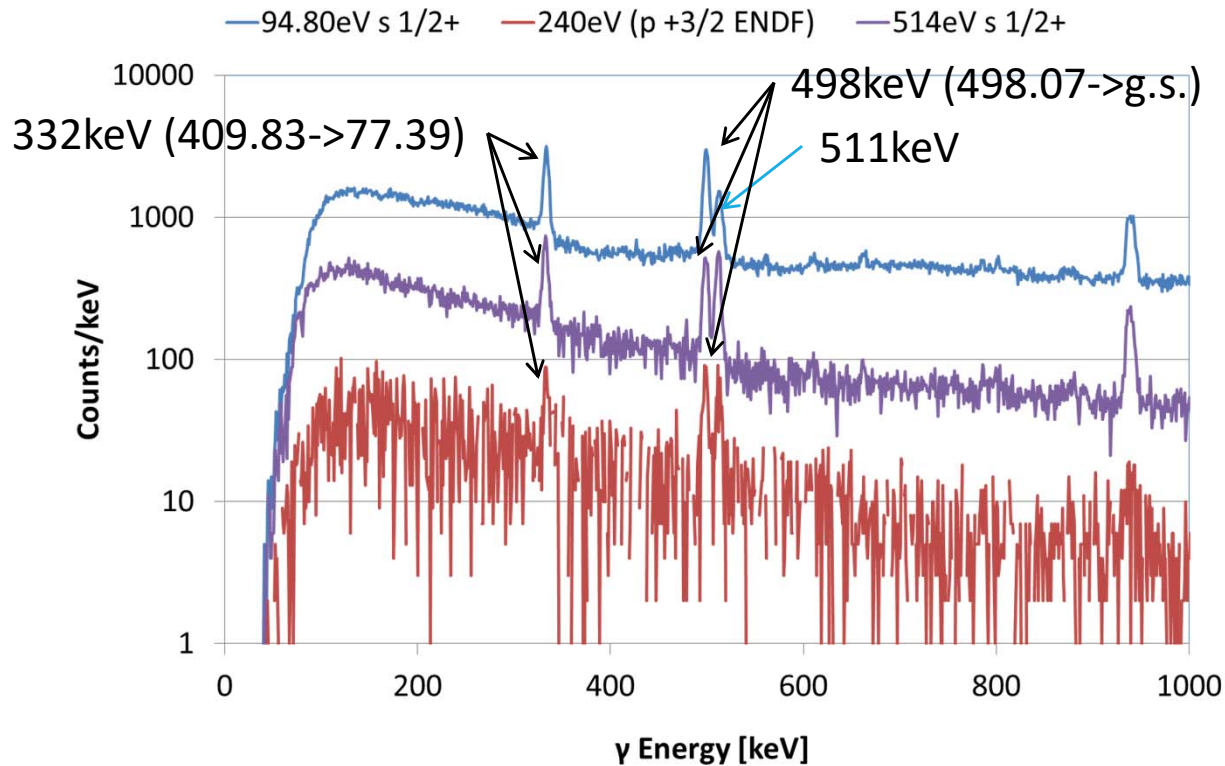


Sample:
 ^{112}Sn (99.6%) 83.8mg,
 Beam Power:
 210kW,
 Measurement Time:
 36 hours

- 14 unknown resonances were observed
- Resonances at 21, 46, and 251 eV were not observed,
- The resonance at 240 eV is listed in ENDF B-VII but not listed in JEBDL 4.0.

Experiments of ^{112}Sn

~ γ -ray spectrum ~



Sn-112 0+

Sn-113 $\frac{1}{2}^+$

The 332- and 498-keV γ -rays are clearly observed in the spectra of 94.80-, 240- and 514-eV resonances.

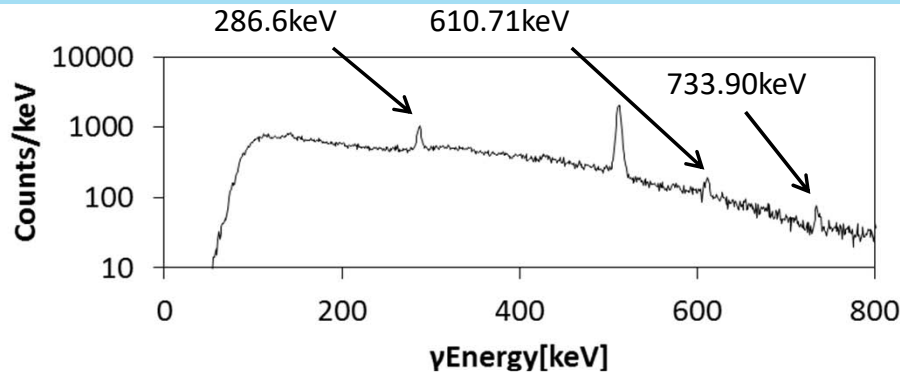
They have already been studied in other reactions. ($^{110}\text{Cd}(\alpha, n\gamma)$, $^{114}\text{Sn}(p, d)\dots$)



The 240-eV resonance is undoubtedly one of the ^{112}Sn resonances.

