Nuclear Data Study for Nuclear Transmutation

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   Nuclear Databases for MAs and LLFPs
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MAs: Minor Actinides (**Np-237, Am-241, Am-242m, Am-243, Cm-244, Cm-245, Cm-246,** etc)

LLFPs: Long-Lived Fission Products (**Se-79, Zr-93, Tc-99, Pd-107, Sn-126, I-129, Cs-135,** etc.)

1. Introduction

The current national policy in Japan for the management and disposal of MAs and LLFPs is a sequence of
vitrification,
interim storage, and then
depositing
underground together with other nuclear wastes.

If MAs and LLFPs are extracted and transmuted into stable nuclides, the environmental loading in the geological repository becomes very small.

The ethical problem that MAs and LLFPs are undesirable property for our far descendants will be solved.

1. Introduction

The nuclear transmutation of MAs and LLFPs is a very attractive subject.

--> Neutron capture and/or fission reactions are the most promising transmutation reactions.

--> Databases on neutron capture and/or fission reaction cross sections for MAs, LLFPs, and related nuclides are indispensable for developing the transmutation technology.

The accuracy of nuclear databases for those nuclides is quite poor at the present time.

--> The improvement of the database accuracy is an urgent task.

2. Present Status of the Accuracy of Nuclear Databases for MAs and LLFPs

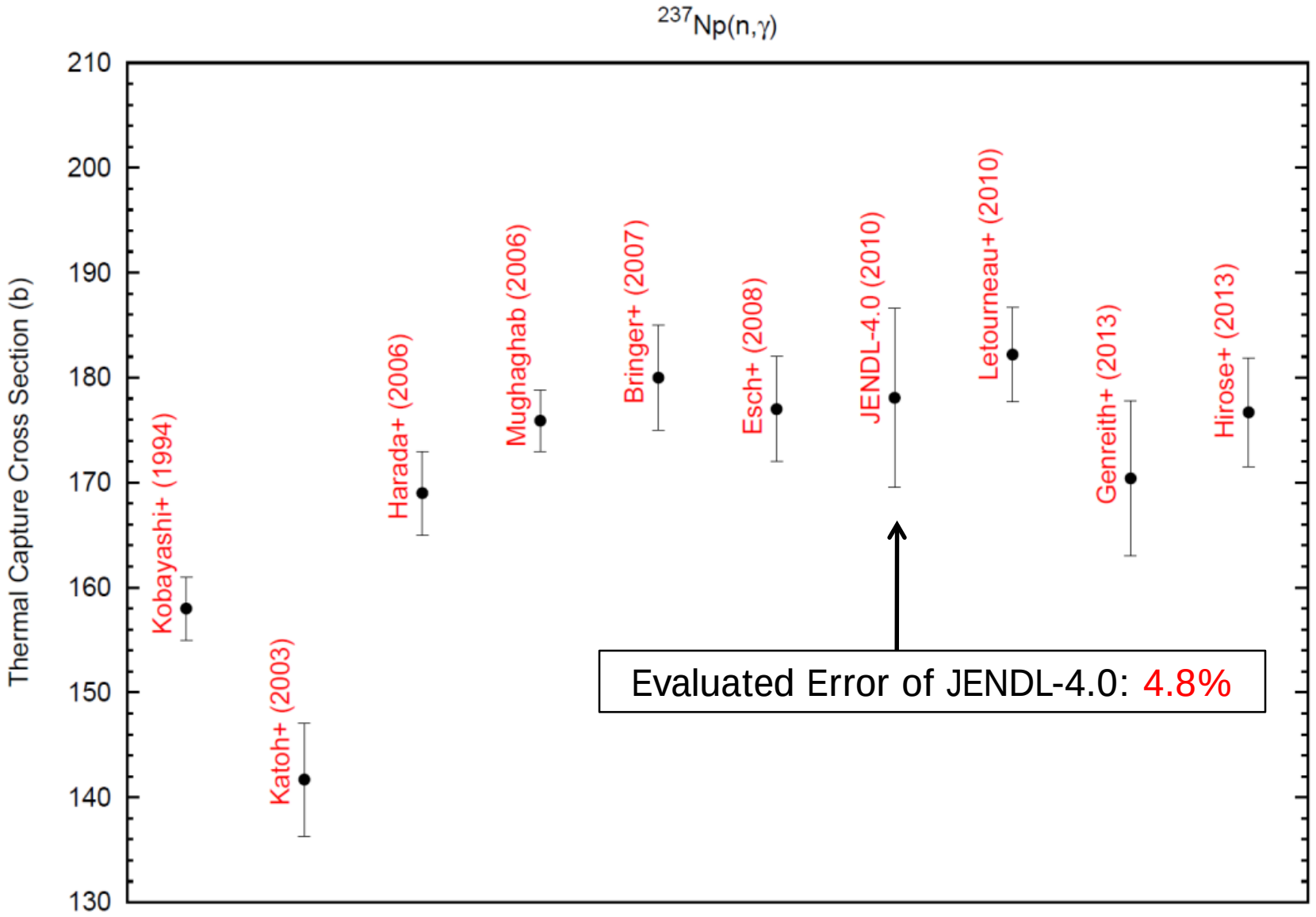
Nuclear Databases in the World

JENDL-4.0: Japanese Evaluated Nuclear Data Library

ENDF/B-VII.1: Evaluated Nuclear Data File

JEFF-3.1.1: Joint Evaluated Fission and Fusion Library

2. Present Status of the Accuracy of Nuclear Databases for MAs and LLFPs



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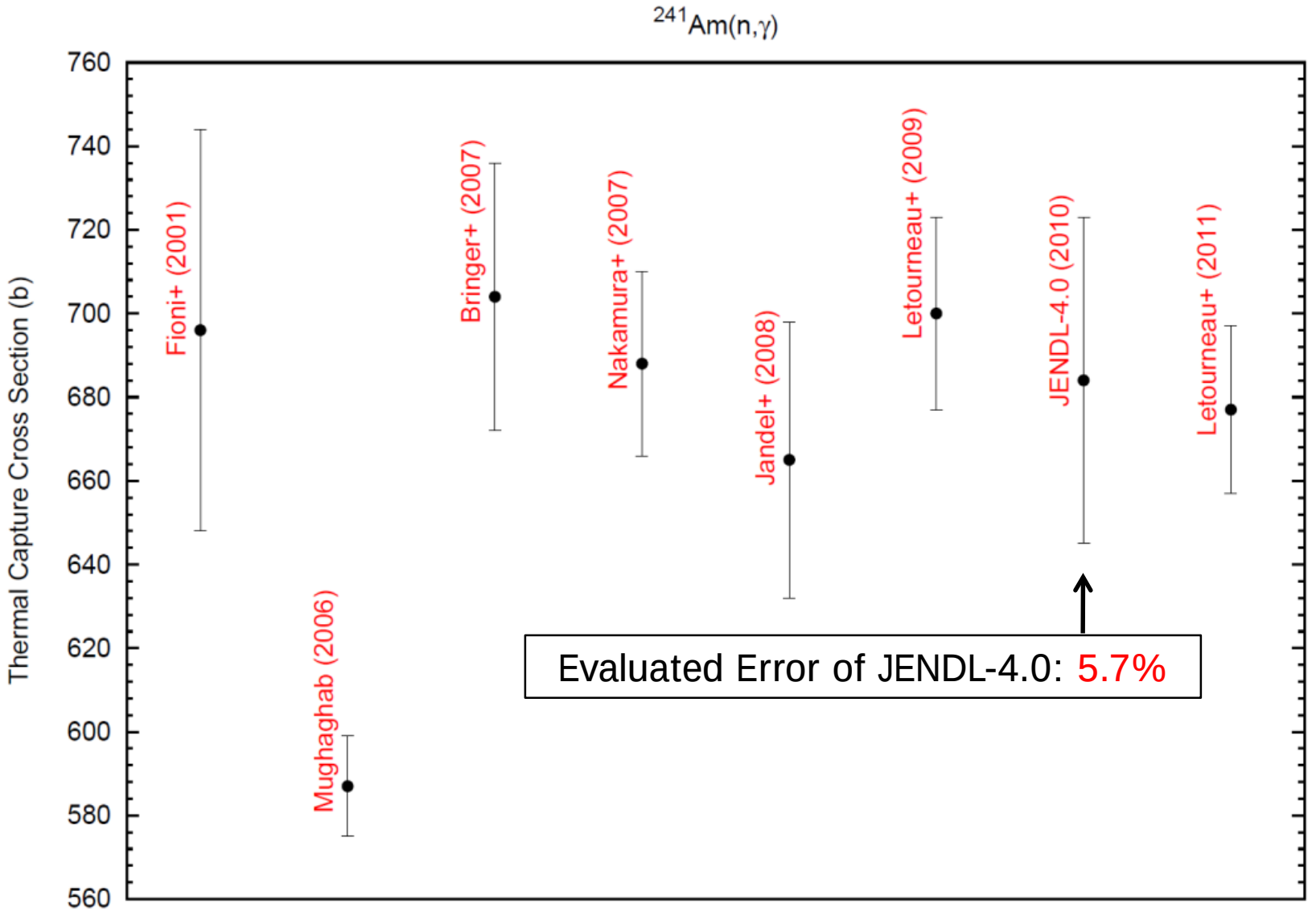

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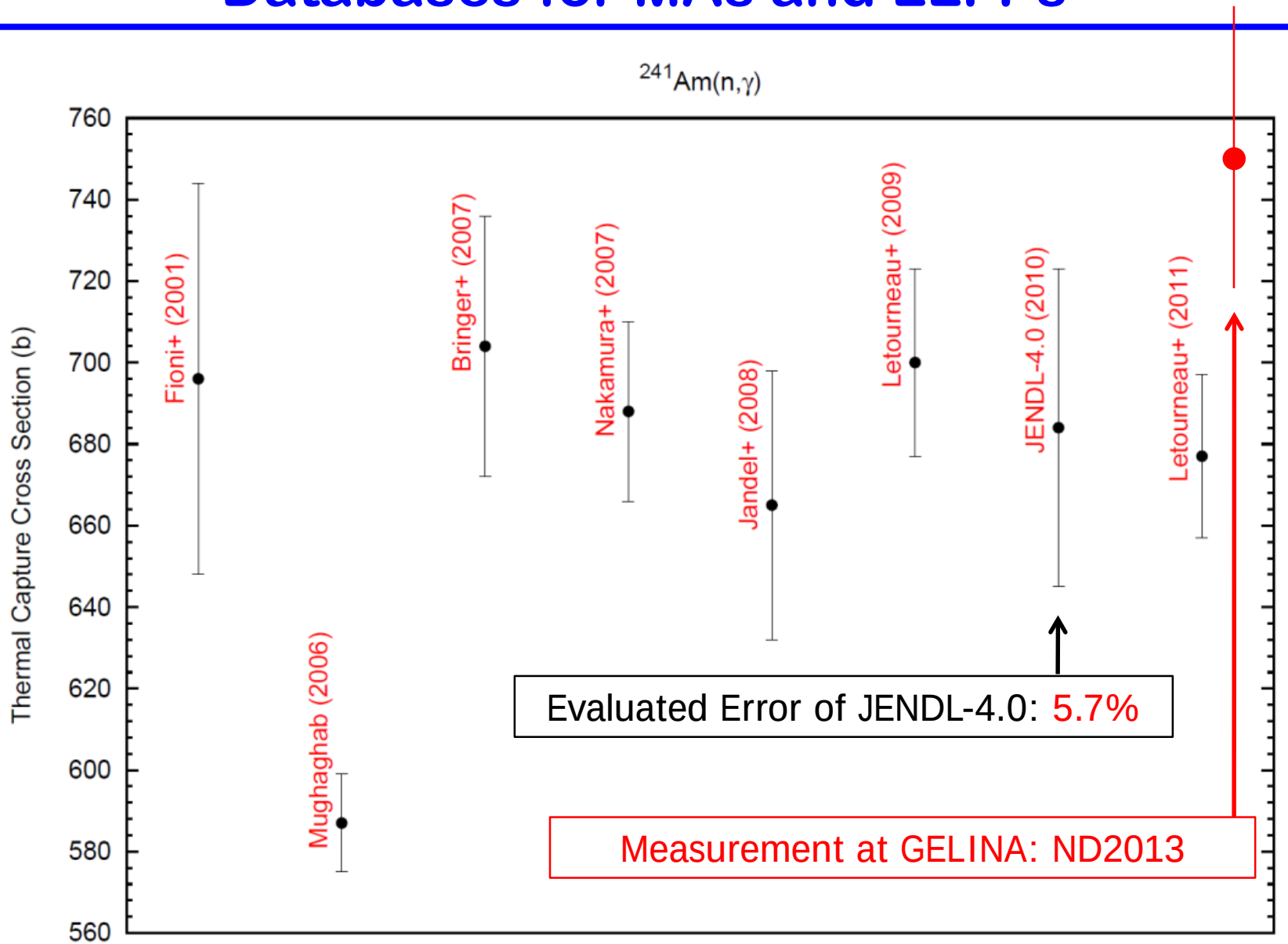

