SANDA WP2/Task 2.2 report

Alberto Mengoni
on behalf of the Task 2.2 partners: ENEA, CIEMAT, JRC-Geel, Uni Lodz, IRSN

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SANDA WP2/Task 2.2 definition

Task 2.2: Neutron capture cross sections
*Task coordinator: **ENEA**, partners: **CIEMAT, JRC, ULODZ, IRSN***

Subtask 2.2.1. Capture measurements of fissile isotopes.
CIEMAT, ULODZ and JRC will perform various cross section measurements at GELINA and n_TOF on the high priority reactions ^{239}Pu(n,g) and ^{239}Pu(n,f). The methodology developed within CHANDA for the absolute measurement of the ^{235}U alfa ratio will be applied to the ^{239}Pu case. A new ionization chamber built by ULODZ will be tested in a ^{239}Pu(n,f) measurement at JRC, which also deliver the ^{239}Pu samples. The combined measurement of the ^{239}Pu(n,g) and ^{239}Pu(n,f) cross sections will be carried out at CERN with the use of the Total Absorption Calorimeter.

Subtask 2.2.2. Capture measurement of stable isotopes.
ENEA will measure the ^{92,94,95}Mo(n,g) cross sections at GELINA and at the n_TOF facility with the high performance total energy detectors developed during the CHANDA project. The impact of the new evaluated nuclear data and their uncertainties will be verified in criticality safety and reactor applications at IRSN as end-user. The data will be part of an evaluation done in WP4 by IRSN.

SANDA WP2/Task 2.2 definition

Deliverable: 2.3
Report on the 239Pu(n,g), 92,94,95Mo(n,g) cross measurements at n_TOF and GELINA
when: month 40 updated to: month 56

Milestones
M.2.11 “Measurement of the 239Pu(n,g) at n_TOF”; M36
M.2.12 “Measurement of the Mo isotopes at GELINA and n_TOF”; M34

Update on the status of the ^{239}Pu data analysis

A. Sanchez-Caballero[1], V. Alcayne[1], J. Andrzejewski[2], D. Cano-Ott[1], J. García-Pérez[1], E. Gónzalez-Romero[1], J. Heyse[3], T. Martínez[1], E. Mendoza[1], J. Perkowski[2], J. Plaza del Olmo[1], A. Plompen[3], P. Schillebeeckx[3], G. Sibbens[3]

[1]CIEMAT, Spain
[2]University of Lodz, Poland
[3]JRC-Geel, Belgium

Accelerator and Research reactor Infrastructures for Education and Learning ARIEL

^{239}Pu production in LWR

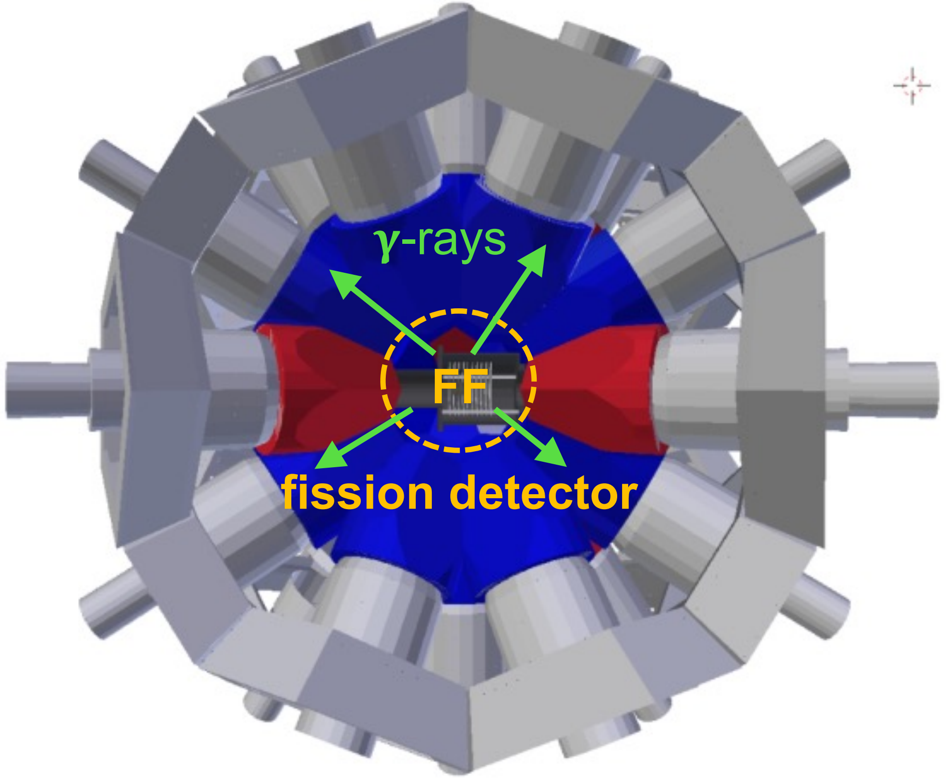
Standard fresh nuclear fuel for thermal reactors has a 5% ^{235}U and a 95% ^{238}U. ^{239}Pu is produced during the reactor operation mainly by neutron captures in ^{238}U + decays of ^{239}U and ^{239}Np. The ^{239}Pu is fissile and thus its neutron induced fission contributes to the power.

LLFP

Experimental technique

Fission tagging: **γ**-rays in coincidence (fission background) and anticoincidence (capture signal) with the fission detector.
J. Balibrea et al. (The n_TOF Collaboration), PRC **102**, 044615, (2020)

$$Y_\gamma = \frac{c_{aco,\gamma} - \frac{1-\epsilon_f^*(E_n)}{\epsilon_f^*(E_n)} c_{tag} - c_{oth,\gamma}}{\epsilon_\gamma \phi_N}$$

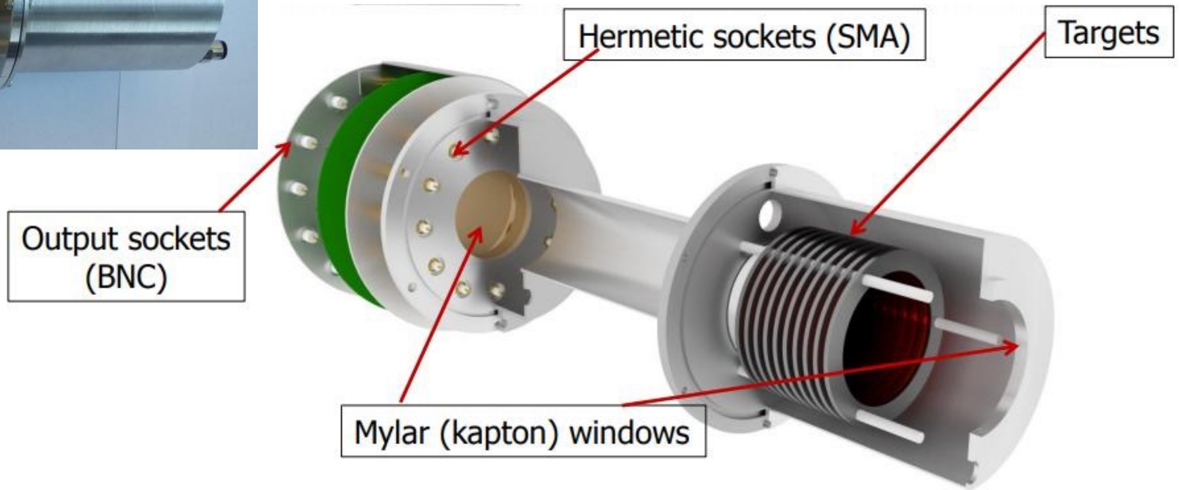


c_{tot} = counts in the TAC.
c_{tag} = counts in the TAC in coincidence. with the ionisation chambers.
c_{oth}= background in the TAC.
ϵ_f^* = fission tagging efficiency.
ϵ_γ = capture detection efficiency.