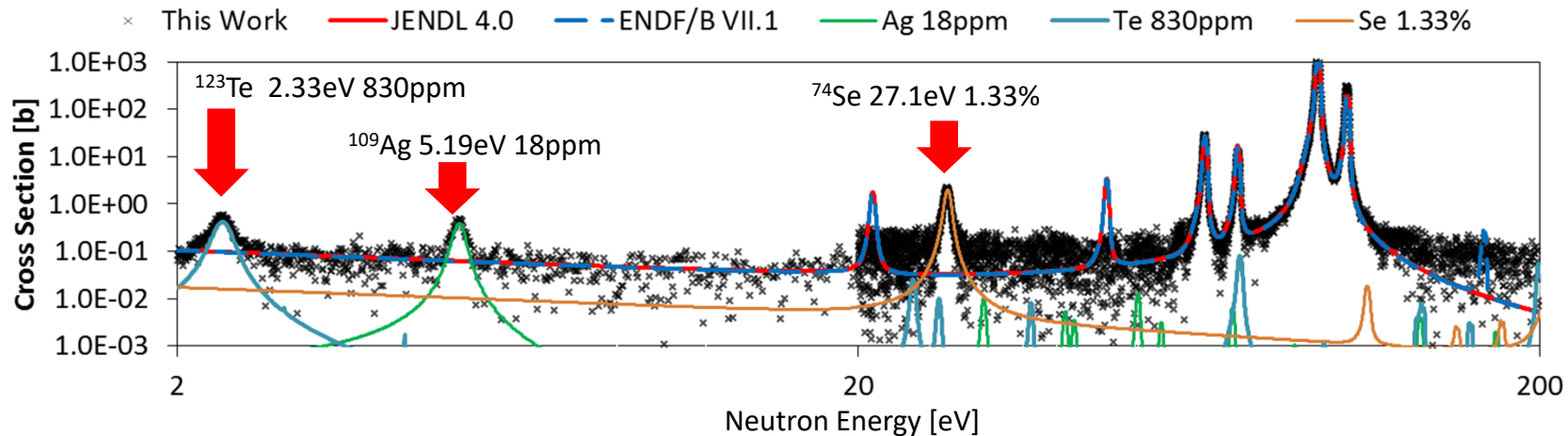
Experiments of ^{112}Sn

~ Origin of the 27.1-eV resonance ~



- Prompt γ -rays of ^{112}Sn are not observed. (332, 498 keV)
- Prompt γ -rays of ^{74}Se (286.57, 610.71 and 733.90 keV) are clearly observed.

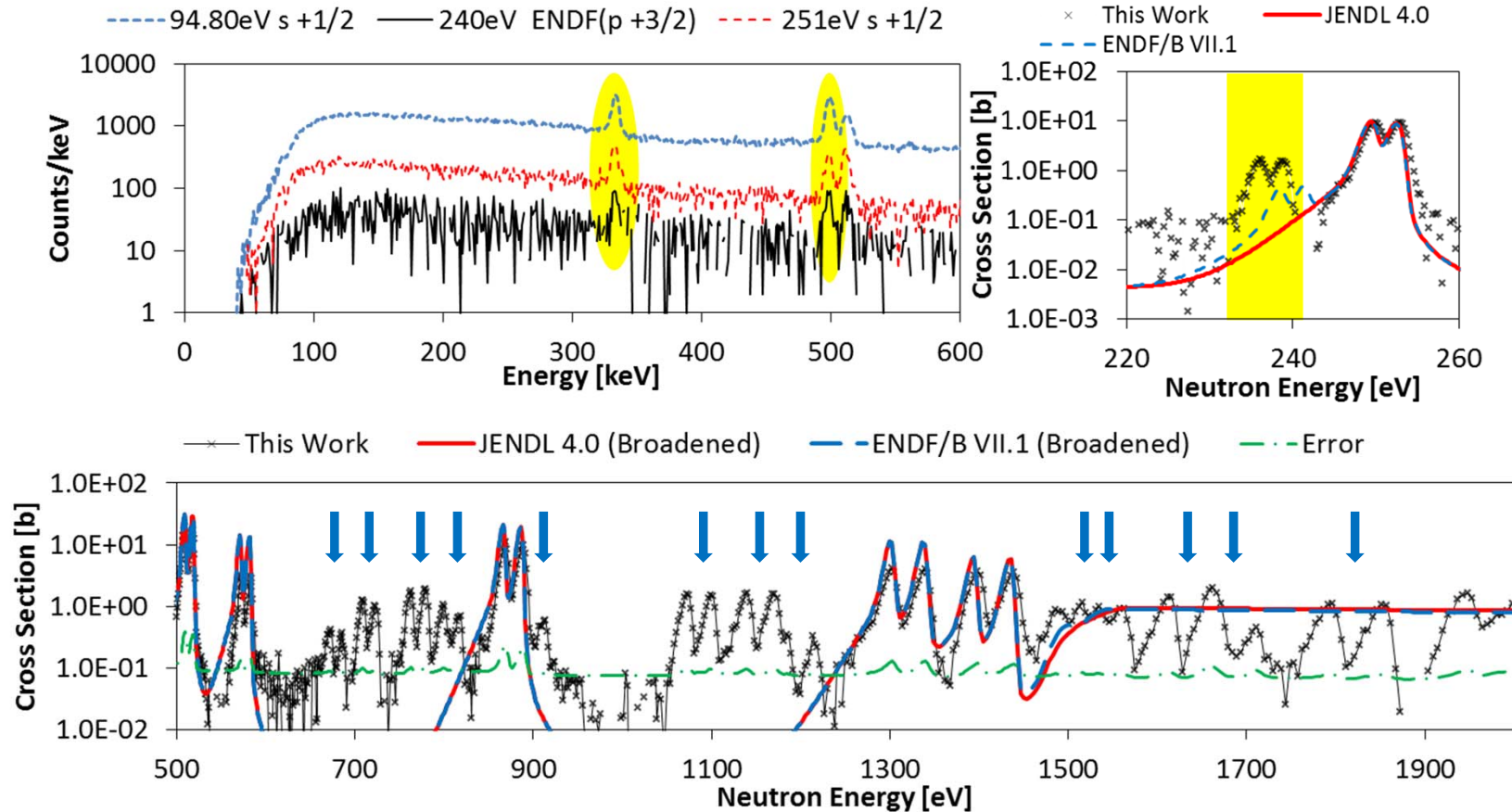
1st resonance of ^{74}Se (27.1eV)



Our sample is contaminated with Se(1.33%), Te(830ppm), and Ag(18ppm). However, these impurities are not listed on the certification sheet.

