Neutron absorption in the Cr isotopes of structural materials affects the criticality of fast reactor assemblies

A. CASANOVAS, B. FERNÁNDEZ, C. GUERRERO,

N. PATRONIS, P. PÉREZ-MAROTO, J. M. QUESADA, M.E. STAMATI, ET AL.

CEIDEN/UPM WORKSHOP ON “IMPACT OF RECENT NUCLEAR DATA EVALUATIONS ON ENERGY AND NON-ENERGY NUCLEAR APPLICATIONS”(MAY 2023)

Motivation: nuclear data for criticality safety **(IAEA)**

NEA Nuclear Data High Priority Request List, HPRL

HPRL Main	High Priority Requests (HPR)	General Requests (GR)	Special Purpose Quantities (SPQ)		New Request	EG-HPRL (SG-C)
			Standard	Dosimetry		

Request ID	98		**Type of the request**	High Priority request	
Target	**Reaction and process**	**Incident Energy**	**Secondary energy or angle**	**Target uncertainty**	**Covariance**
24-CR-53	(n,g) SIG	1 keV-100 keV		8-10	Y
Field	**Subfield**	**Created date**	**Accepted date**	**Ongoing action**	**Archived Date**
Fission		20-JAN-18	05-FEB-18	Y	

Send a comment on this request to NEA.

Requester: Dr Roberto CAPOTE NOY at IAEA, AUT
Email: roberto.capotenoy@iaea.org

Project (context):

Impact:

Neutron absorption in the Cr isotopes of structural materials affects the criticality of fast reactor assemblies [Koscheev2017]. These cross sections are also of interest for stellar nucleosynthesis [Kadonis10].

Accuracy:

8-10% in average cross-sections and calculated MACS at 10, 30, 100 keV.

Selected criticality benchmarks with large amounts of Cr (e.g., PU-MET-INTER-002, and HEU-COMP-INTER-005/4=KBR-15/Cr) show large criticality changes of the order of 1000 pcm due to 30% change in Cr-53 capture in the region from 1 keV up to 100 keV [Trkov2018]. On the other side different evaluations (e.g., BROND-3.1, ENDF/B-VII.1, ENDF/B-VIII.0 and JEFF-3.3) for Cr-53(n,g) are discrepant by 30% in the same energy region. For Cr-50, evaluated files show better agreement at those energies but they are lower than Mughabghab evaluation of the resonance integral by 35%. These discrepancies are not reflected in estimated uncertainty of the evaluated files (e.g., JEFF-3.3 uncertainty is around 10% which is inconsistent with the observed spread in evaluations). Due to these differences we request new capture data with 8-10% uncertainty to discriminate between different evaluations and improve the C/E for benchmarks containing Chromium and/or SS.

Justification document:

Criticality benchmarks can test different components of stainless steel (SS), including Cr which is a large component of some SS. Currently, a large part of the uncertainty in SS capture seems to be driven by uncertainty in Cr capture [Koscheev2017]. Indeed, some benchmarks highly sensitive to Cr (as a component of SS) indicate a need for much higher capture in Cr for both Pu and U fueled critical assemblies (e.g., HEU-COMP-INTER-005/4=KBR-15/Cr and PU-MET-INTER-002=ZPR-6/10).

- Stainless Steel is often used as a **structural material in nuclear reactors** and contains between **11-26% of chromium**
- There are **serious discrepancies** (~30%) between the different evaluated data of **^{50}Cr and ^{53}Cr capture cross section**, which is not present in the corresponding estimated uncertainties
- **OECD NEA-HPRL** (High Priority Request List)
  → **^{50,53}Cr(n,γ) within 8-10% at 1 to 100 keV**

Previous talk link: Edinburgh'22

Motivation: nuclear data for criticality safety (IAEA)

Criticality benchmarks can test different components of stainless steel (SS), including Cr which is a large component of some SS. Currently, **a large part of the uncertainty in SS capture seems to be driven by uncertainty in Cr capture**. Indeed, some benchmarks highly sensitive to Cr (as a component of SS) indicate a need for much higher capture in Cr for both Pu and U fueled critical assemblies (e.g., HEU-COMP-INTER-005/4=KBR-15/Cr and PU-MET-INTER-002=ZPR-6/10).

Capture in natural Cr is driven by capture on Cr-50 and especially in odd Cr-53.

For Cr-53(n,g) there is a very large spread in MACS(30) values in different libraries compared to recommended KADoNiS 1.0 value of 41 +/- 10 mb (the latter is 25% larger). Existing measurements from the 70s are even larger being close to 60 mb with 30% uncertainty. Finally, the re-evaluation for ENDF/B-VIII.0 of the ORNL TOF measurement on enriched Cr-53 target contradicts the increase suggested in Koscheev et al. (2017) where preliminary data have been used.

Note ~50% discrepancies in resonance integrals (in barns) between evaluated libraries and ATLAS [Mughabghab2006] for both Cr-50(n,g) and Cr-53(n,g)

Such contradictions need to be resolved thanks to new measurements and evaluation.

Sensitivity of selected benchmarks to Cr-53 and Cr-50 capture

Andrej Trkov, Oscar Cabellos and Roberto Capote

January 2018

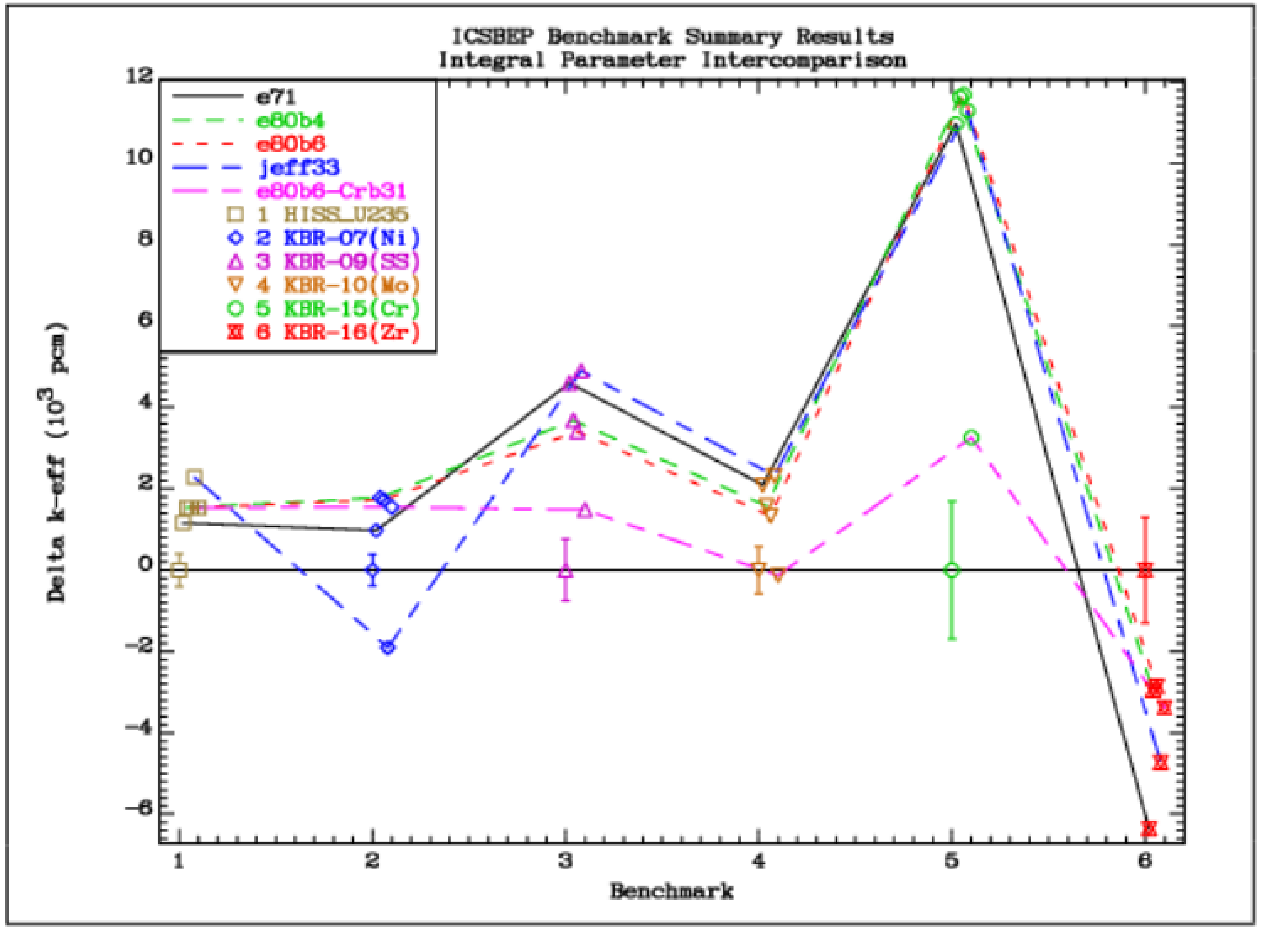
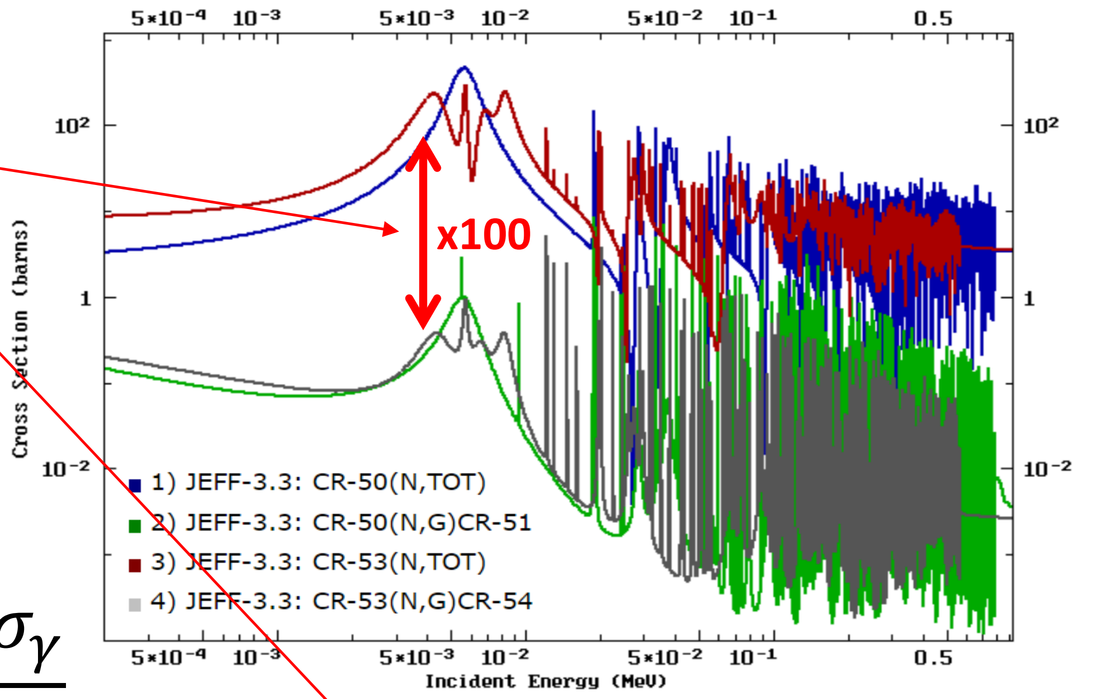
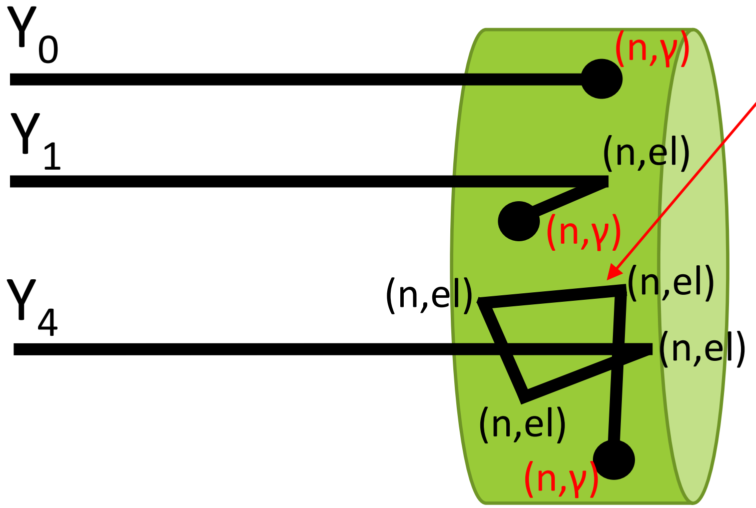