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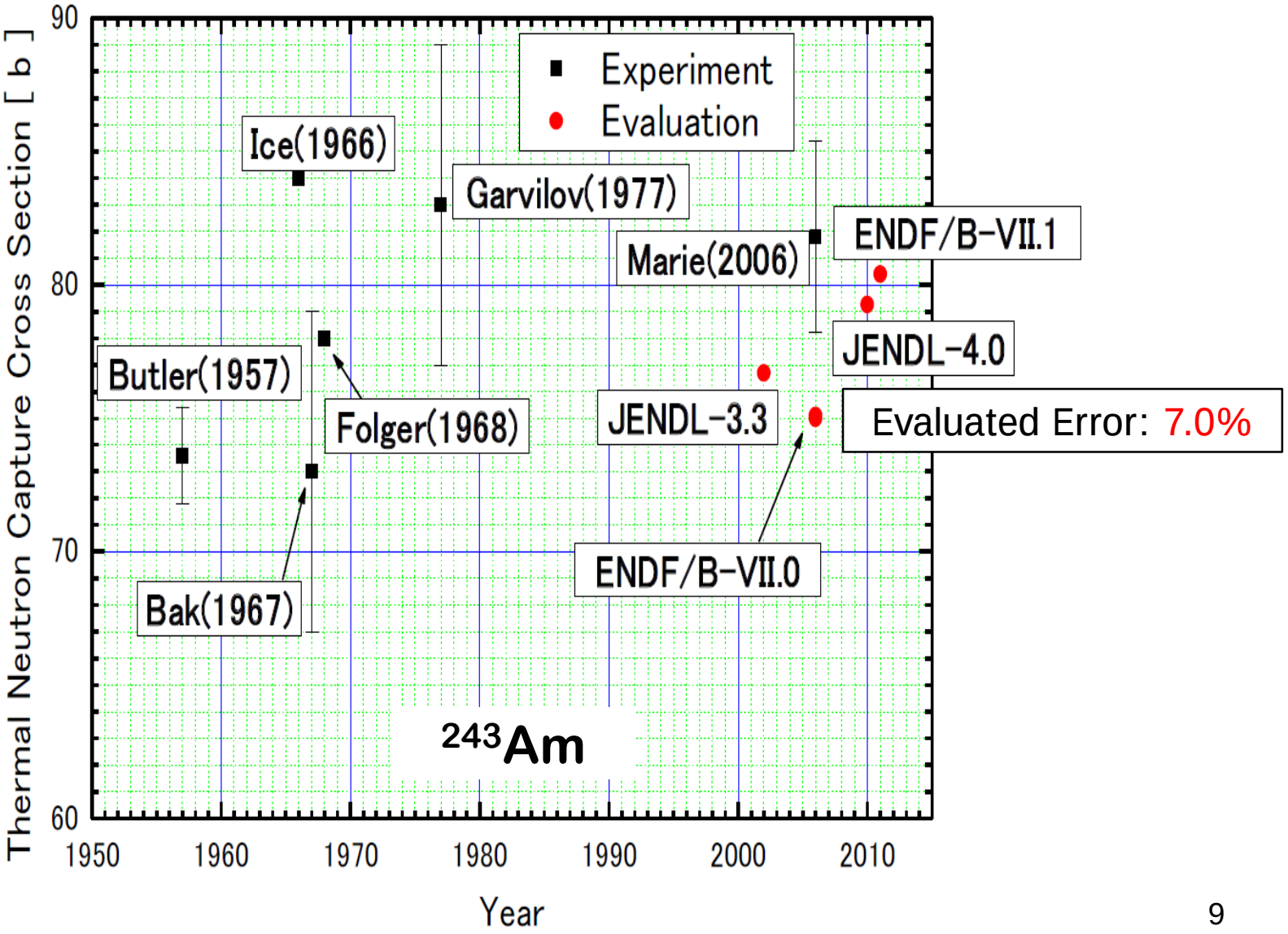

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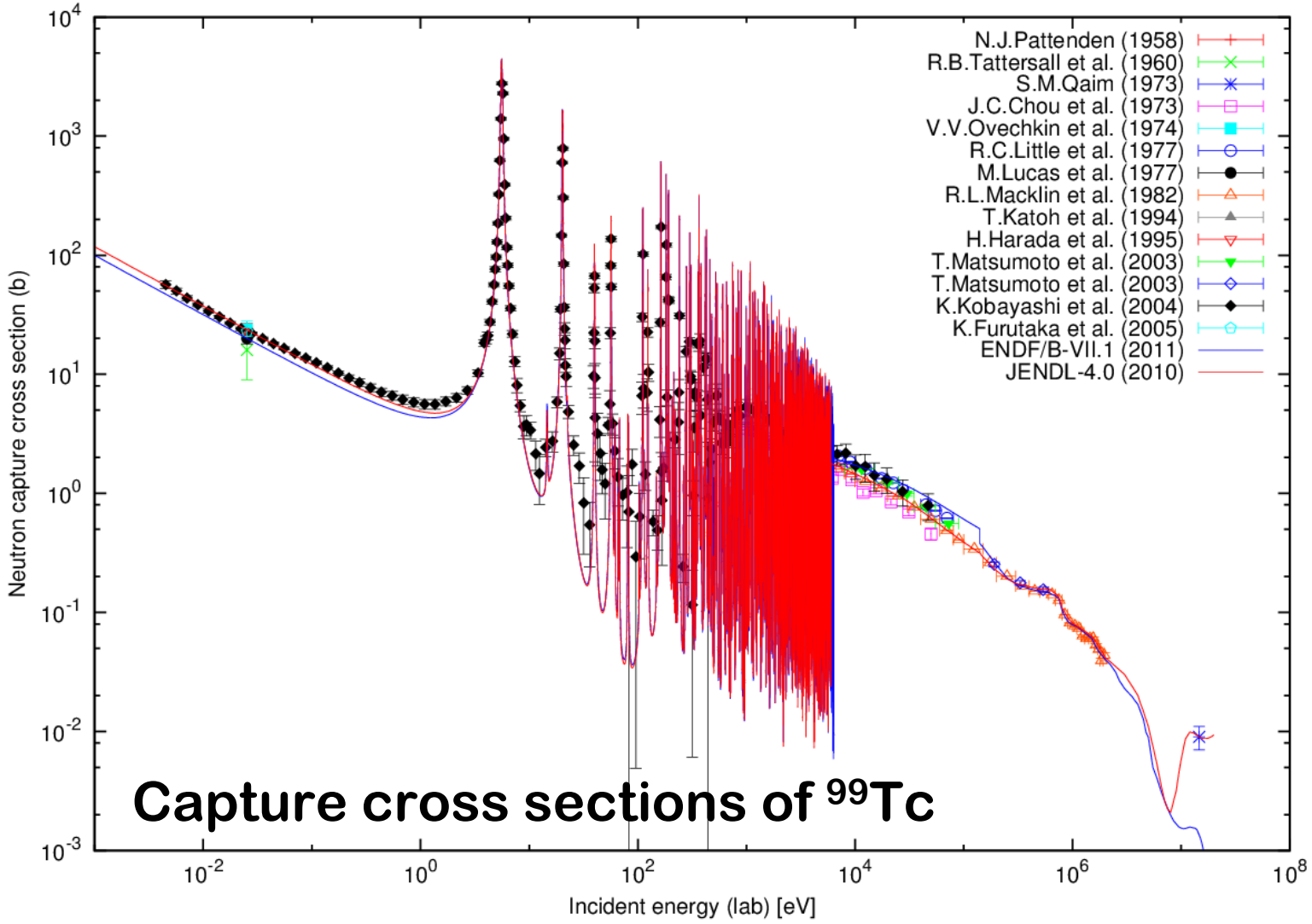

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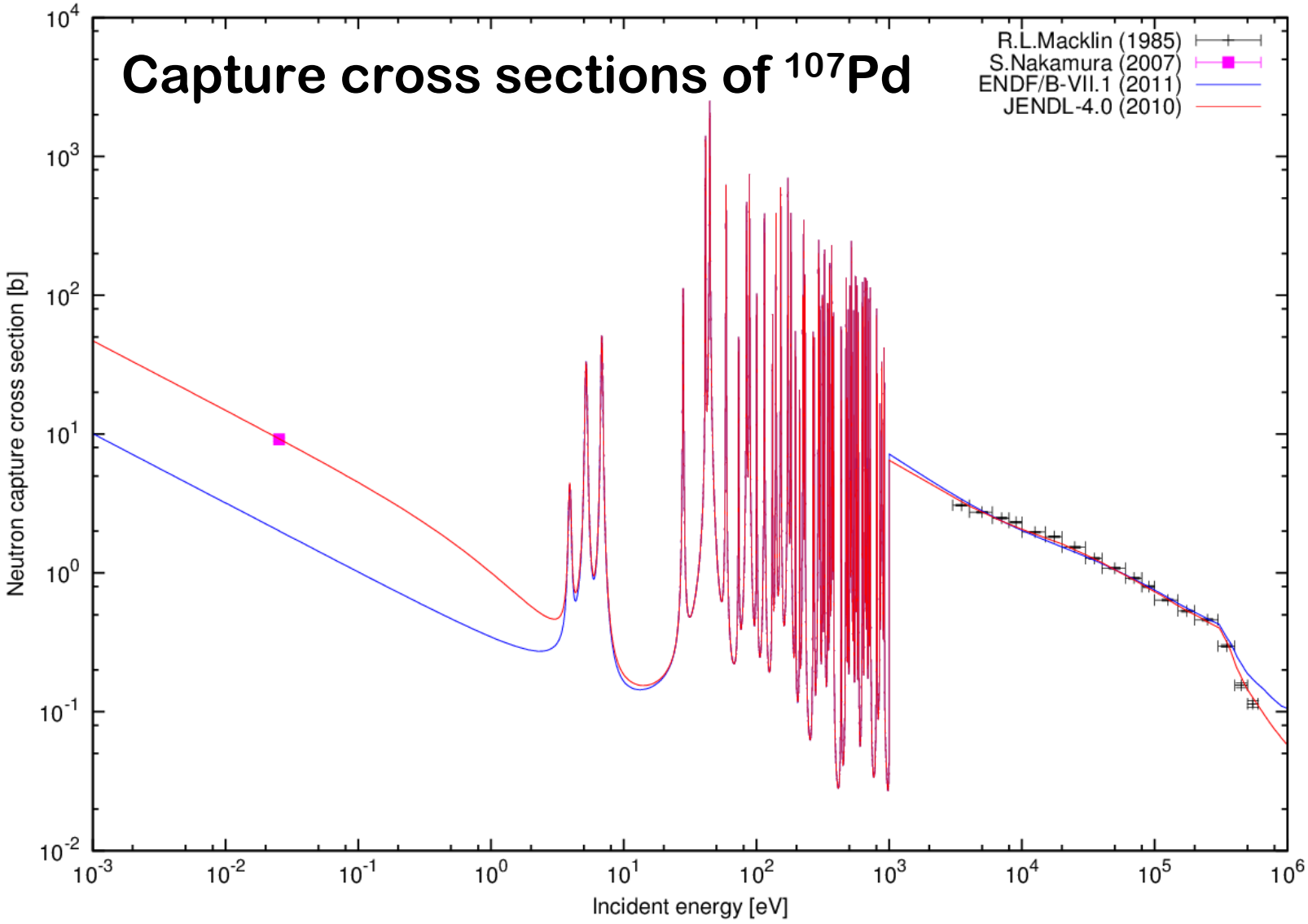

