

Neutron capture and total cross measurements on $^{94,95,96}\text{Mo}$ at n_TOF and GELINA

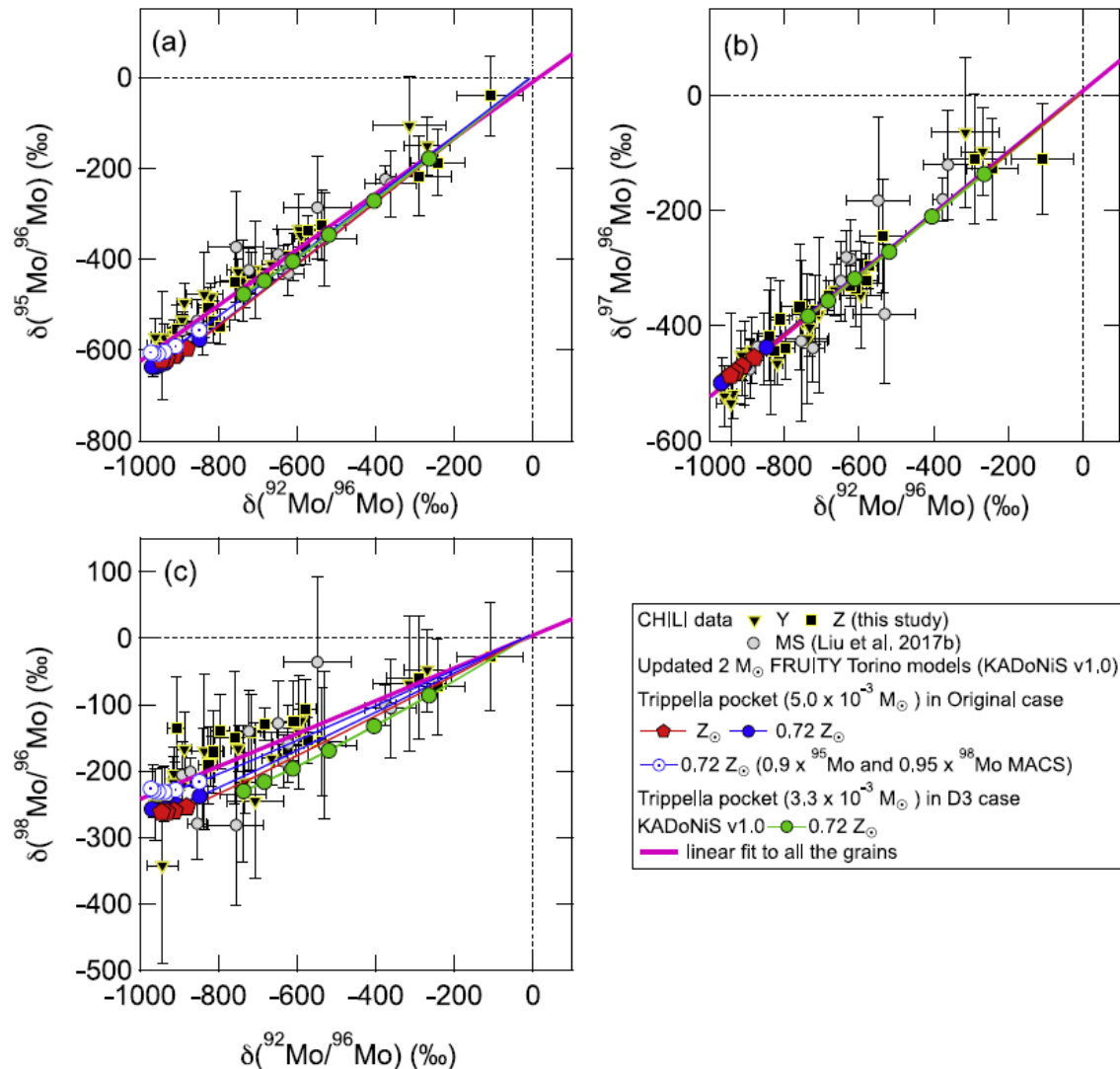
RICCARDO MUCCIOLA

Importance of molybdenum



- Fission product in nuclear power plants;
- Nucleosynthesis of heavy elements: pollution in presolar SiC grains;
- Transport casks, irradiated fuel storage;
- Research reactors and Accident Tolerant Fuels.

Presolar grain composition



- Comparison of SiC grains composition versus stellar model (FRUITY Torino model)
- MACS form KADoNiS v1.0
- Slight discrepancy between model and isotopic composition
- Possible overestimation of MACS in KADoNiS.

N. Liu, et al., ApJ 881 (2019) 28.

SANDA project

SANDA WP2:

Task 2.2: Neutron capture cross sections

Subtask 2.2.1. Capture measurements of fissile isotopes

Combined measurement of the $^{239}\text{Pu}(n,\gamma)$ and $^{239}\text{Pu}(n,f)$ cross sections at GELINA and n_TOF.

Subtask 2.2.2. Capture measurement of stable isotopes

$^{92,94,95}\text{Mo}(n,\gamma)$ cross sections at GELINA and n_TOF .

Objective of experiments

Improve capture cross section accuracy for neutron energies from thermal (10 meV) to hundreds keV

Submit results to EXFOR to improve nuclear data libraires (ENDF, JEFF, JENDL ecc.)

Experimental campaigns

Transmission measurement

- Carried out at GELINA
- Total cross section measurement
- Natural and enriched samples
- 10m and 50m flight path

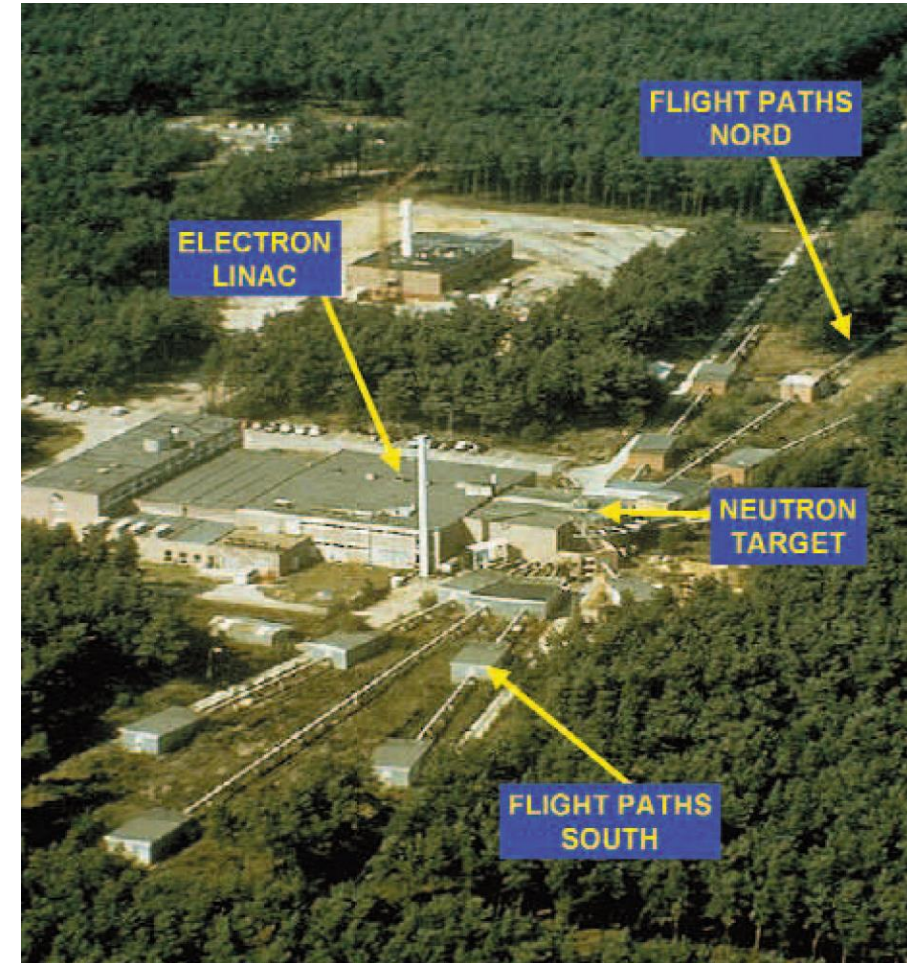
Radiative capture measurement

- Carried out at GELINA and n_TOF
- Neutron capture cross section
- Both experimental areas of n_TOF
- 10m station of GELINA

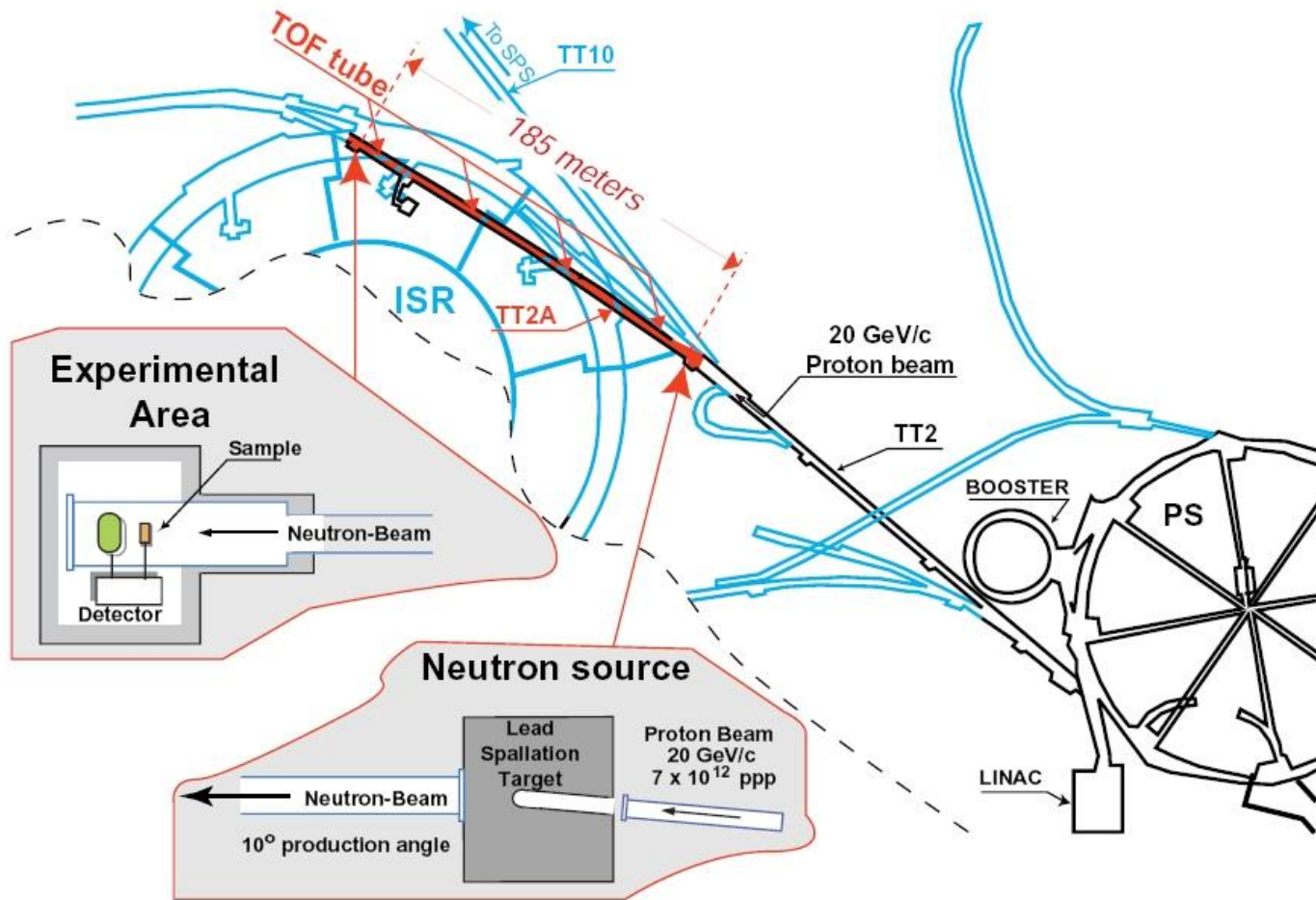
Facilities and technique

GELINA

- Located at JRC-Geel
- Multi-user time-of-flight facility
- Electron beam produced by LINAC (E = 140 MeV)
- Rotating uranium target
- Production of neutrons via (γ,n) or (γ,f)
- Pulsed neutron source (10 meV < E < 20 MeV)
- Water moderators



n_TOF



- Located at CERN
- Neutron beam produced using PS proton on lead target
- Production of neutrons via spallation
- Pulsed neutron source ($10 \text{ meV} < E < 1 \text{ GeV}$)
- Three experimental areas (EAR1, EAR2 and NEAR)