A new fission chamber

A new multi target fission chamber has been built taking into account the following important characteristics:

1. **Low mass** intercepting the neutron beam, to minimize the background in the TAC due to captures and elastically scattered neutrons.
2. Good **discrimination** between **alphas (2 MBq/mg)** and **fission fragments (5 mm gap)**.
3. Small **pile-up** effects.

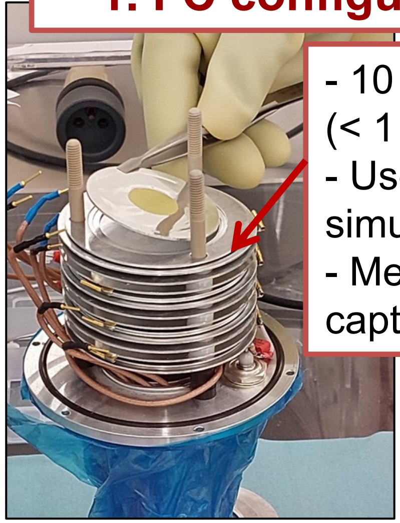


UNIVERSITY OF LODZ

A quick reminder

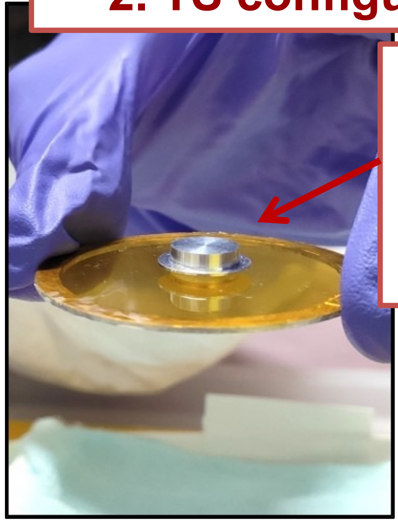
- In 2022 we **measured** the **^{239}Pu(n,γ)** and **^{239}Pu(n,f) (α-ratio) cross-sections in EAR1**. The experiment consisted in **two different setups**: Fission Chamber configuration (thin samples) and Thick Sample configuration.

1. FC configuration



- 10 thin Pu samples (< 1 mg).
- Use FICH + TAC simultaneously.
- Measure fission and capture (up to 1 keV).

2. TS configuration



- 1 thick sample (~100 mg).
- Use only TAC.
- Measure capture above 1 keV.

- **Re-processed** of the entire exp. dataset with a refined version of the new Pulse Shape Analysis routine was performed at the beginning of 2023 (see ^{239}Pu presentation at the n_TOF Collaboration meeting in May 2023). Improvements in the preliminary results.
- **BaF_2 time and energy calibrations** were performed and validated with Monte Carlo simulations.
- Pileup/Dead-time analysis performed.

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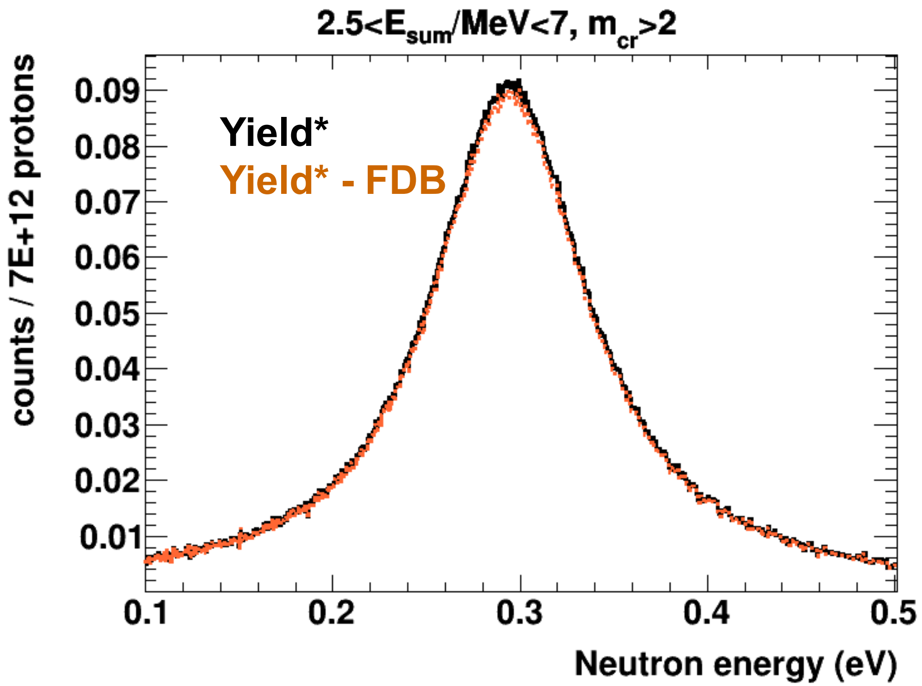
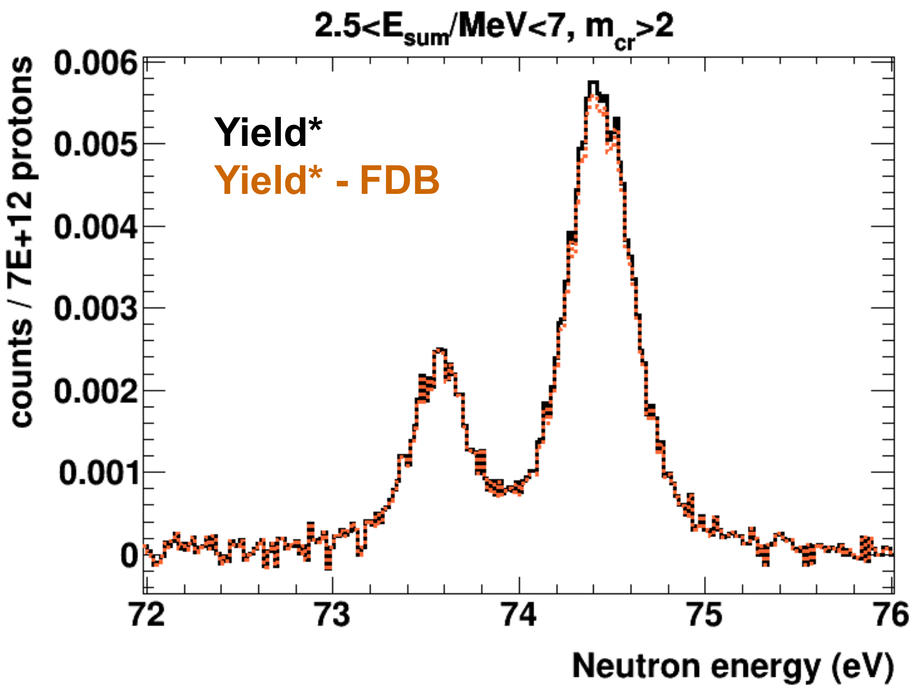
The *PFB effect* in the final capture yield (1/2)

Comparing the yield* (before dividing by neutron flux) with and without the post-fission background for the TAC event conditions of the analysis. In general, the effect is small, reaching up to ~3% change in some resonances.