Figure 1: Comparison of the differences between the calculated k_{inf} values and the reference benchmark values for the HISS and KBR benchmarks. e80b6 results correspond to the ENDF/B-VIII.beta5 library. e80b6-Crb31 results correspond to the Brond-3.1 Cr evaluation with all other elements taken from the ENDF/B-VIII.beta5 library. A large impact of Cr data on these benchmarks is evident.

Why the discrepancies?

- The main problem for measuring Cr(n,γ) is the large **neutron multiple-scattering effects**
- In the previous measurements thick samples were used, aiming for good statistics in a very wide energy range





$$Y_0 = (1 - e^{n\sigma_t})\frac{\sigma_\gamma}{\sigma_t}$$

Capture yield (captures/neutron) $\rightarrow Y = Y_0 + Y_1 + Y_2 + Y_3 \ldots$

$Y_0 + Y_1$: Analytical (accurate)

$Y_2 + Y_3 \ldots$: Numerical (aproximate)

How to improve σ(n,γ) down to a few %?

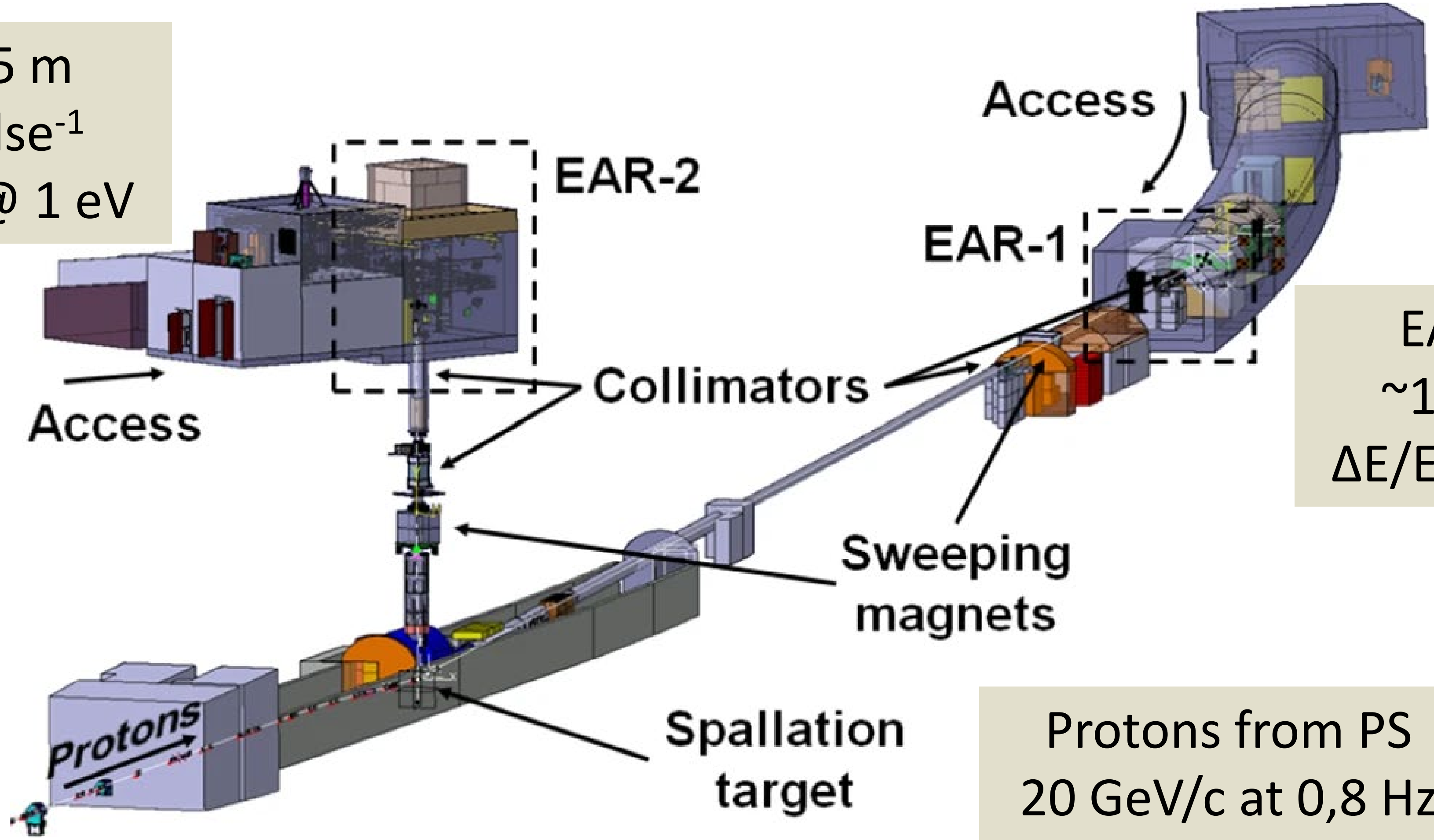
- Enriched (expensive and scarce) material with high purity → 94,6% ^{50}Cr & 97,7% ^{53}Cr
- Controlling multiple-scattering effects:
  - Very thin/thin sample approach
  - C_6D_6 detectors (low sensitivity to scattered neutrons)
- Complementing with ^{50}Cr activation measurement → HiSPANoS@CNA

Experiment	Beer (1975)	Stieglitz (1971)	Brusegan (1986)	Kenny (1977)	Guber (2011)	**This work (2022)**
Facility	FZK	RPI	GELINA	ORELA	ORELA	**n_TOF**
L (m)	0,7	27	60	40	40	**185**
Energy (keV)	1-300	1-200	1-200	1-200	0,01-600	**1-100**
Density ^{50}Cr (10^{-3} at/barns)	18	8	7	5/8	-	**0,6/1,9**
Density ^{53}Cr (10^{-3} at/barns)	14	14	12/60	8/12	14	**1,2/6**

Our "thicks" are thinner than all previous → lower multiple interaction corrections

The neutron_TOF facility at CERN

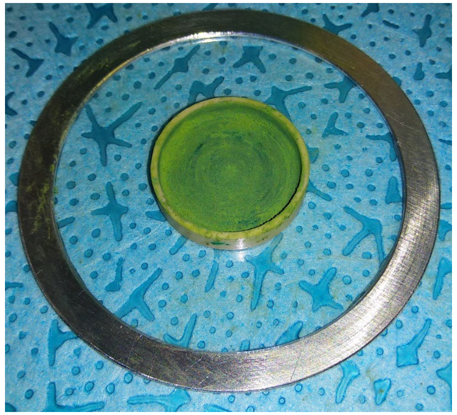
EAR2 -> 18,5 m
~10^6 $cm^{-2}pulse^{-1}$
$\Delta E/E$ ~ $4 \cdot 10^{-3}$ @ 1 eV



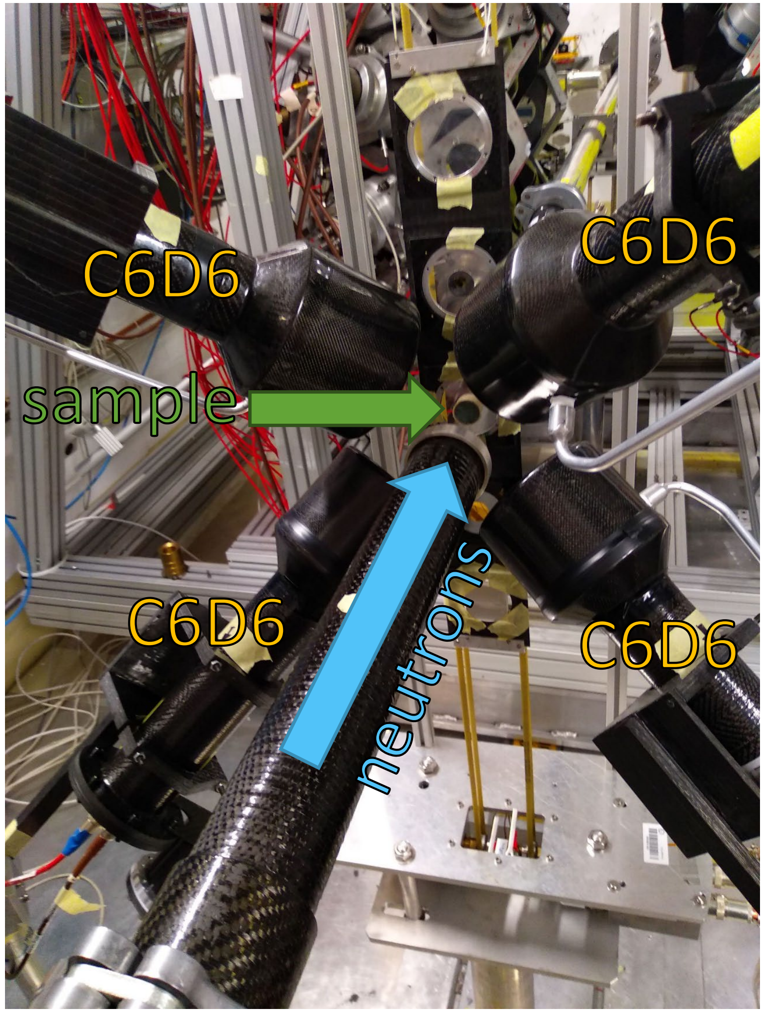
EAR1 -> 185 m
~10^4 $cm^{-2}pulse^{-1}$
$\Delta E/E$ ~ $5 \cdot 10^{-4}$ @ 1 eV