Time-of-flight technique

$$E_n = mc^2(\gamma - 1) \approx \frac{1}{2}mv^2$$

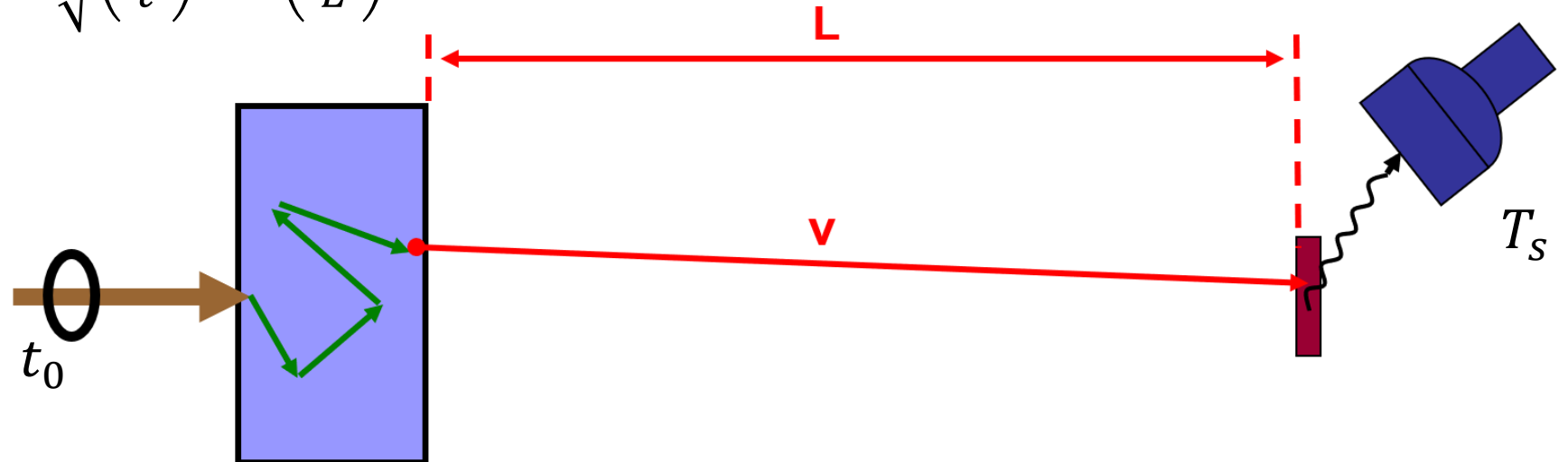
$$v = \frac{L}{t}$$

Flight path

Time-of-flight

$$t = (T_s - t_0) - (t_\gamma - L/c)$$

$$\frac{\Delta E}{E} = (1 + \gamma)\gamma \frac{\Delta v}{v} \approx 2 \frac{\Delta v}{v} = 2 \sqrt{\left(\frac{\Delta t}{t}\right)^2 + \left(\frac{\Delta L}{L}\right)^2}$$



Experimental measurements

Transmission

Percentage of neutrons that traverses a samples without interacting with it

- Related to total cross section:

$$T = N \frac{C_{in}(t) - KB_{in}(t)}{C_{out}(t) - KB_{out}(t)} = \frac{\varphi_n e^{-n\sigma_{tot}}}{\varphi_n} = e^{-n\sigma_{tot}}$$

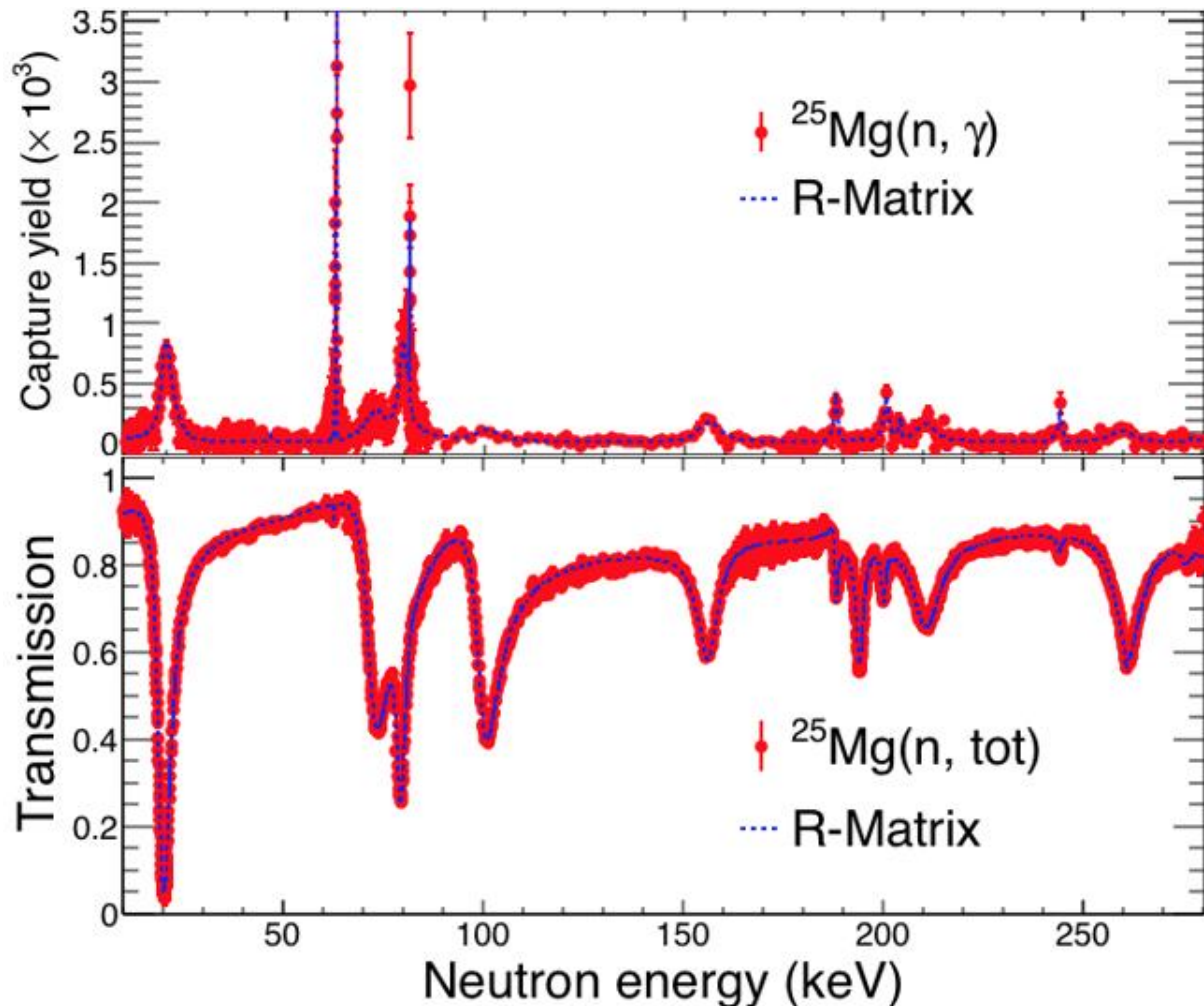
Radiative capture (capture yield)

Percentage of neutrons that undergoes capture reaction in the sample

- Related to capture cross section via:

$$Y_{exp} = N \frac{C_\gamma(t) - B_\gamma(t)}{C_\varphi(t) - B_\varphi(t)} Y_\varphi = (1 - T) \frac{e^{-n\sigma_\gamma}}{e^{-n\sigma_{tot}}}$$

Resonance Shape Analysis



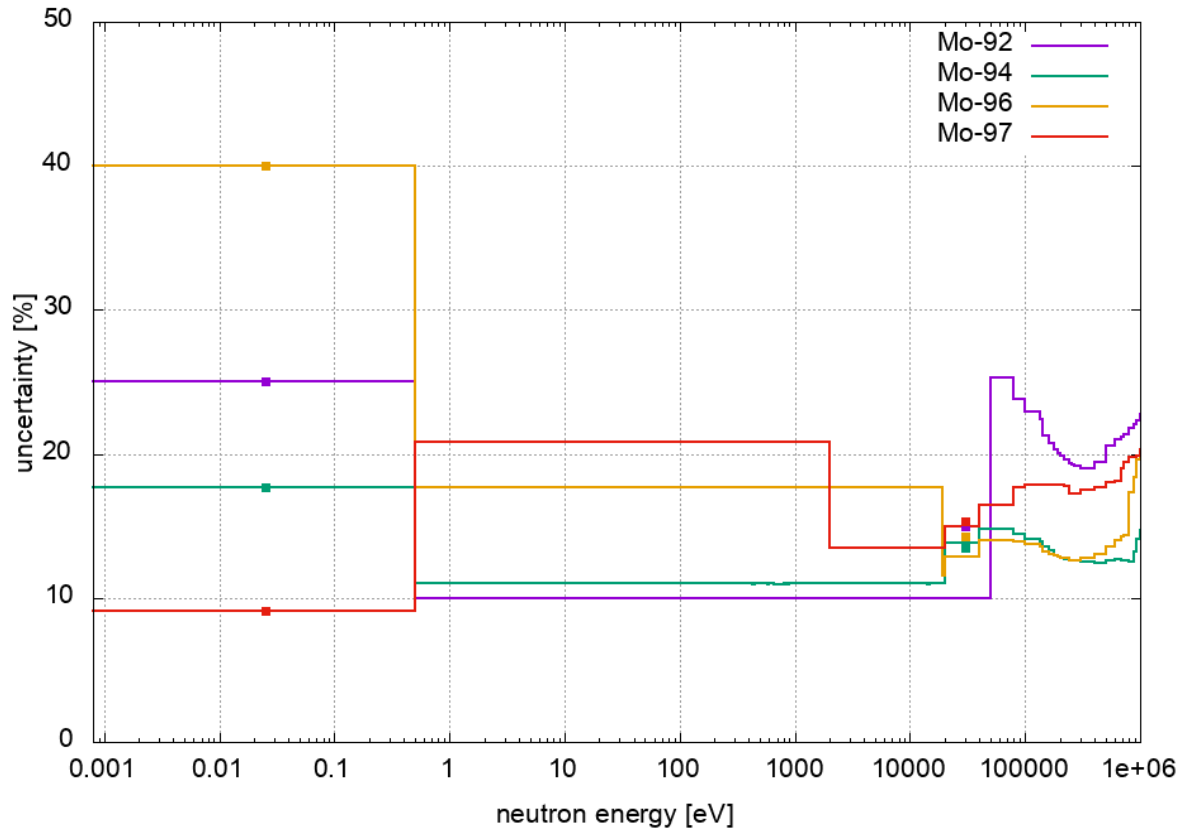
- Determination of the resonance parameter $E_0, \Gamma_\gamma, \Gamma_n$
- Simultaneous fit of transmission and capture data
- Fit performed using R-Matrix formalism