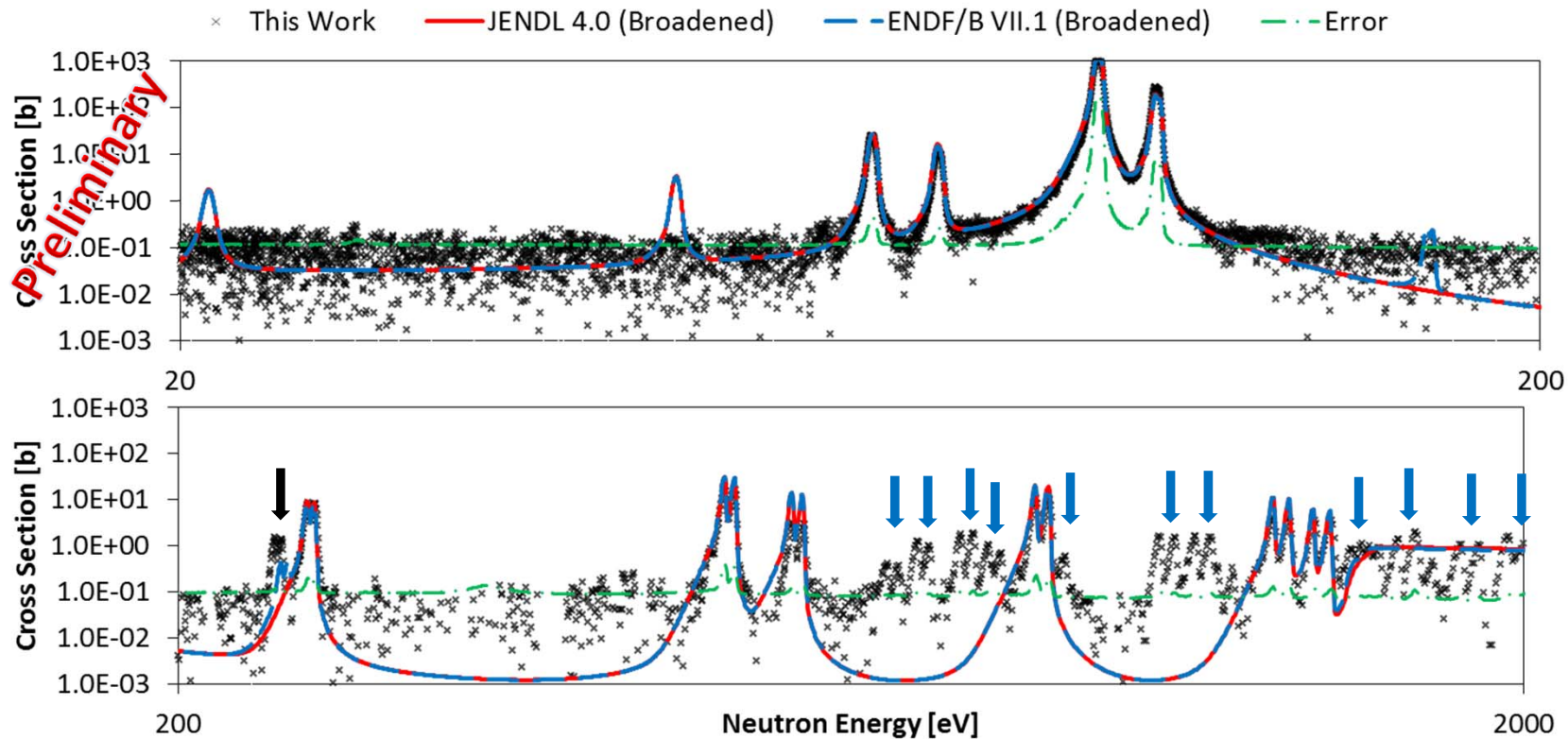
Experiments of ^{112}Sn

~ Origin of the 236.6-eV resonance ~



Origin of the 27.1-eV resonance

With subtracting the influences of the chemical and isotopic impurities, preliminary cross section for ^{112}Sn was obtained.



We found 3 miss assigned resonances and 13 new resonances.

Compilation at JAEA

JAEA is responsible for the compilation of the following papers:

- Published by JAEA members
- Experiments in J-PARC facilities.

In 2022, 6 papers were compiled at JAEA.

23741: Cm-244,246(n,g) in J-PARC by Dr. Kawase

23748: Am-243(n,g) in J-PARC by Mr. Kodama

23745: Np-237(n,g) in J-PARC by Dr. Rovira

23749: Tc-99(n,g) in YAYOI(Reactor) by Dr. Nakamura

23601: Np-237(n,g) in J-PARC in keV by Dr. Rovira

23602: Nb-93(n,g),(n,tot) in J-PARC by Mr. Endo

I still have some duties. I would like to compile them ASAP.

EXFOR Editor

EXFOR Editor is very useful and powerful tool for me.

- It is very easy to set large data.

In TOF experiments, we have to set huge number of data points.

The editor can import a text file with separator of commas (CSV format).

But I have two request.

Request for EXFOR Editor ①

DAQ: CAEN V1724 Digitizer (14bit, 100MHz)	23741	1	28
SAMPLE The 244Cm sample and the 246Cm sample were used. The	23741	1	29
244Cm sample and the 246Cm sample contained curium	23741	1	30
oxide with weight of 0.6 and 2.1 mg.	23741	1	31
The isotopic and chemical impurities (mol%):	23741	1	32
244Cm sample 246Cm sample	23741	1	33
244Cm 87.3 +-1.7 21.9 +-0.4	23741	1	34
245Cm 3.4 +-0.4 1.1 +-0.3	23741	1	35
246Cm 9.3 +-0.4 64.1 +-1.4	23741	1	36
247Cm 3.1 +-0.4	23741	1	37
248Cm 9.8 +-0.3	23741	1	38
240Pu 46.9 +-0.9 10.5 +-0.2	23741	1	39
242Pu 0.102+-0.005	23741	1	40
243Am 0.47+-0.05 1.02 +-0.10	23741	1	41
Each sample was sealed in an aluminum case with an	23741	1	42
outer diameter of 9 mm and a wall thickness of 0.5 mm.	23741	1	43
For the calibration of the time-to-energy conversion	23741	1	44
function, a thin 197Au sample was used.	23741	1	45
CORRECTION Dead Time Correction was applied with a paralyzable	23741	1	46
dead time model	23741	1	47

anti-coincidence shields.	23741	1	27
DAQ: CAEN V1724 Digitizer (14bit, 100MHz)	23741	1	28
SAMPLE The 244Cm sample and the 246Cm sample were used. The			
244Cm sample and the 246Cm sample contained curium			
oxide with weight of 0.6 and 2.1 mg. The isotopic and			
chemical impurities (mol%) : 244Cm sample 246Cm			
sample 244Cm 87.3 +-1.7 21.9 +-0.4 245Cm 3.4 +-0.4			
1.1 +-0.3 246Cm 9.3 +-0.4 64.1 +-1.4 247Cm 3.1 +-0.4			
248Cm 9.8 +-0.3 240Pu 46.9 +-0.9 10.5 +-0.2 242Pu			
0.102+-0.005 243Am 0.47+-0.05 1.02 +-0.10 Each sample			
was sealed in an aluminum case with an outer diameter			
of 9 mm and a wall thickness of 0.5 mm. For the			
calibration of the time-to-energy conversion function,			
a thin 197Au sample was used.			
CORRECTION Dead Time Correction was applied with a paralyzable	23741	1	46
dead time model.	23741	1	47
Correction factors for self-shielding and multiple	23741	1	48



to a 7-mm diameter at the sample position. 23741 1 21

Target Description

SAMPLE: The 244Cm sample and the 246Cm sample were used. The 244Cm sample and the 246Cm sample contained curium oxide with weight of 0.6 and 2.1 mg. The isotopic and chemical impurities (mol%): 244Cm sample 246Cm sample