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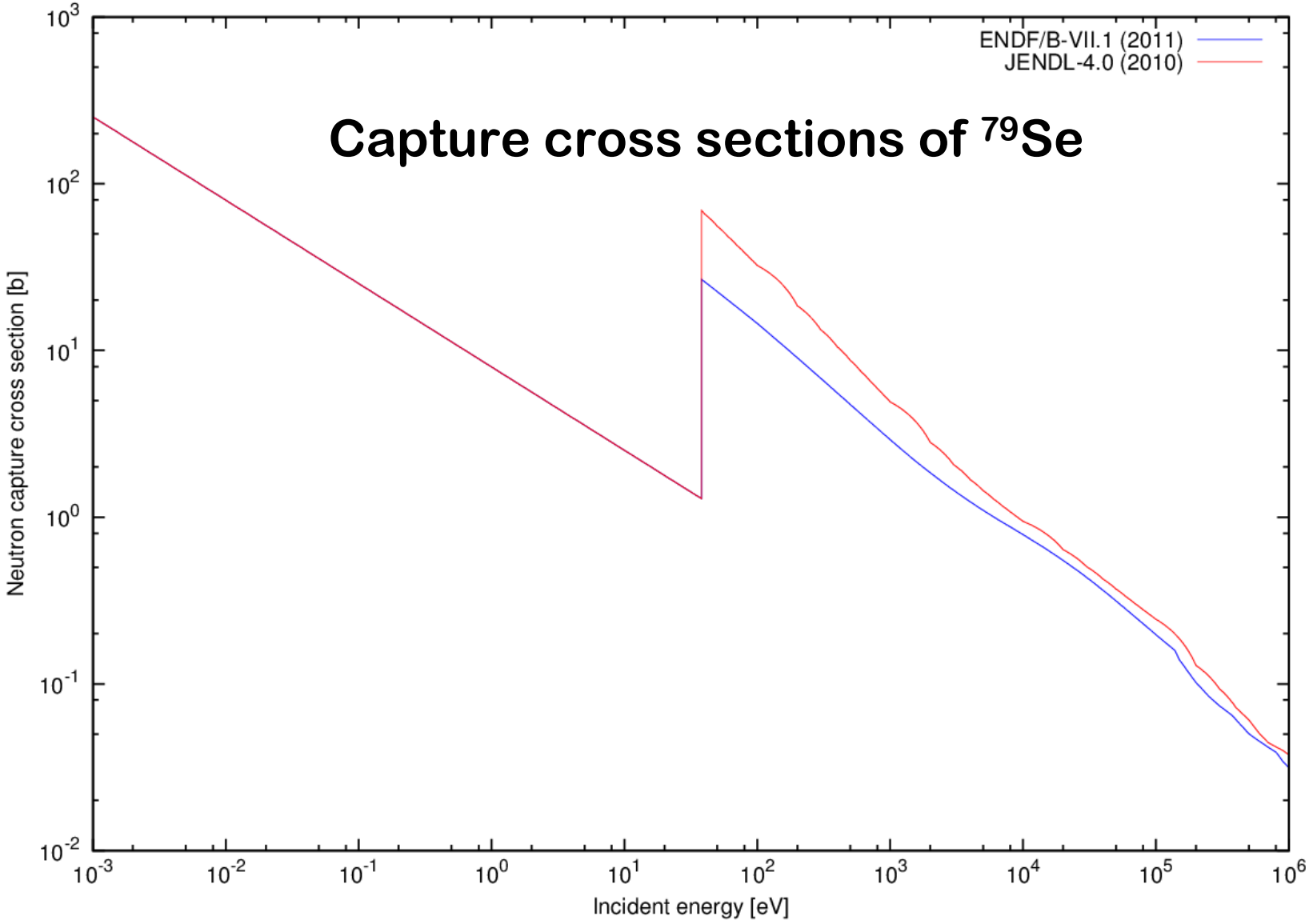


2. Present Status of the Accuracy of Nuclear Databases for MAs and LLFPs

Accuracy at the present moment:

5% - 100%

Accuracy needed for the development of Nuclear Transmutation Technology:

3% - 5%

3. Cross Section Measurements for MAs and LLFPs

Problems in Measurements for MAs and LLFPs

- Isotopic and Chemical Impurities in Samples*
  (Preparation of High-Purity Sample is essential.)
  --> Impurity identification is necessary.
  --> Measurements are required for Impurities.

- Radioactivity of Sample
  --> Small amount of sample
  --> Intense and Pulsed Neutron Source

*Samples of MAs and LLFPs are chemically extracted from spent fuel. No isotopic separation is usually performed.

3. Cross Section Measurements for MAs and LLFPs

- Intense and Pulsed neutron sources:

  # Electron Linear Accelerators: Photo-Neutron Sources

  ORELA, GELINA, KURRI-LINAC, etc. (40 - 200 MeV)

  0.01 - 0.001 neutron/electron

  # Proton Linear Accelerators (+ Synchrotron): Spallation Neutron Sources

  LANCE, n_TOF, J-PARC/MLF, etc. (0.8 - 20 GeV)

  20 - 300 neutrons/proton

3. Cross Section Measurements for MAs and LLFPs

Spallation Neutron Sources

- **LANSCE**: 0.8 GeV protons + W target (80 KW)

  LANSCE: Los Alamos Neutron Science Center

  Lujan Center

- **n_TOF** at CERN: 20 GeV + Pb target (9 KW)

- **MLF** at **J-PARC**: 3 GeV protons + Hg target (>200 KW)

  J-PARC: Japan Proton Accelerator Research Complex

  MLF: Material and Life Sciences Experimental Facilities

3. Cross Section Measurements for MAs and LLFPs

- LANSCE (Lujan)

Capture, Fission and Inelastic Cross Sections, DDX

- n_TOF

Capture and Fission Cross Sections

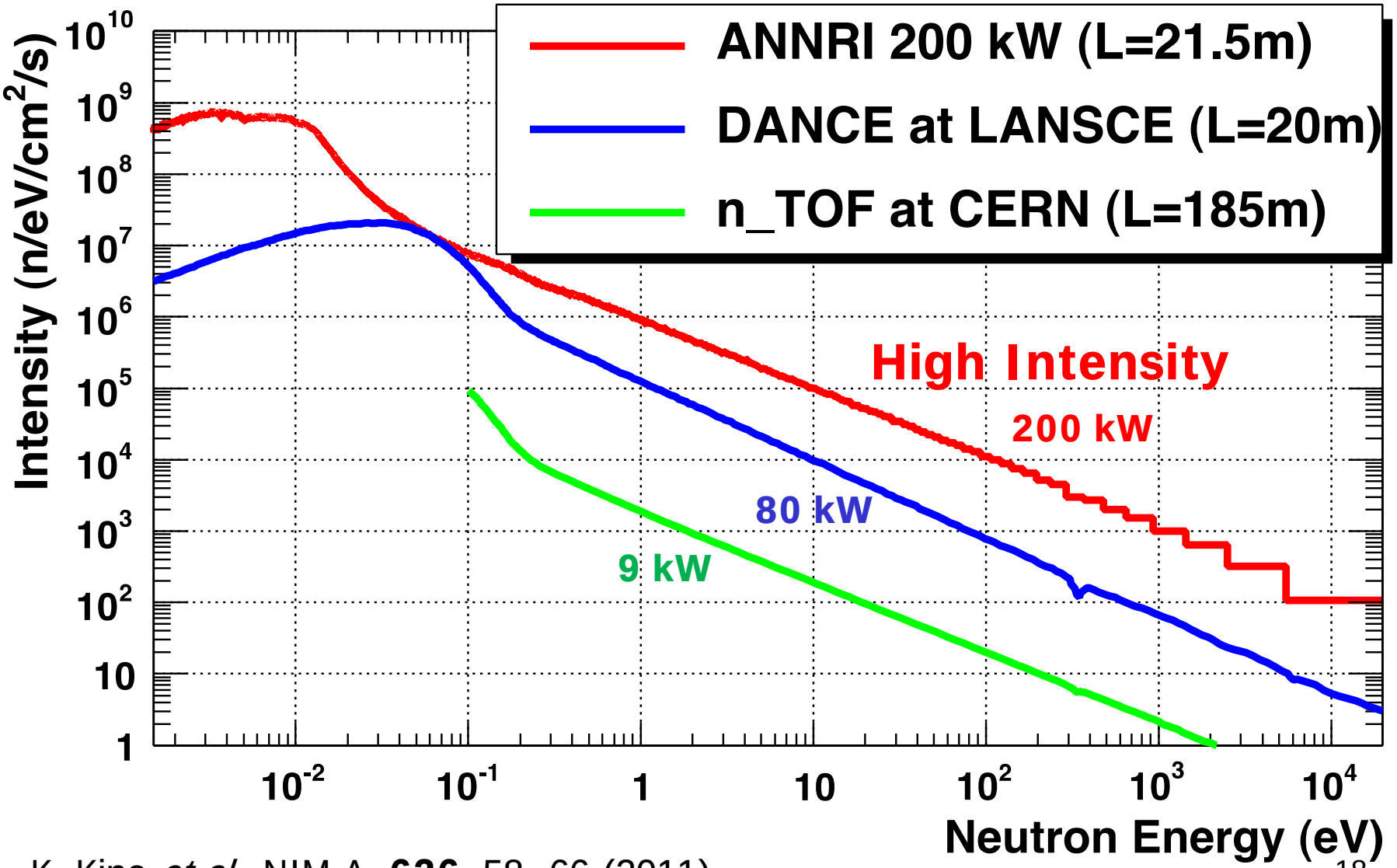
- ANNRI* at J-PARC/MLF

Capture Cross Sections of MAs, LLFPs, etc.

 # Th, U, Pu and unsealed RI are not usable in MLF

***** ANNRI**:** Accurate Neutron-Nucleus Reaction Measurement Instrument

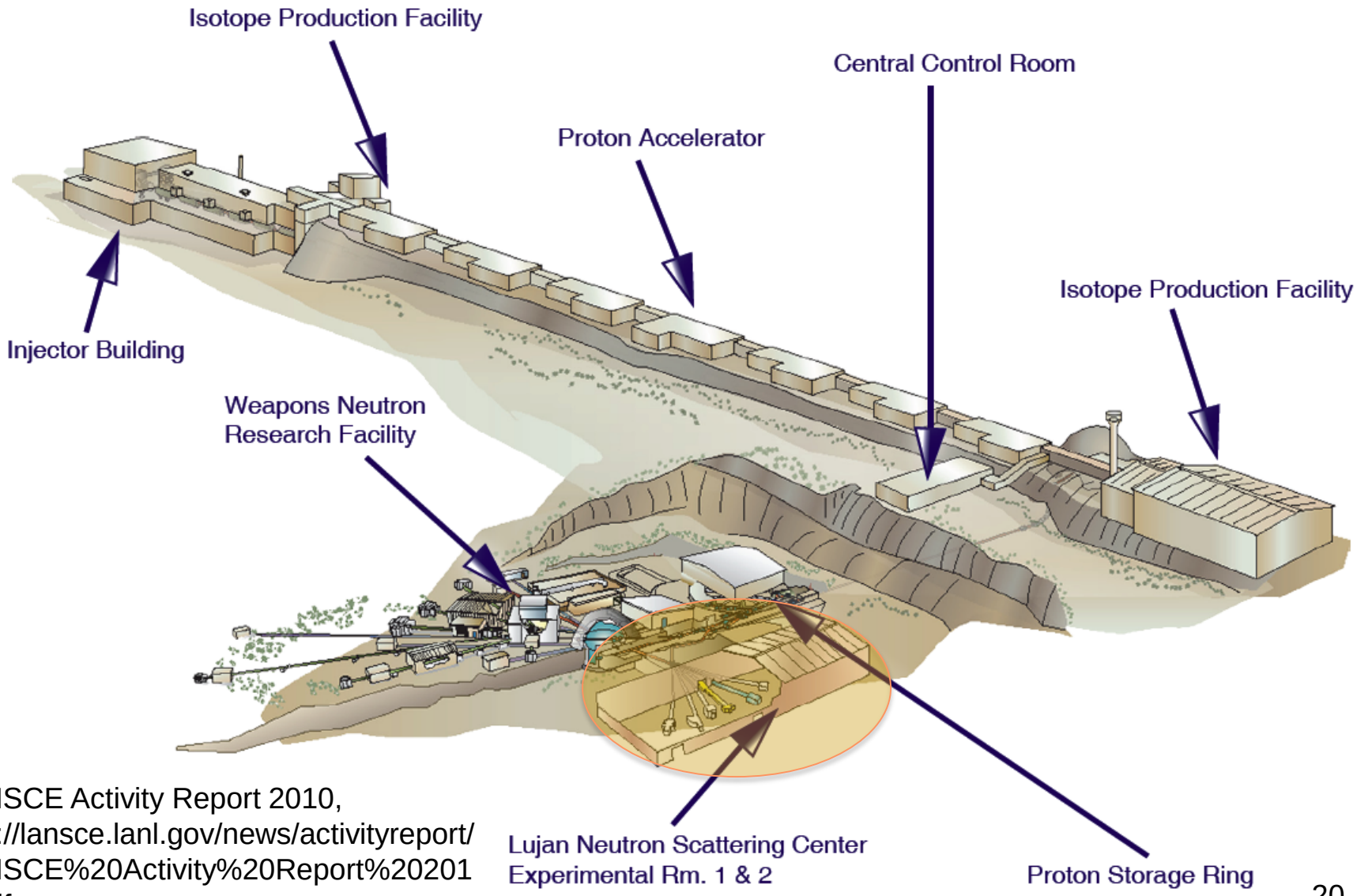
3. Cross Section Measurements for MAs and LLFPs