In the thermal resonance, the effect of subtracting this background is of <u>2.2%</u>



Here, about a <u>2.7%</u>



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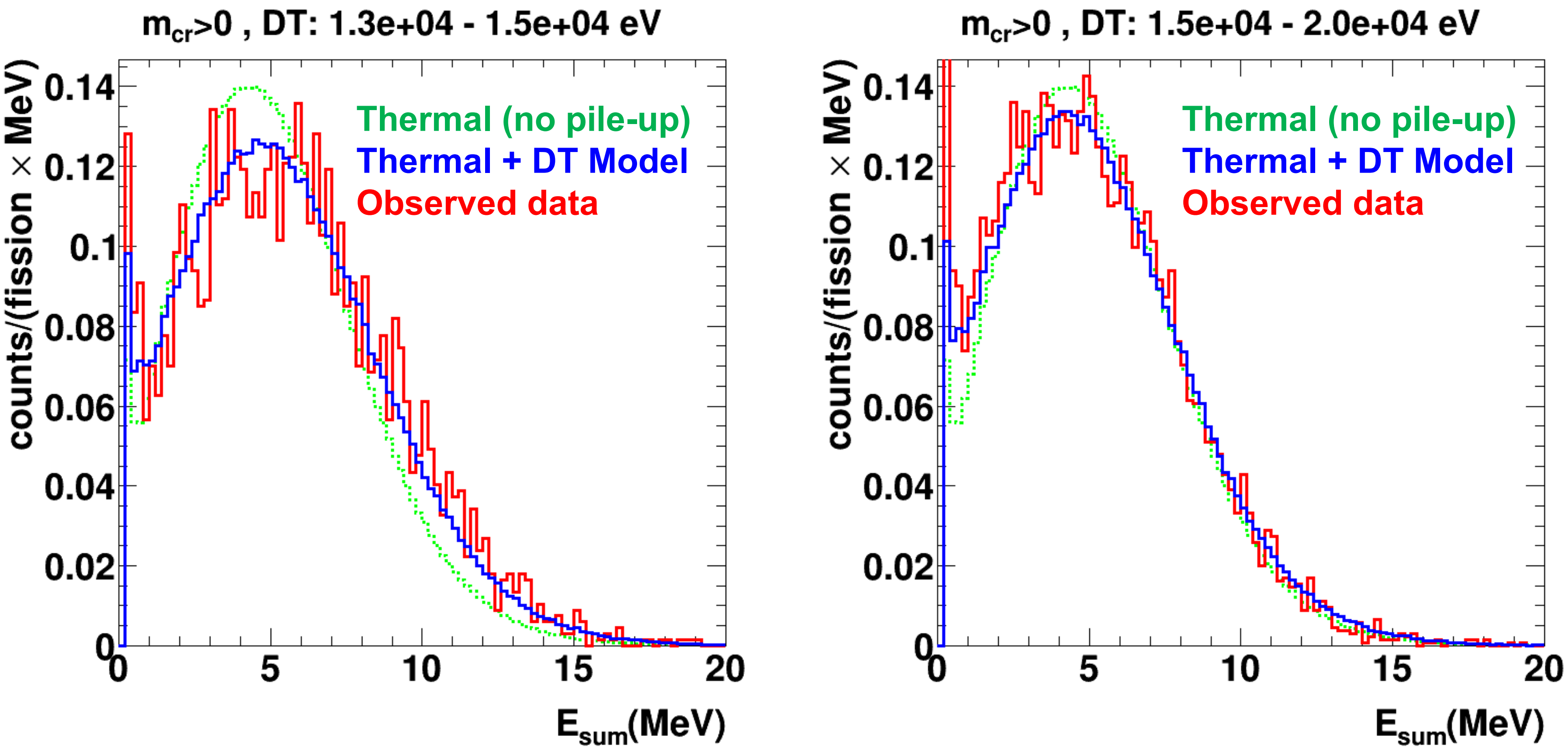
Evaluation of TOF DT model (2/2)

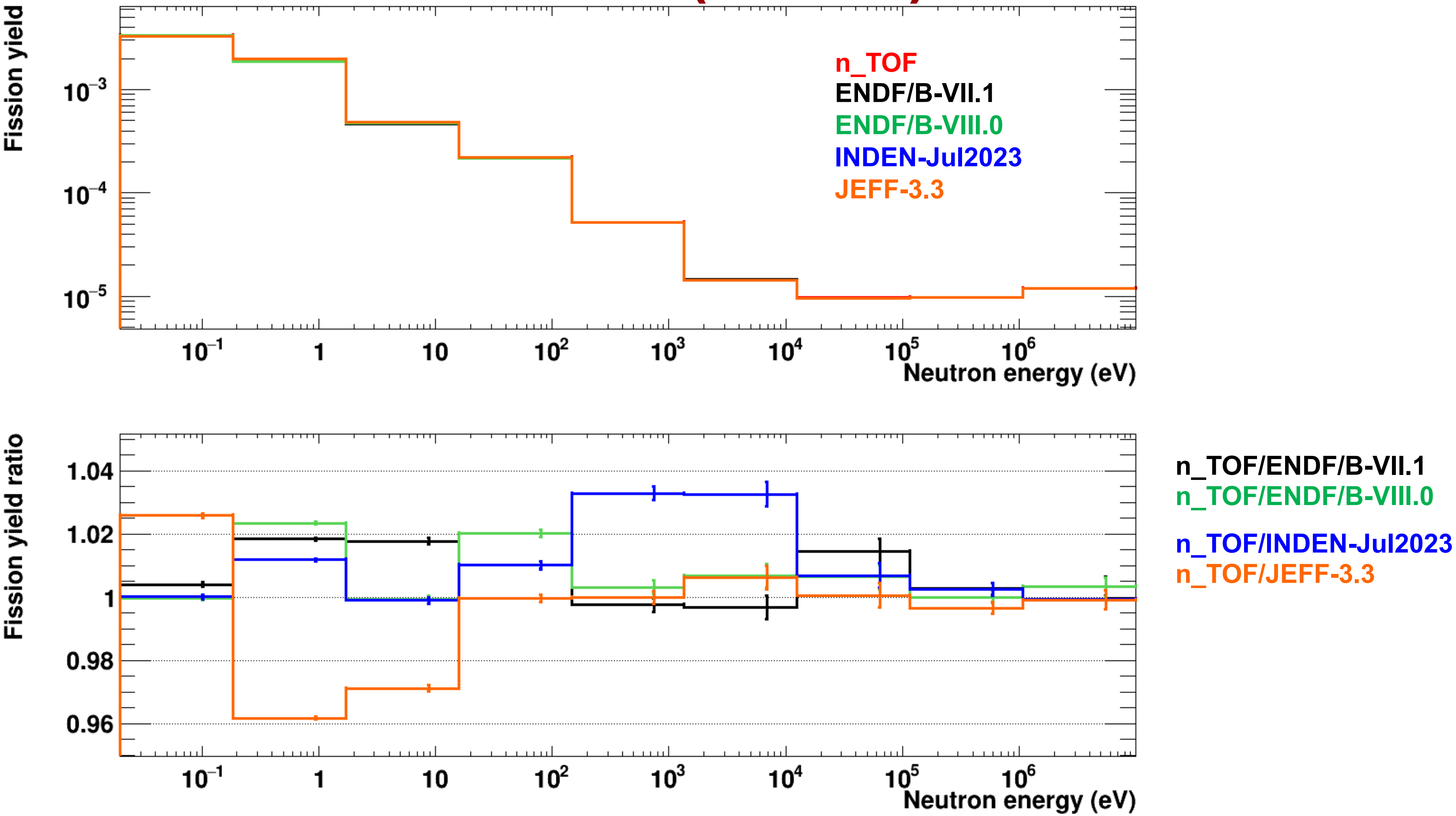
We can **validate** this Dead Time model using the **experimental data**.