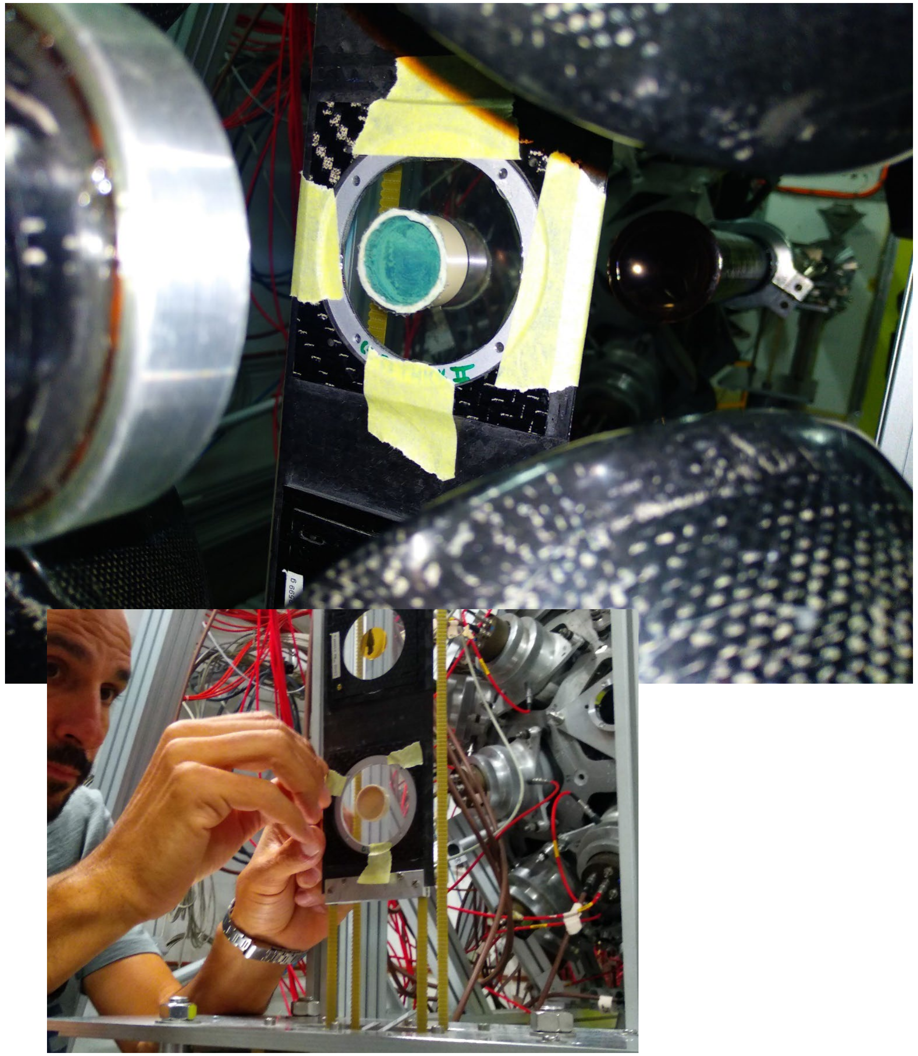
Protons from PS
20 GeV/c at 0,8 Hz
~$850 \cdot 10^{10}$ ppp

Samples and detector set-up (EAR1)