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

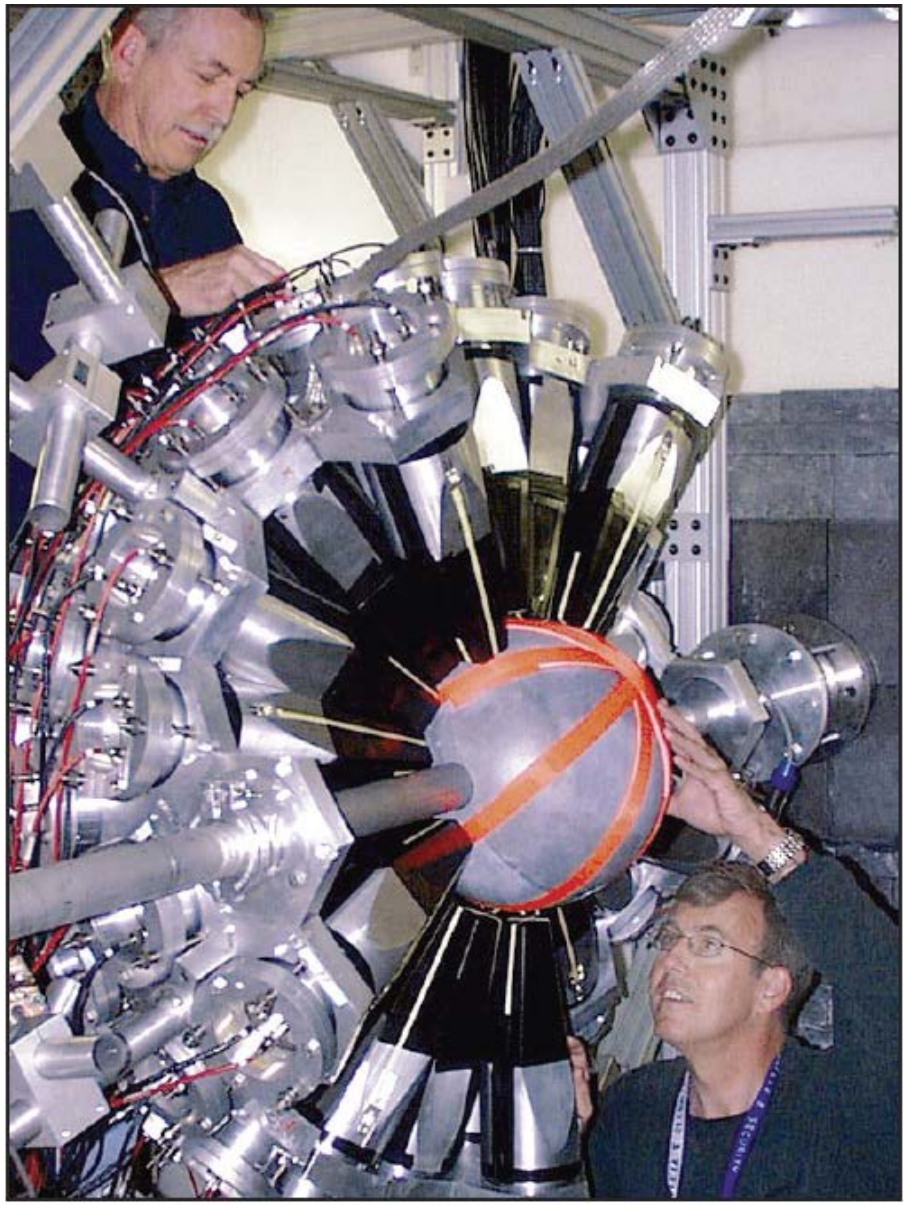
3. Cross Section Measurements for MAs and LLFPs

3. Cross Section Measurements for MAs and LLFPs



LANSCE Activity Report 2010, http://lansce.lanl.gov/news/activityreport/LANSCE%20Activity%20Report%202010.pdf

3. Cross Section Measurements for MAs and LLFPs



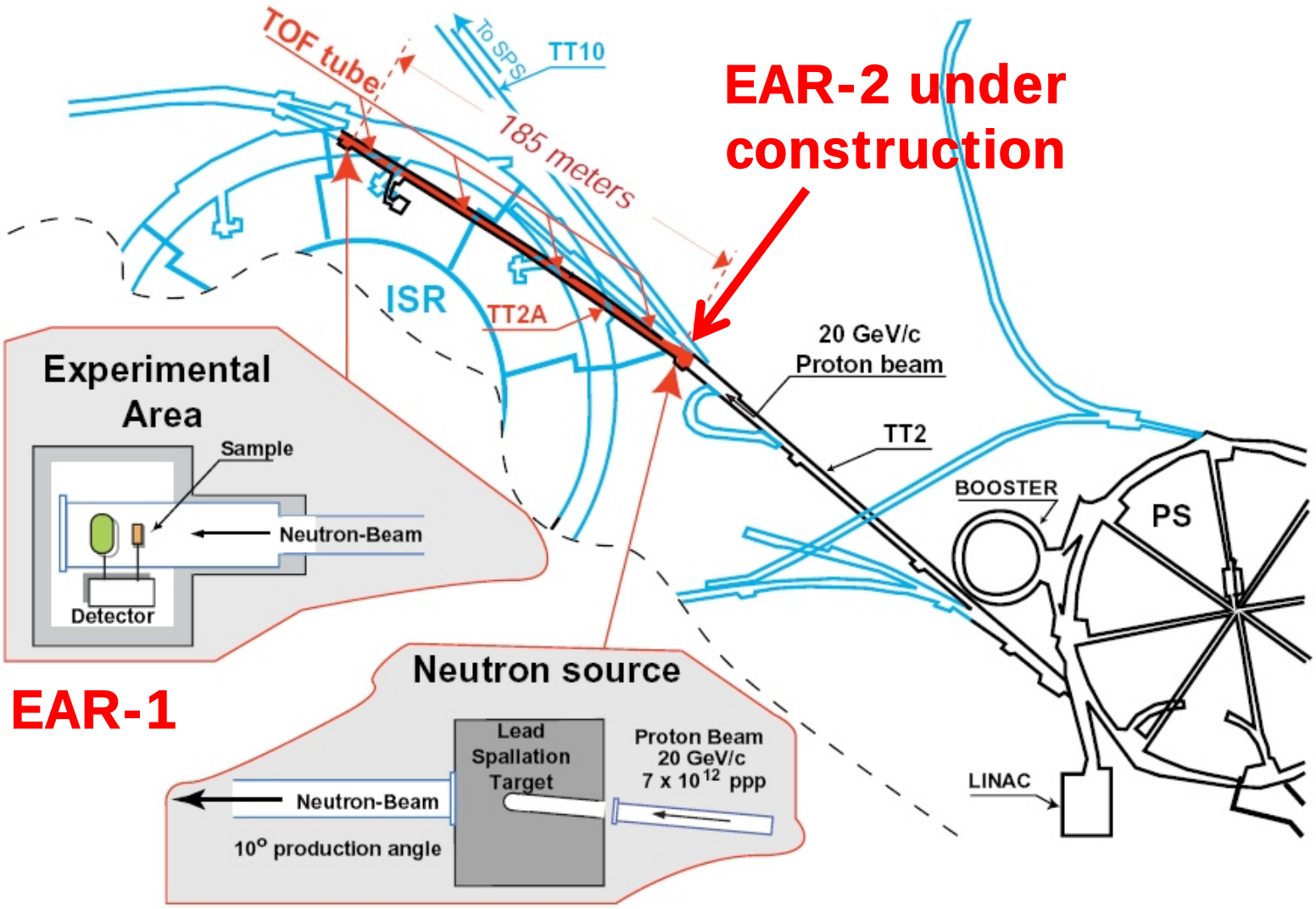
DANCE

Detector for Advanced Capture Experiments

Specifications	
Moderator	Water (2nd Tier)
Flight Path	20 m to sample
Beam Size at Sample Location	1 cm diameter
Sample Size	> 100 micrograms
Gamma Detection	160 BaF_2 scintillators
Flux Monitoring	^{6}Li, BF_3, ^{235}U fission
Typical Experiment Duration	10 days

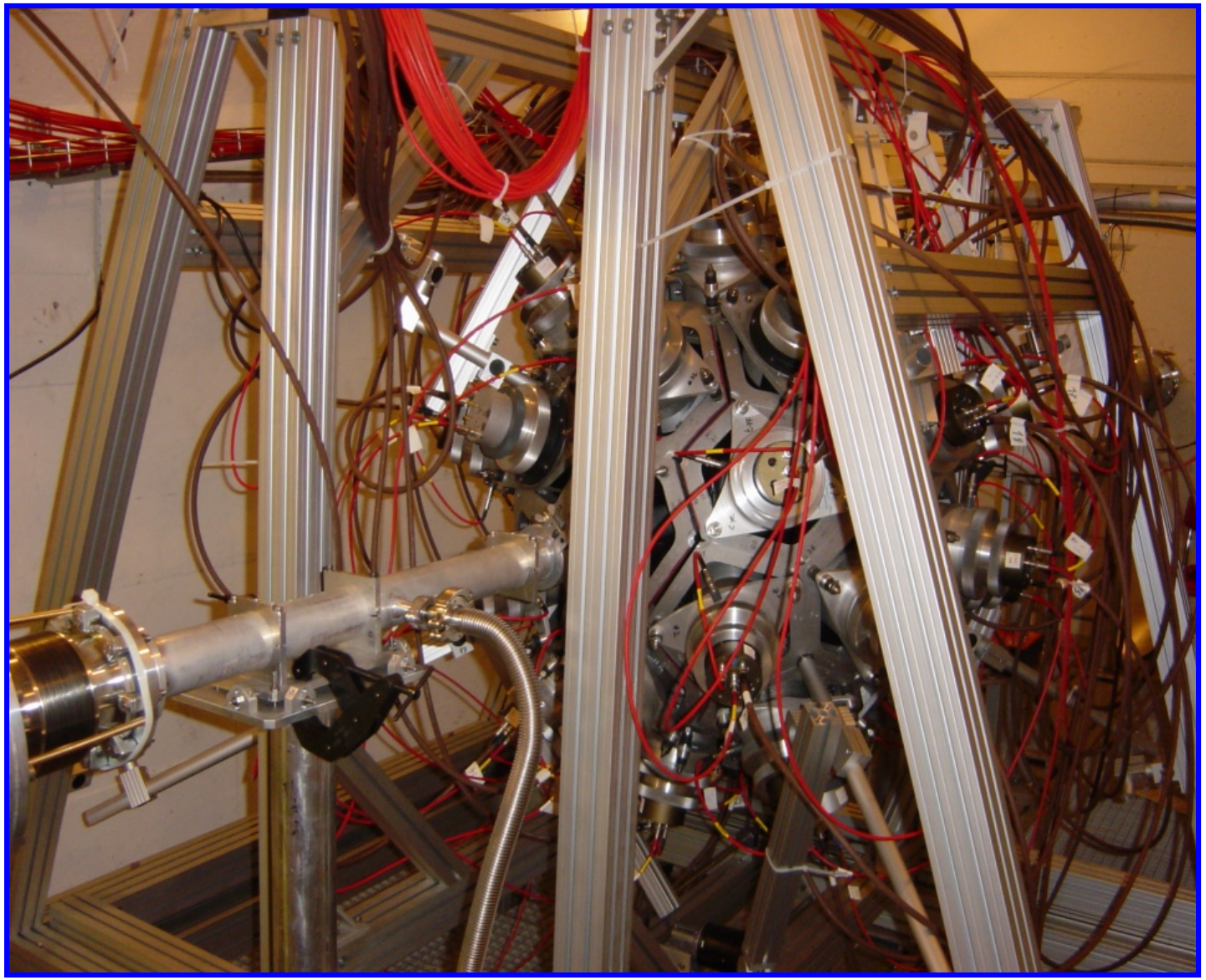
http://lansce.lanl.gov/lujan/instruments/DANCE/pdfs/DANCE.pdf