The fission tagged E_{sum} spectrum



The prediction of the DT model agrees with observed data even for high neutron energies

Comparison with evaluations (1 BPD)



Conclusions

- Significant progress in the ^{239}Pu data analysis have been done in the last months, including a better understanding and characterization of the backgrounds for the capture measurement, determination and validation of dead-time and pile-up models, etc.
- Determination of the neutron flux for the ^{239}Pu measurement, including the boron concentration correction and the beam intersection factor calculation for a wide neutron energy range.
- These improvements allow us to provide a **^{239}Pu(n,f) yield** that agrees with evaluations within ~2% (integrating in 1 bin per decade) **from 0.02 eV to 10 MeV in one single measurement**.
- The fission yield is ready to be released (paper in preparation).
- The analysis of ^{239}Pu(n,γ) data is in progress.

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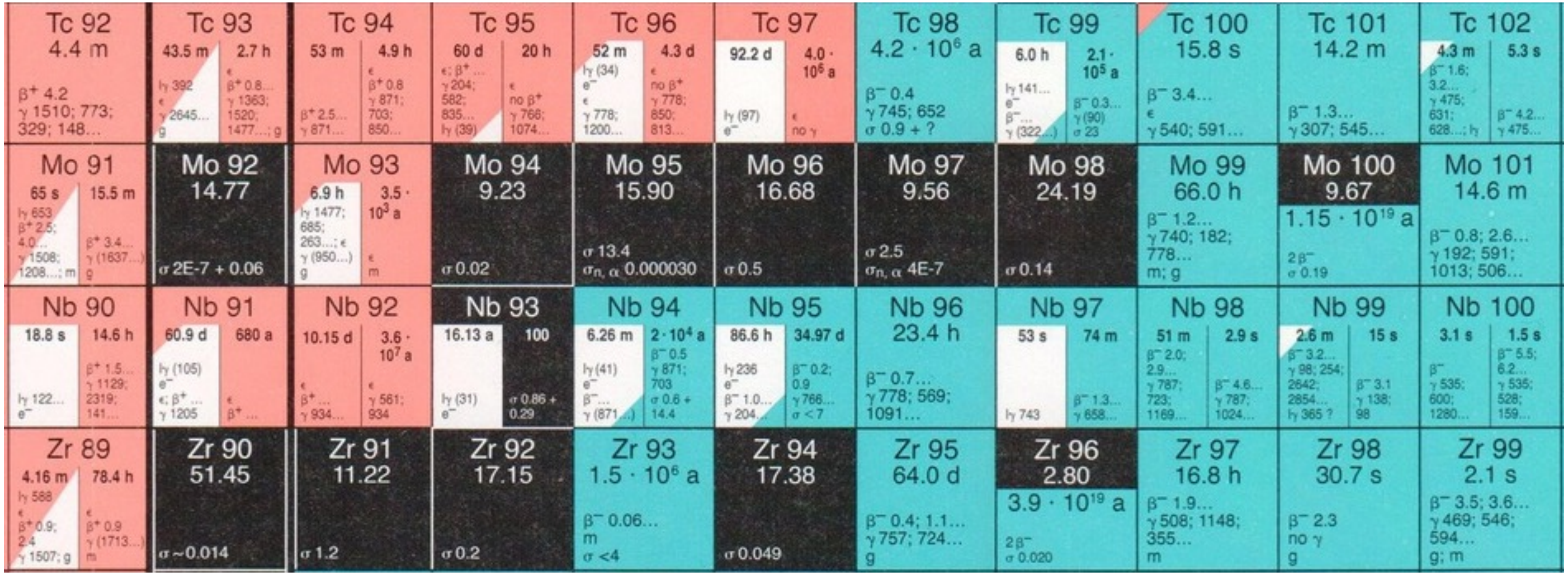
SANDA WP2/Task 2.2 report

^{94,95,96}Mo measurements

Molybdenum is relevant for **nuclear astrophysics** and **nuclear technology** and presently **known with large uncertainties**.



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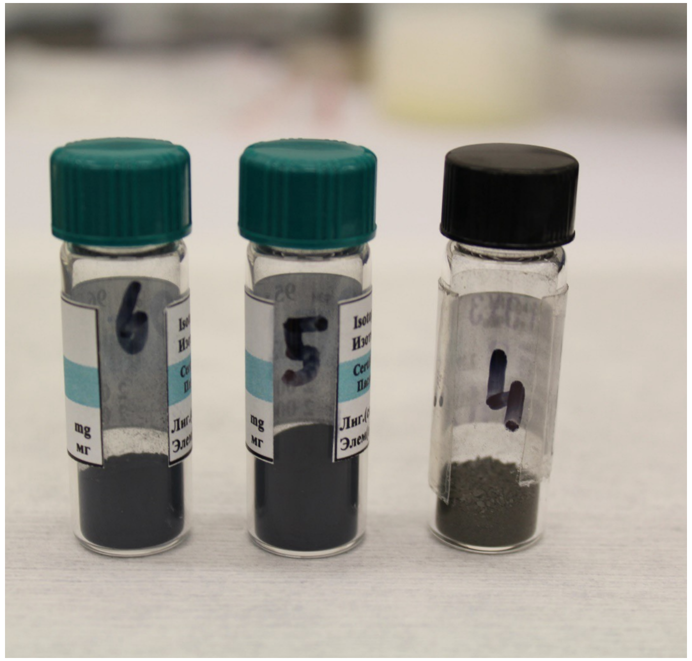
Enriched pellets preparation

To avoid the background coming from aluminum capsule three pressed pellets were prepared using enriched powder:

- Pellets prepared at JRC-Geel;
- Self sustaining pellets of ~ 2g;
- Additional ^{nat}Mo samples prepared using powder with different grain sizes;



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Mo pellet samples

Atomic %	^{92}Mo	^{94}Mo	^{95}Mo	^{96}Mo	^{97}Mo	^{98}Mo	^{100}Mo
^{94}Mo	0,63%	98,97%	0,36%	0,01%	0,01%	0,01%	0,01%
^{95}Mo	0,31%	0,69%	95,40%	2,24%	0,51%	0,65%	0,20%
^{96}Mo	0,28%	0,24%	1,01%	95,90%	1,00%	1,32%	0,25%

Isotope	Mass (mg)	Areal density (atoms/b)
^{94}Mo	1,952.6	3,9592E-03
^{95}Mo	1,974.5	3,9558E-03
^{96}Mo	1,917.5	3,8064E-03
^{nat}Mo-5 µm	2,014.0	4,0059E-03
^{nat}Mo-350 µm	1,989.0	3,9584E-03

Measurements

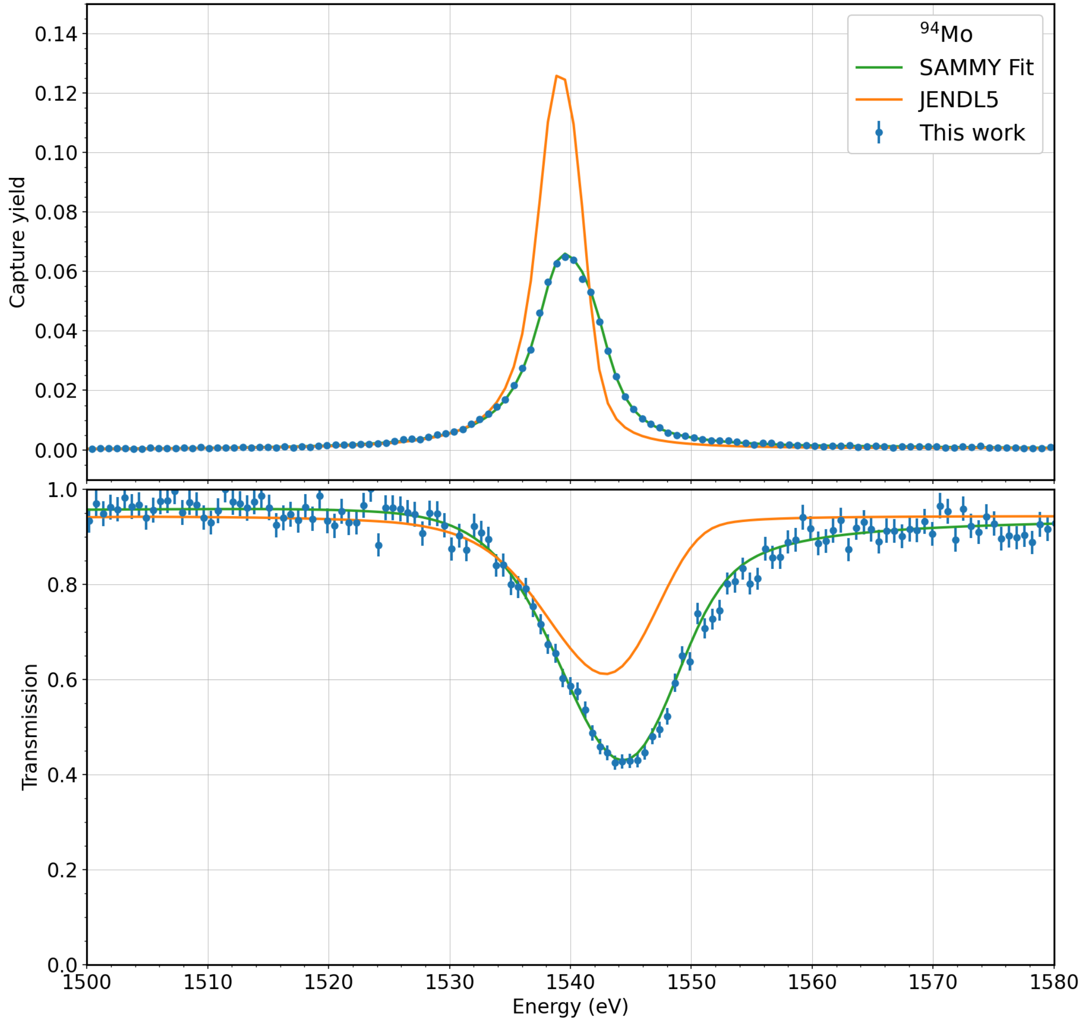
EAR2_2021	EAR1_2022	EAR2_2022
1.7 10^{18} protons	6.0 10^{18} protons	1.7 10^{18} protons
3 B6D6, 1 L6D6, 1 STED	4 C6D6	8 STED, 2 L6D6, 1 DSTI
Powder sample in aluminum canning	Pressed pellets in plastic bags	Pressed pellets in plastic bags

+ additional transmission measurement with enriched pellets at 10m station of GELINA

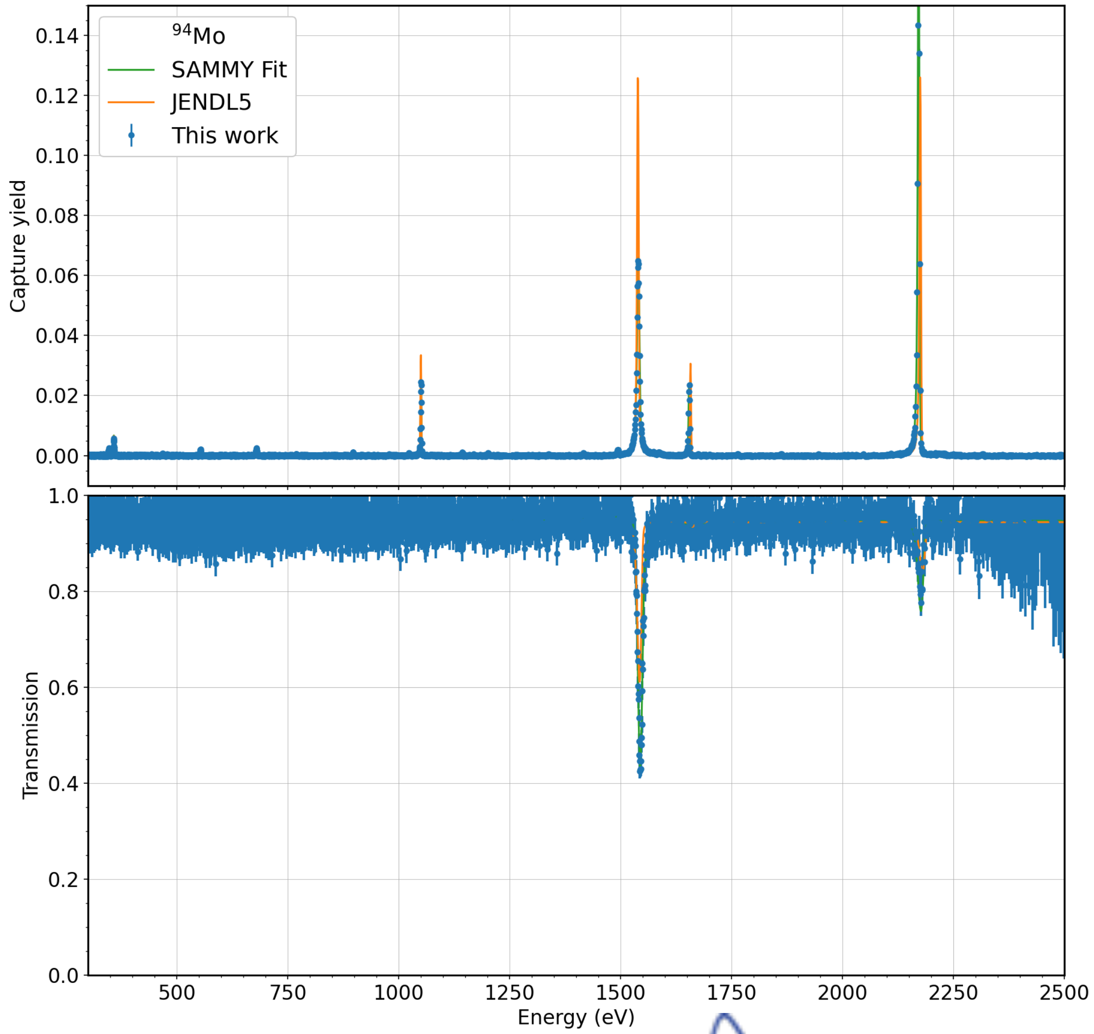
+ transmission measurements with natural samples at 50m station of GELINA

