Horia Hulubei National Institute for R&D
in Physics and Nuclear Engineering

EUROPEAN ORGANIZATION FOR NUCLEAR RESEARCH

Proposal to the ISOLDE and Neutron Time-of-Flight Committee

The neutron capture cross section of ^{124}Sn and its impact on neutrinoless double β decay searches

September 25, 2024

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Why do we need $^{124}Sn(n,\gamma)$ cross section data?

Motivation A

A few groups around the world search for the $0\nu\beta\beta$ of $^{124}Sn \dashrightarrow {}^{124}Te$:

India (In progress)

Lab: *Indian Neutrino Observatory (INO) and TATA institute*

Detector: Sn cryogenic superconducting bolometer (TIN.TIN -The India based TIN detector)

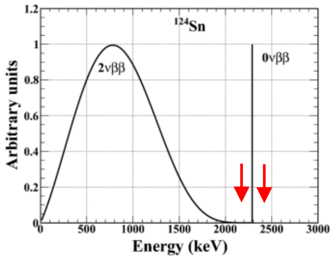
Nanal, Vandana.EPJ Web of Conferences, EDP Sciences 66 (2014)

South Korea (Done)

Lab: *YangYang Underground Laboratory (Y2L)-2009*

Detector : tin-loaded liquid scintillator for an active source-detector technique

Hwang, M. J., et al., Astroparticle Physics 31.6 (2009): 412-416.



Singh, M.K., et al. Indian J Phys 94, 1263-1270 (2020).

Background assessment of $0\nu\beta\beta$ decay searches

- ^{124}Sn is one of the promising $0\nu\beta\beta$ candidates
- They measure the $0\nu\beta\beta$ decay peak which is at an energy equal to the Q-value of the reaction: 2292.7(4) keV
- Golden channel: decay rate <-> direct access to neutrino mass & CP violating Majorana phases (can not be probed by ν oscillations experiments)
- $0\nu\beta\beta$ is a second order weak interaction process and the event rates are very low ($T_{1/2} > 10^{17}$ y)*
- They are extremely sensitive to **background signals which can mimic the signal of interest** -> neutron-induced background in the Q-value region*

*Dawson, J., et al., Physical Review C 78.3 (2008): 035503

Gupta, G., et al., Applied Radiation and Isotopes 158 (2020): 108923

Motivation A

Background assessment of $0\nu\beta\beta$ decay experiments

↓

Background: Neutrons

Cosmic ray-induced neutrons

Rocks (^{nat}Th, ^{nat}U) -> spontaneous fission neutrons
-> (α,n)-induced neutrons

Motivation A

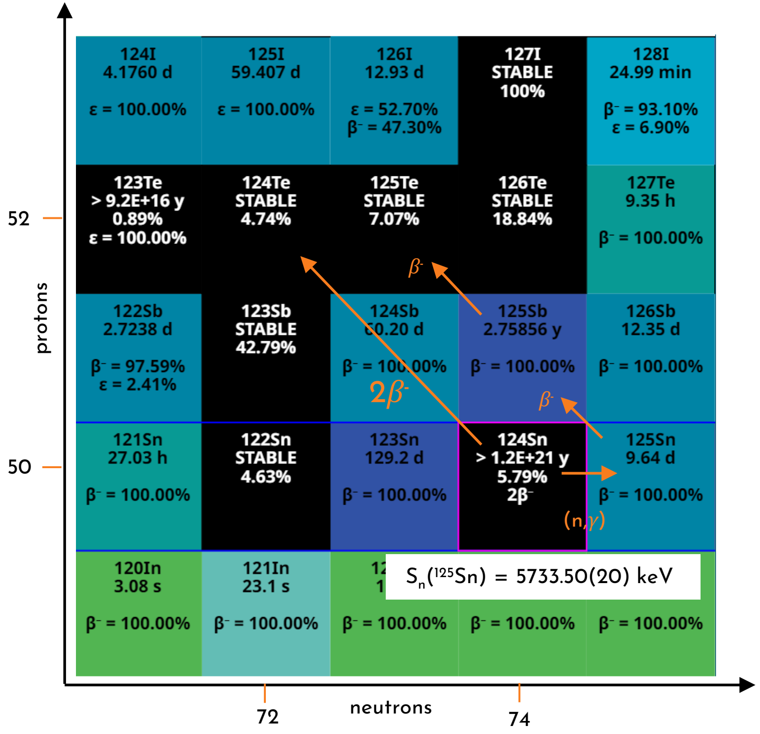
Background assessment of $0\nu\beta\beta$ decay experiments

ISSUE: γ rays following neutron capture on ^{124}Sn can mimic the $0\nu\beta\beta$ decay signal!!!