Parametrization of cross section using resonance parameters

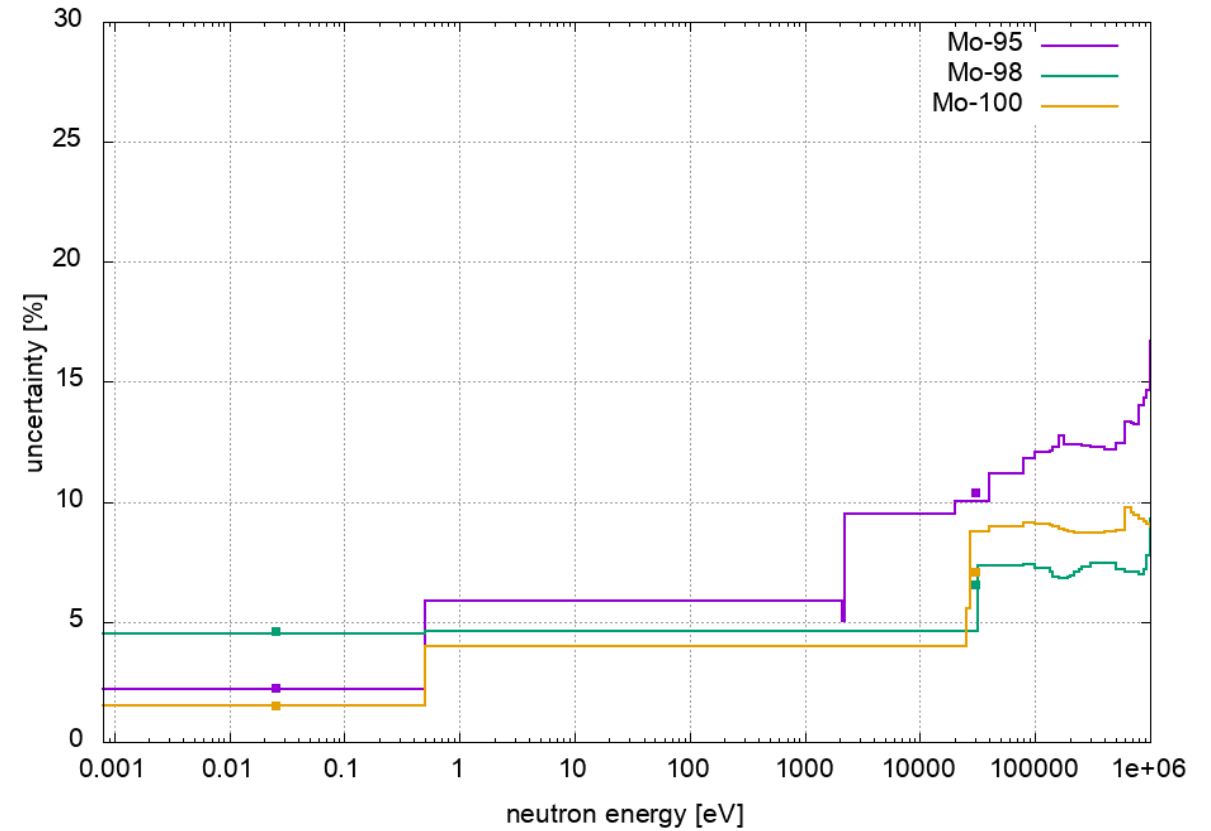
Resonance parameters evaluation

Cross section uncertainties in ENDF/B-VIII

Capture cross section uncertainties - ENDF/B-VIII.0 data set



Capture cross section uncertainties - ENDF/B-VIII.0 data set



Improved RP for $^{94,95,96}\text{Mo}$, natMo

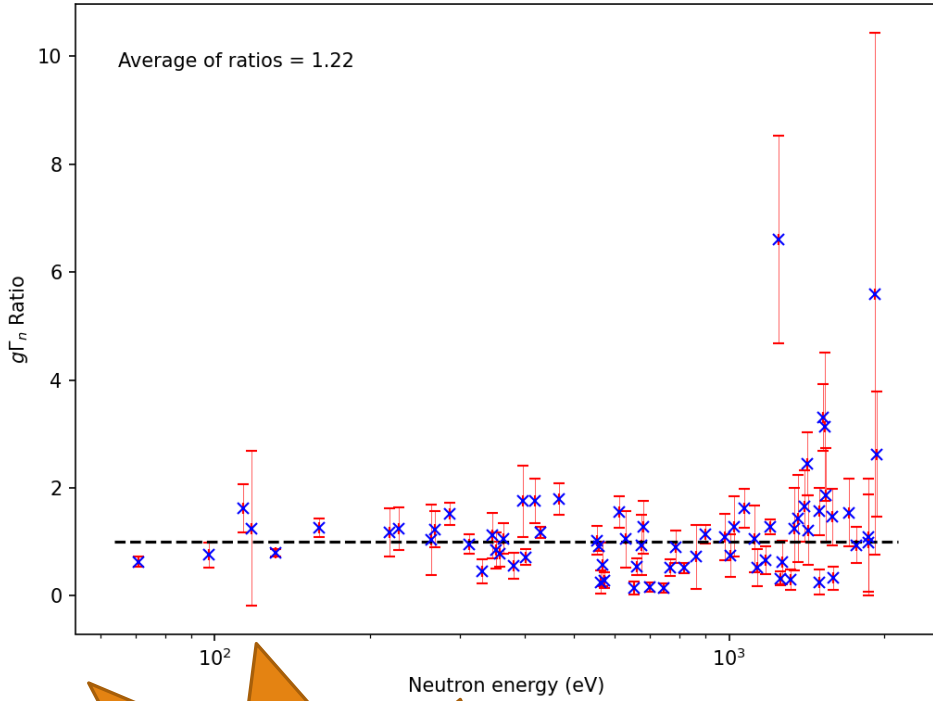
- 1) Study transmission and capture data for Mo reported in the literature:
 - compilation of resonance parameters based on these data
 - 2) Transmission cross section measurements using $^{\text{nat}}\text{Mo}$ samples at 50m GELINA:
 - adjust the compiled resonance parameter file by RSA with REFIT
 - 3) Experiments with enriched $^{94,95,96}\text{Mo}$ samples:
 - Transmission and capture measurements at GELINA
 - Capture measurements at n_TOF
- Final resonance parameter file by a simultaneous analysis of GELINA and n_TOF data

Mo literature study

Transmission			Capture		
Wang	^{nat}Mo	POHANG (<200 eV)	Weigmann	^{nat}Mo	GELINA (<25 keV)
Pevzner	$^{92,94,95,96,97,98,100}\text{Mo}$	DUBNA (<10 keV)	Weigmann	$^{92,94,95,96,97,98,100}\text{Mo}$	GELINA (<5 keV)
Wynchank	^{nat}Mo	Columbia Univ. (<5 keV)	Musgrove	$^{92,94,95,96,97,98,100}\text{Mo}$	ORELA (>3keV)
Shwe	$^{95,97}\text{Mo}$, ^{nat}Mo	Argonne (<1.5 keV)	Wasson	^{92}Mo	ORELA (<30 keV)
Chrien	^{98}Mo	ORELA (<50 keV)			
Babich	^{98}Mo	90m chopper (<2.5 keV)			
Leinweber	^{nat}Mo	RPI (<2 keV)			
Wasson	^{92}Mo	ORELA (<30 keV)			
Weigmann	^{100}Mo	ORELA (<4keV)			

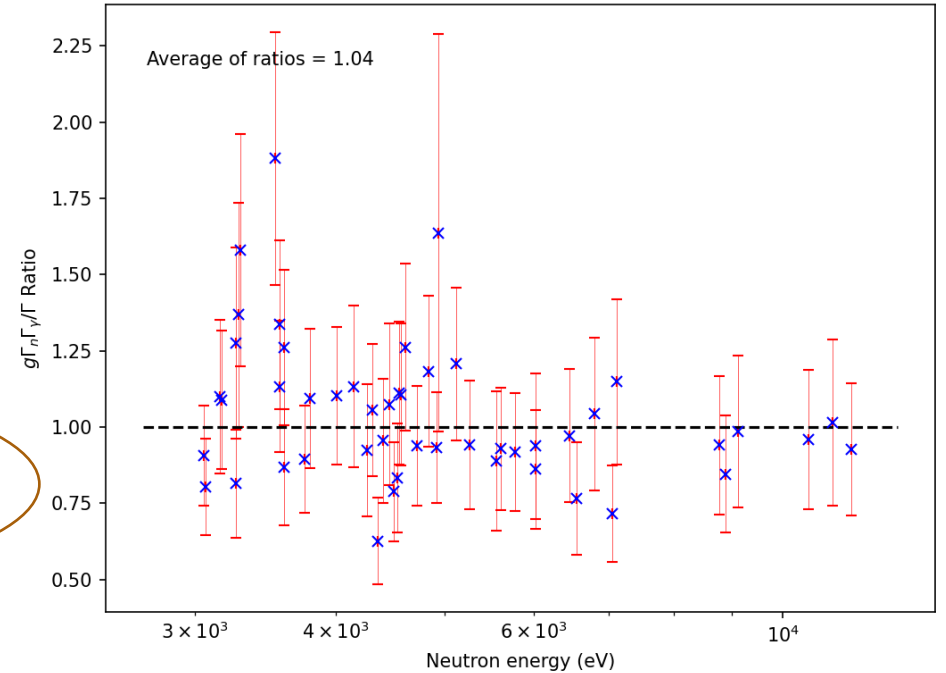
Mo literature comparison

Leinweber-Wynchank



Ratio of neutron width for transmission

Weigmann-Musgrove



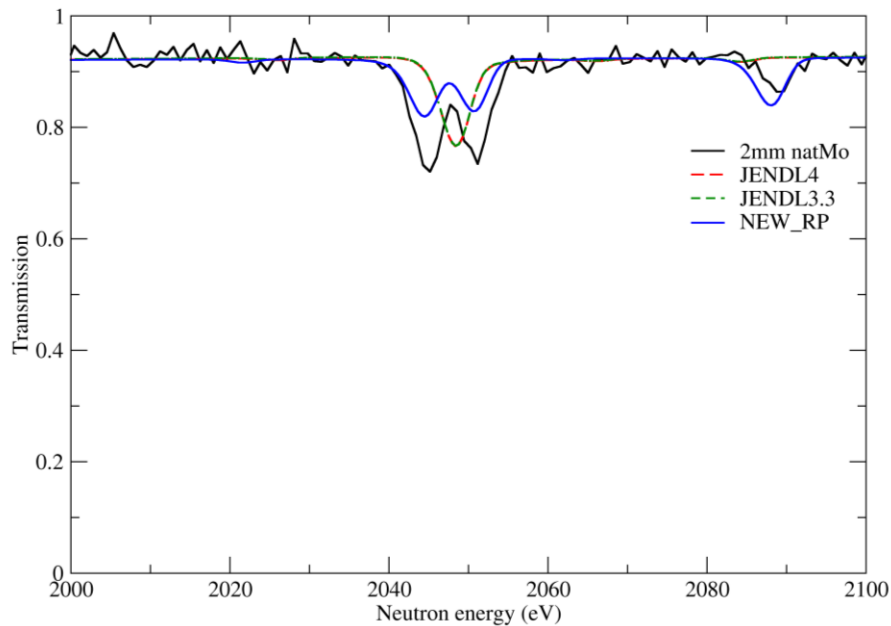
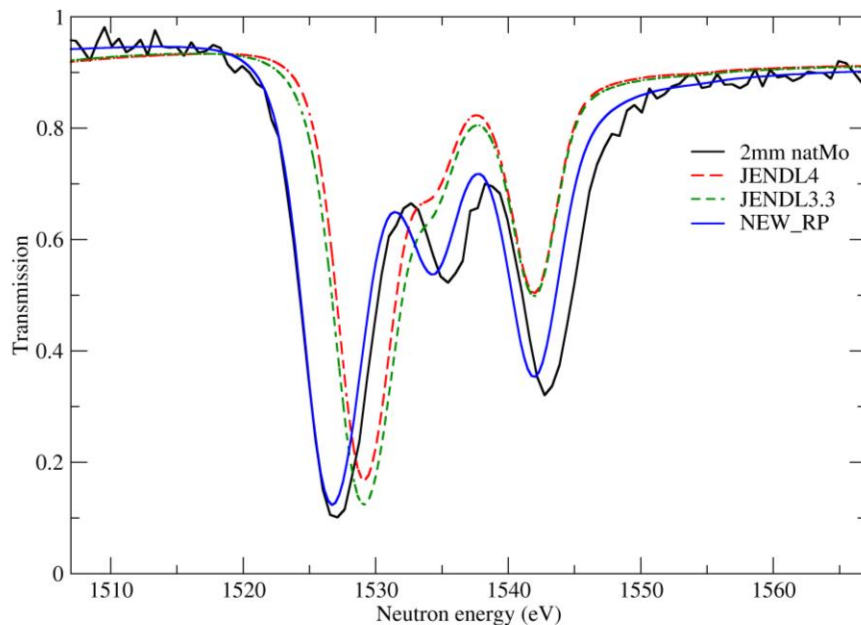
Ratio of resonance kernel for capture