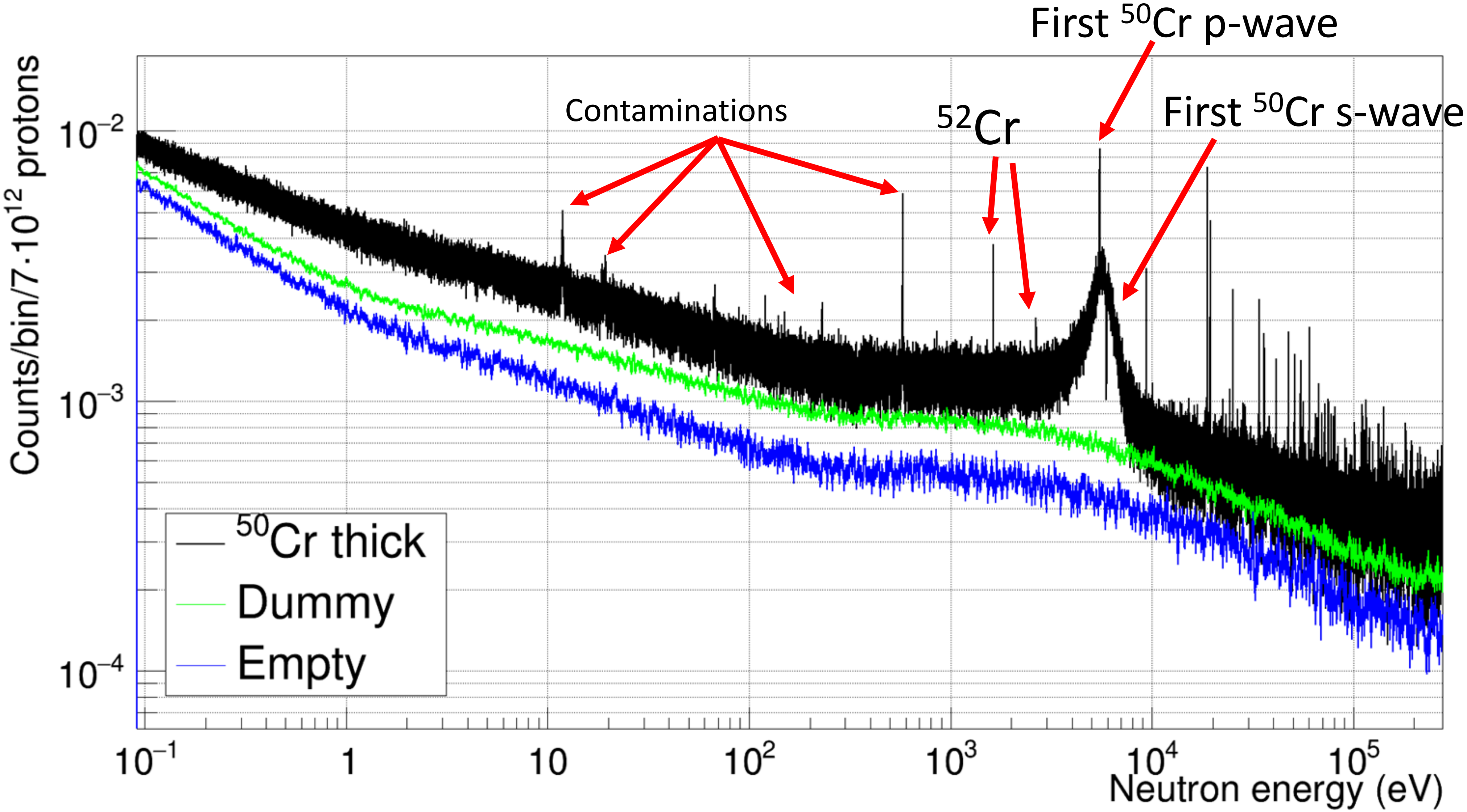


Cr_2O_3 powder pressed in a PEEK capsule & Al holder

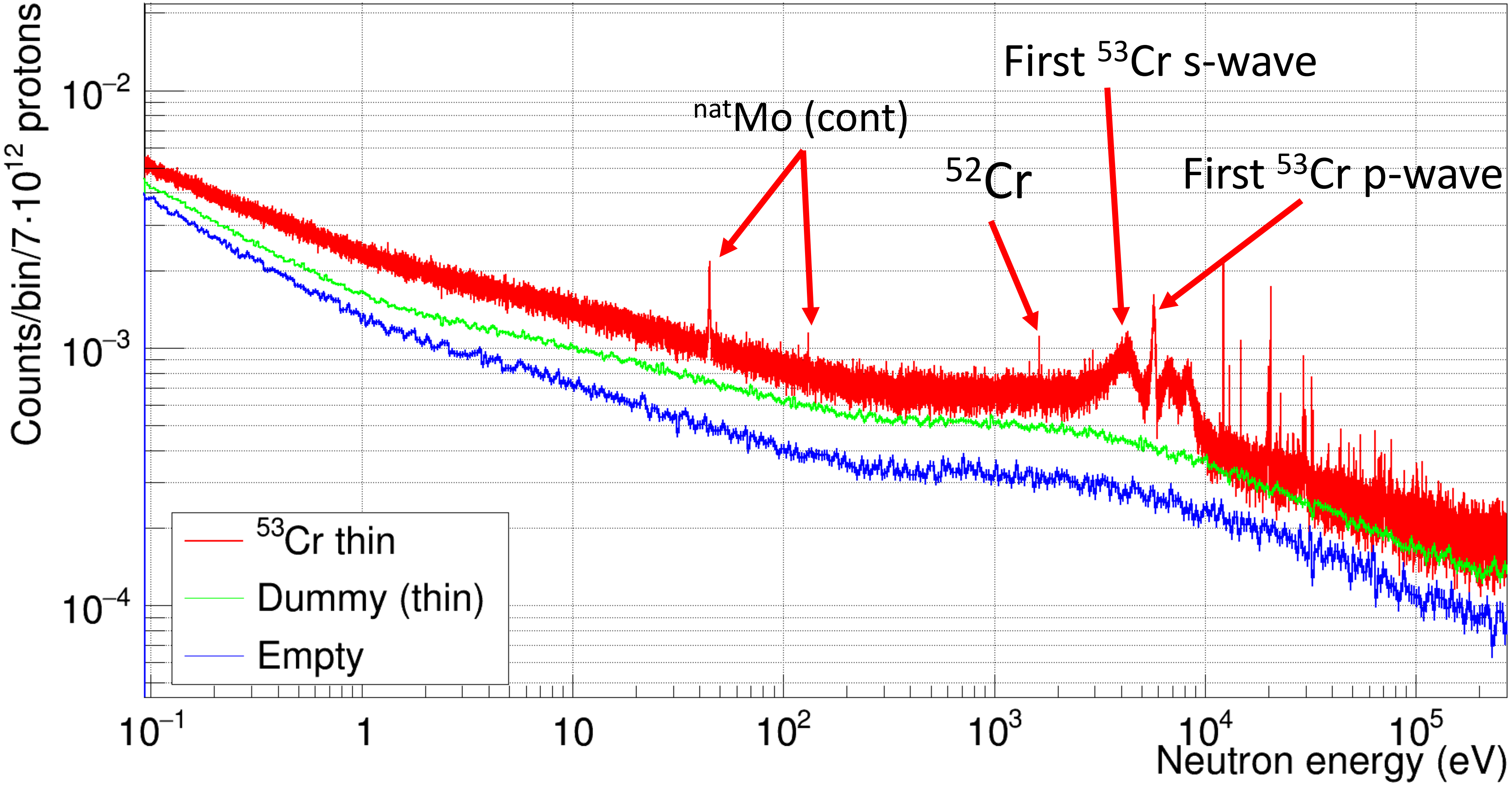




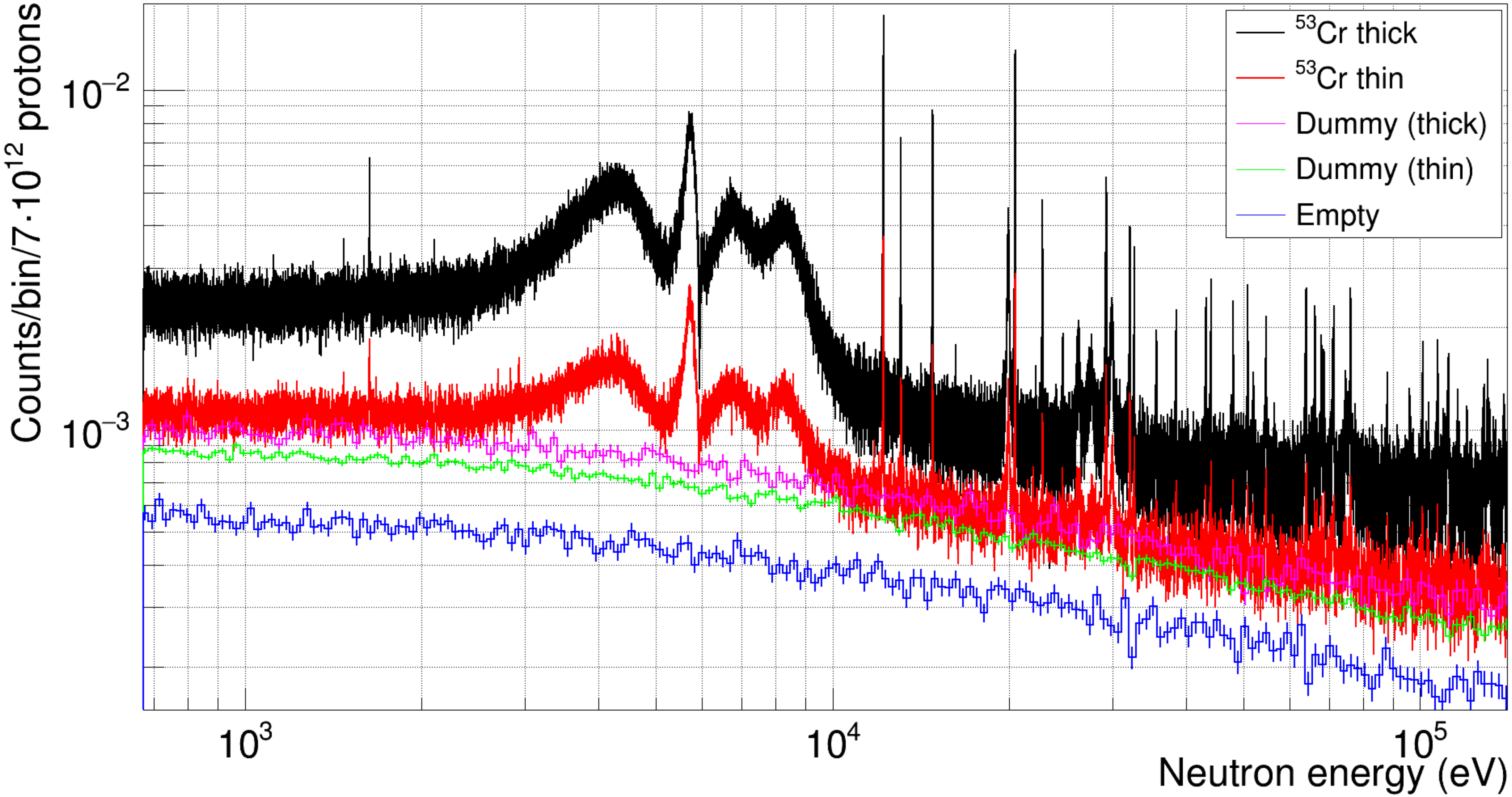
Preliminary results (^{50}Cr-thick)



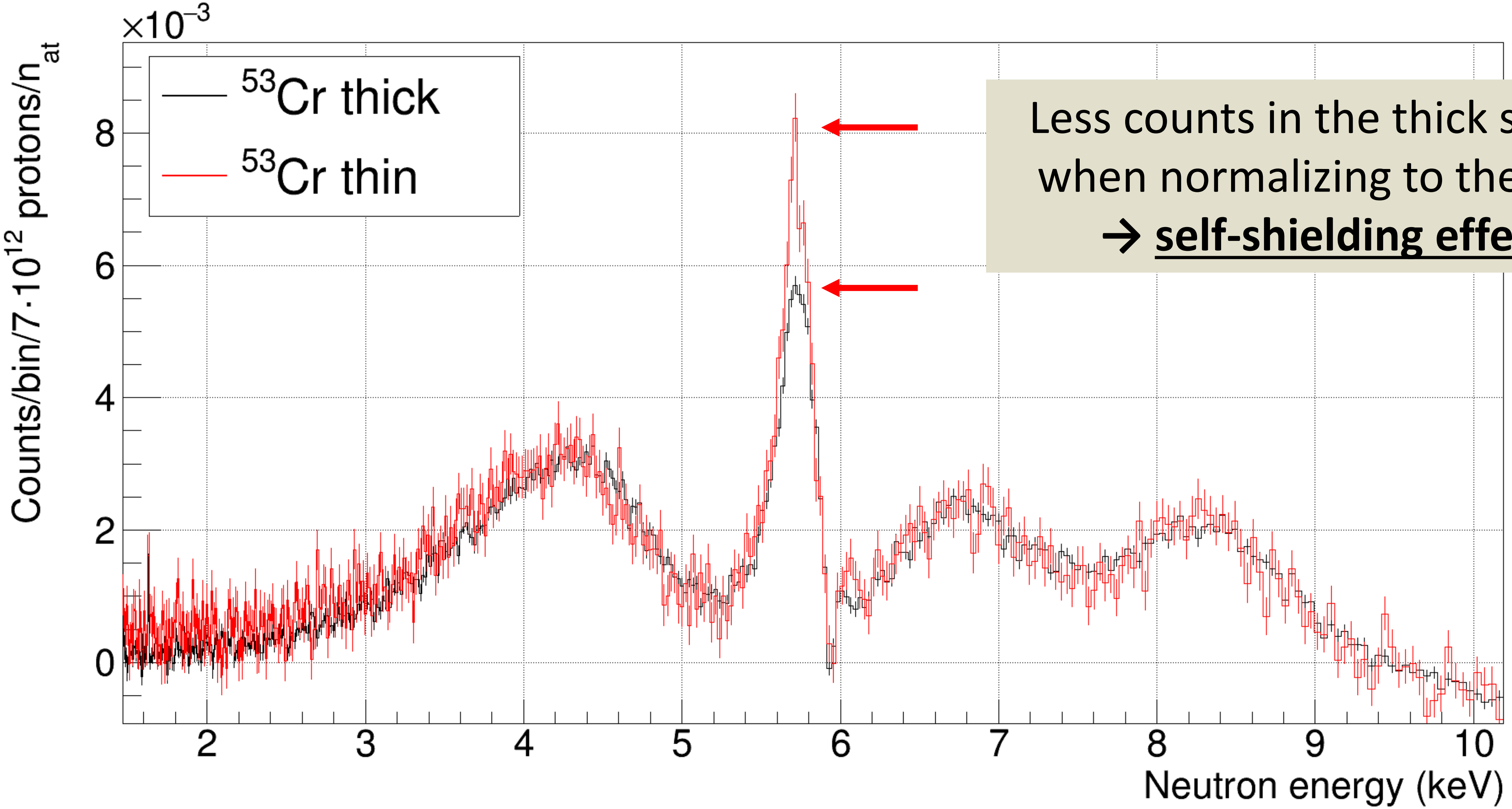
Preliminary results (^{53}Cr-thin)



Preliminary results (^{53}Cr: thin vs. thick)



Preliminary results (^{53}Cr: thin vs. thick)

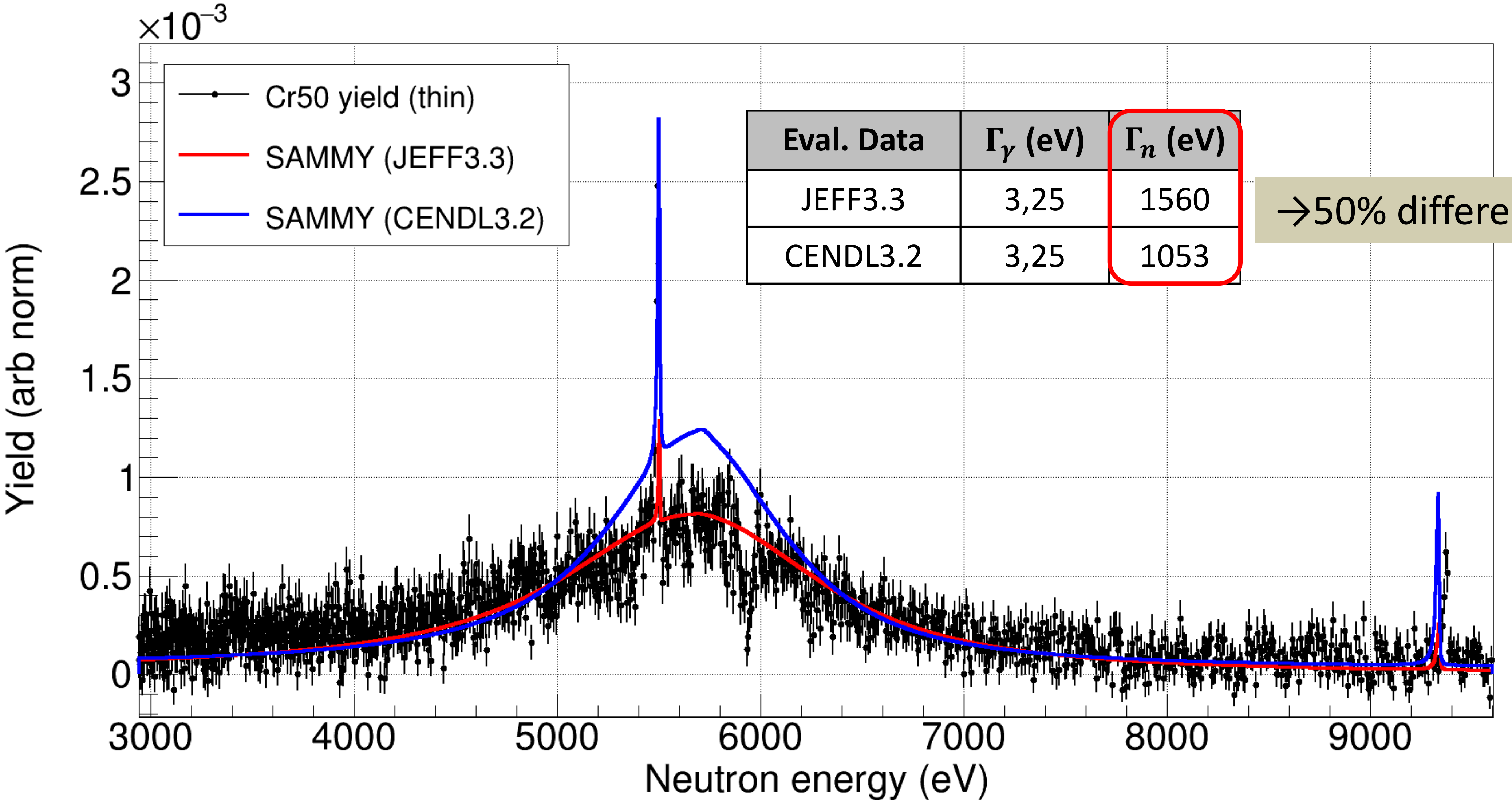


Less counts in the thick sample when normalizing to the mass
→ **self-shielding effects**