- Literature* shows that, after activating a ^{124}Sn sample with a neutron thermal flux and measuring the delayed γ following neutron capture and subsequent β^- decays with a HPGe detector, a strong summing peak of 2288.2 keV has been seen*

$$Q_{0\nu\beta\beta} = 2292.7\ \text{keV}$$

- Also, a (worrying) 30% simulation vs. experiment difference was observed*



*Gupta, G., et al., Applied Radiation and Isotopes 158 (2020): 108923

Why do we need ^{124}Sn(n,γ) cross section data?

Motivation B

Problem in reactor fuel depletion calculations for ^{125}Sb:

Annals of Nuclear Energy 161 (2021) 108441

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Annals of Nuclear Energy

journal homepage: www.elsevier.com/locate/anucene

ELSEVIER

Review

Monte Carlo neutronics benchmarks on nuclear fuel depletion: A review

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A B S T R A C T

Monte Carlo (MC) neutronics codes are used widely for academic and industrial needs. Several schemes of coupling MC neutronics code with isotope generation and depletion code exist, which are used for performing nuclear fuel depletion simulations. These simulations can estimate the inventory of isotopes in neutron irradiated nuclear reactor fuel. However, the accuracy of these simulations shall be validated through experiments. MC codes are seldom validated by isotopic benchmarks compared to criticality benchmarks. This work compiles and analyzes the fuel depletion benchmarks and validations used to analyze the performance of MC-based fuel depletion neutronics codes. Analyses of these benchmarks and validations showed that the computed concentrations of ^{133}Cs, ^{135}Cs, ^{137}Cs, ^{148}Nd, ^{239}Pu, ^{240}Pu, and ^{241}Pu in the irradiated fuel by the depletion codes agreed with the measured values within 10% error. However, the computed concentrations of ^{125}Sb, ^{242}Cm, ^{243}Cm, ^{244}Cm, ^{245}Cm, and ^{246}Cm had errors more than 15% compared to the measured values. Ventina depletion code showed the most accurate predictions for the greatest number of isotope concentrations compared to ORIGEN2 and CINDER90.

Motivation B

NUREG/CR-6798
ORNL/TM-2001/259

Isotopic Analysis of High-Burnup PWR Spent Fuel Samples From the Takahama-3 Reactor

Manuscript Completed: May 2002
Date Published: January 2003

Table 12 Comparison of analyses and calculations of Takahama-3 SF95-3

Nuclide	SF95-3 Measured	SF95-3 SAS2H	SF95-3 HELIOS	C/E SAS2H	C/E HELIOS
	Measured[a] g/MgU				
^{234}U	1.873E+02	2.401E+02	1.92E+02	1.28	1.02
^{235}U	1.326E+04	1.299E+04	1.36E+04	0.98	1.02
^{236}U	4.911E+03	4.904E+03	4.95E+03	1.00	1.01
^{238}U	9.338E+05	9.340E+05	9.34E+05	1.00	1.00
^{238}Pu	1.539E+02	1.491E+02	1.53E+02	0.97	1.00
^{239}Pu	6.194E+03	6.043E+03	6.54E+03	0.98	1.06
^{240}Pu	2.186E+03	2.219E+03	2.21E+03	1.02	1.01
^{241}Pu	1.486E+03	1.413E+03	1.51E+03	0.95	1.01
^{242}Pu	4.516E+02	4.571E+02	4.16E+02	1.01	0.92
^{241}Am	3.310E+01	3.732E+01	3.93E+01	1.13	1.19
^{242m}Am	7.877E−01	8.130E−01	8.14E−01	1.03	1.03
^{243}Am	8.047E+01	9.360E+01	7.98E+01	1.16	0.99
^{242}Cm	1.964E+01	1.178E+01	1.64E+01	0.60	0.83
^{243}Cm	3.720E−01	3.006E−01	3.02E−01	0.81	0.81
^{244}Cm	2.562E+01	2.507E+01	2.36E+01	0.98	0.92
^{245}Cm	1.396E+00	8.783E-01	1.30E+00	0.63	0.93
^{246}Cm	1.049E−01	9.553E−02	8.97E−02	0.91	0.86
^{137}Cs	1.347E+03	1.338E+03	1.31E+03	0.99	0.97
^{134}Cs	1.404E+02	1.204E+02	1.06E+02	0.86	0.76
^{154}Eu	2.525E+01	2.476E+01	2.76E+01	0.98	1.09
^{144}Ce	4.560E+02	4.359E+02	4.22E+02	0.96	0.92
^{125}Sb	3.733E+00	7.952E+00	1.01E+01	2.13	2.69
^{106}Ru	1.360E+02	1.713E+02	1.67E+02	1.26	1.23
^{142}Nd	2.116E+01	1.802E+01	N/A	0.85	N/A
^{143}Nd	9.299E+02	9.046E+02	9.10E+02	0.97	0.98
^{144}Nd	9.347E+02	9.457E+02	9.29E+02	1.01	0.99
^{145}Nd	7.392E+02	7.395E+02	7.35E+02	1.00	0.99
^{146}Nd	7.340E+02	7.418E+02	7.34E+02	1.01	1.00
^{148}Nd	3.979E+02	3.946E+02	3.97E+02	0.99	1.00
^{150}Nd	1.896E+02	1.866E+02	1.87E+02	0.98	0.99
Burnup (GWd/MTU)[b]	35.42				