Check parameters consistency

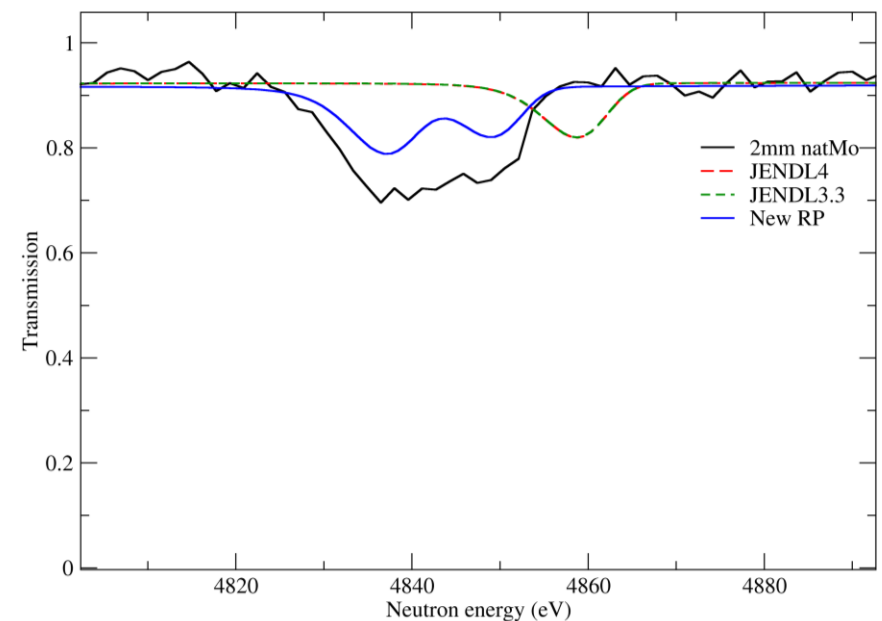
RP compilation from literature

- 1) Define consistent energy scale:
Weigmann et al. (capture experiments at GELINA)
 - 2) Select $g\Gamma_n$ reference:
E < 2keV: Leinweber
E > 2keV: Whynchank
 - 3) Select $\frac{g\Gamma_\gamma\Gamma_n}{\Gamma}$ reference:
Weigmann
Musgrove for odd isotopes and E>3keV
- Compilation of RP file from literature data
 - ^{nat}Mo transmission measurements at GELINA to validate and improve RP file

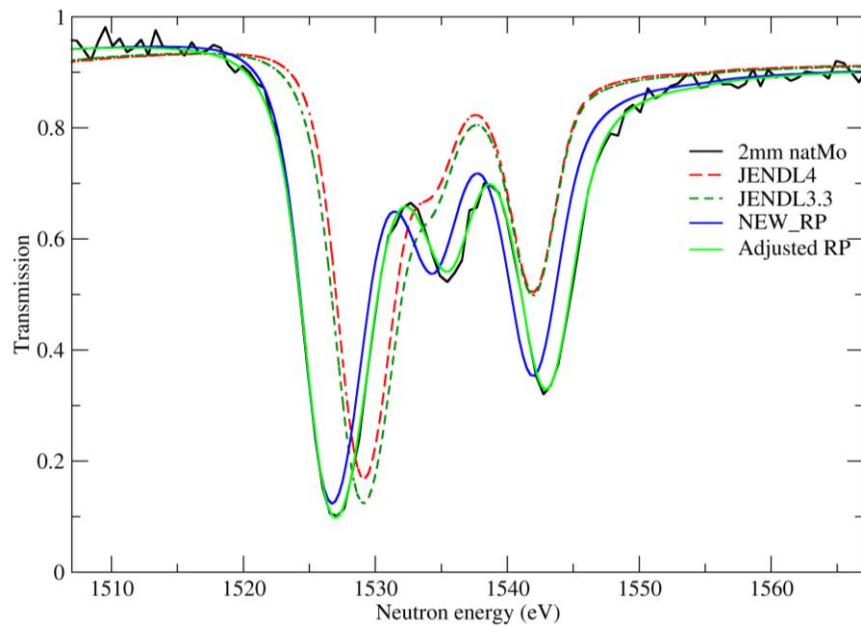
Validation of compiled RP file



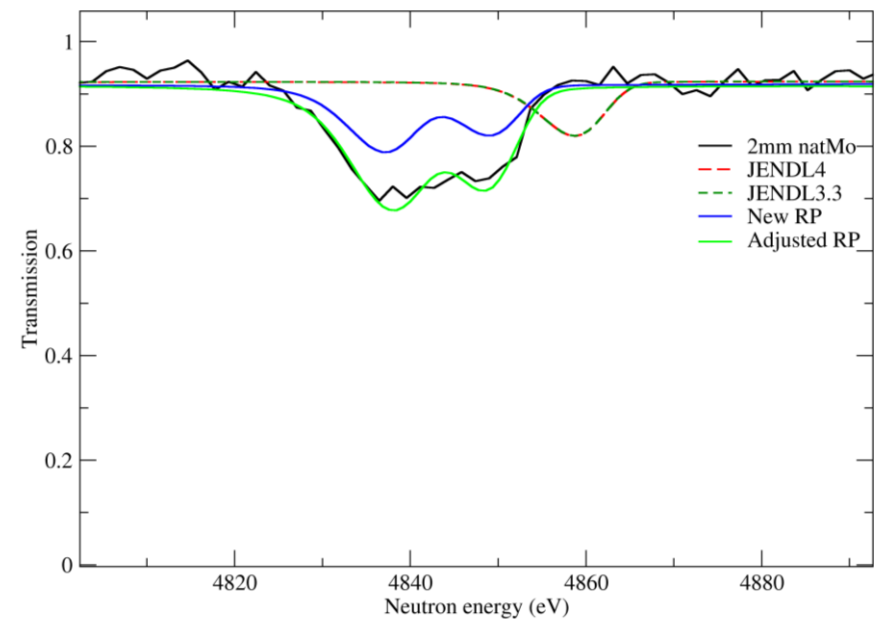
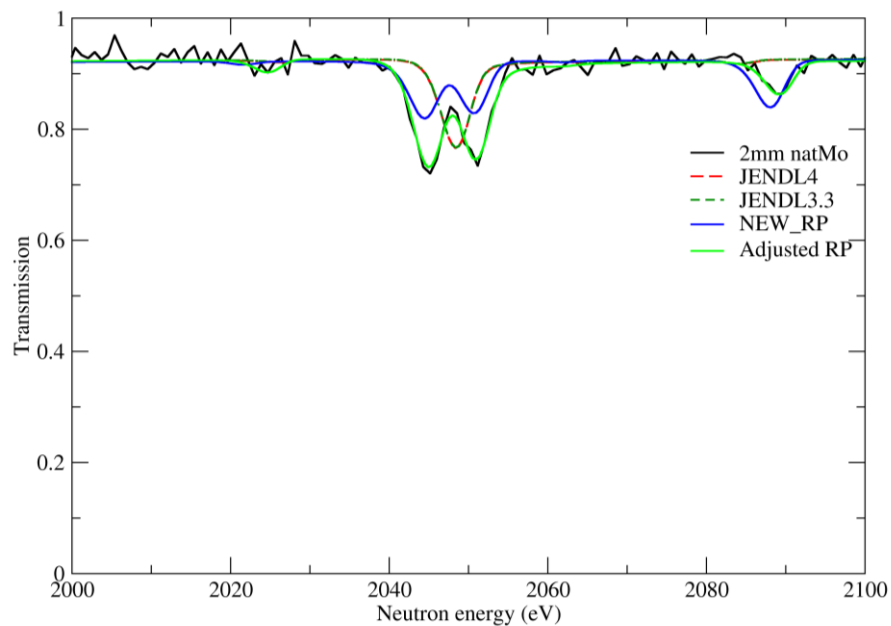
- RP file verified by transmission data (50 m) of 2mm and 5mm thick ^{nat}Mo samples
- Missing resonances in libraries reported in literature data
- Literature parameters more consistent with transmission data
- **New RP file improve data description.**



Improvement of RP file



- RP file improved by an adjustment to transmission data using REFIT
- Fit of resonances up to 5 keV
- Final paper ready for submission



Dissemination of results

- Results of resonance parameters compilation submitted for publication
- EXFOR submission under preparation
- Collaboration with additional experiment performed in another facility

Evaluated data file will be proposed for new version of JEFF!

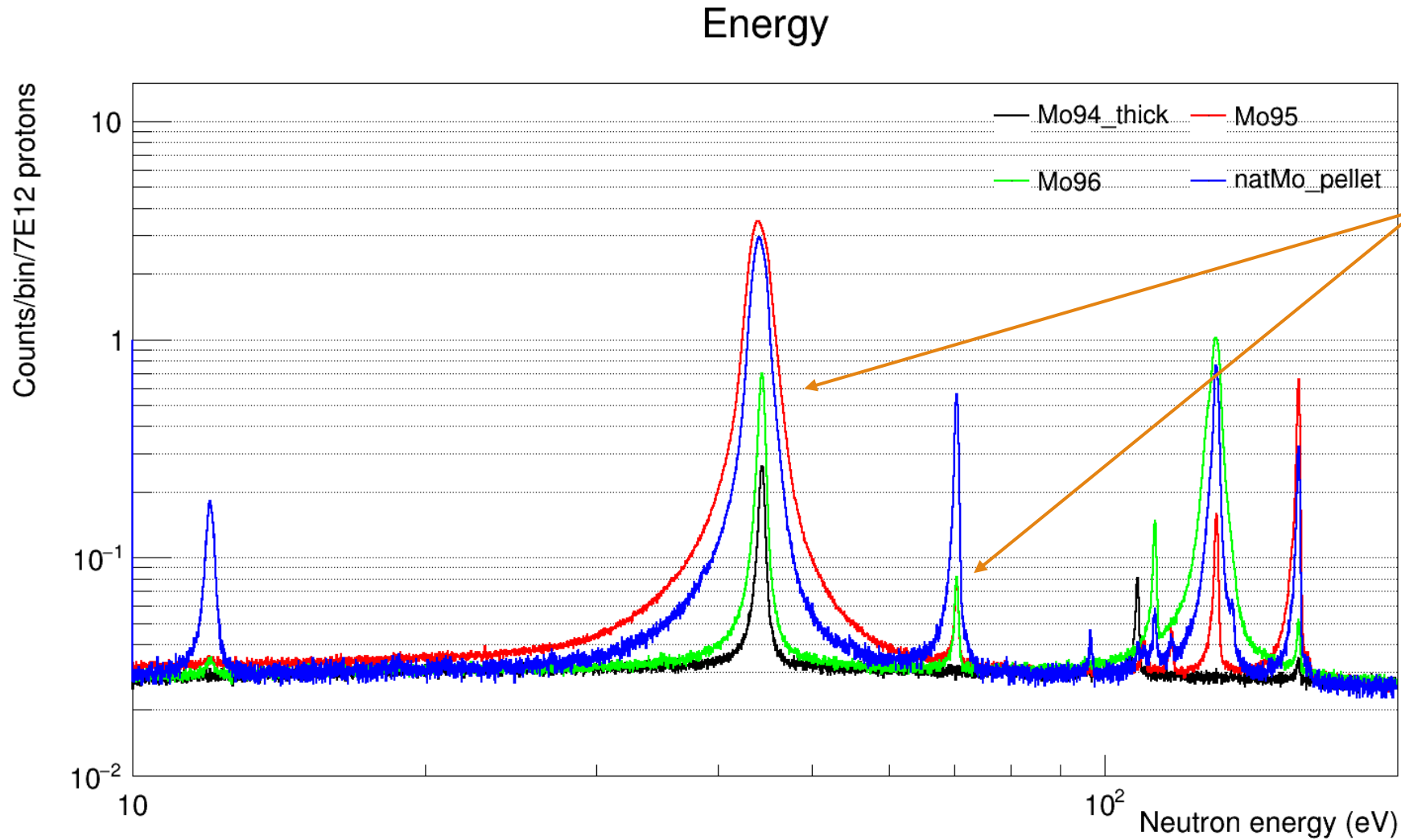
Enriched samples campaign

Mo powder @ n_TOF

- Metallic powder in metallic capsules;
- Capsule fixed to mylar disk using Kapton foil;
- 2g of powder available for each isotope;
- Capture measurements performed at n_TOF in October 2021.