3. Cross Section Measurements for MAs and LLFPs



"http://pceet075.cern.ch/ "CERN n_TOF Facility: Performance Report"

3. Cross Section Measurements for MAs and LLFPs



TAC: Total Absorption Calorimeter

40 BaF_2

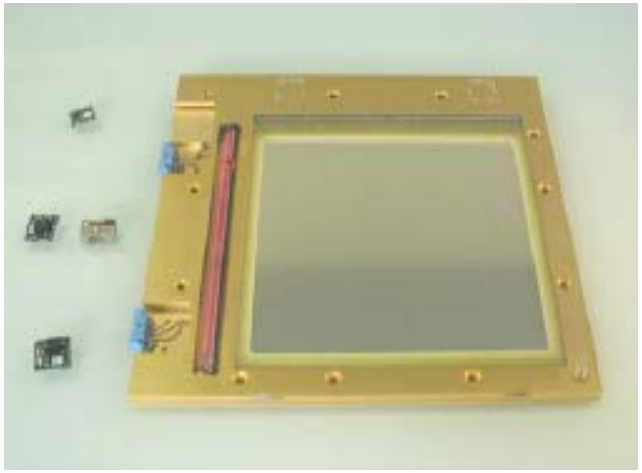
3. Cross Section Measurements for MAs and LLFPs

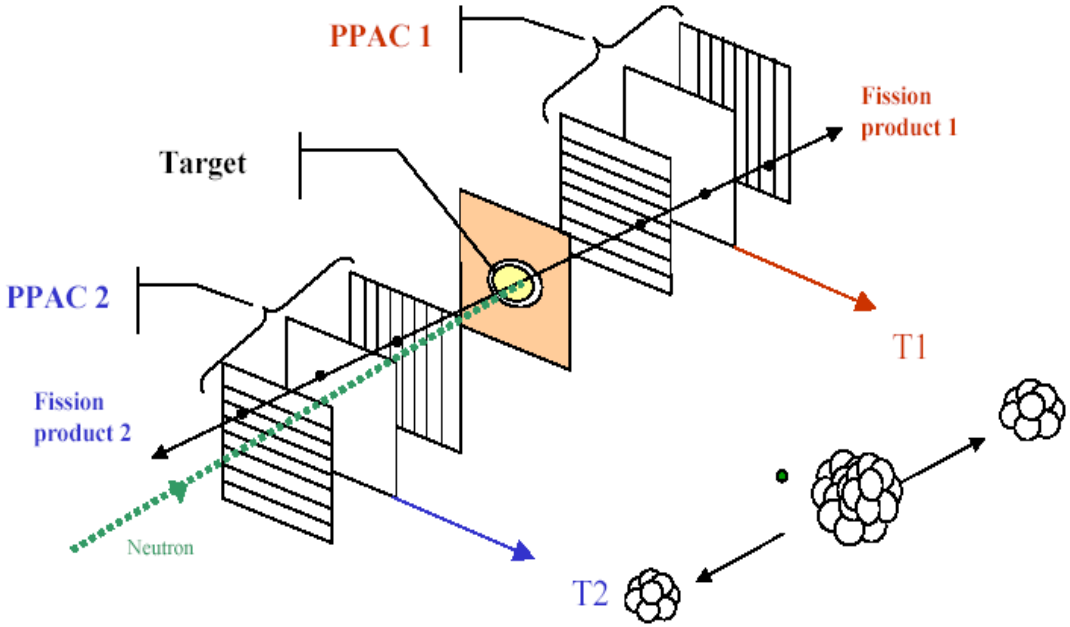
Parameters

Dimensions: 20x20 cm^2
Isobutane gas: 7 mbar
High Voltage: 500-600 V
Gap between electrodes: 3mm
Electrode thickness: 1.5 μm (Mylar+Al)
Deposit thickness: 100-300 $\mu g/cm^2$
Backing thickness: 0.1 μm (Al)
1.5 μm (Mylar)
Fission event identification:
T_1 in coincidence with T_2
1 anode: Mylar foil Al-plated
2 cathodes with 2mm strips
Delay between strips: 3 ns

☺ Position sensitive
☹ 5 channels/detector

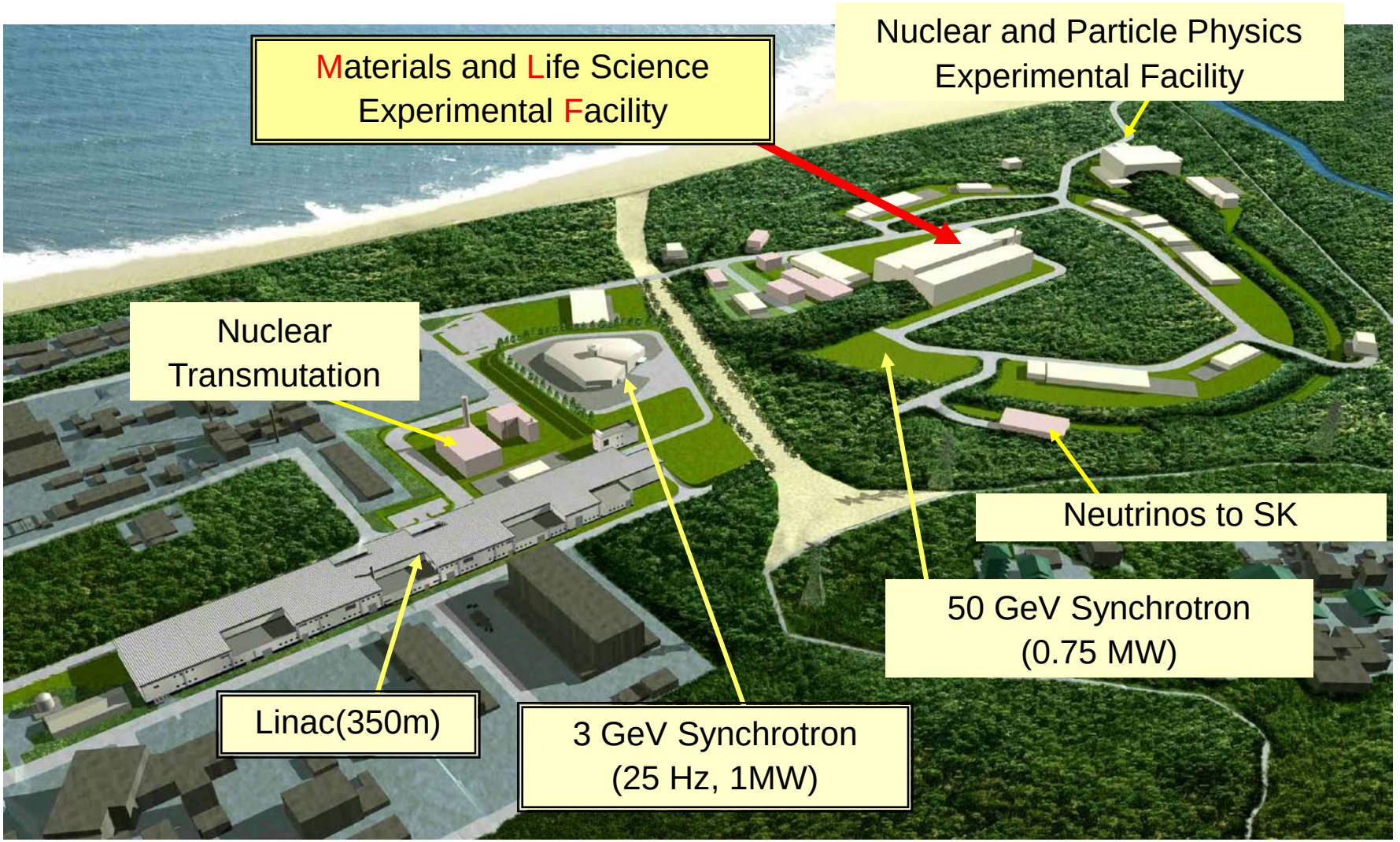
Construction by:
IN2P3 (IPN Orsay)