[a] At discharge, except for ^{239}Pu which includes contribution from ^{239}Np precursor.
[b] Burnup estimated using ^{148}Nd analysis.

Motivation B

Comparison of Gamma Spectroscopy Measurements to Simulation

	Measured Activity (Ci/g DUO_2)	Standard Deviation (Ci/g DUO_2)	Simulation Activity (Ci/g DUO_2)	S/E[a]
^{144}Ce	9.60E−02	2.29E−03	8.43E−02	0.88 ± 0.09
^{134}Cs	2.01E−03	5.24E−05	2.21E−03	1.10 ± 0.11
^{137}Cs	1.41E−02	2.32E−04	1.32E−02	0.94 ± 0.10
^{154}Eu	1.80E−04	1.12E−05	2.00E−04	1.11 ± 0.13
^{125}Sb	1.16E−03	4.63E−05	1.75E−03	1.51 ± 0.16
^{95}Zr	6.53E−03	1.96E−04	6.00E−03	0.92 ± 0.10

[a] S/E = simulation/measurement.

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DOI: http://dx.doi.org/10.13182/NT16-76

ANS

Experimental and Computational Forensics Characterization of Weapons-Grade Plutonium Produced in a Fast Reactor Neutron Environment

The final step in this part of the investigation was to compare the quantities of the various fission products predicted from the MCNPX burnup simulation to those measured using gamma spectroscopy. Both the simulated and measured values were normalized to the mass of DUO_2 in order to account for differences in the simulated mass and the mass of the actual sample. These comparative data are shown in Table V. As can be seen in Table V, the difference between the simulated and measured values for most of the isotopes is equal to or less than 12%; however, the activity predicted for ^{125}Sb was over 50% larger than the measured activity. Upon further investigation, it was discovered that ^{125}Sb is a particularly troublesome nuclide to

Motivation B

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Experimental and Computational Forensics Characterization of Weapons-Grade Plutonium Produced in a Fast Reactor Neutron Environment

中图分类号:TL32 文献标志码:A 文章编号:1000-6931(2022)05-0952-09
doi:10.7538/yzk.2021.youxian.0872