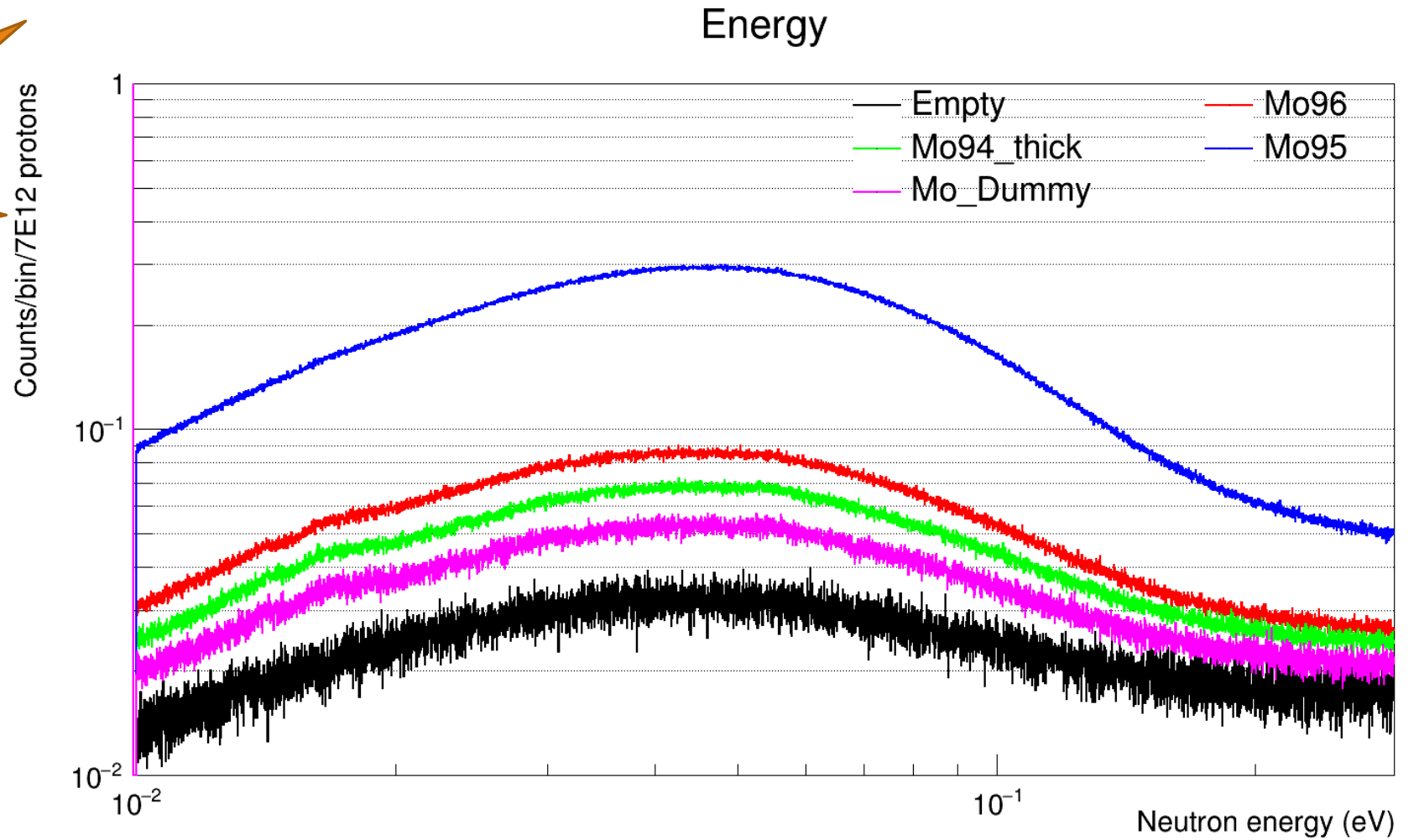
Energy spectra



Contaminants effect visible

Energy spectra

Good signal to noise ratio at thermal energy

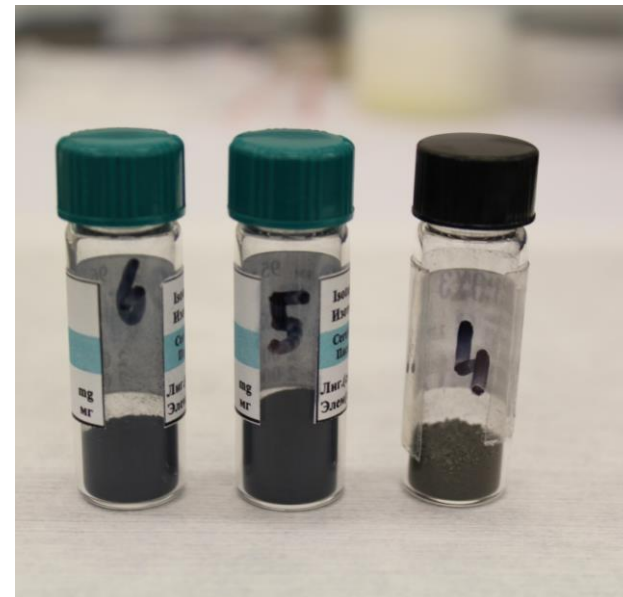


Enriched pellets preparation

- Pressed pellets prepared using enriched powder
- Pellets prepared at JRC-Geel
- Self sustaining pellets of $\sim 2\text{g}$
- Additional ^{nat}Mo samples prepared using powder with different grain sizes
- Samples used in EAR1 campaign at n_TOF

Samples prepared

^{94}Mo	^{95}Mo	^{96}Mo
99%	95%	96%



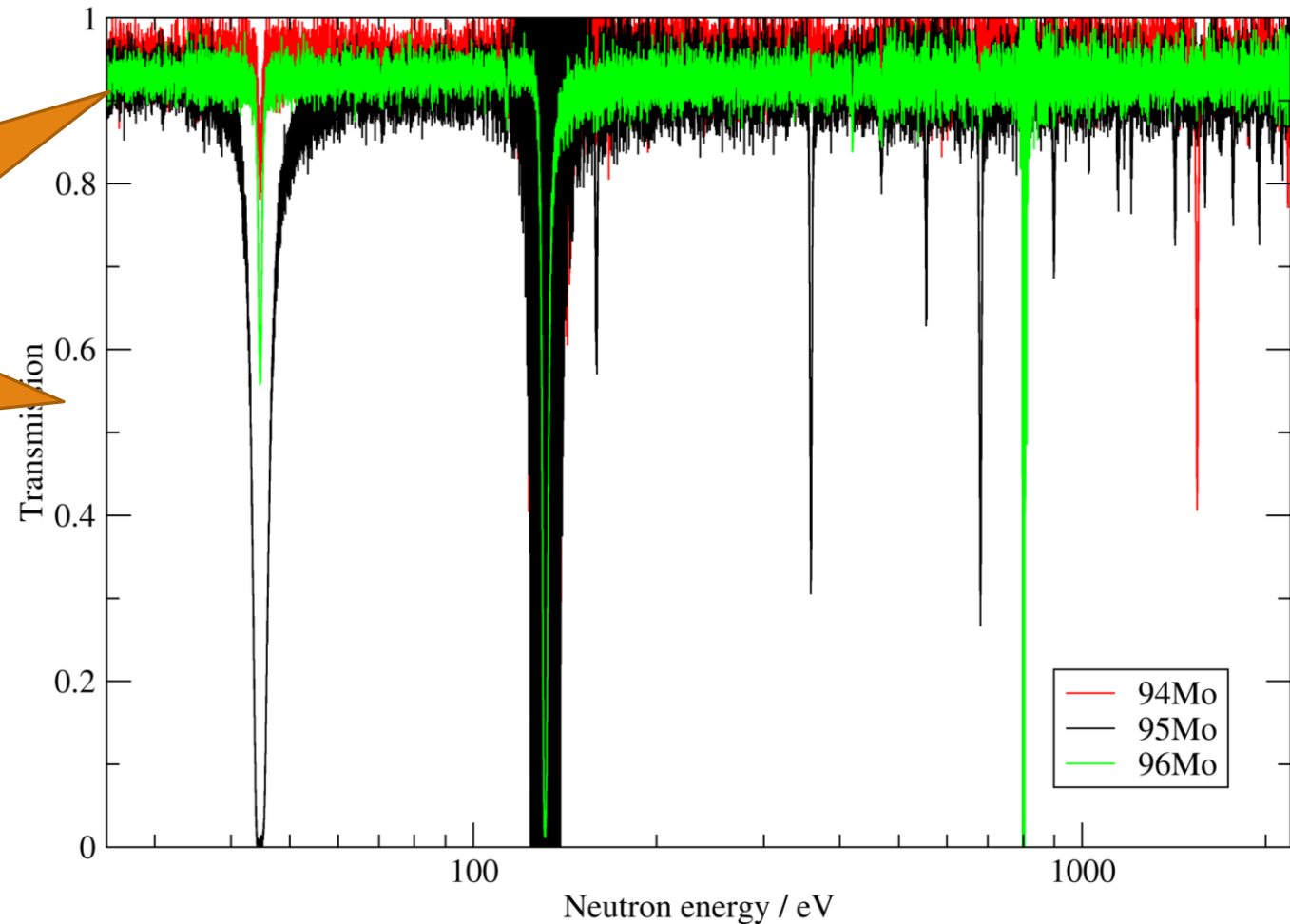
Mo samples

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 (g)	Areal density (atoms/b)
⁹⁴ Mo	1,9526	3,9592E-03
⁹⁵ Mo	1,9745	3,9558E-03
⁹⁶ Mo	1,9175	3,8064E-03
natMo-5 μm	2,014	4,0059E-03
natMo-350 μm	1,989	3,9584E-03

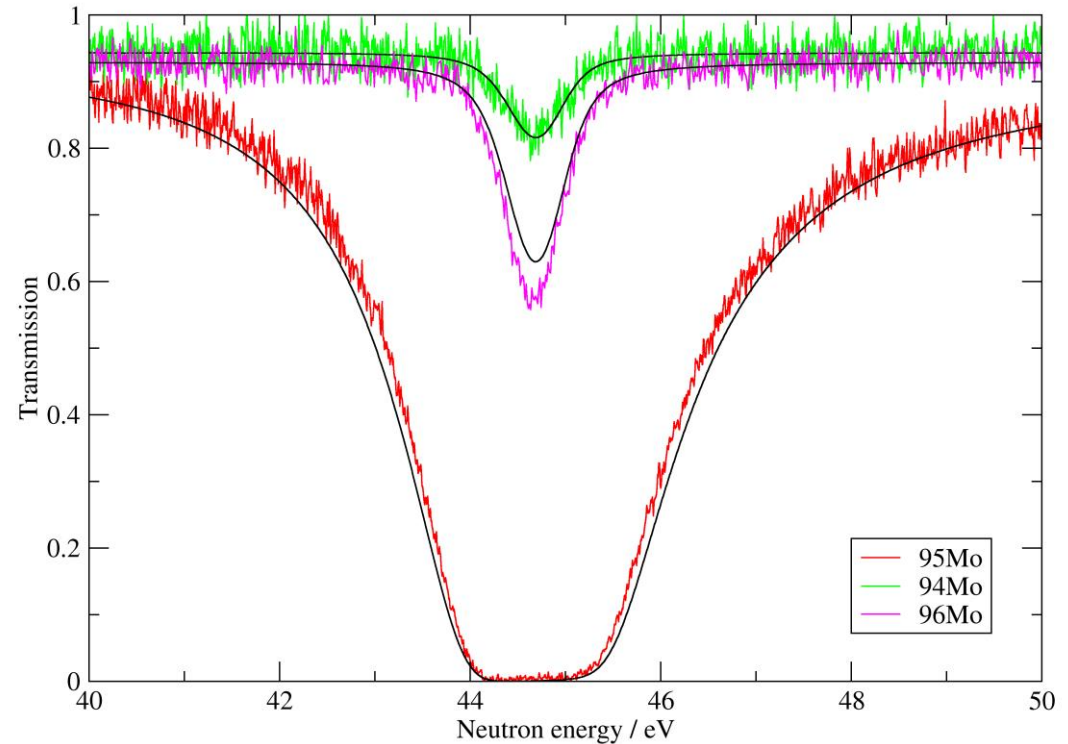
Transmission with enriched Mo

Transmission measurement at 10m with enriched samples performed



Transmission with enriched Mo

- Preliminary results of transmission @10m for enriched pellets;
- Resonance parameters from new compilation;
- Deviation on ^{95}Mo content from declared abundance;
- Abundance of biggest contaminants fitted with REFIT.