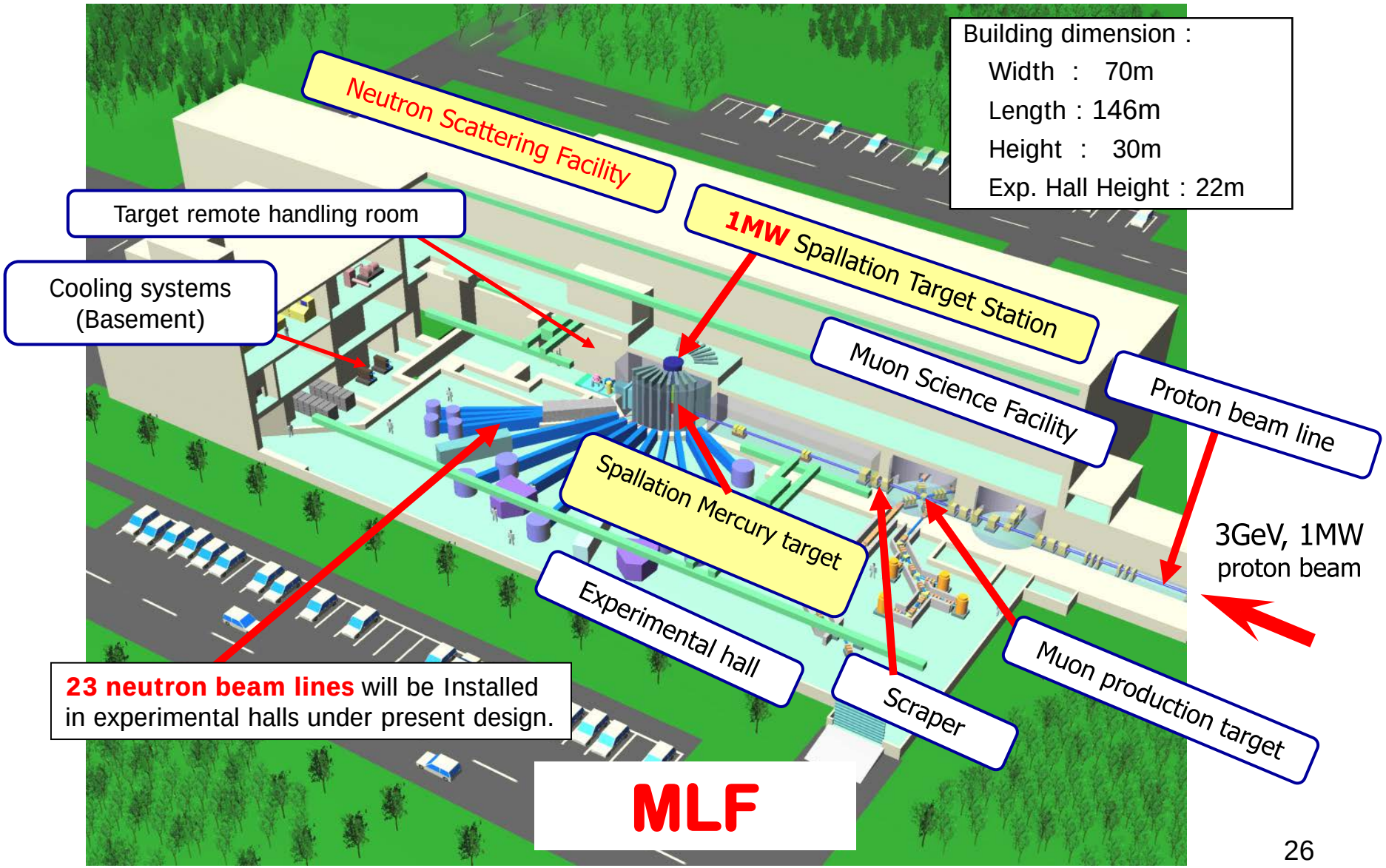
Parallel Plate Avalanche Counters PPACs

3. Cross Section Measurements for MAs and LLFPs



J-PARC

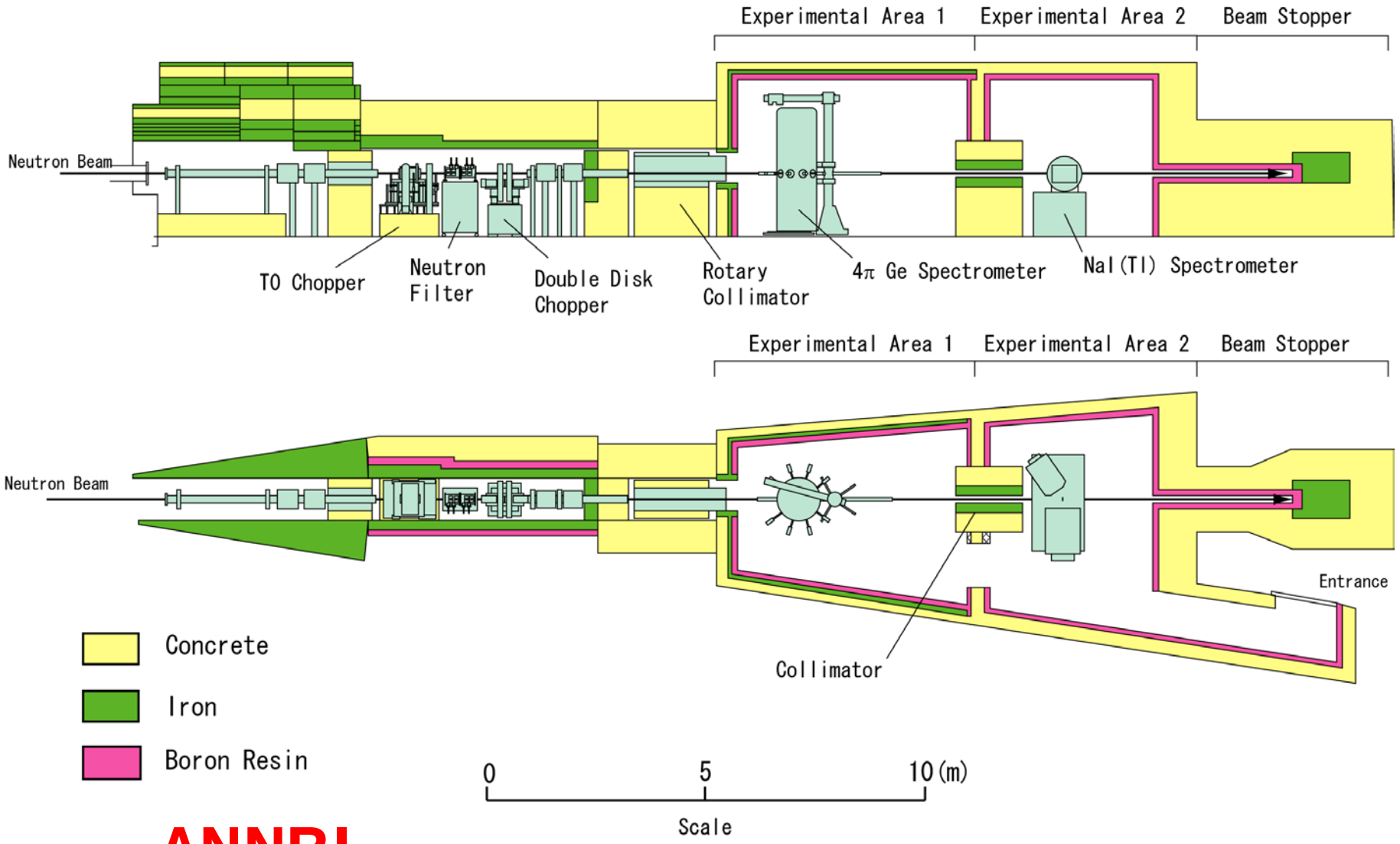
3. Cross Section Measurements for MAs and LLFPs



3. Cross Section Measurements for MAs and LLFPs

ANNRI (July, 2009)

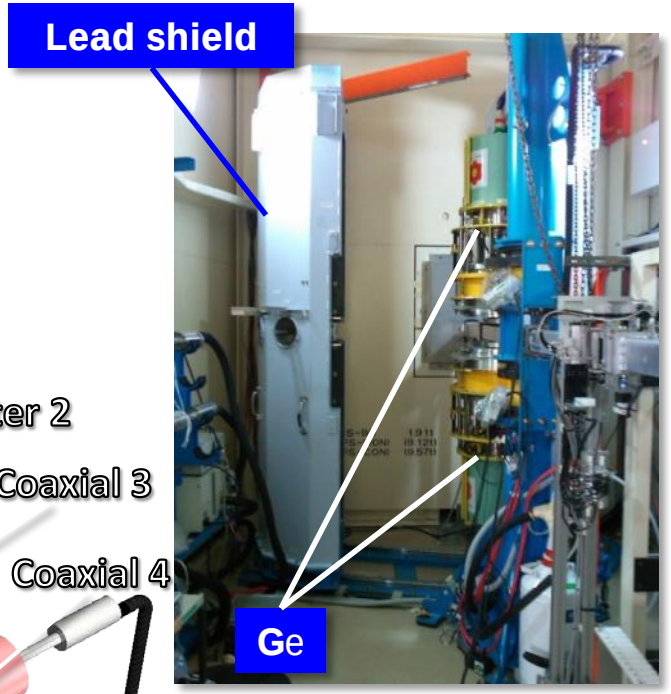
3. Cross Section Measurements for MAs and LLFPs

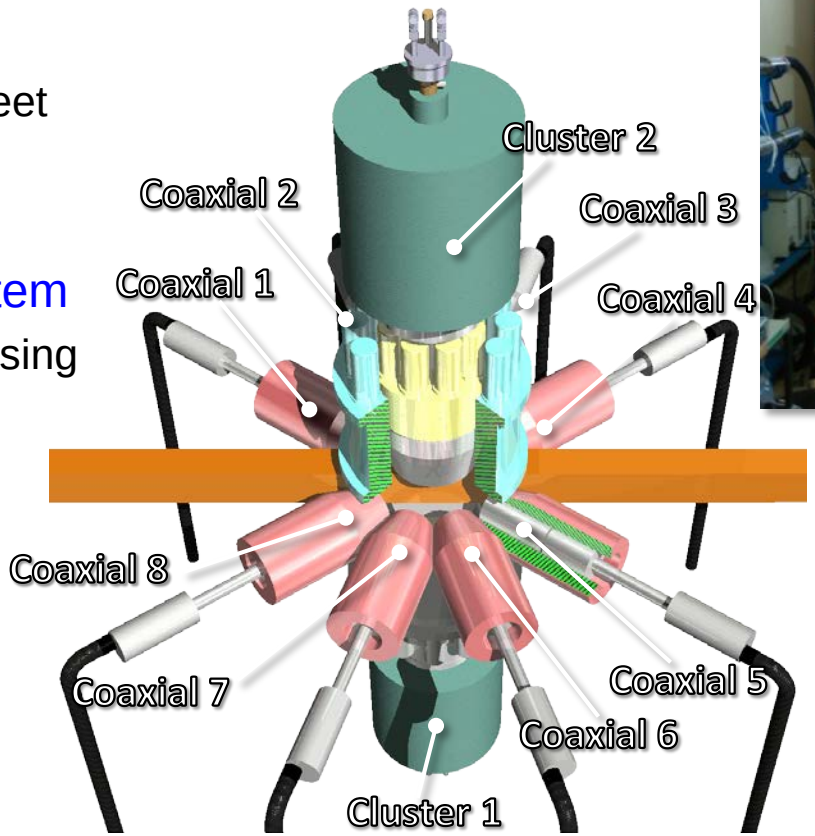


ANNRI

3. Cross Section Measurements for MAs and LLFPs

- Large Ge detector array
  - Two cluster Ge detectors: 7 crystals for each
  - Eight coaxial Ge detectors
  - BGO anti-Compton detectors
- Shielding
  - Borated rubber sheet
  - ^{6}LiH
  - Lead shields
- Data acquisition system
  - Digital data processing
  - Pulse height and TOF are recorded sequentially
  - Time resolution = 10 nsec

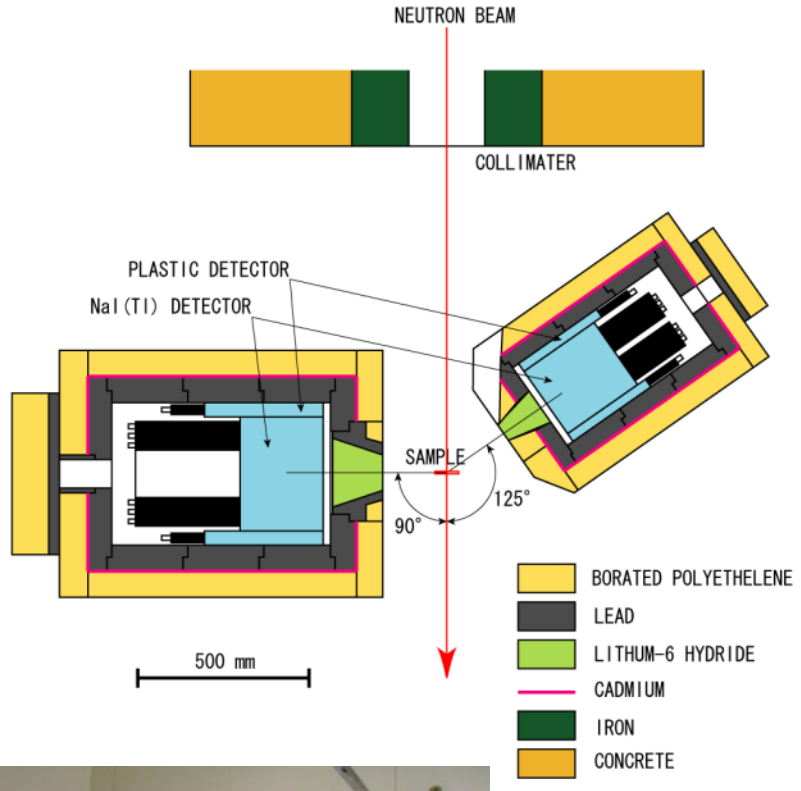


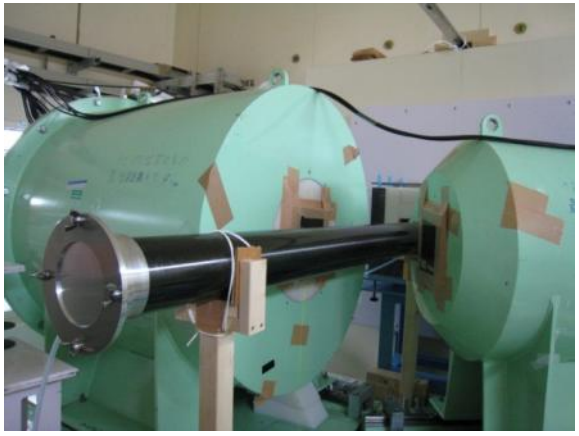


4π Ge Spectrometer

3. Cross Section Measurements for MAs and LLFPs

- Use for:
  - Complementary use to Ge array
  - Measurement in the high energy range
- Detectors
  - 90° detector: 13” diam. × 8” long
  - 125° detector: 8” diam. × 8” long
- Shielding
  - Borated polyethylene, Pb, ^{6}LiH, Cd
- Data acquisition
  - Multi-stop time digitizer
  - TOF, pulse height, pulse width are recorded sequentially