Research on Benchmarking Method of Burnup Database

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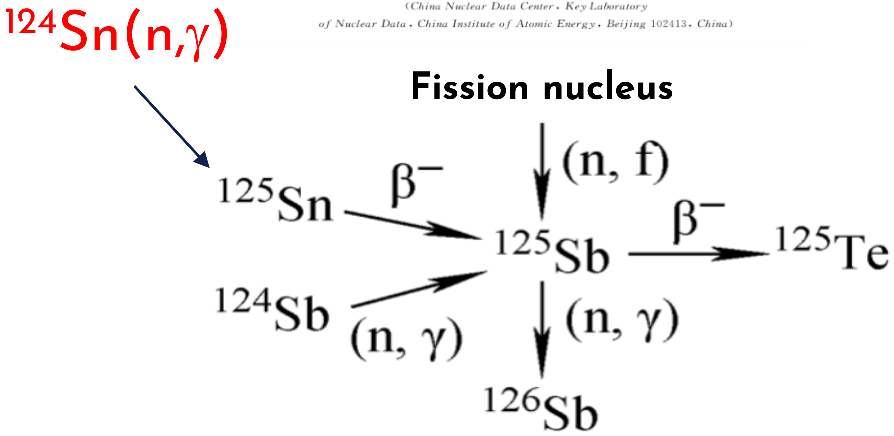
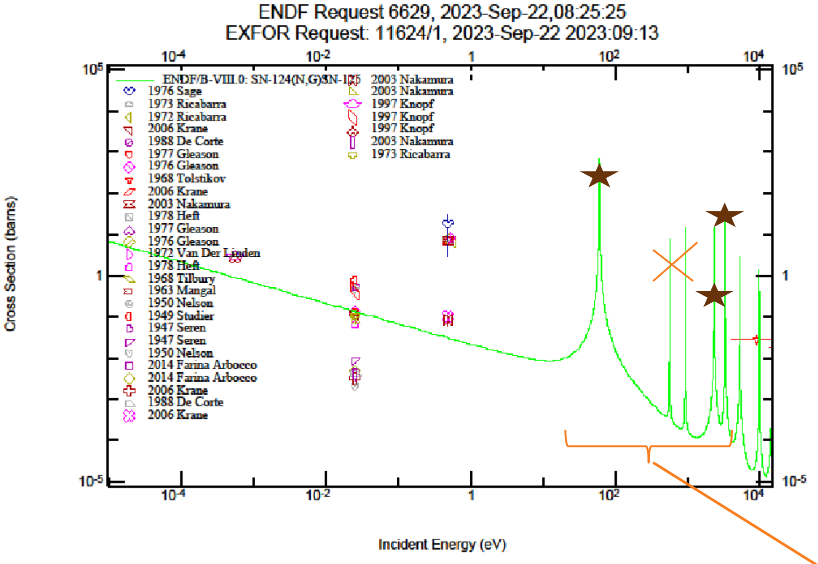
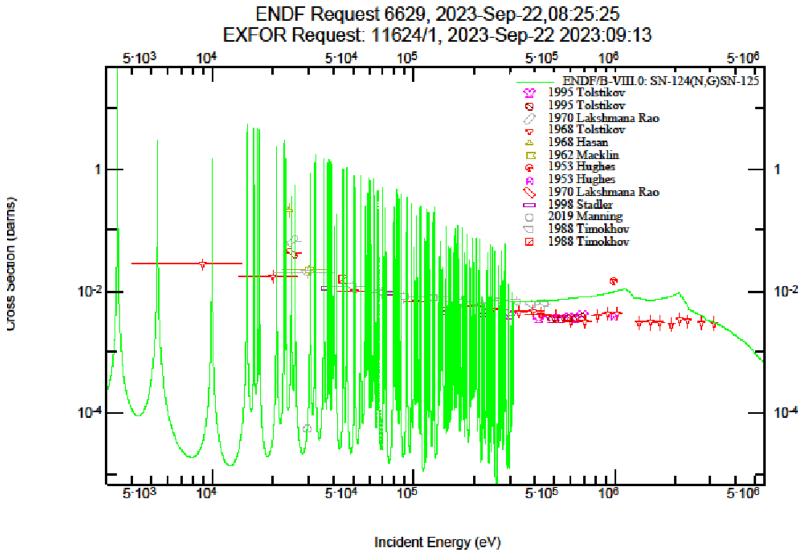