Spell Checking Compress Clear

Data Modifier = RAB (SF8 under REACTION Keyword)

Nuclide Field Search Add

Abundance Field NAT= Add New Record

Current Position: SUBENTRY 23741001 BIB section

OK Cancel Help EXFOR-Help

248Cm 9.8 +-0.3 23741 1 38

Once edited the text using the editor, line break positions are moved. Is there line feed code in the editor?
 Yes: Please tell me how to use it.
 No: Please add line feed code or accept multi (continuous) spaces.

Request for EXFOR Editor ②

@Decay Data Sec.

	flux spectrum.				23749	1	38
DECAY-DATA	(43-TC-100,15.27SEC,DG,539.59,0.066,				23749	1	39
		DG,590.83,0.055)			23749	1	40
	(79-AU-198,2.6943D,DG,411.80,0.9562)				23749	1	41
	(21-SC-46,83.79D,DG,889.28,0.99984,				23749	1	42
		DG,1120.55,0.99987)			23749	1	43
	(29-CU-64,12.700HR,DG,1345.84,0.00473)				23749	1	44
	(25-MN-56,2.57878HR,DG,846.77,0.9887)				23749	1	45
Miss	(13-AL-28,9.458MIN,DG,843.76,41.8,				23749	1	46
		1014.44,0.280)			23749	1	47
	(49-IN-115-M,4.486H,DG,336.24,0.459)				23749	1	48
HISTORY	(20220915C) Compile by A. Kimura.				23749	1	49
ENDBIB	47				23749	1	50
NOCOMMON	0				23749	1	51

Correct:
(12-MG-27,9.458MIN,DG,843.76,0.718,

Could you install a checking system for the decay data section?

The values (half-life, decay energy, and Intensity) are mostly the same (within a few %).

Thank you for your kind Attention!!



Experiments of $^{244,246}\text{Cm}$

~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 \pm 1.7	27.5 \pm 0.5
^{245}Cm	2.71 \pm 0.34	1.06 \pm 0.28
^{246}Cm	7.22 \pm 0.34	59.4 \pm 1.3
^{247}Cm	N.D.	2.9 \pm 0.4
^{248}Cm	N.D.	9.10 \pm 0.24

Samples:

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

Net weight = 0.6 mg

Activity = 1.8 GBq

Measurement Periods: 64 hours

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

Net weight = 2.1 mg

Activity = 12.1 MBq (^{244}Cm : 1.7GBq)

Measurement Periods: 94 hours

Both of the samples

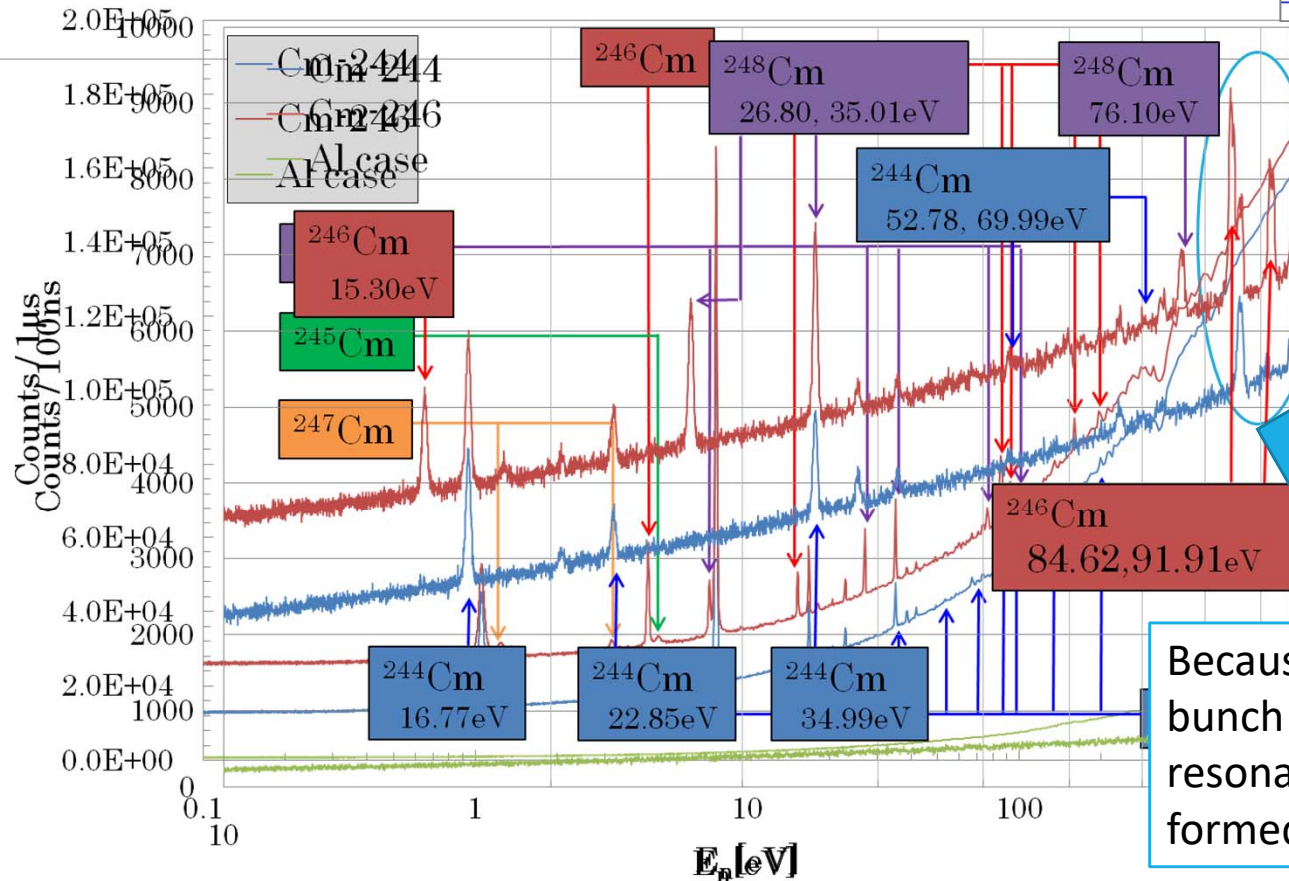
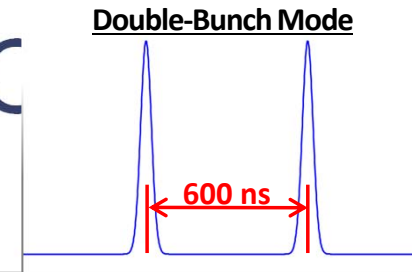
Chemical form = CmO_2