Transmission with enriched Mo

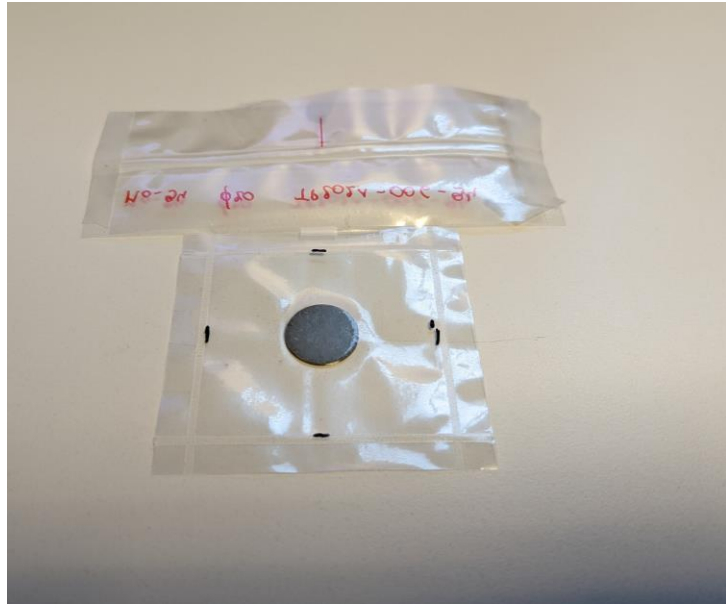
Nominal	⁹² Mo	⁹⁴ Mo	⁹⁵ Mo	⁹⁶ Mo	⁹⁷ Mo	⁹⁸ Mo	¹⁰⁰ Mo
⁹⁴ Mo	0,63%	98,97%	0,36%	0,01%	0,01%	0,01%	0,01%
⁹⁵ Mo	0,31%	0,69%	95,40%	2,24%	0,51%	0,65%	0,20%
⁹⁶ Mo	0,28%	0,24%	1,01%	95,90%	1,00%	1,32%	0,25%



Fitted	⁹² Mo	⁹⁴ Mo	⁹⁵ Mo	⁹⁶ Mo	⁹⁷ Mo	⁹⁸ Mo	¹⁰⁰ Mo
⁹⁴ Mo	0,63%	97,61%	0,35%	0,01%	0,01%	0,01%	0,01%
⁹⁵ Mo	0,31%	0,69%	84,10%	2,24%	0,51%	0,65%	0,20%
⁹⁶ Mo	0,28%	0,24%	1,25%	94,50%	1,08%	1,00%	0,25%

EAR1 samples

^{94}Mo



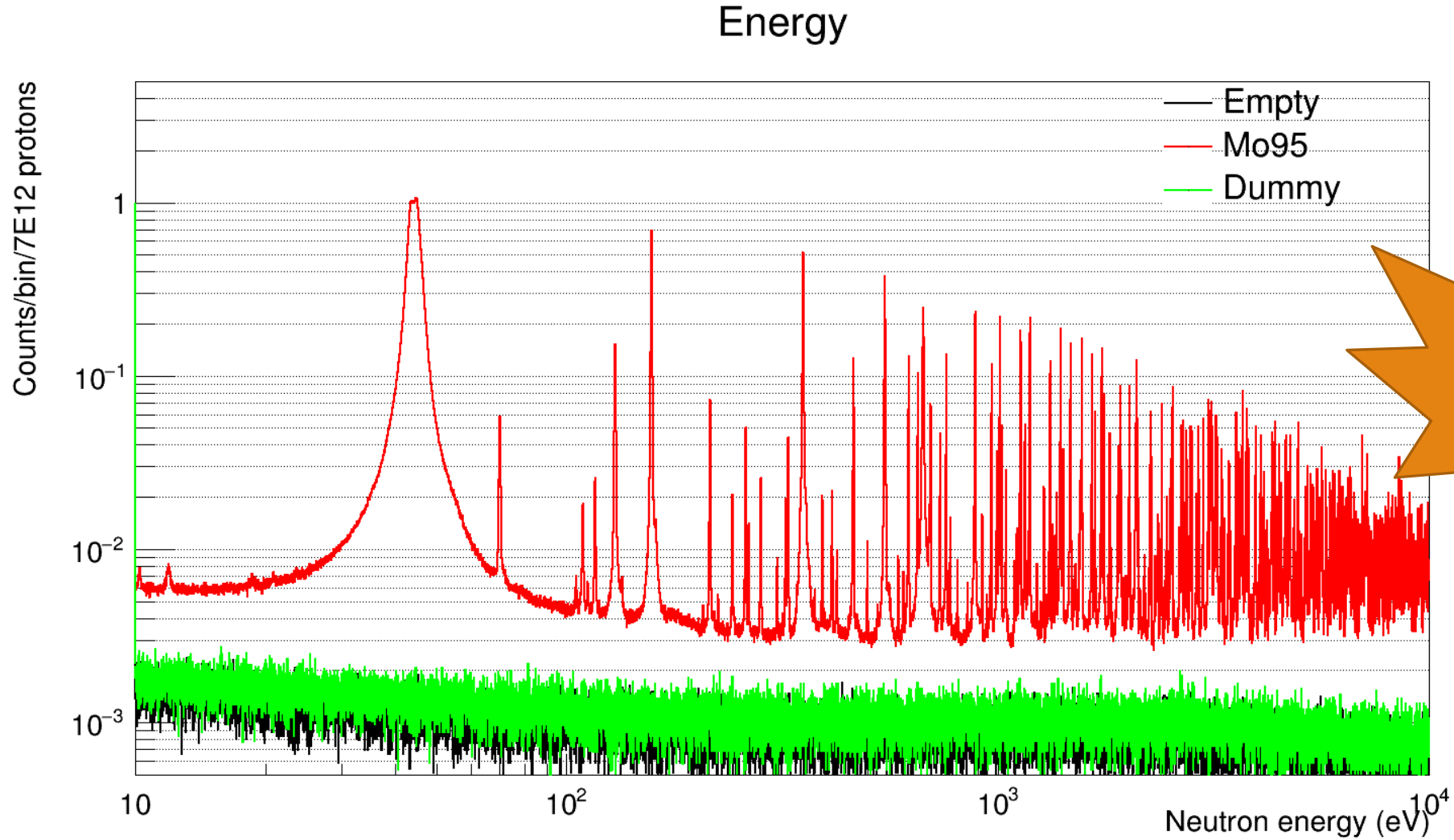
^{95}Mo



^{96}Mo



EAR1 measurements



First results
of EAR1
campaign

What is done:

- Compilation and validation of new resonance parameters file for all molybdenum isotopes
- Preparation of article describing the recommended resonance parameters
- Capture measurements at n_TOF (EAR2) and transmission measurements at GELINA using enriched samples

What is left to do:

- Full analysis of capture and transmission data of enriched samples
- Additional capture measurements at n_TOF (EAR1) and GELINA
- Preparation of results for EXFOR submission

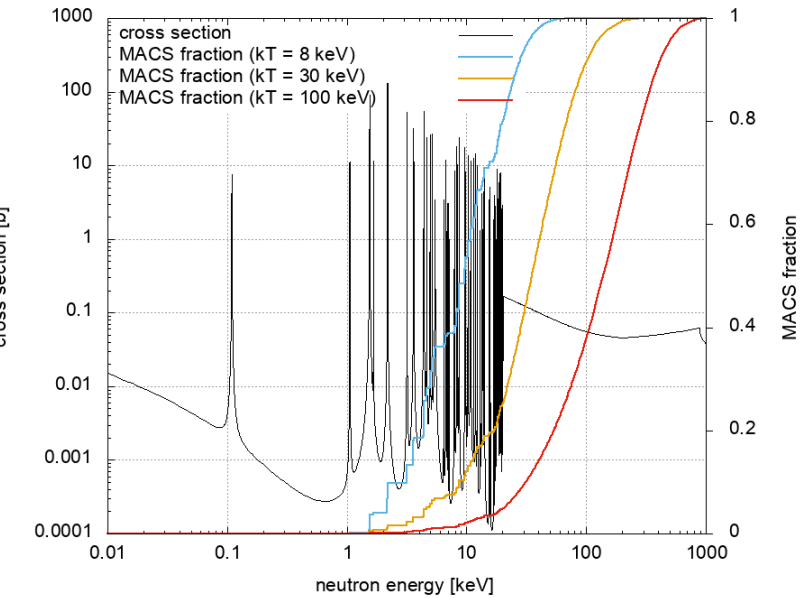
Thank you for your attention!

This project has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 847594 (ARIEL).