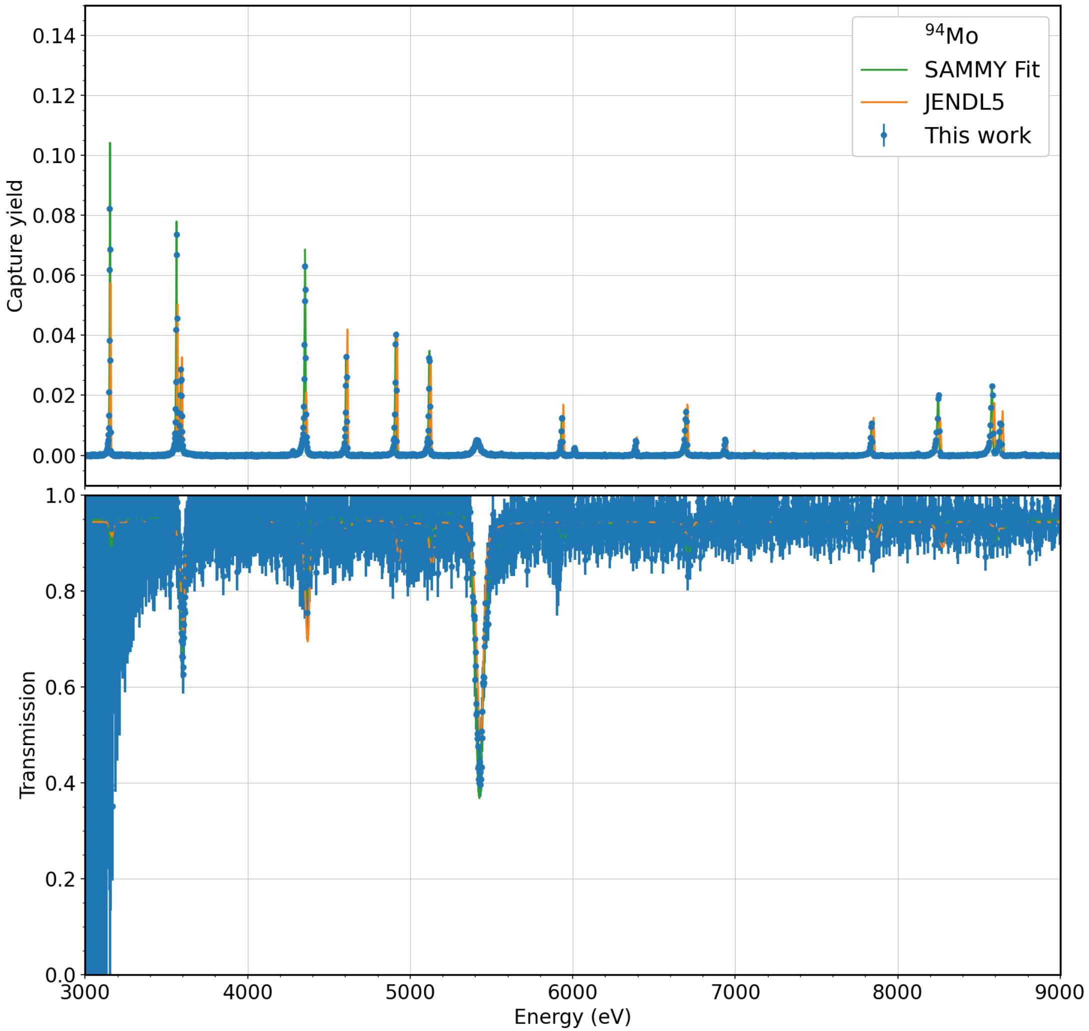
Resonance parameters ^{94}Mo

- Resonance parameters have been adjusted in all the resolved resonance region (<21 keV);
- Extended resolved resonance region up to 75 keV
- Example of fit showed here compared to the calculation performed with JENDL5 parameters
- Good agreement between transmission and capture data with enriched samples