Figure: Burnup chain adjacent to 125Sb

Present status of ^{124}Sn(n,γ) cross section data

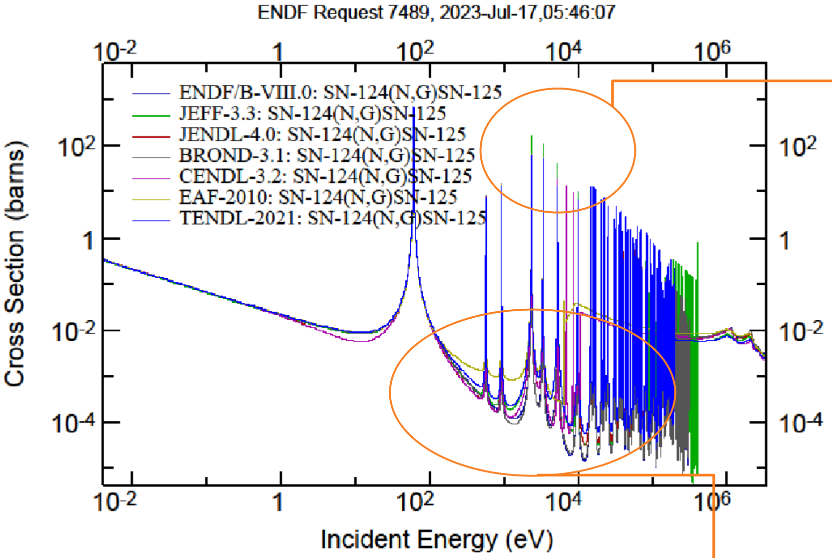


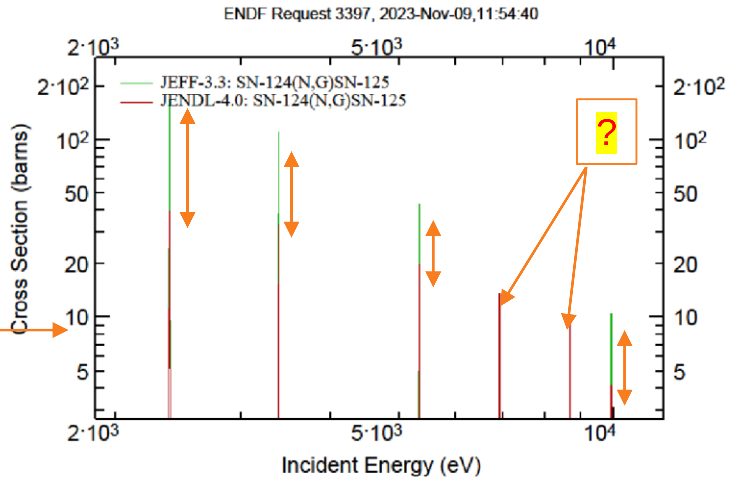


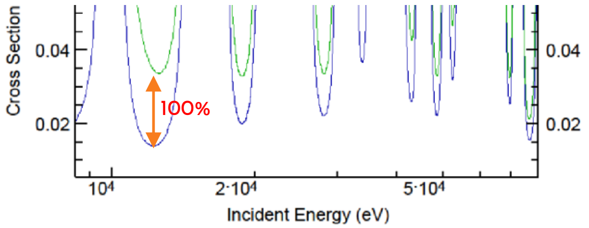
HPGe-based measurement

A. Kimura et al., EPJ Web of Conferences 146, 11031 (2017) in ANNRI at MLF/J-PARC (ND2016 proceedings)

Present status of ^{124}Sn

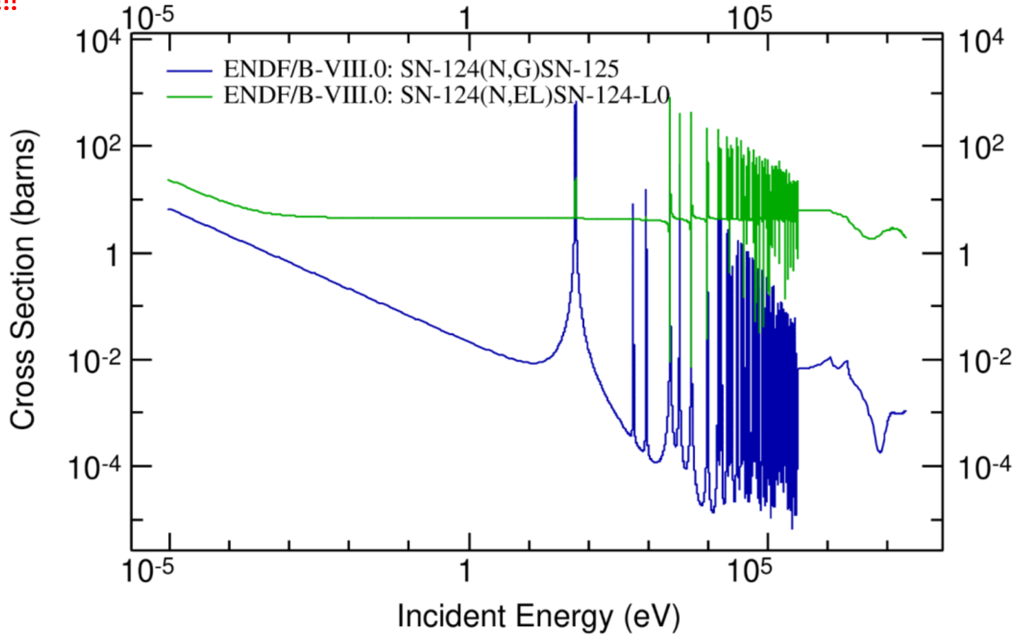






Experimental Details and Difficulties

Elastic to capture ratio -> very unfavorable!!!