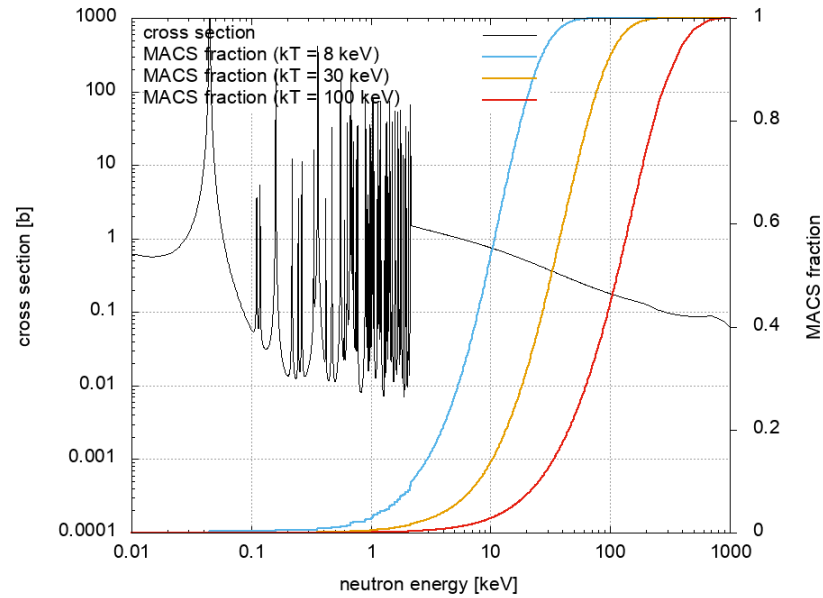
Backup

MACS fractions

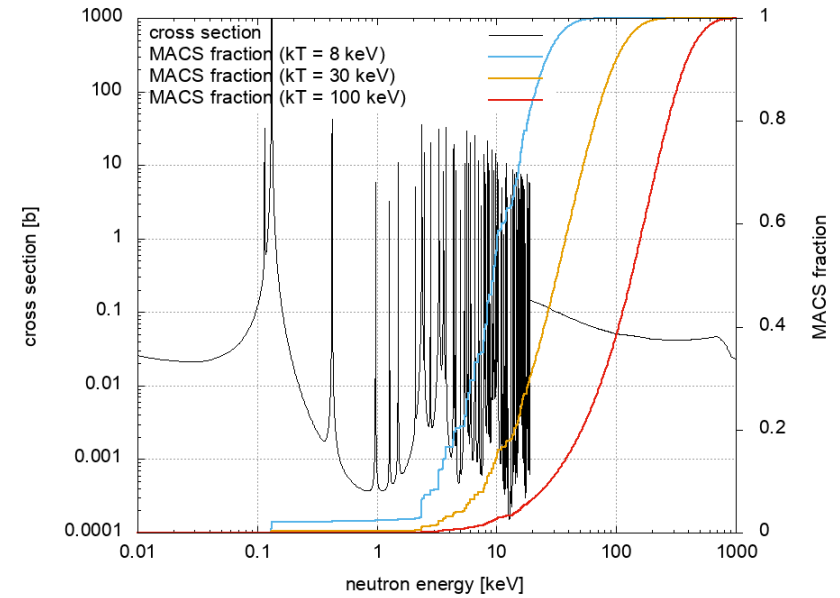
Mo-94



Mo-95

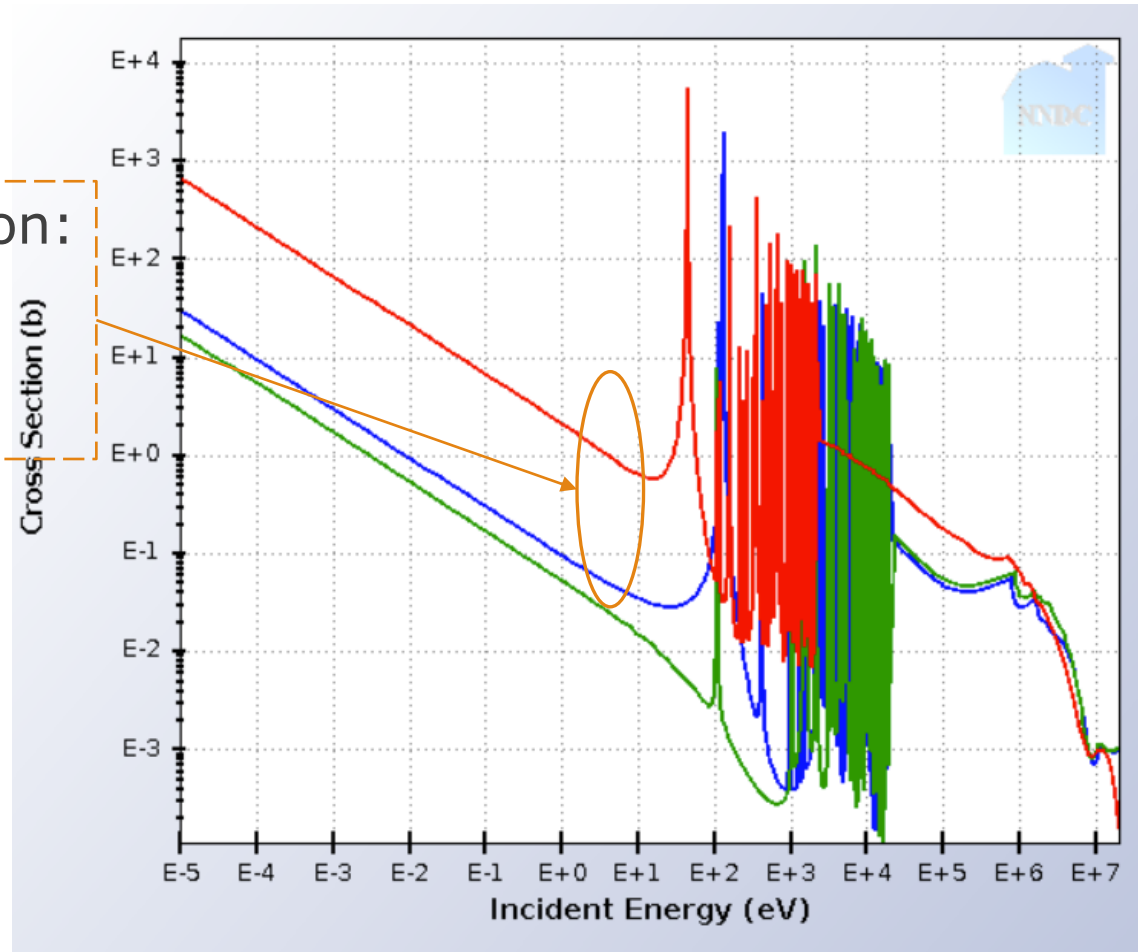


Mo-96



Capture cross section ENDF/B-VIII

Thermal cross section:
 $^{94}\text{Mo} \sim 350\text{mb}$
 $^{95}\text{Mo} \sim 13\text{b}$
 $^{96}\text{Mo} \sim 620\text{mb}$



— ^{94}Mo
— ^{95}Mo
— ^{96}Mo

Libraries sources

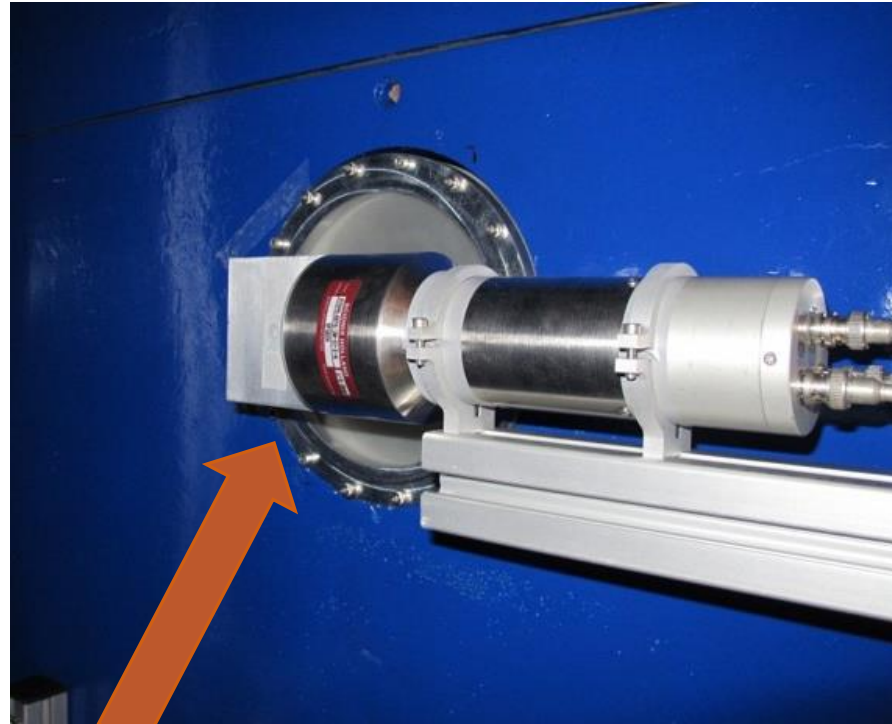
Isotope	JENDL-3.3	JENDL-4	ENDF-B/VIII	JEFF-3.3
⁹² Mo	Wasson, Weigmann, Musgrove	Wasson, Weigmann, Musgrove	Mughabghab	JENDL-4
⁹⁴ Mo	Weigmann, Musgrove	Weigmann, Musgrove, Wang	JENDL-3.3	JENDL-4
⁹⁵ Mo	Weigmann, Shwe	Weigmann, Shwe, Wang	Mughabghab	Mughabghab
⁹⁶ Mo	Weigmann, Musgrove	Weigmann, Musgrove, Wang	JENDL-3.3	JENDL-4
⁹⁷ Mo	Weigmann, Shwe	Weigmann, Shwe, Wang	JENDL-3.3	JENDL-4
⁹⁸ Mo	Weigmann, Musgrove, Chrien	Weigmann, Musgrove, Chrien, Babich, Wang	JENDL-3.3	JENDL-4
¹⁰⁰ Mo	Weigmann, Musgrove, Weigmann	Weigmann, Musgrove, Weigmann, Wang	JENDL-3.3	JENDL-4

Backup - ^{nat}Mo abundances

Isotope	Abundance
⁹² Mo	14.84%
⁹⁴ Mo	9.25%
⁹⁵ Mo	15.92%
⁹⁶ Mo	16.68%
⁹⁷ Mo	9.55%
⁹⁸ Mo	24.13%
¹⁰⁰ Mo	9.63%

Detection system

- Li glass scintillators
- Enriched to 95% in ^6Li
- Placed inside metallic “castle” to reduce background
- Amplitude and time signals
- Time resolution 4,21 ns



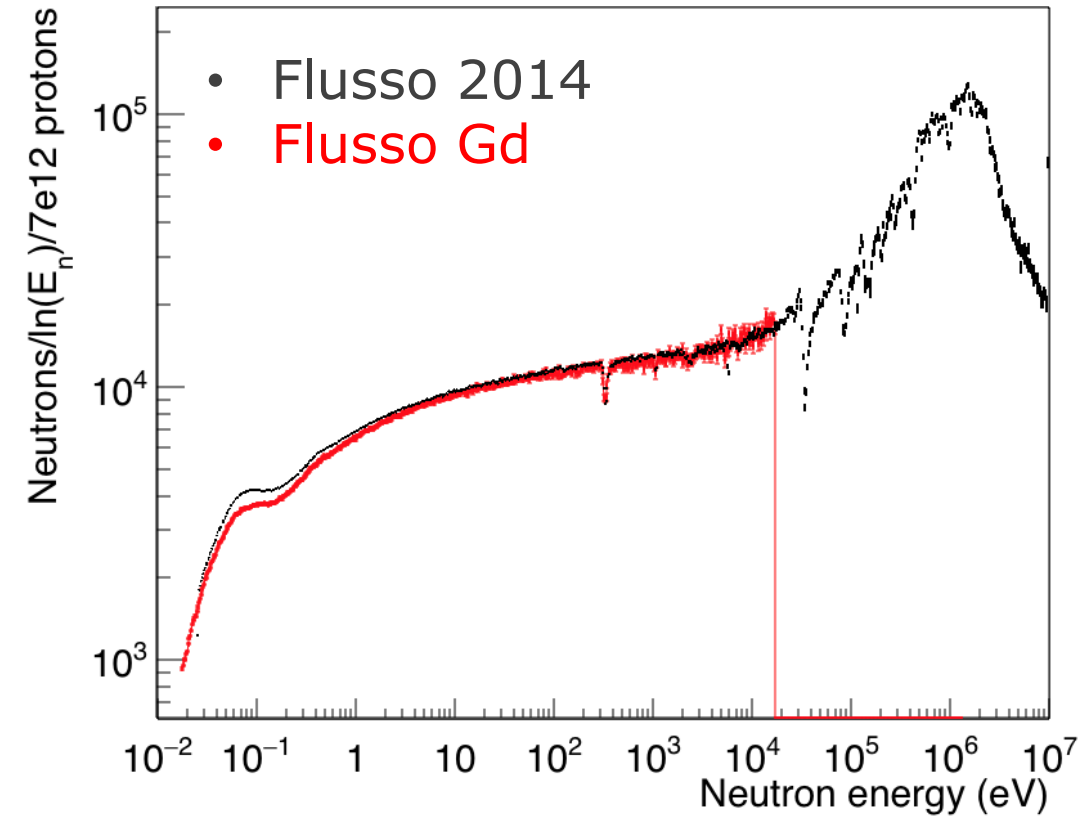
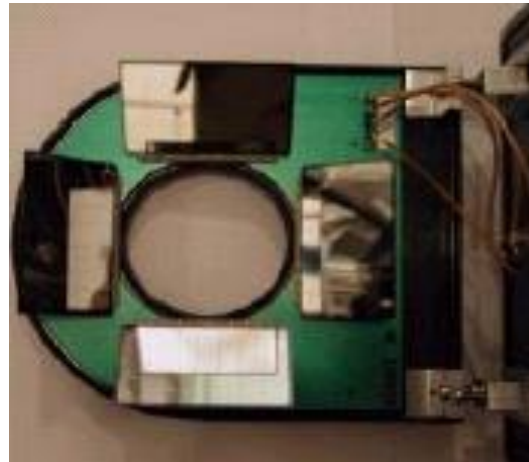
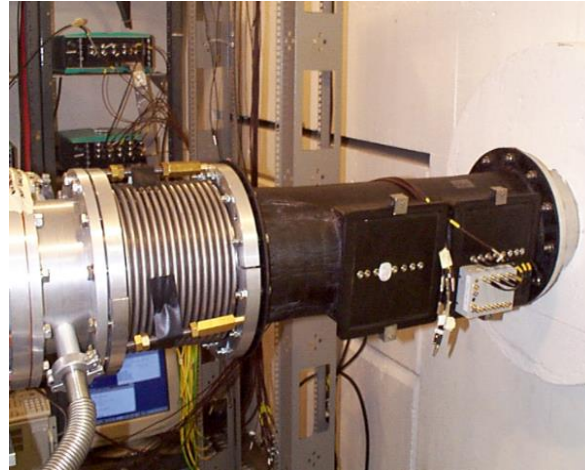
Scintillator