Preliminary yield (^{50}Cr)

- We have obtained a preliminary “yield” to check our experimental data.
- To do so:
  1. Only background from dummy considered
  2. Normalize to the main resonance
  3. Resolution Function <u>NOT</u> included
  4. Fixed flight path + fine tunning added “manually” for each resonance
  5. Compared with SAMMY calculations with JEFF3.3 & CENDL3.2 (then INDEN & others)

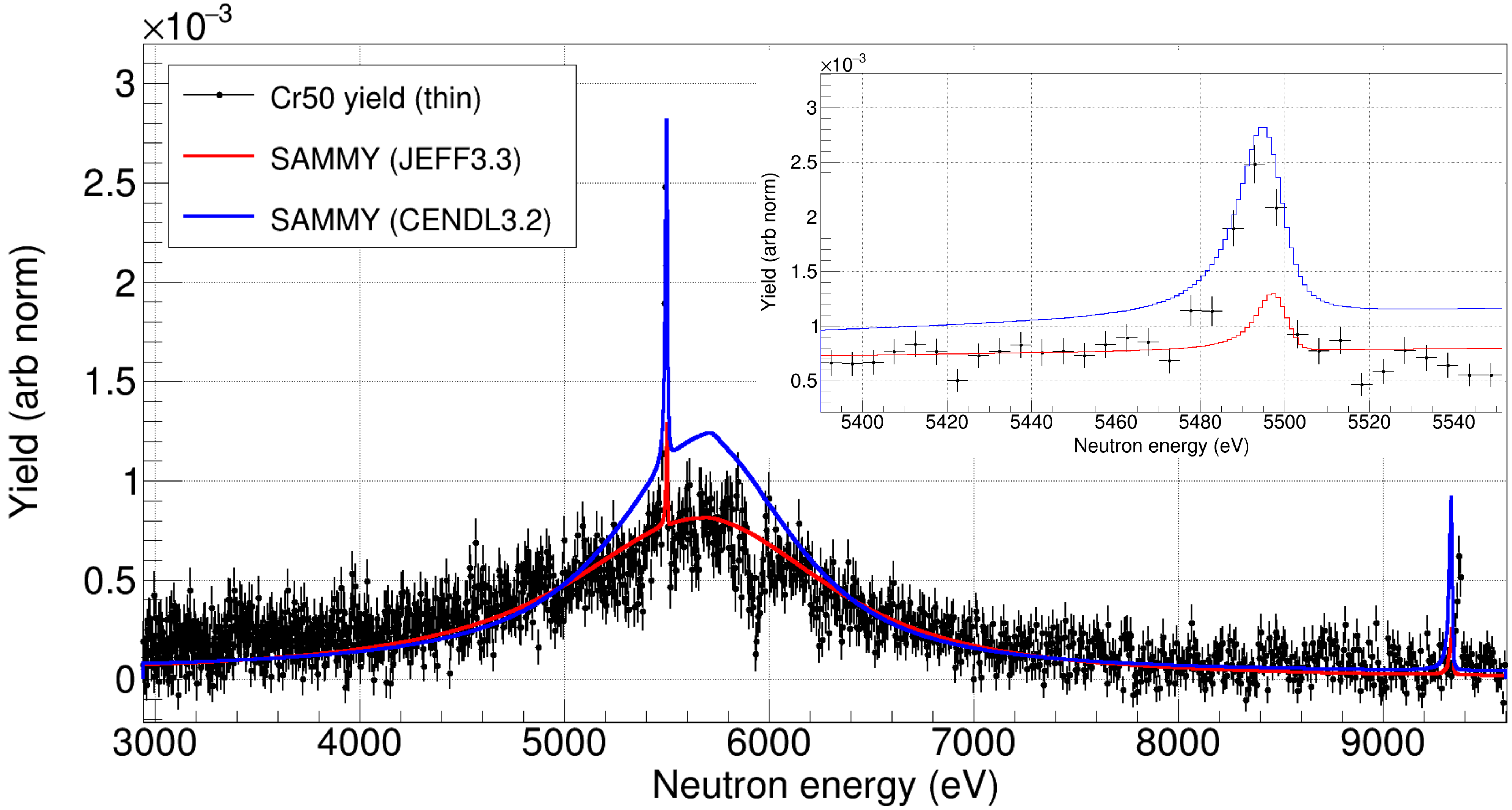
Preliminary yield (^{50}Cr)



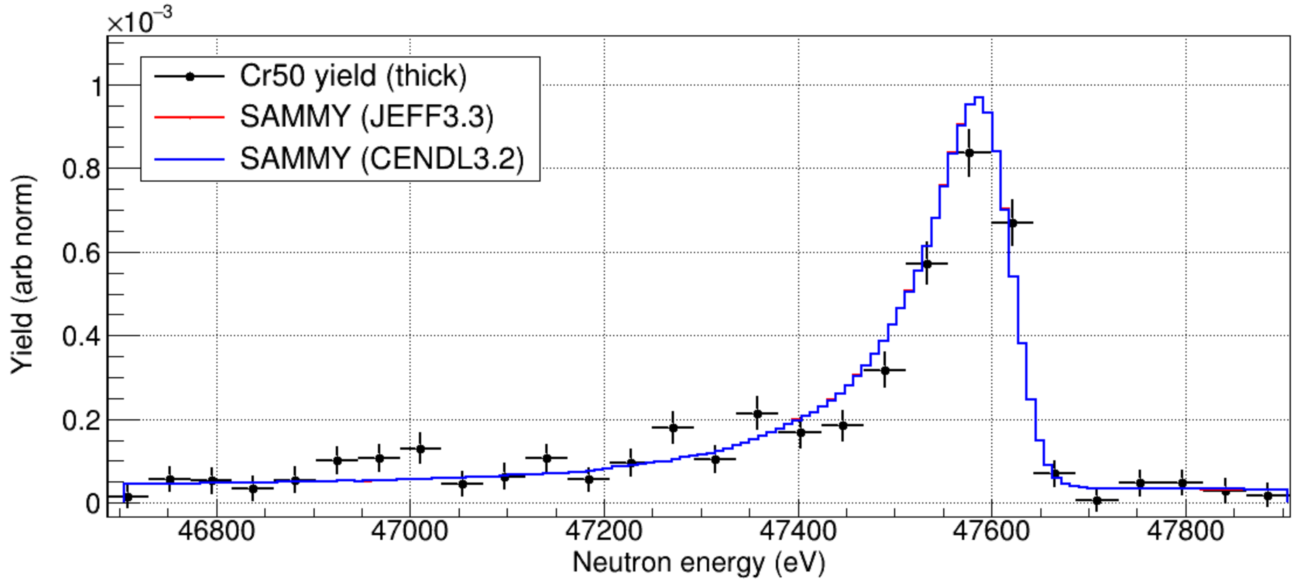
Eval. Data	Γ_γ (eV)	Γ_n (eV)
JEFF3.3	3,25	1560
CENDL3.2	3,25	1053

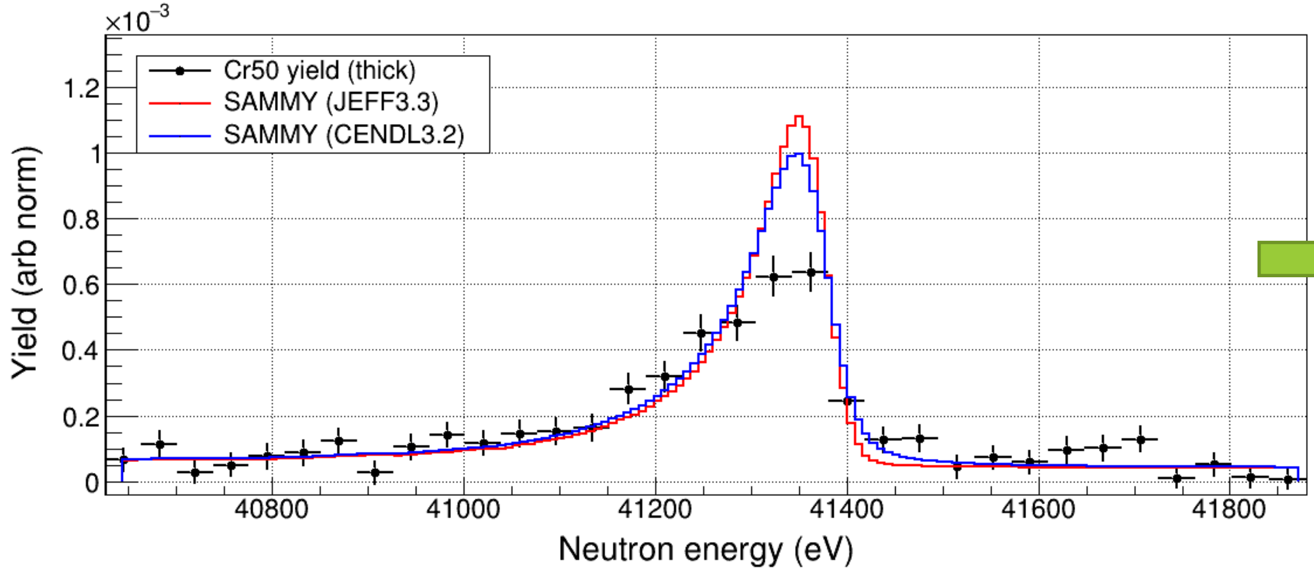
→50% difference

Preliminary yield (^{50}Cr)



Preliminary yield (^{53}Cr)