NaI(Tl) Spectrometer

4. A Japanese Nuclear Data Project

Title of Project

Systematic Study on Neutron Capture Reaction Cross Sections for the Technological Development of Nuclear Transmutation of Long-Lived Nuclear Wastes

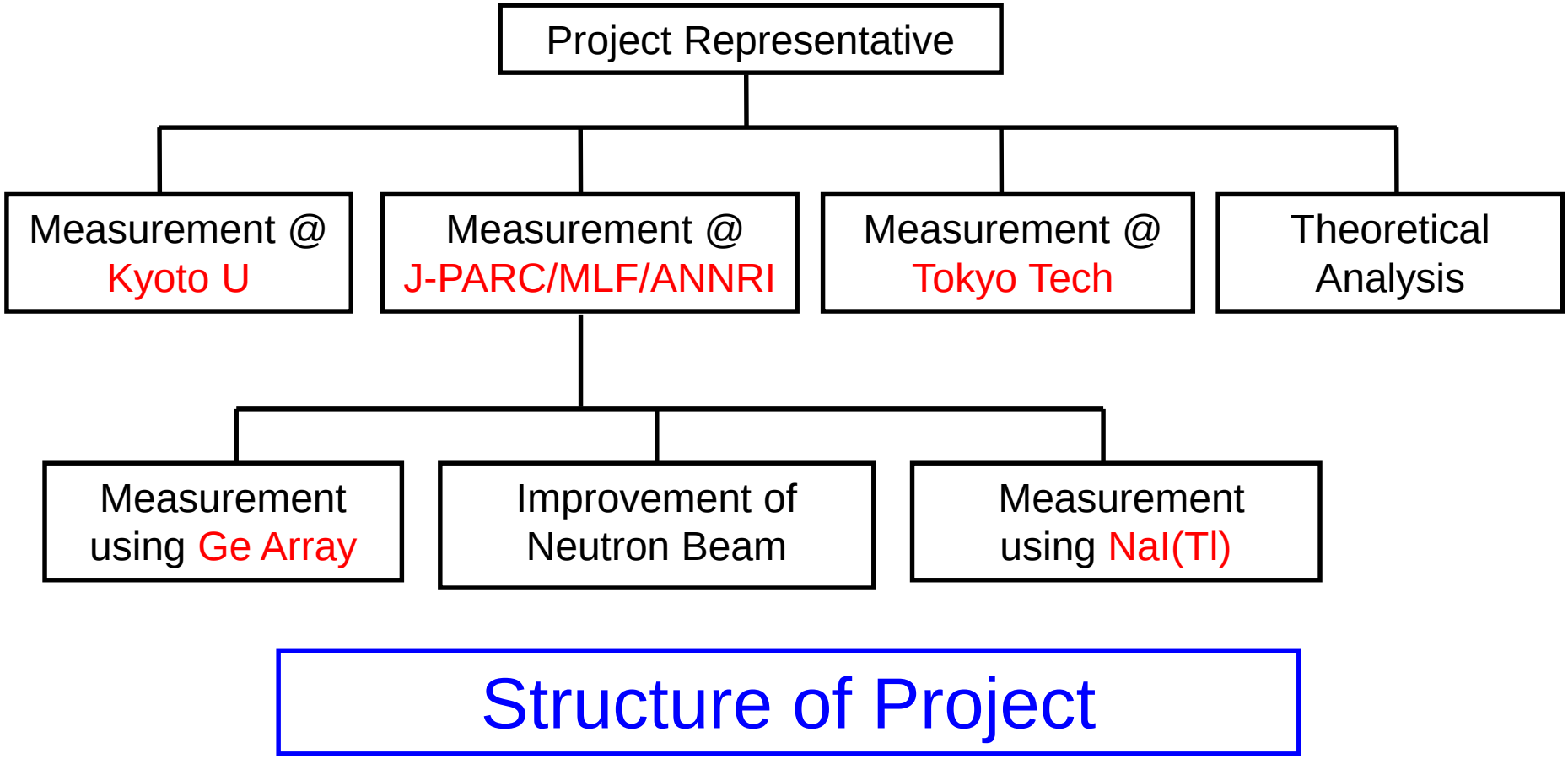
Objective of Project

To contribute to the improvement of nuclear database accuracy, by making the precise measurements of capture cross sections of Long-Lived Nuclear Wastes (LLNW), analyzing the measured results theoretically, elucidating the capture reaction mechanism of LLNW, and supplying reliable calculated capture cross sections for all LLNW and in the whole neutron energy region.

Term of Project

From June, 2010 to March, 2015

4. A Japanese Nuclear Data Project



Structure of Project

4. A Japanese Nuclear Data Project

- Radioactive samples of MAs and LLFPs were prepared.
- All samples are sealed in Al or Ti containers.

Radioactive Samples

Samples of important stable isotopes were also prepared.

MA

	Half Life	Decay Mode	Activity (Bq)	Net Weight (mg)
Np-237	2.1×10^6 y	α	26M 5.2M 1M	1000 200 38
Am-241	4.3×10^2 y	α	0.95G	7.5
Am-243	7.4×10^3 y	α	0.95G	128
Cm-244	1.8×10 y	α	1.8G × 6	0.6 × 6
Cm-246	4.4×10^3 y	α	1.8G × 4	1.1 × 4

LLFP

	Half Life	Decay Mode	Activity (Bq)	Net Weight (mg)
Zr-93	1.5×10^6 y	β^-	47M	472
Tc-99	2.1×10^5 y	β^-	50M	78
Pd-107	6.5×10^6 y	β^-	380k	20
I-129	1.6×10^3 y	β^-	3M	404

Experiments of ^{244,246}Cm
- Samples and Measurement Conditions -

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

	^{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 hours

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

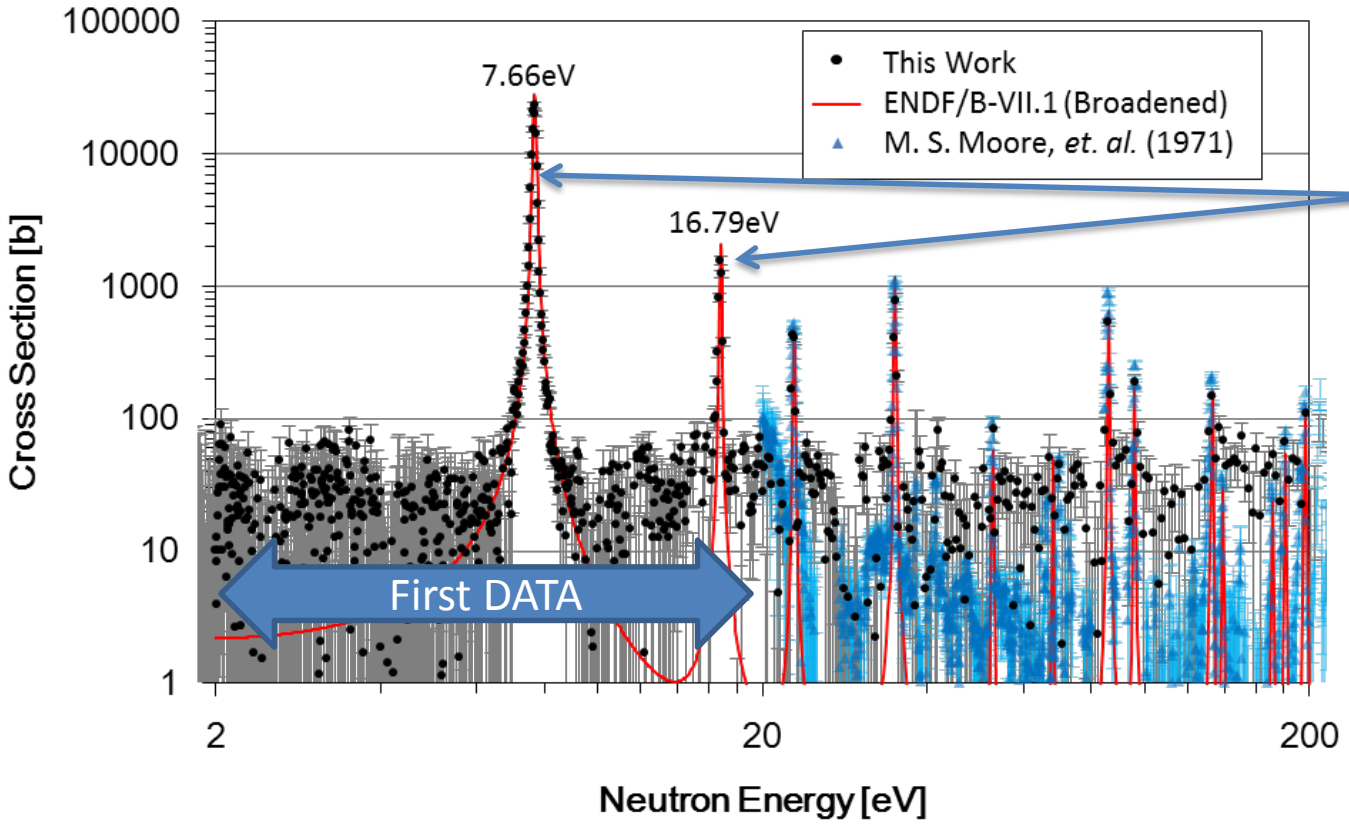
Container = Al capsule

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

Capture Cross Sections of ^{244}Cm

A. Kimura et al., J. Nucl. Sci. Technol., 49, 708-724 (2012)

Only one set of cross section data for ^{244}Cm (n,γ) was reported in 1971[1].



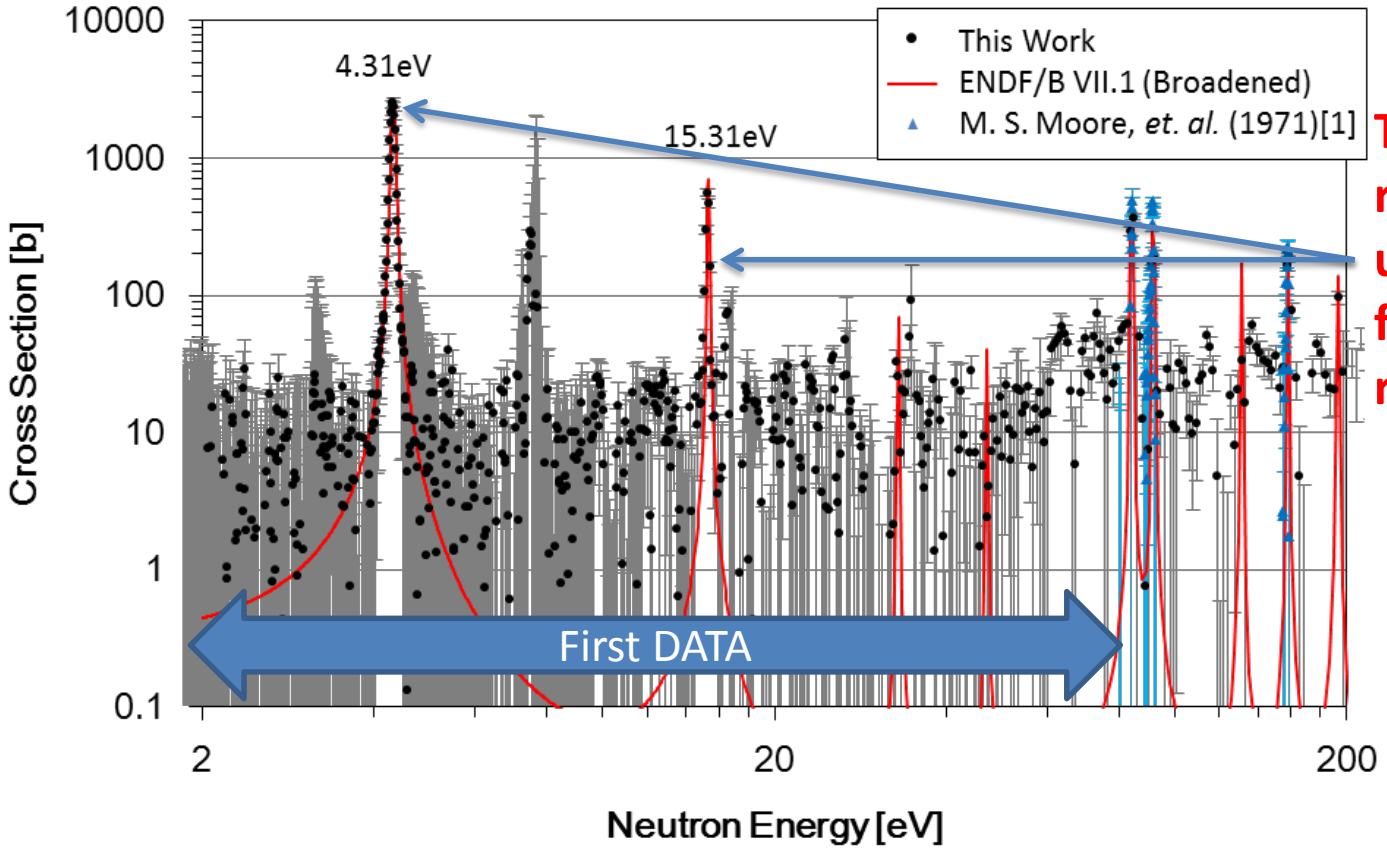
The results of the resonance peaks under 20-eV are also the first experimental results in the world.