Resonance parameters ^{94}Mo





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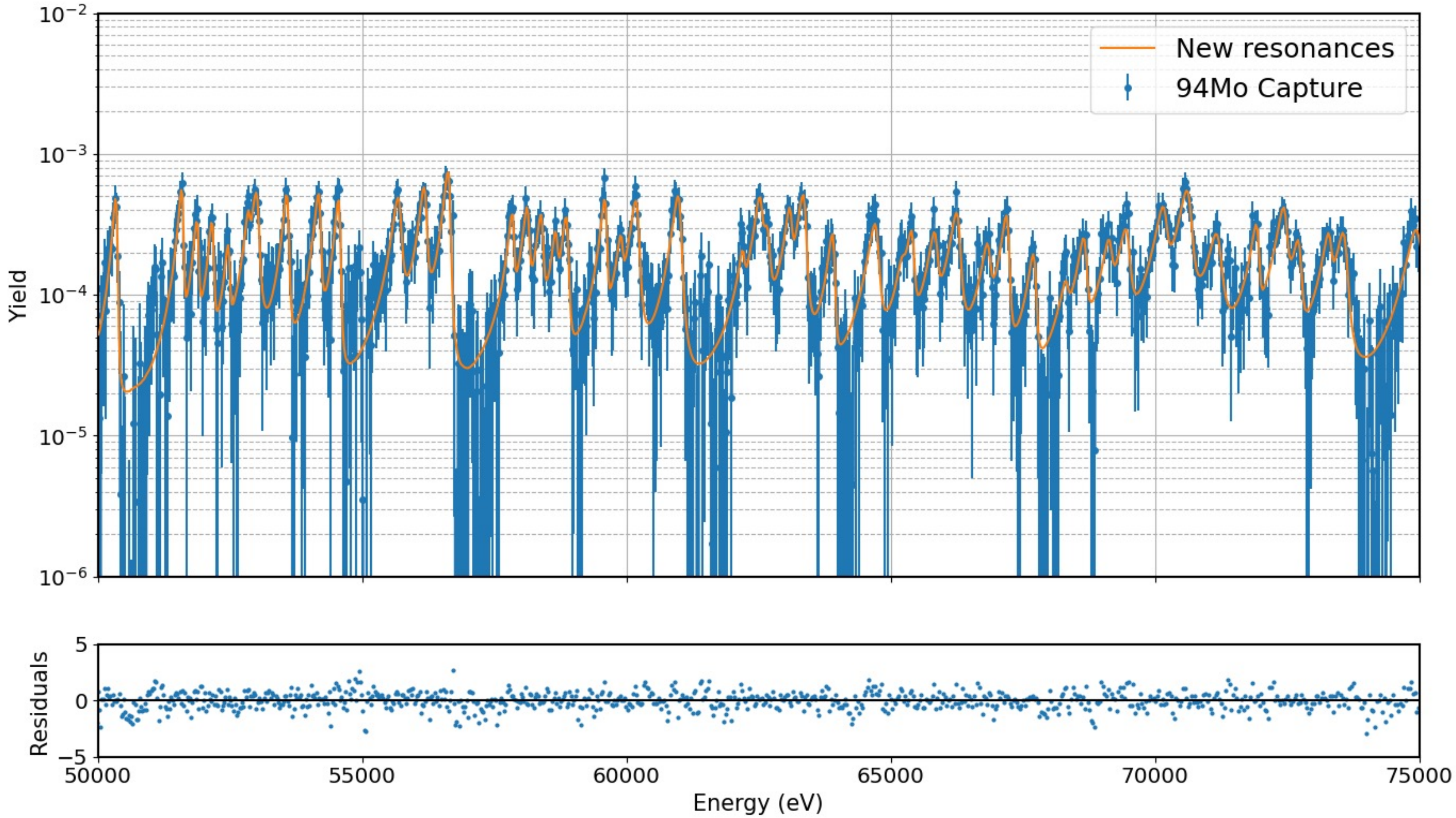
Resonance parameters ^{94}Mo

- Extended resolved resonance region up to 75 keV using data from capture measurements,
- New resonances not present in literature.



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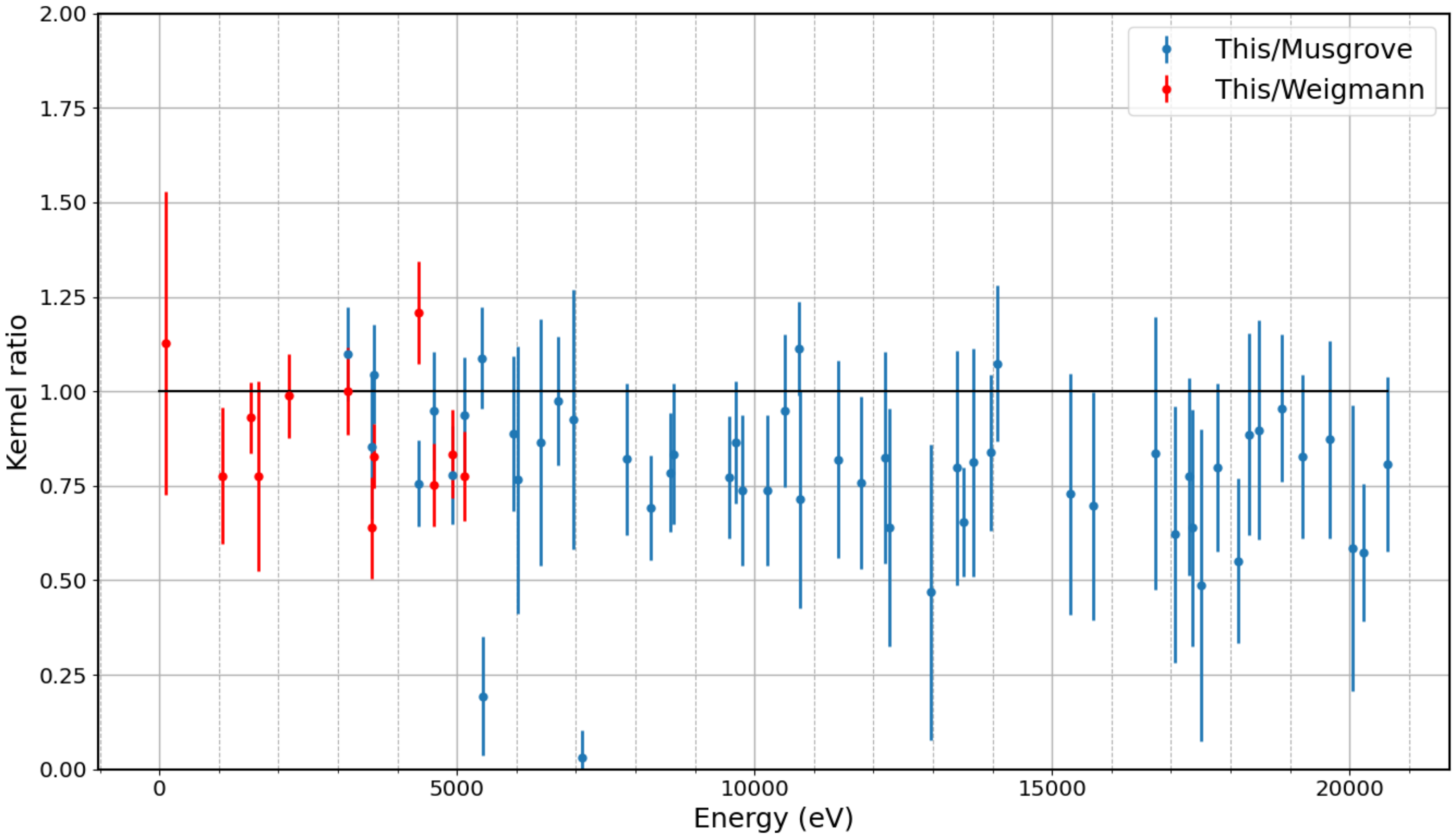
Resonance parameters ^{94}Mo

J	L	Energy (eV)	Unc_E	W_Capture (meV)	Unc_Cap	Width_ n (meV)	Unc_ n
-0.5	1	108.7365	2.29E-03	158.837	4.69049	0.180556	1.22E-03
-1.5	1	1051.963	1.48E-02	237.578	25.6533	2.35311	3.02E-02
0.5	0	1542.773	1.16E-02	124.952	0.568967	1673.86	8.59281
-1.5	1	1657.322	2.08E-02	169.781	30.3225	4.65519	6.62E-02
-1.5	1	2175.49	1.01E-02	159.592	1.06928	340.652	4.81211
⋮							
-1.5	1	9576.481	0.109357	122.857	2.46143	673.324	68.231
0.5	0	9689.416	0.184379	98.0503	2.40078	2383.27	162.983
-1.5	1	9797.066	0.132802	95.4524	7.68889	230.418	44.3515