Container = Al capsule

For the background estimation, a dummy case (Al 278mg) and a blank sample was measured for done for 48 and 44 hours.

To reduce air scattering, the air in the beam duct was replaced with helium.

Experiments of $^{244,246}\text{Cm}$ ~TOF Spectrum~



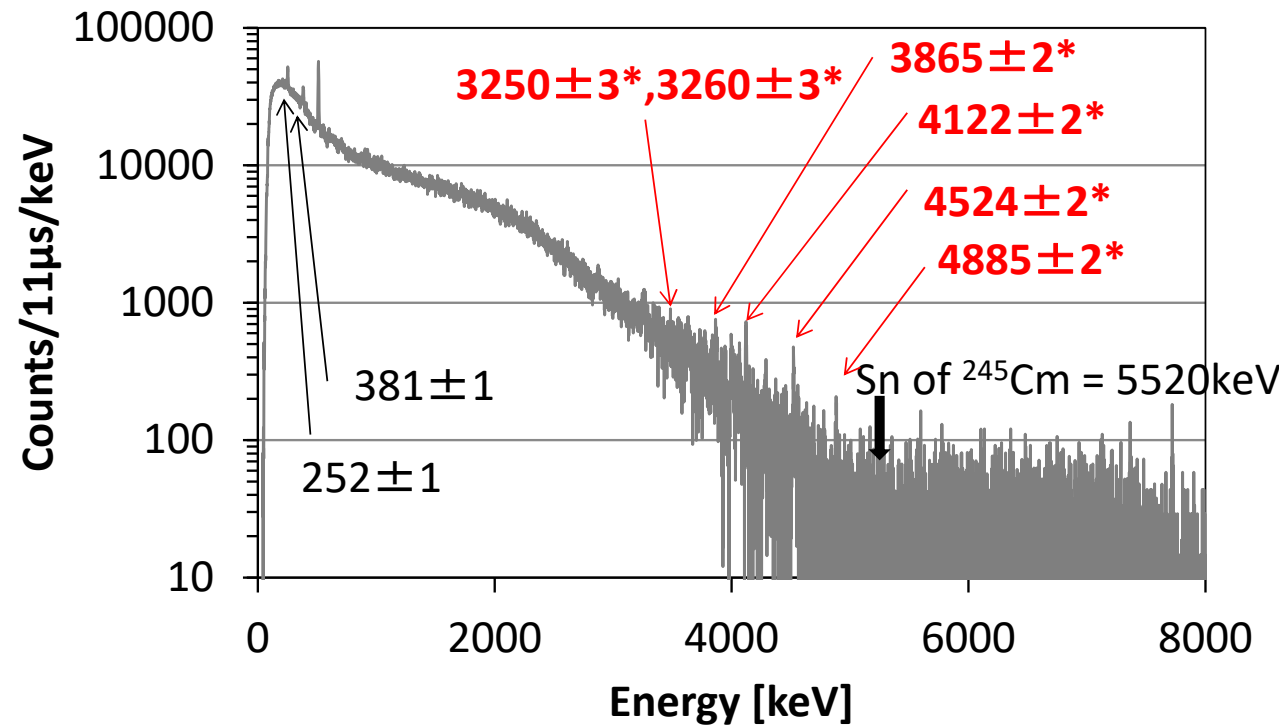
Because of the double-bunch mode, resonance peaks were formed double peaks.

This graph shows TOF spectra of the ^{244}Cm , the ^{246}Cm sample, and the dummy case