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

Capture Cross Sections of ^{246}Cm

A. Kimura et al., J. Nucl. Sci. Technol., 49, 708-724 (2012)

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

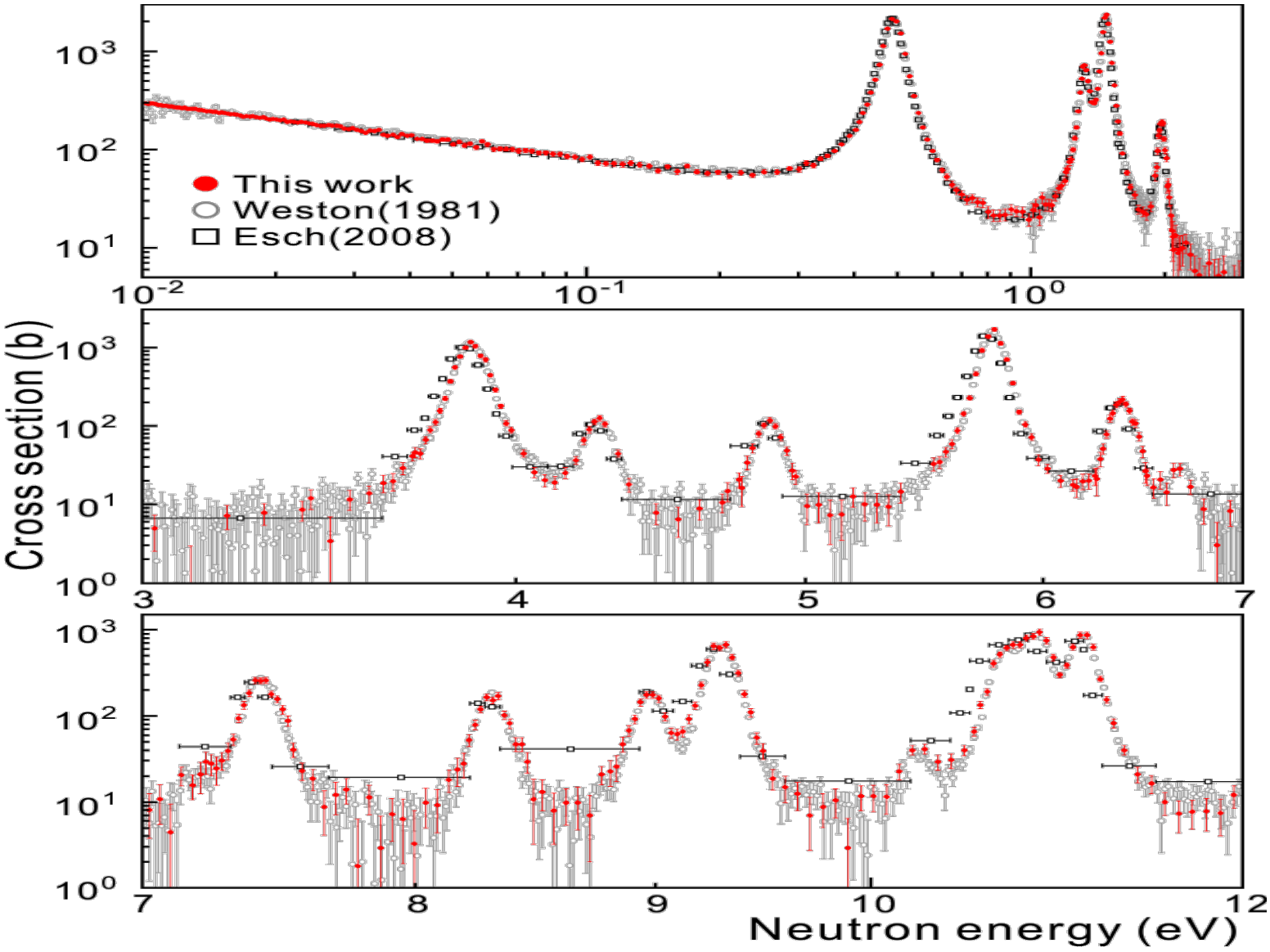


The results of the resonance peaks under 50-eV are the first experimental results in the world.

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

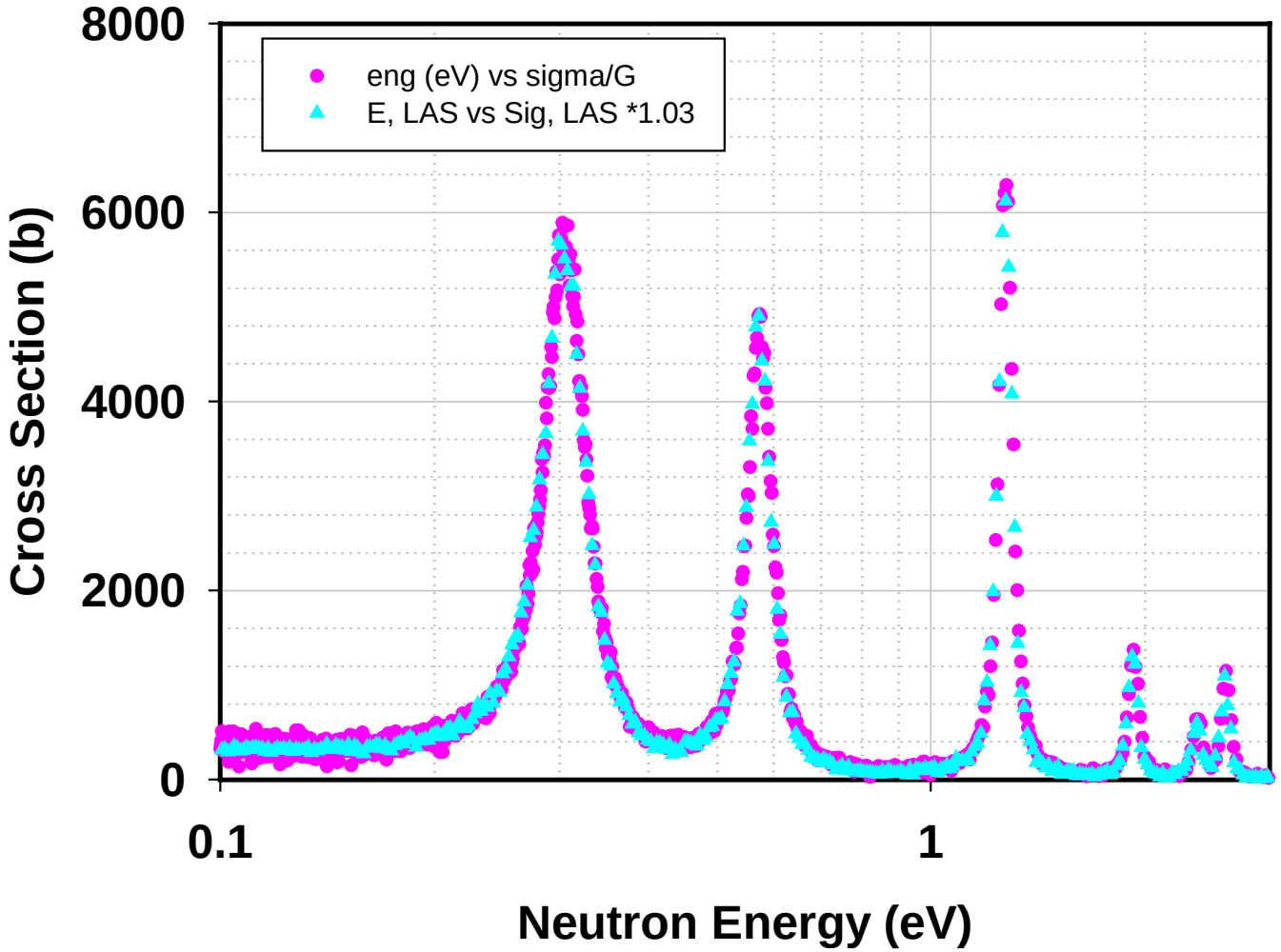
Capture Cross Sections of ^{237}Np

K. Hirose et al., J. Nucl. Sci. Technol., 50, 188-200 (2013)



Capture Cross Sections of ^{241}Am

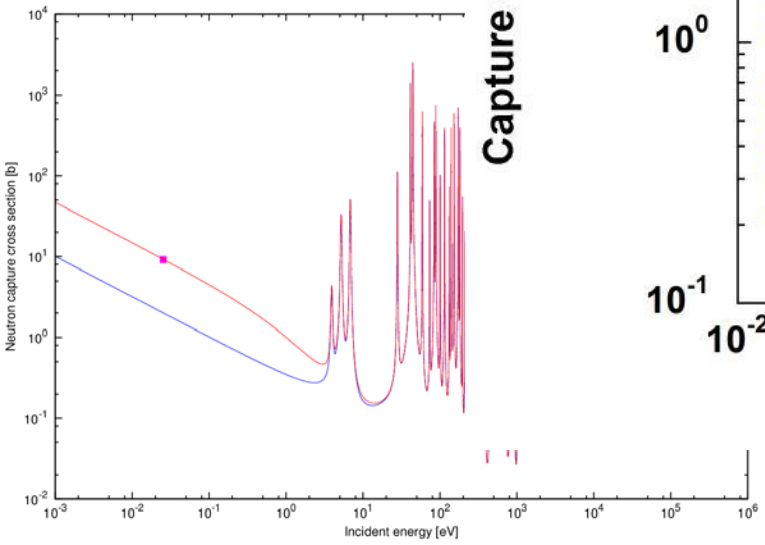
H. Harada et al., ND2013



665 [b] at 25.3 meV (DANCE) was normalized as 684.87 [b]

Capture Cross Sections of ^{107}Pd

S. Nakamura et al., ND2013



Capture Cross Section (b)

This Work
~10.0(b) @25.3meV

6.8-eV Pd-107

3.9 & 5.2-eV Res.
should be rejected
3.9-eV Res. --> ^{105}Pd
5.2-eV Res. --> ^{109}Ag

c.f. 9.16±0.27(b)
Lower limit of σ_0

by Prompt γ-ray Analysis
S.Nakamura *et al.*
J.Nucl.Sci.,44,103(2007)

This Work
JENDL-4.0

Neutron Energy (eV)

5. Summary

It was explained that databases on neutron capture and/or fission reaction cross sections for MAs, LLFPs, and related nuclides were indispensable for developing the nuclear transmutation technology.

The present status of the accuracy of nuclear databases for MAs and LLFPs was briefly reviewed.

A general view of cross section measurements for MAs and LLFPs using SNSs was taken.

A Japanese nuclear data project was explained.

Thank you for your attention

Collaborators

Hokkaido Univ.: Y. Kiyanagi (moved to Nagoya Univ.), K. Kino

Japan Atomic Energy Agency: H. Harada, S. Nakamura, A. Kimura, N. Iwamoto

Tokyo Institute of Technology: T. Katabuchi

Kyoto University: J. Hori, H. Yashima

This Work was supported by JSPS KAKENHI Grant Number 22226016.