Experimental Details and Difficulties

Elastic to capture ratio -> very unfavorable!!!
Neutron energy resolution is not paramount -> AVERAGE CROSS SECTION (above 20 keV)

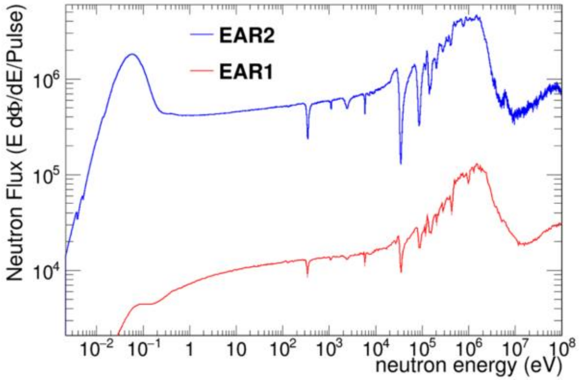


Fig. 15. Comparison between the evaluated neutron flux in EAR2 (blue) and in EAR1 (red). The increase at the new measuring station is on average a factor 40.

Table: Average distance in-between resonances against energy resolution at EAR1 versus EAR2

Neutron Energy range	No. of resonances (ENDF/B-VIII.0)	Avg. distance between Capture Resonances	Energy resolution in EAR1 *	Energy resolution in EAR2 *
50 eV-10 keV	7	1200 eV	10 eV	200 eV
10-100 keV	67	1300 eV	300 eV	3000 eV
100-200 keV	68	1500 eV	800 eV	-
200-314 keV	47	2400 eV	1200 eV	-

Goal: resolve resonances up to 15-20 keV

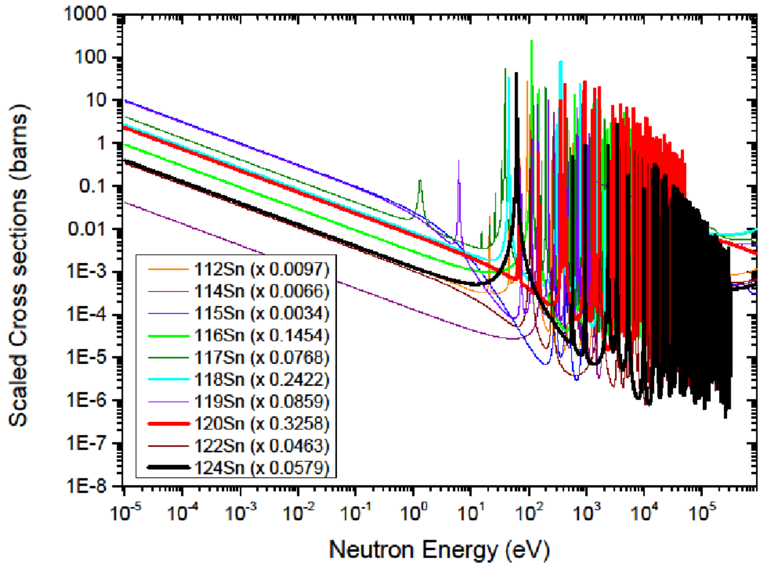
*Guerrero, C., et al. The European Physical Journal A 49 (2013): 1-15.

*Lerendegui-Marco, J., et al. The European Physical Journal A 52 (2016): 1-10.

Experimental Details and Difficulties

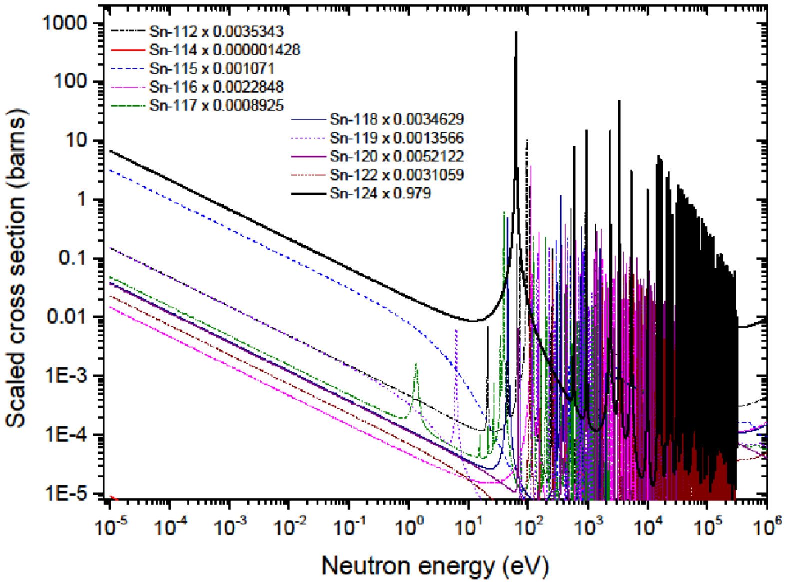
Enriched sample -> paramount!