Experiments of $^{244,246}\text{Cm}$

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

Cm-244

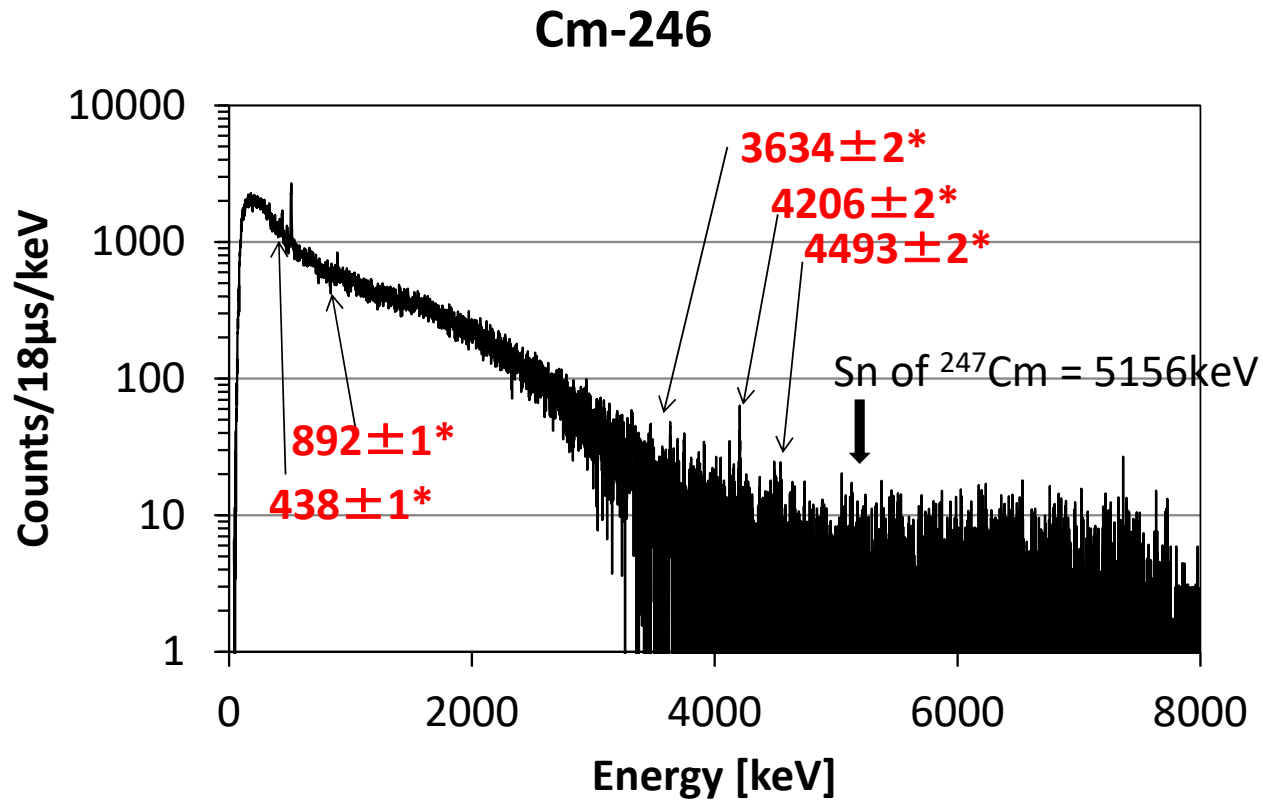


The 252.4- and 380.8-keV γ -rays have already been studied in α decay of ^{249}Cf , electron capture decay of ^{245}Bk , and β -decay of ^{245}Am .

The other γ -rays were **previously unknown γ rays**.

Experiments of $^{244,246}\text{Cm}$

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



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

VIENNA EXFOR WORKSHOP (13-16 DEC.)

Experiments of $^{244,246}\text{Cm}$

~Analysis~

The data were analyzed with the procedure.

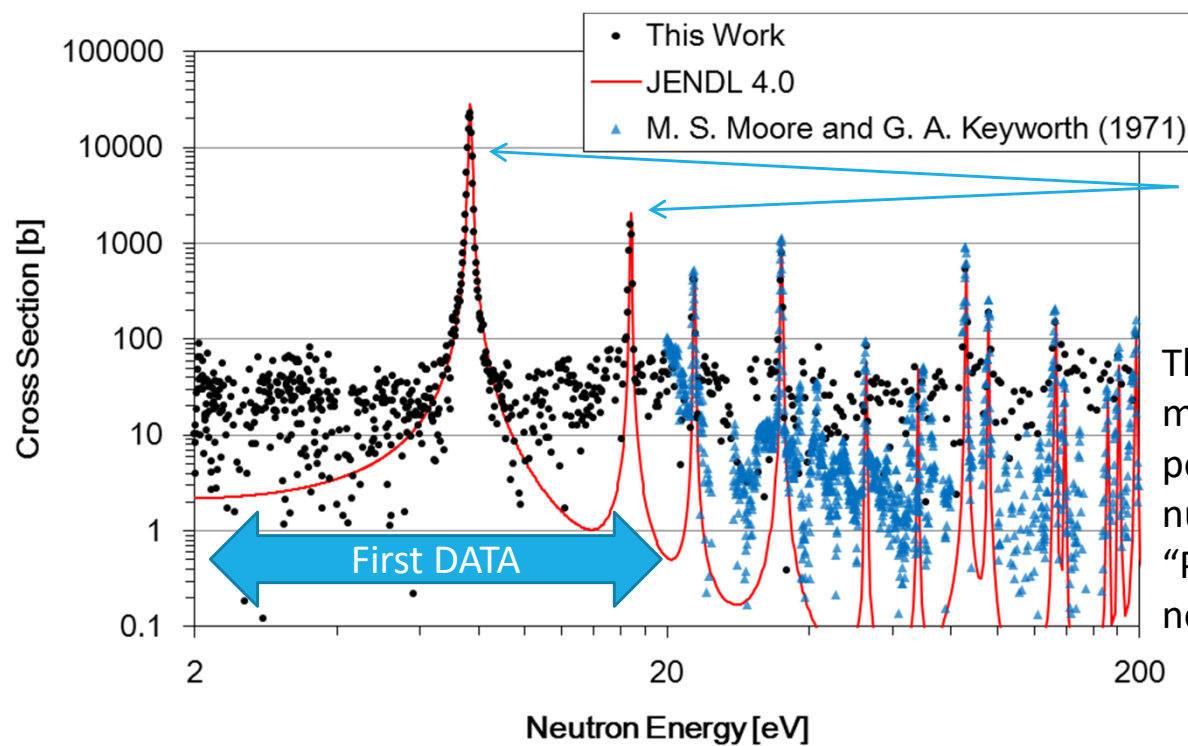
- **Dead-Time Correction**
- **Background Estimation and Subtraction**
- **Self-Shielding and Multiple-Scattering Correction**
- **Normalization (Using the 1st resonance of ^{240}Pu)**
- **Evaluation and Subtraction of Influence of Fission Events**
- **Evaluation and Subtraction of Influence of Impurities**