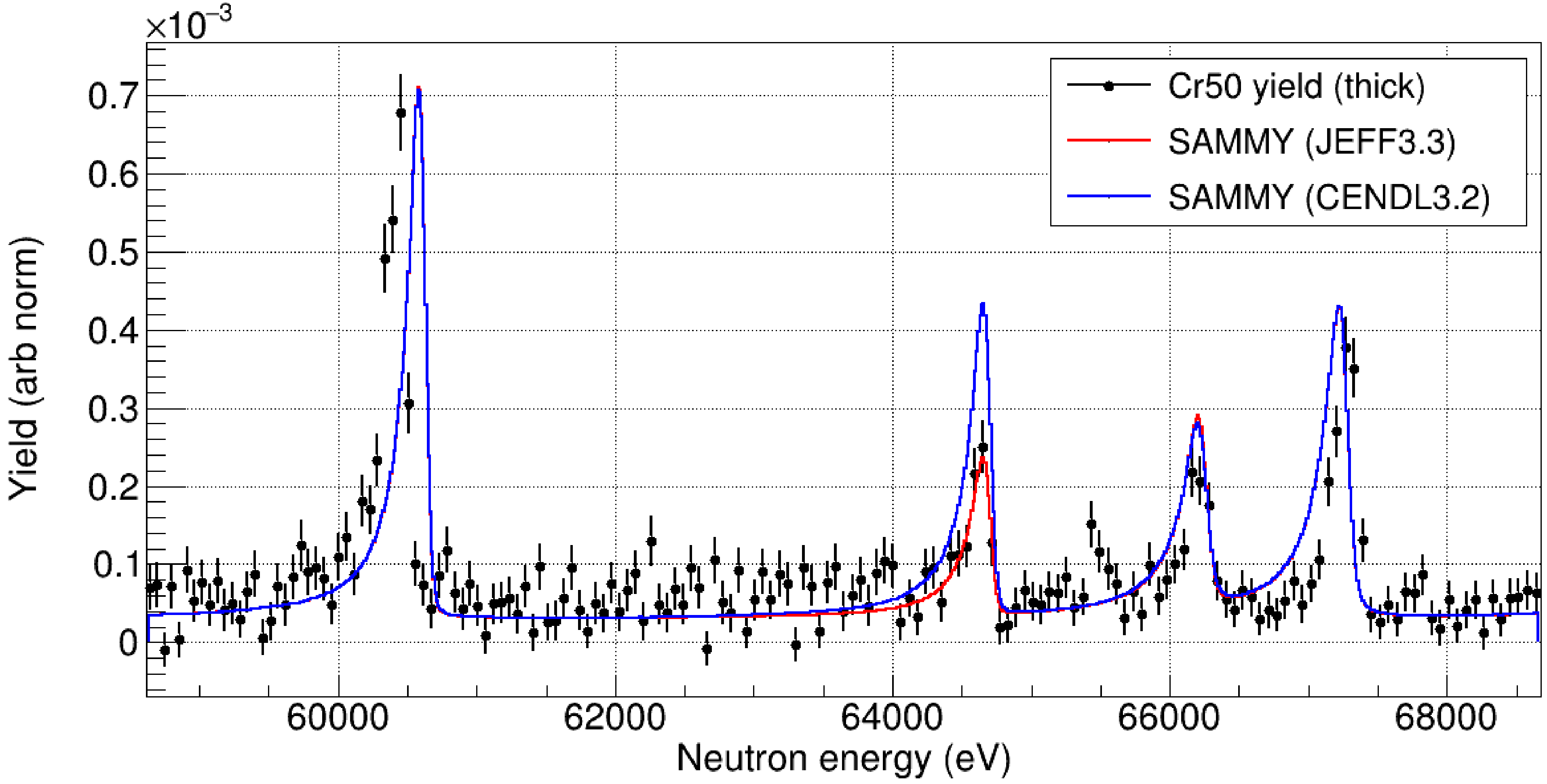




- Just tunning the normalization and the energy of the resonances, the shape of the yield matches the one obtained with SAMMY.
- This is the case for most of the resonances, for a few of them there are differences → bigger scattering contribution?

Eval. Data	Γ_γ (eV)	Γ_n (eV)
JEFF3.3	0,43	3,5
CENDL3.2	0,43	18,1

Preliminary yield (^{53}Cr)

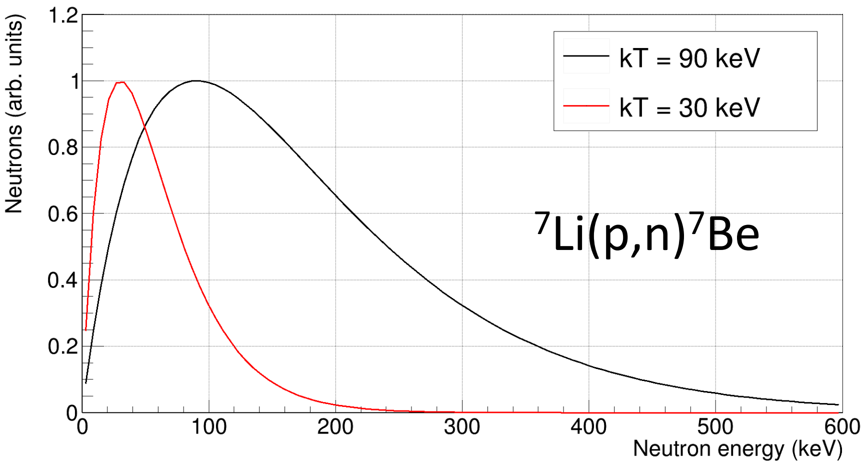


- In general, if there are discrepancies between evaluations, we agree with JEFF3.3
- A new measurement to solve the problem was indeed necessary.
- Let's wait for the results of our data!

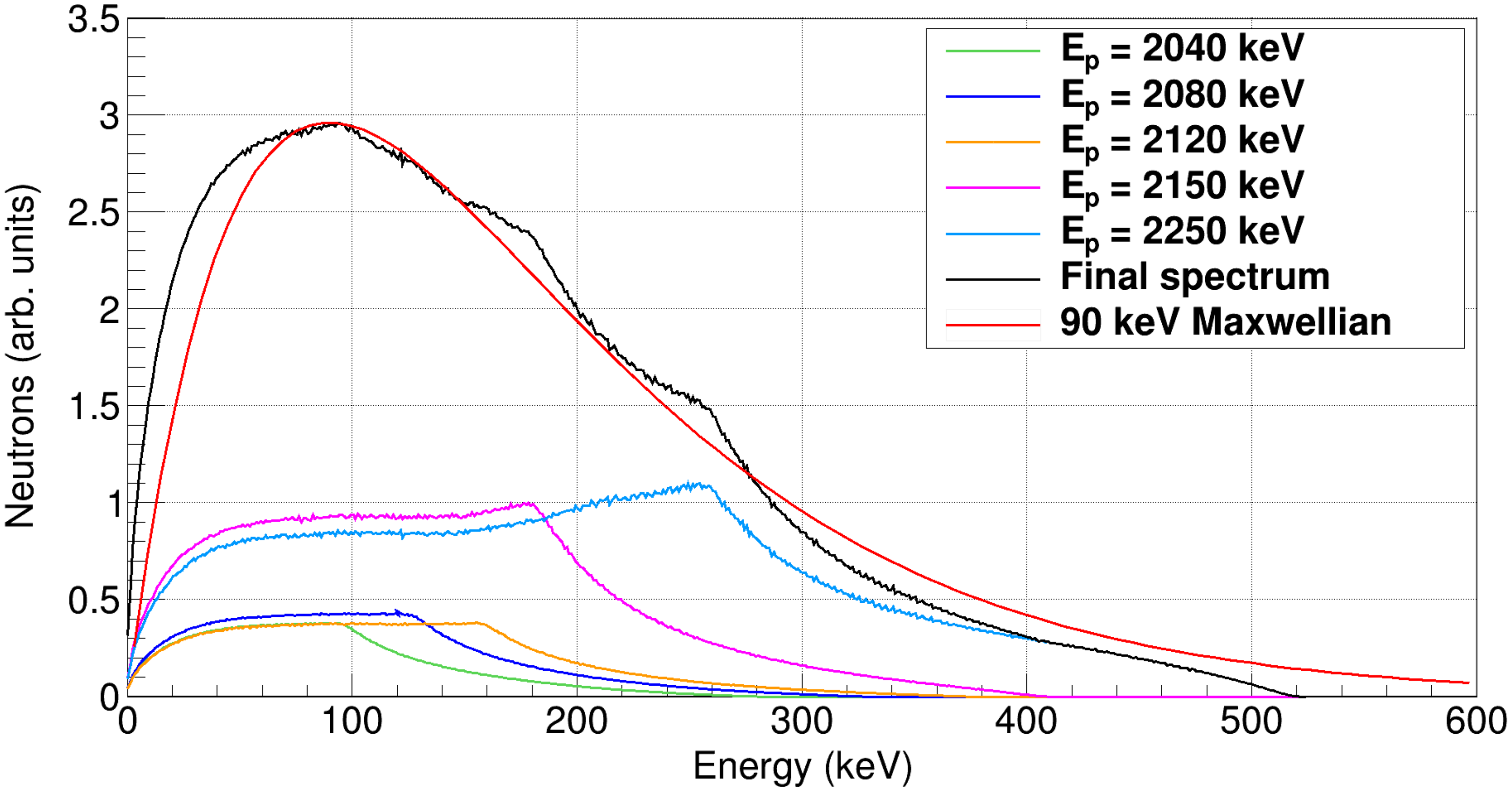
^{50}Cr MACS measurement at HiSPANoS@CNA

	Time of flight technique	Neutron activation
Energy and resonance shape	Very well defined	Limited “resolution” (MB distribution)
Absolute value	Susceptible to systematic effects	Very accurate (“easily” ~5%)

- An integral measurement can be very helpful with the analysis.
- ***“Development of a 90 keV Maxwellian neutron spectrum and measurement of the 30 & 90 keV ^{50}Cr MACS for criticality safety”*** (H2020-ARIEL Transnational Access).



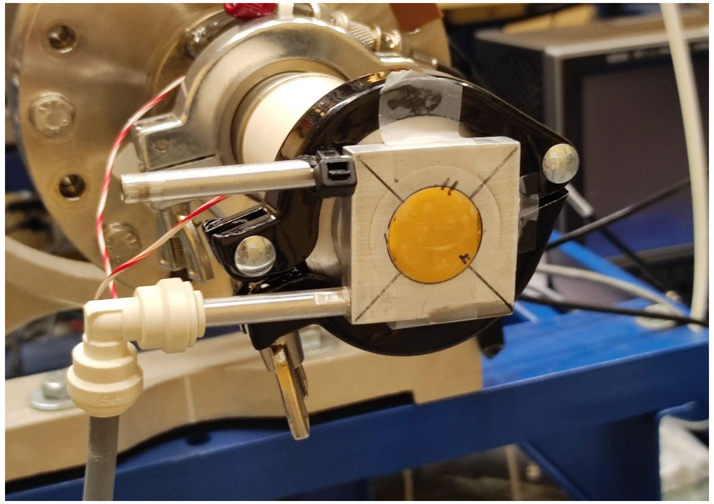
^{50}Cr activation: 90 keV neutron distribution



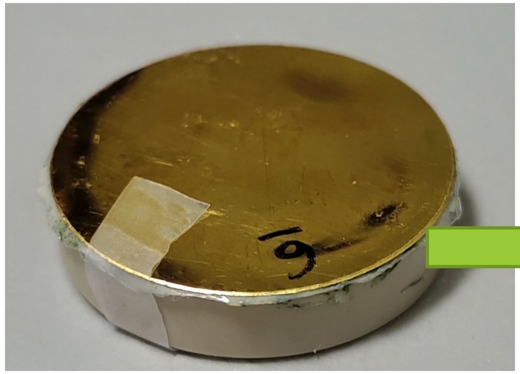
- A 30 keV spectrum can be produced with E_p = 1912 keV.
- For the 90 keV spectrum we need a linear combination of different proton energies.