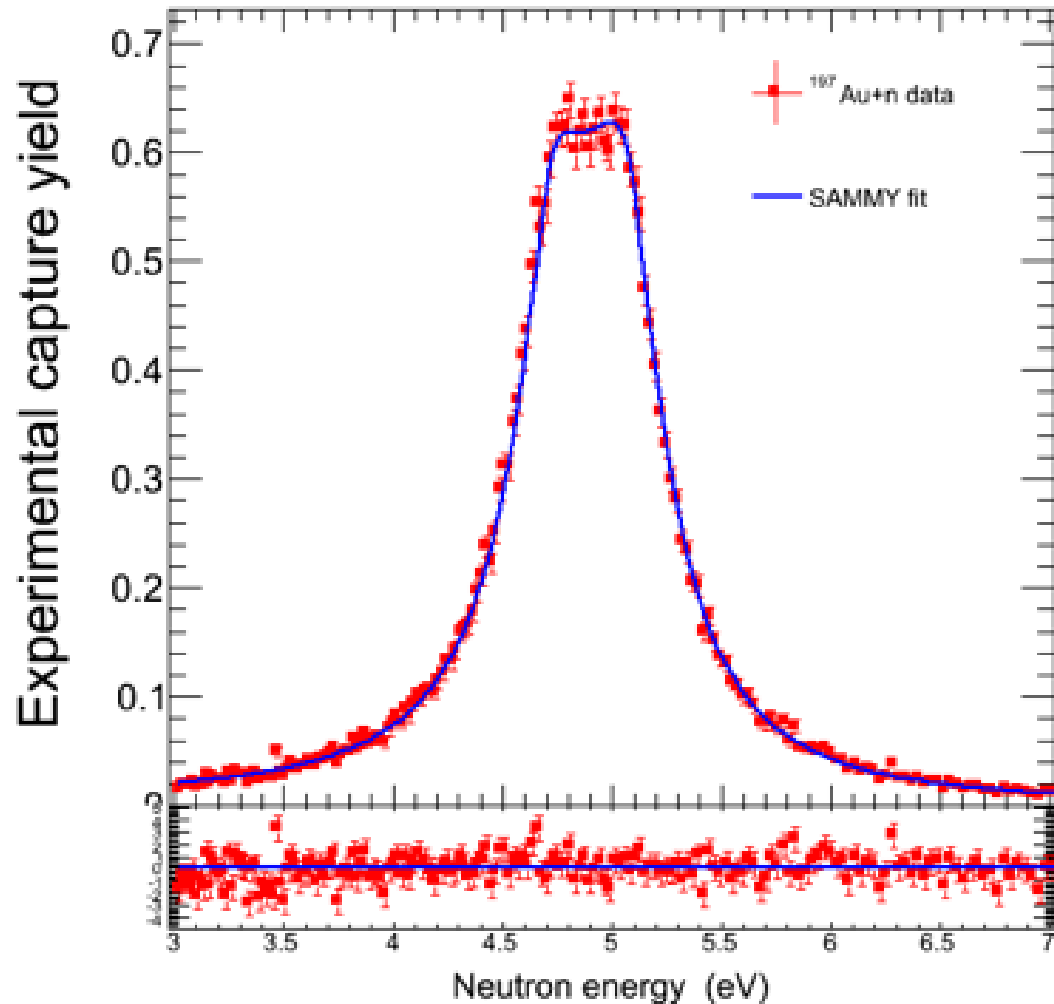
Castle

Neutron flux monitor

- Neutron flux continuously monitored
- SiMON (Silicon MONitor) in beam
- Silicon detector facing mylar foil coated in lithium
- Minimal reduction of neutron flux



Normalization



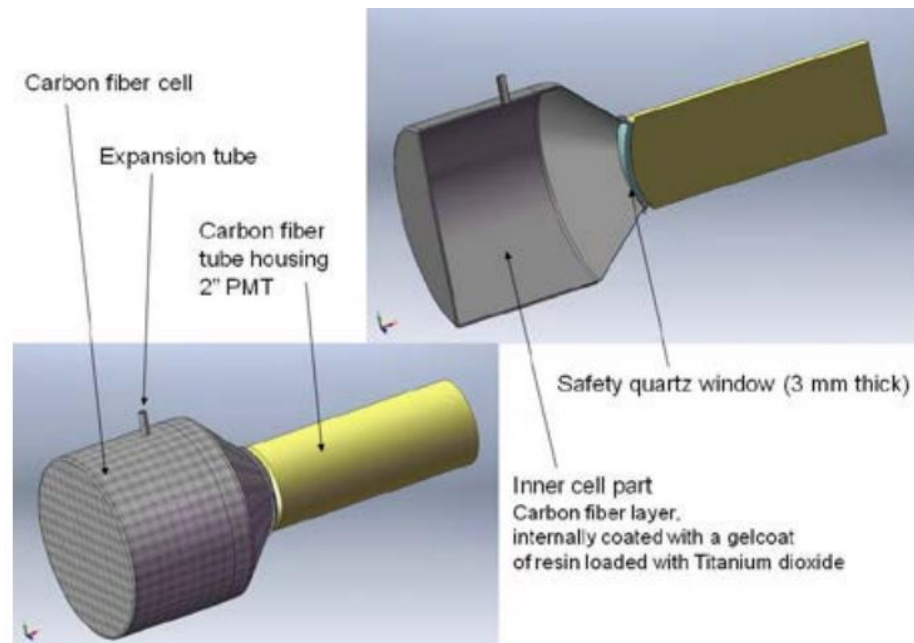
- Au sample
- Black resonance at 4.9 eV
- $\Gamma_\gamma \gg \Gamma_n$
- $Y_c \approx 1$



Extract normalization factor from saturated resonance

Capture detectors

- Five gamma detectors
 - 4 C6D6 liquid scintillators
 - 1 sTED prototype
- Low sensitivity to scattered neutrons
- Fast recovery from gamma flash



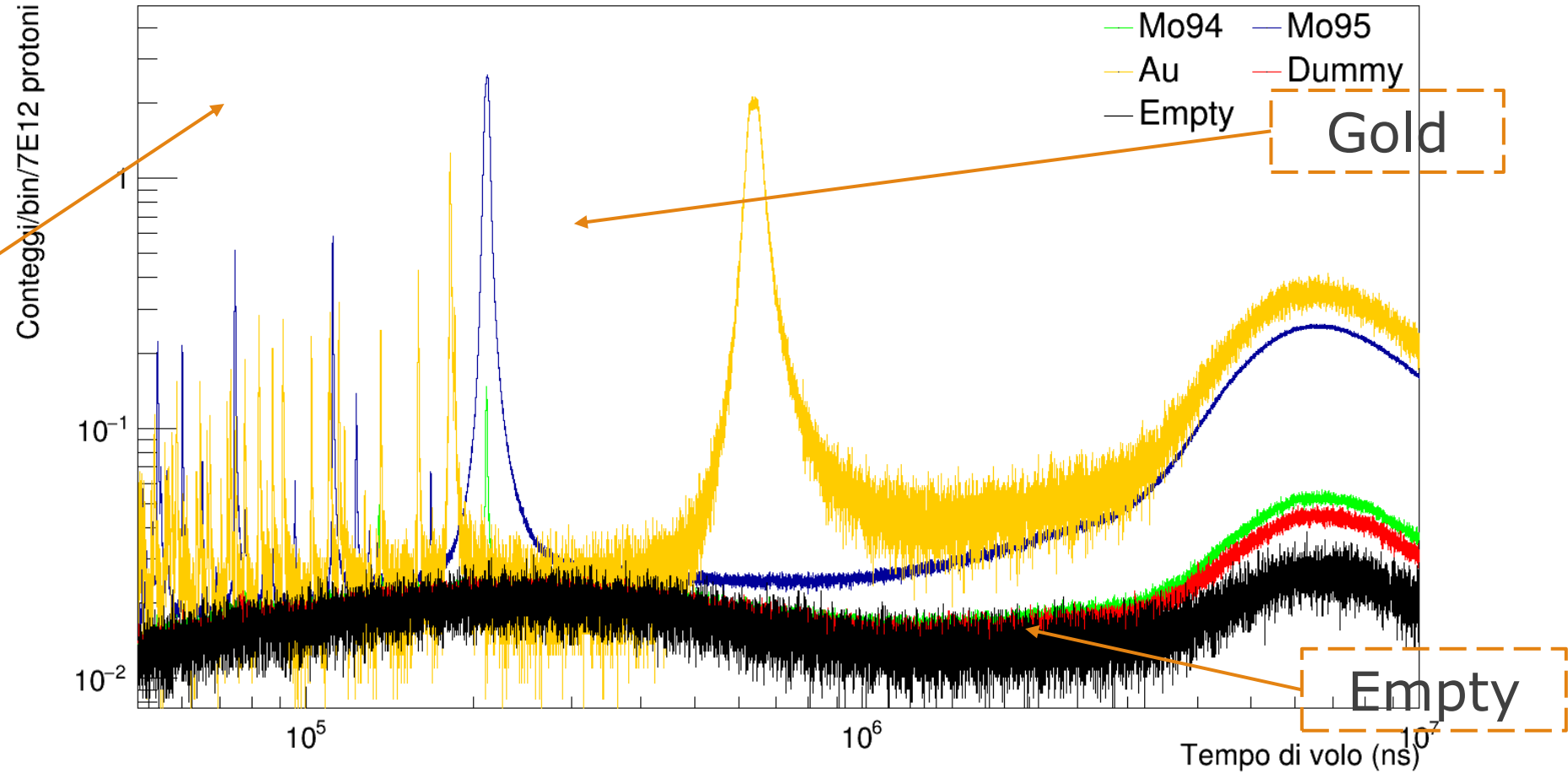
Backup - EAR2 samples

Sample	Mass	Areal density
^{94}Mo	1737,5 mg	3,47E-3
^{95}Mo	929,2 mg	1,86E-3
^{96}Mo	1611 mg	3,22E-3
natMo pellet	2003,3 mg	4,00E-3
natMo powder	985,7 mg	1,97E-3



Time of flight spectra

Time-of-Flight



Backup – EAR2 Measurements

Sample	Protons
⁹⁴ Mo (thick)	4,9E17 (2,8E17)
⁹⁵ Mo	3,7E17
⁹⁶ Mo	4,2E17
natMo	2,1E17
Au	7,3E15
Dummy	1,1E17
Empty	1,1E17
Pb	4,3E16
Filters (Ag, Bi, Cd)	7,3E16
TOTAL	1,84E18