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

Experiments of $^{244,246}\text{Cm}$

~Cross Section of ^{244}Cm ~

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



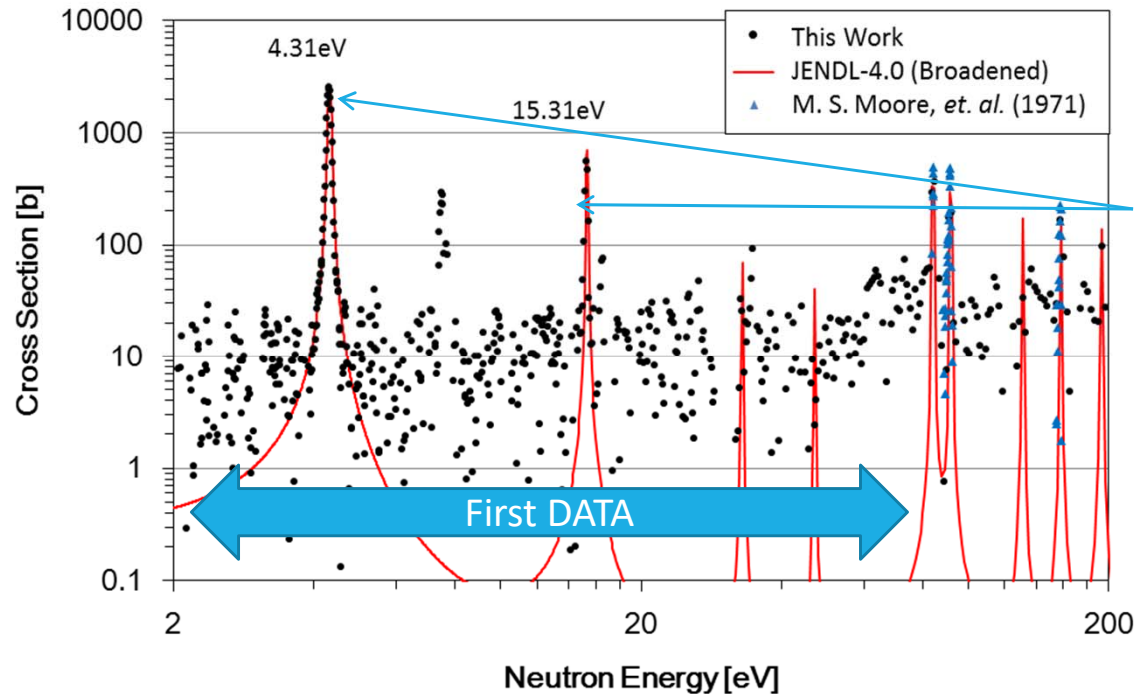
The results of the resonance peaks under 20-eV are also the first experimental results in the world.

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

Experiments of $^{244,246}\text{Cm}$

~Cross Section of ^{246}Cm ~

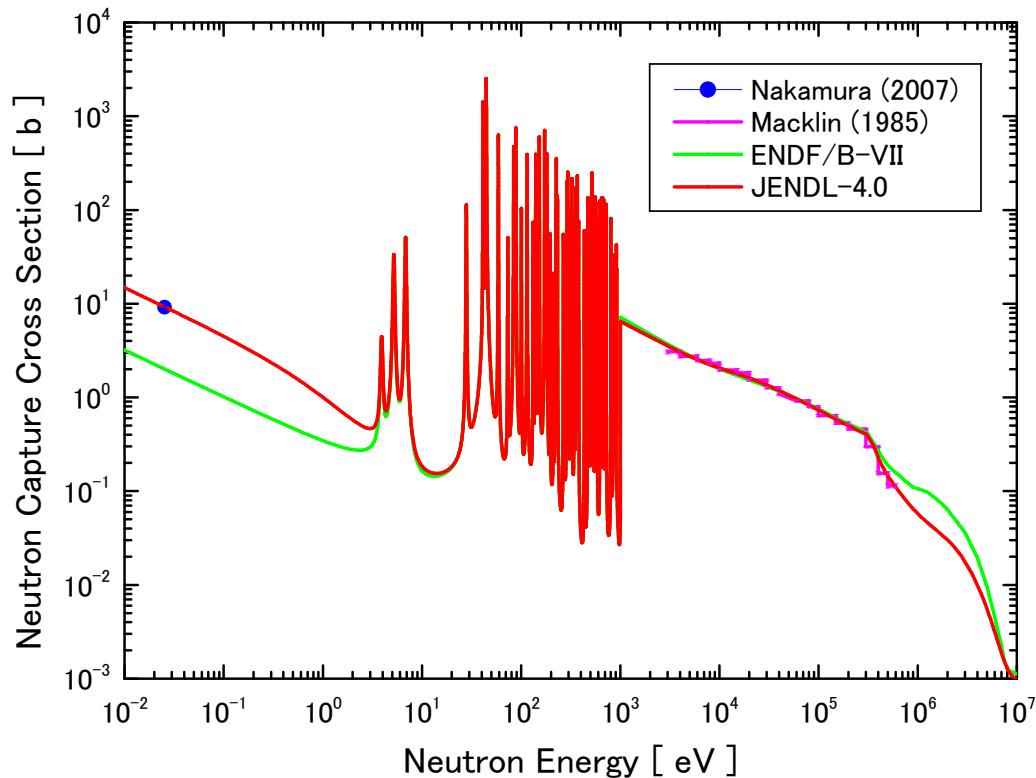
Only one neutron-capture cross-section data of ^{246}Cm (n,γ) was made in 1971[1].



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

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

Present status of nuclear data measurement



Pd-107(LLFP)

At thermal energy:

- Prompt gamma Nakamura(JAEA) (2007)

keV region:

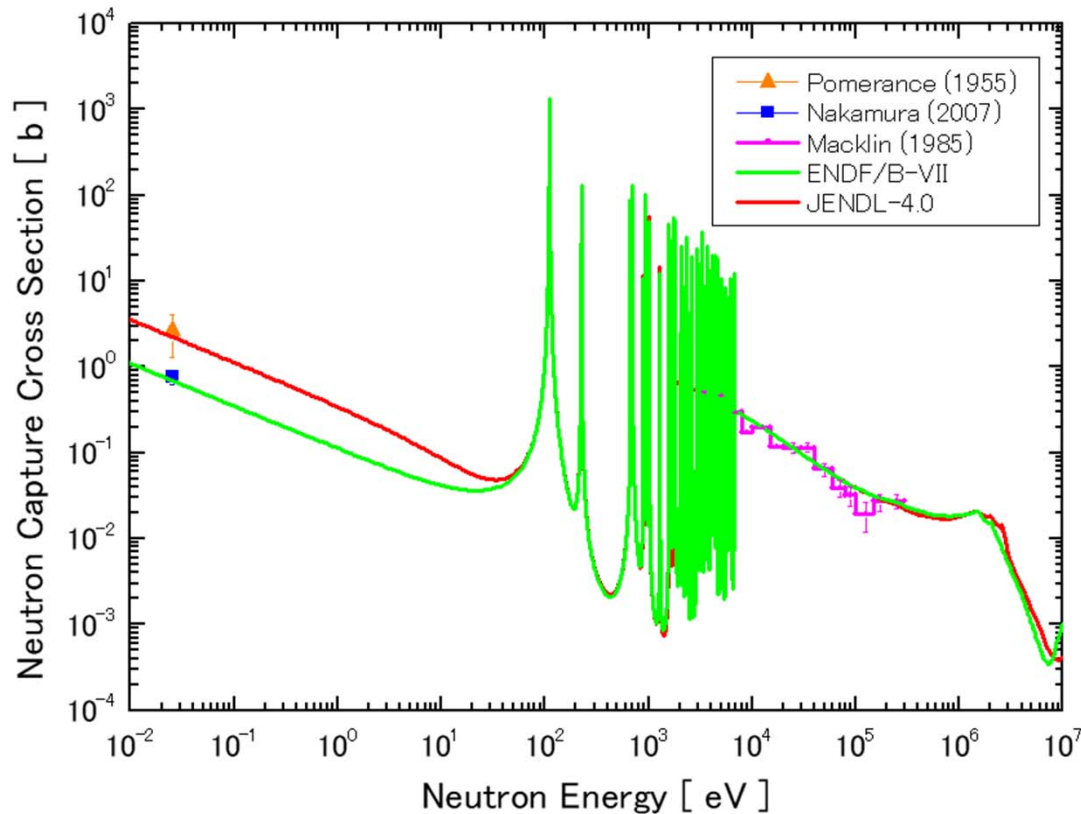
- TOF ORELA (1985) (C_6D_6)