^{50}Cr activation: set-up

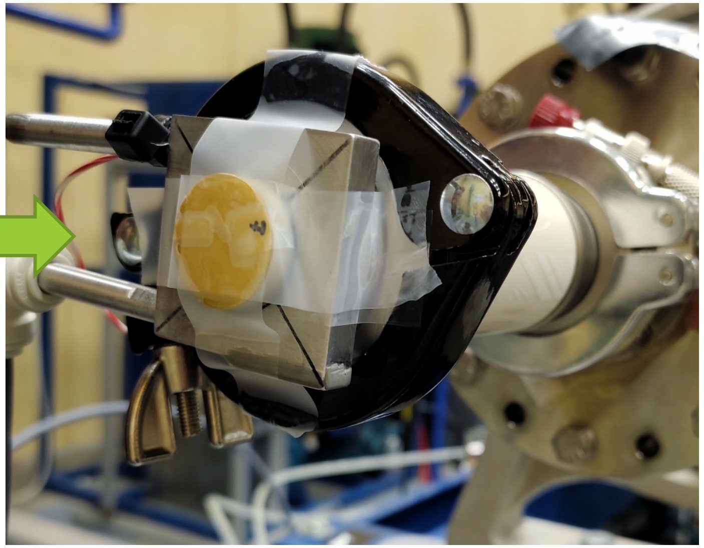
Metallic Li for higher production → cooled target

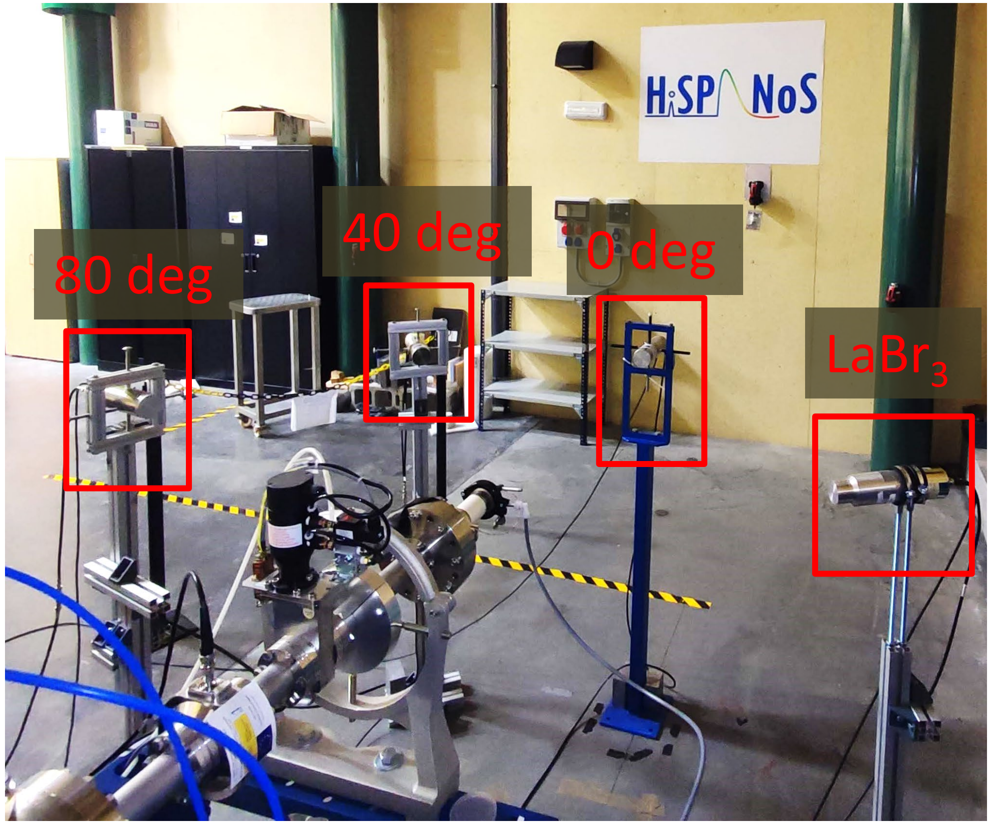


^{197}Au irradiation for activation checks



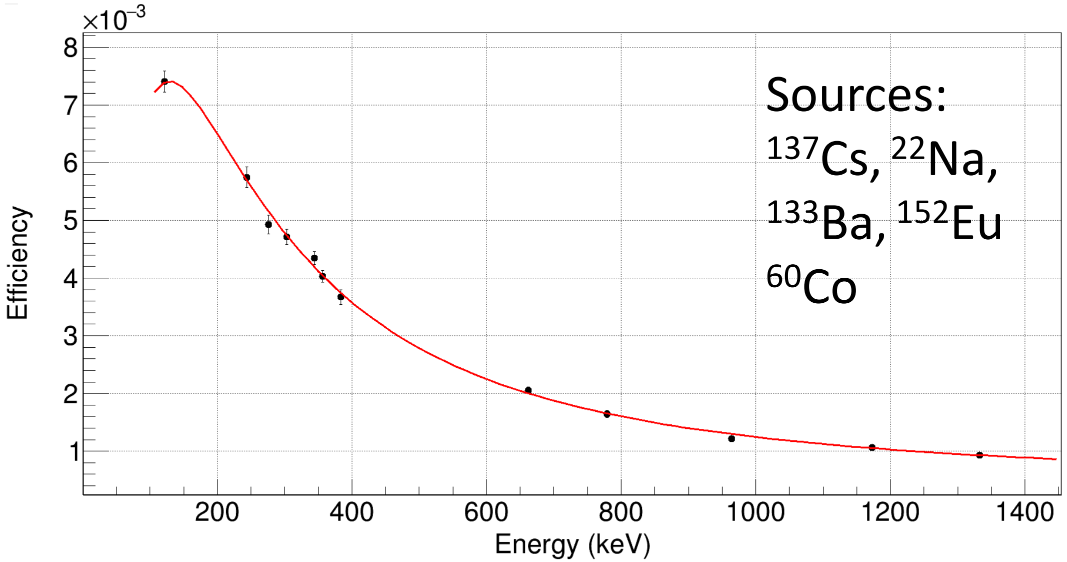
^{197}Au + ^{50}Cr + ^{197}Au sample

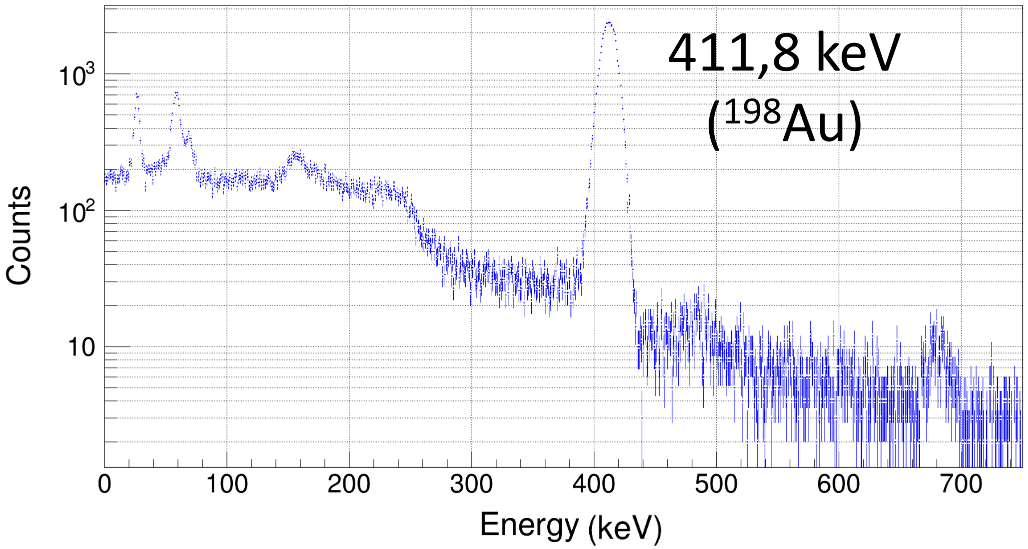


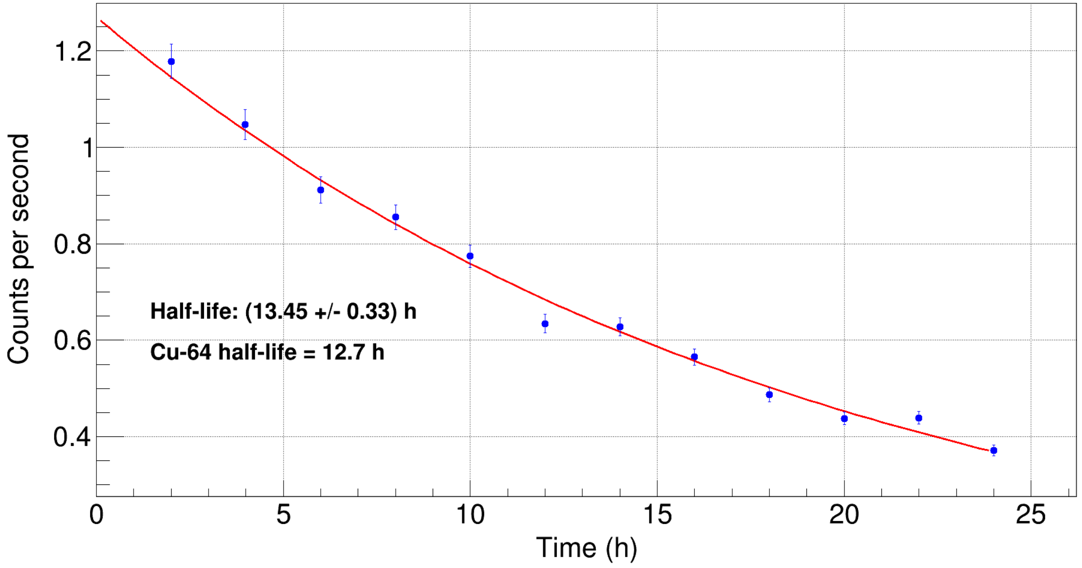


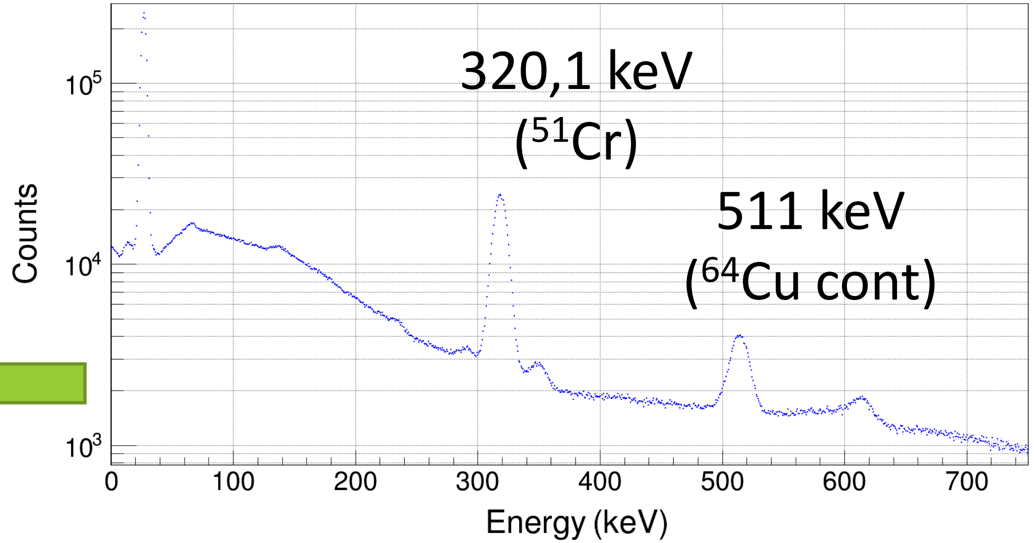
- 3 Lithium-glass neutron monitors
- 1 $LaBr_3$ for ^{7}Be decay
- 1 $LaBr_3$ for ^{198}Au and ^{51}Cr decay