^{nat}Sn



Comparison of ^{124}Sn resonances (in black) with the resonances from other Sn isotopes (cross section data from ENDF/B-VIII.0)

Enriched ^{124}Sn



Scaled cross section associated to the other tin isotopes (scaled by the abundances of the ^{124}Sn enriched sample we plan to use)

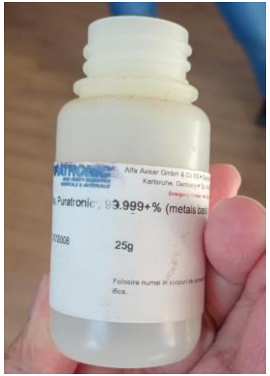
Experimental Details and Difficulties

Available @ Nuclear Physics Institute
Czech Academy of Sciences (I. Tomandl, F. Marek)

Sample(s) Details

- 3 disks (1 g each)
- 97.9% enrichment
- 10 mm diameter
- 1.8 mm thickness

- 25 g (natural Tin rod)
- 99.999% Purity
- 13 mm diameter

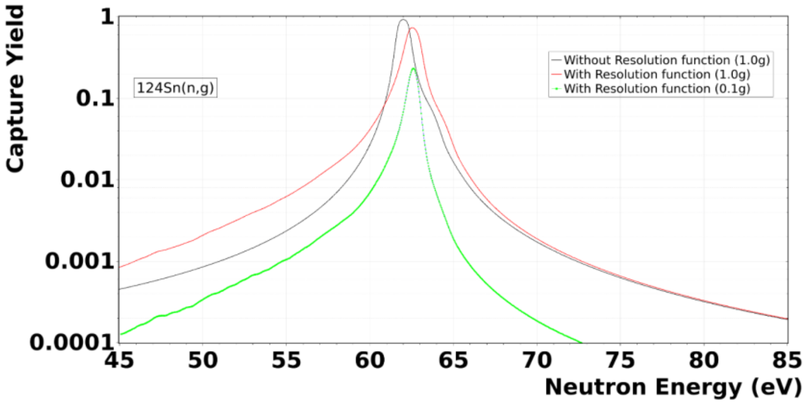


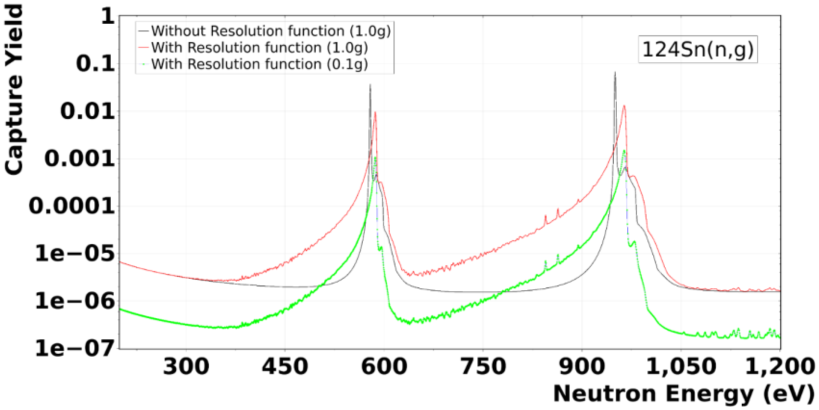
Available @ IFIN-HH Target lab

Experimental Details and Difficulties [EAR2]

Resolution function and multiple scattering -> SAMMY-based calculations

- In EAR2 : 1.0 g sample (0.00618 at/b)
- In EAR2 : 0.1 g sample (0.000618 at/b)