⇒ Only 2 data sets are available.
There are large discrepancy
between the evaluated data sets.

The capture cross sections were
deduced from other reactions.

If we have some data sets

^{93}Zr (LLFP, $T_{1/2}=1.5 \times 10^6$ years)



σ_{th}

Num. of exp. data : 2

- Reactor Neutron (1955, 2007)

Unresolved Resonance Region

Num. of exp. data : 1

- TOF
ORELA (1985: C_6D_6)

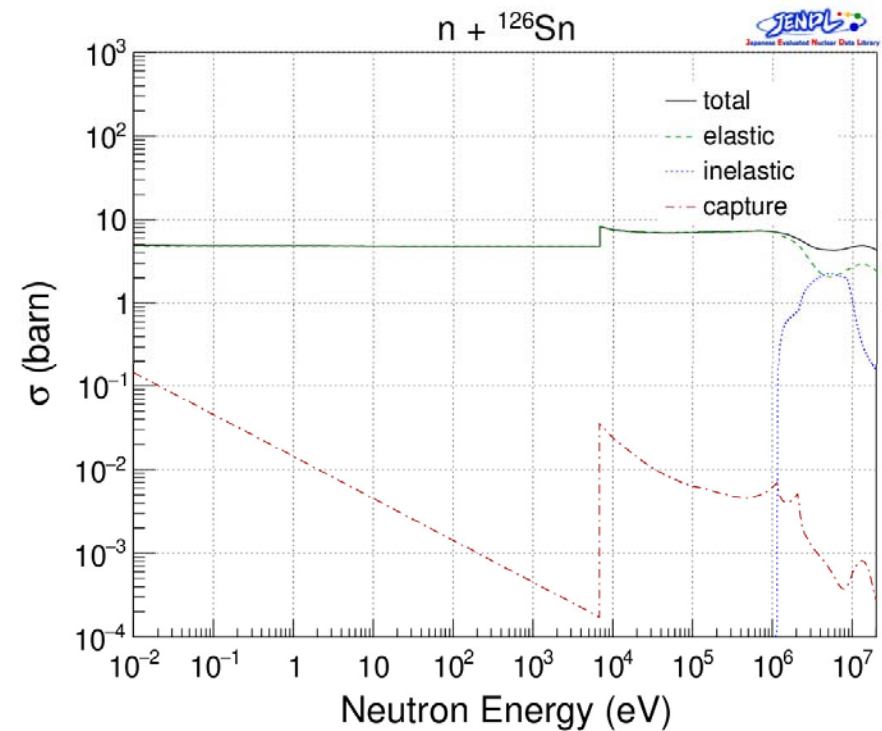
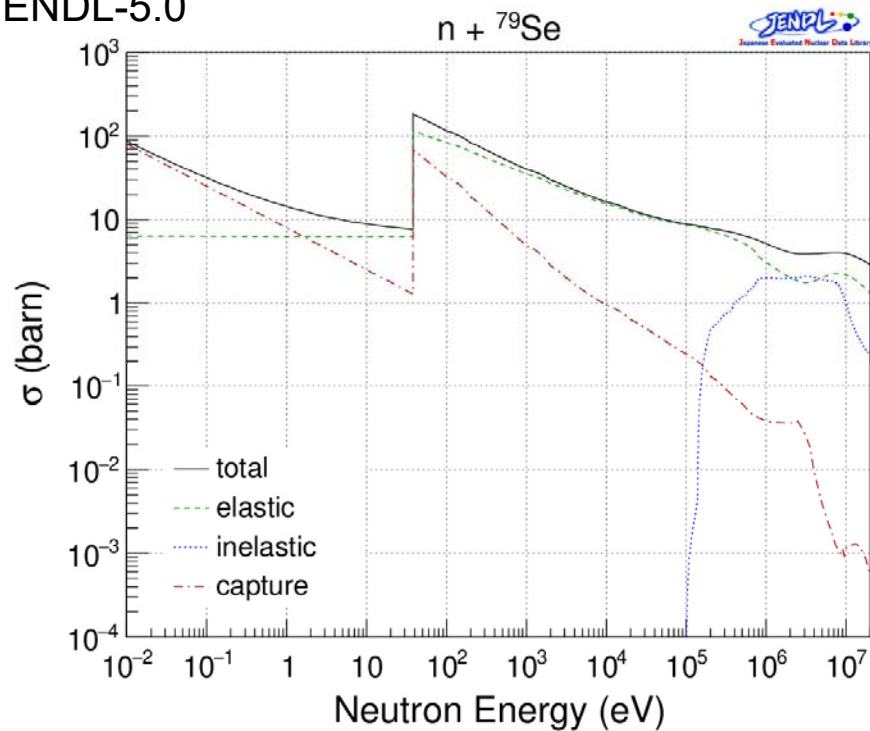
Resonance Region

No experimental data

Se-79, Sn-126 (LLFP)

No experimental data

JENDL-5.0



Cross sections were deduced from theoretical calculations.