^{50}Cr activation: preliminary results

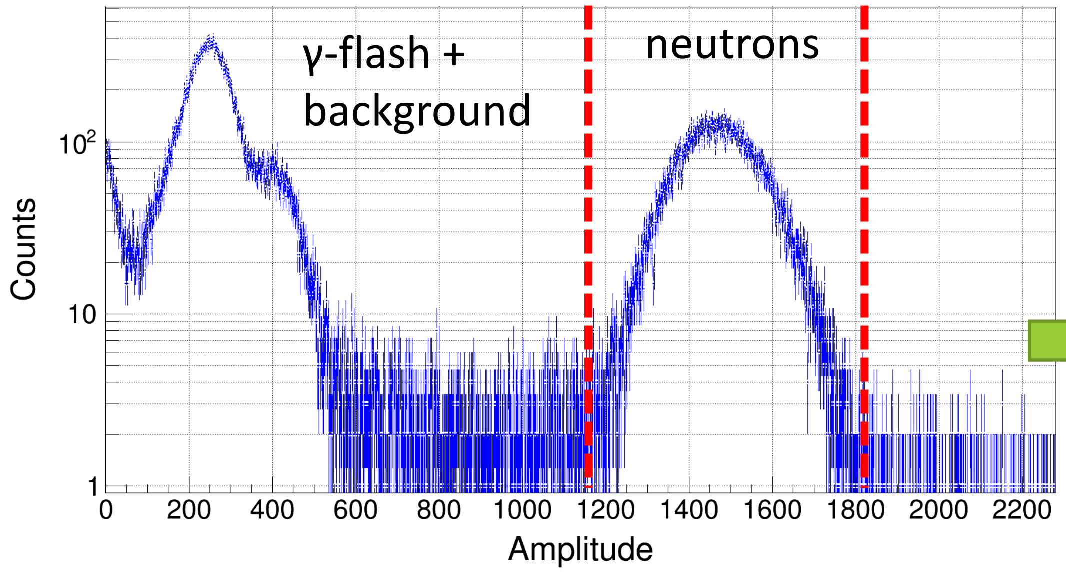


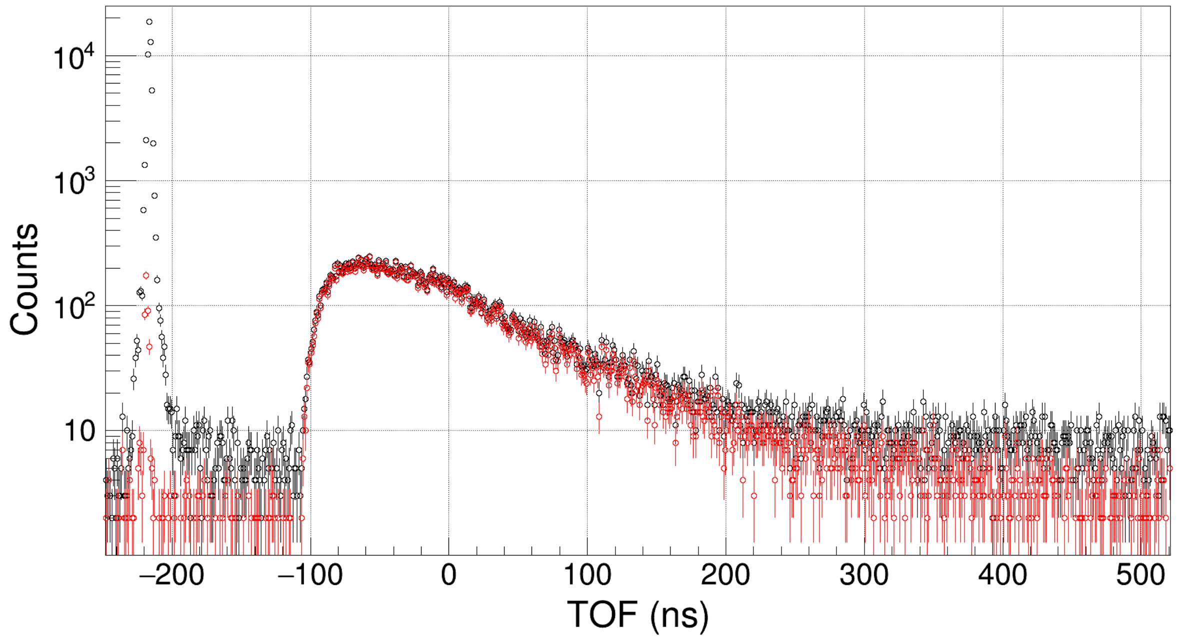






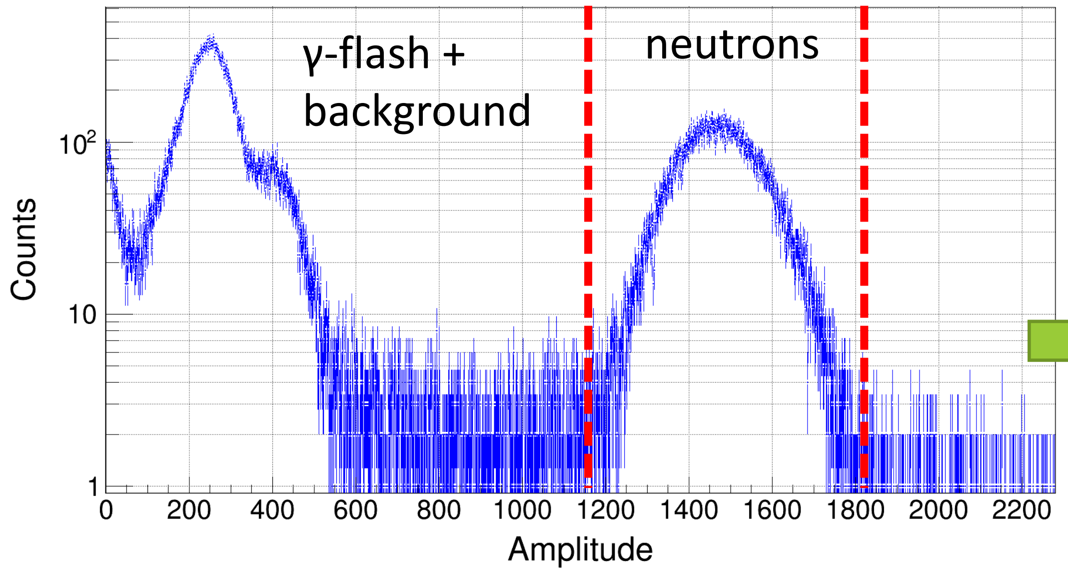
^{50}Cr activation: preliminary results

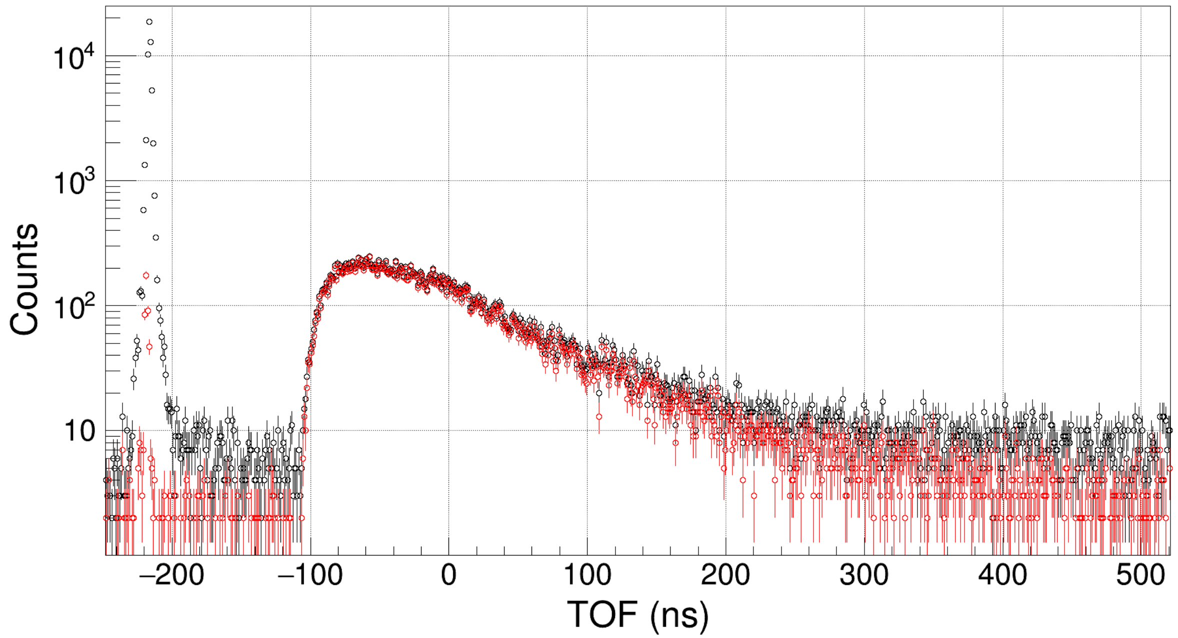




- Every neutron flux measured with Li-glass detectors at 3 angles.
- Flight paths of 50 cm (E_p = 1912 & 2080 keV) and 100 cm (higher energies).
- Cuts in signal amplitude eliminate the gamma flash and background

^{50}Cr activation: preliminary results





- Every neutron flux measured with Lithium-glass detectors at 3 angles.
- Flight paths of 0,5m (Ep = 1912 & 2080 keV) and 1m (the rest).
- With cuts in deposited energy we remove the gamma flash.
- A lot of work ahead!

$$SACS = \frac{N_{^{198}Au}}{N_{^{7}Be}{}^{nat}}$$

$$MACS = \frac{2}{\sqrt{\pi}} \frac{\langle \sigma_{MB} \rangle}{\langle \sigma_{\Phi} \rangle} SACS$$

Less than 3% difference*

HiSPANoS SACS	584 mb
HiSPANoS MACS	631 mb
KADONIS MACS	648 mb

*(Uncertainties to be estimated)

Summary & Outlook

- IAEA request responded: improving $^{50,53}Cr$ $\sigma(n,\gamma)$ to 8-10% accuracy at 1-100 keV
- Two experiments:
  - n_TOF@CERN, Summer'22 (H2020-ARIEL Scientific Visit).
  - HiSPANoS@CNA, March'23 (H2020-ARIEL Transnational Access).
- Preliminary results are very promising.

NEXT STEPS:

- **n_TOF experiment data analysis**
  - Identify (and correct?) systematic effects
  - Implement Pulse Height Weighting Function
  - Resonance analysis with SAMMY
- **HISPANOS activation data analysis:**
  - Study MACS at 30 and 90 keV for ^{197}Au
  - Extract MACS at 30 and 90 keV for ^{50}Cr

Thank you!

Carlos GUERRERO
cguerrero4@us.es