Capture kernels for ^{94}Mo



- The preliminary kernels obtained with SAMMY were compared to the ones in literature (Weigmann and Musgrove capture measurements);
- Main measurements used in libraries;
- Systematic deviation of around 20% observed

Presentation and article

- Journal articles
  - *R. Mucciola et al., Evaluation of resonance parameters for neutron interactions with molybdenum, NIMB* ***531*** *(2022) 100*
  - *R. Mucciola et al., Neutron capture and total cross-section measurements on ^{94,95,96}Mo at n_TOF and GELINA, EPJ Web of Conferences* ***284****, 01031 (2023)*
- Contributions to seminar and conferences
  - *Nuclei in the cosmos XVII, 17-22 September 2023, Daejeon (Korea). Poster presentation*
  - *GIANTS XI, 20-21 October 2022, Caserta (Italy). Invited talk*
  - *ND 2022, 24-29 July 2022, Sacramento (USA). Oral presentation*
  - 13th Torino Workshop on AGB stars, 19-24 June 2022, Perugia (Italy). Oral presentation

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Overall status of the ^{94,95,96}Mo measurements

	Transmission measurement	Capture measurement	Transmission data analysis	Capture data analysis
Mo-94	Performed at 10 m station GELINA	Performed at EAR1 and EAR2 n_TOF	Preliminary resonance parameters	EAR1 preliminary resonance parameters, EAR2 analysis ongoing
Mo-95	Performed at 10 m station GELINA	Performed at EAR1 and EAR2 n_TOF	Transmission spectra obtained	EAR1 yield obtained, EAR2 analysis ongoing
Mo-96	Performed at 10 m station GELINA	Performed at EAR1 and EAR2 n_TOF	Transmission spectra obtained	EAR1 yield obtained, EAR2 analysis ongoing

SANDA WP2/Task 2.2 report summary 1/2

^{239}Pu(n,γ) neutron capture cross section and α ratio

- ✓ proposal for measurement at n_TOF submitted and approved by the CERN Research Board (December 2020)
- ✓ sample preparation procedure agreed between CERN and JRC-Geel

- The ionization chamber that is used for these measurements will be tested at JRC-Geel during 2021
- Measurements to be performed at n_TOF during 2022

SANDA WP2/Task 2.2 report summary 1/2

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SANDA WP2/Task 2.2 report summary 2/2

$^{94,95,96}Mo(n,\gamma)$ neutron capture and total cross section measurements

- ✓ proposal for measurement at n_TOF submitted and approved by the CERN Research Board (December 2020)
- ✓ sample orders issued to Neonest AB, Sweden. Expected delivery Q1-2021

- Total cross section measurements to be performed at JRC-Geel (GELINA) during 2021
- Measurements to be performed at n_TOF during 2022

SANDA WP2/Task 2.2 report summary 2/2

^{94,95,96}Mo(n,γ) neutron capture and total cross section measurements

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The END

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SANDA GENERAL MEETING
PROGRESS OF IRSN ON WP2&4 WORK PACKAGE
9-11 FEBRUARY 2021

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Working package WP2 & 4 – Measurement and assessment of nuclear data

ASSESSMENT OF MOLYBDENUM NUCLEAR DATA

- Target: produce Mo evaluation with covariance for RP and cross sections
  - JEFF-33, ENDF/B-VIII.0: covariance for cross sections BUT no covariance for RP
  - JENDL-4.0: no covariance for RP and cross sections
- Resonance parameters retrieved from JEFF-3.1.1, JEFF-3.3, JENDL-4.0, ENDF/B-VII.1 and ENDF/B-VIII.0 evaluations
- Identification of available differential measurements in EXFOR
  - Lack of data for ^{95}Mo
- Use of JENDL-4.0 resonance parameters to initiate the evaluation process
  - Correspondence between spin groups and channel spins addressed
- Transmission and capture measurements of ^{nat}Mo at J-PARC (Japan) on various samples
  - ANNRI experimental device
  - 0.1, 0.5, 2 mm thick for capture and 0.5, 5 mm thick for transmission

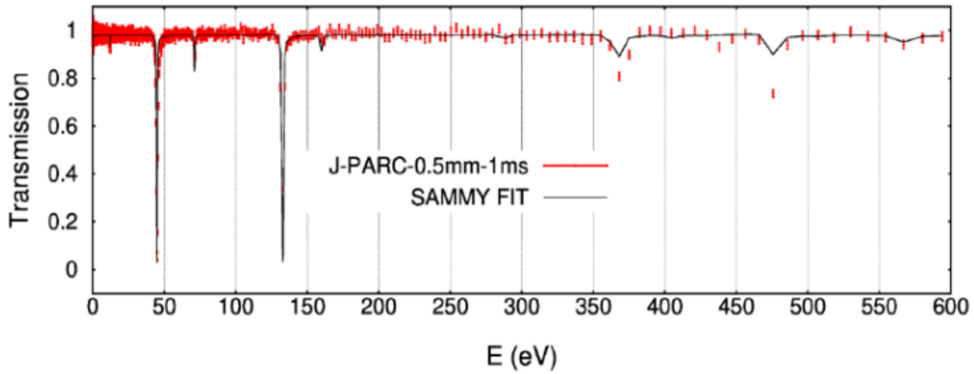
Isotope	Composition (%)	Thermal Cross Section (barns)	Resonance Integral (barns)
^{92}Mo	14.84	0.08±0.02	0.83
^{94}Mo	9.25	0.34±0.02	1.12
^{95}Mo	15.92	13.4±0.3	118±7
^{96}Mo	16.68	0.5±0.3	17±3
^{97}Mo	9.55	2.2±0.2	14.4±3.0
^{98}Mo	24.13	0.130±0.006	6.7±0.3
^{100}Mo	9.63	0.199±0.002	3.76±0.15

Library	Lower limit RR (eV)	Upper limit RR (eV)	Lower limit URR (eV)	Upper limit URR (eV)
JEFF-3.3	0	2141.2	0	206269
ENDF/B-VIII.0	0	2141.2	0	206269
JENDL-4.0	0	2000.0	0	400000

Working package WP2 & 4 – Measurement and assessment of nuclear data

ASSESSMENT OF MOLYBDENUM NUCLEAR DATA

- Fit of experimental data with SAMMY code (R-matrix)
  - Preliminary evaluation: sequential fit
    - Resolved resonance description
      - Use of $\chi 2$ as figure of merit
      - Generation of covariance matrix
      - Updated RP and RP covariance data at each step
  - Use of 0.5 mm sample in 0-600 eV
    - Low data resolution above 350 eV



- n_TOF and GELINA measurements of enriched Mo are planned/underway.

- Additional data: a) RPI transmission data for isotopes (^{95}Mo, ^{96}Mo, ^{98}Mo, ^{100}Mo); b) Transmission and capture data for ^{95}Mo from LANL

➡ **Need of enriched samples**