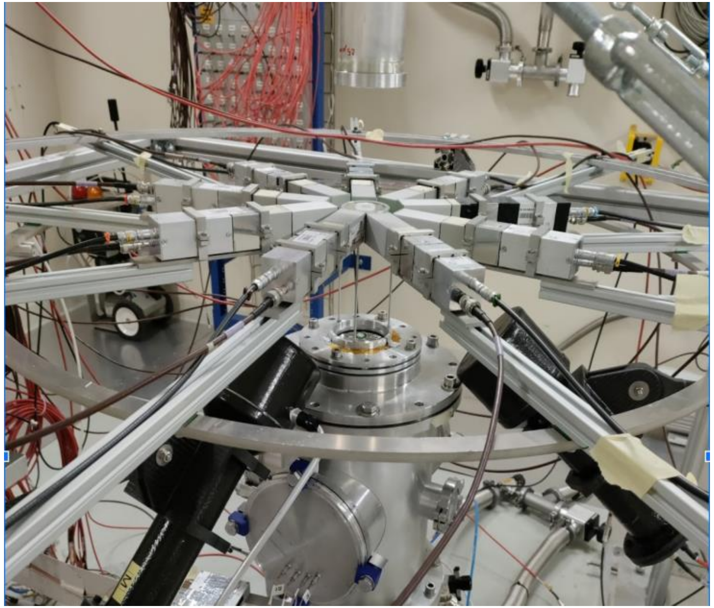




Experimental Details [EAR2]

How will we measure?

- Setup of 9 sTED detectors
- Measure from thermal to highest reachable energy by these detectors in EAR2
- Use an enriched sample: 97.9% of ^{124}Sn
- Thin-thick approach:
  - Up to 15-20 keV-> thick (1.0 g)
  - Average xs above 20 keV-> thick (3.0 g)
  - First resonance -> thin (0.1 g)
- **Ancillary**: also irradiate ^{nat}Sn, ^{197}Au, ^{nat}C, ^{nat}Pb samples + Empty



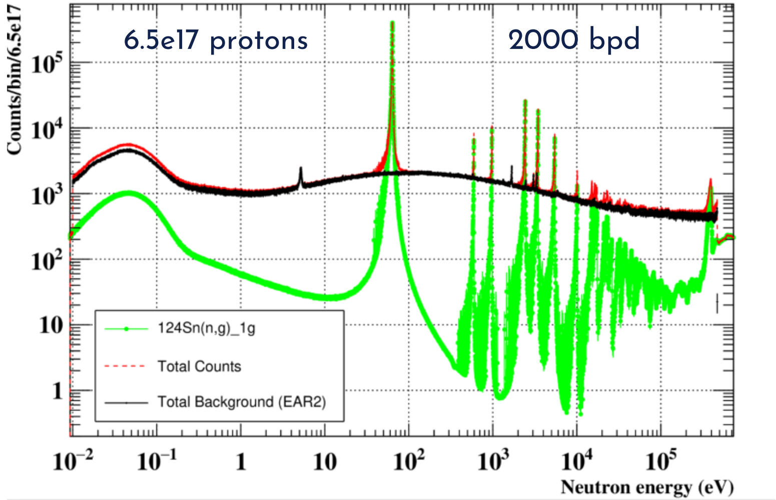
The 9 sTED's setup used in EAR2 for the $^{209}Bi(n,\gamma)$ campaign

Proton Request [EAR2]

Counts estimation:

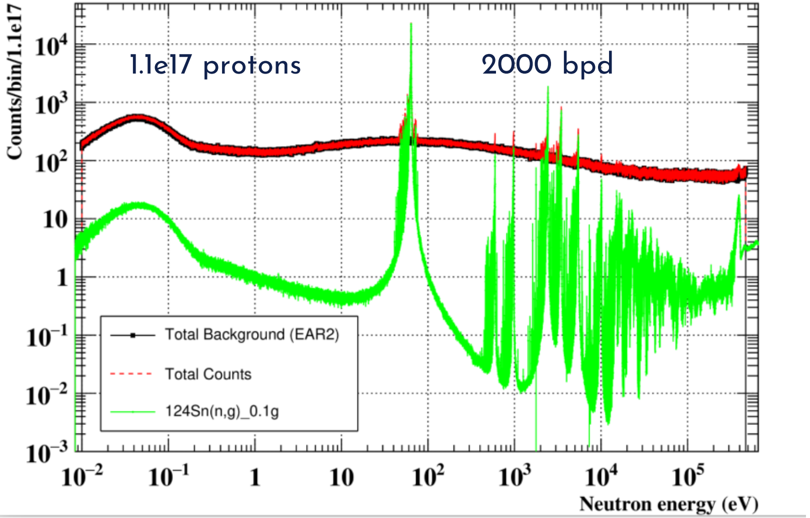
- 1.0 g sample, 0.00618 at/b (10 mm-diameter)

Eff-4.5% (9 sTEDs), JEFF-3.3



- 0.1 g sample, 0.000618 at/b (10 mm-diameter)

Eff-4.5% (9 sTEDs), JEFF-3.3



Total background = empty+in-beam γ+elastic

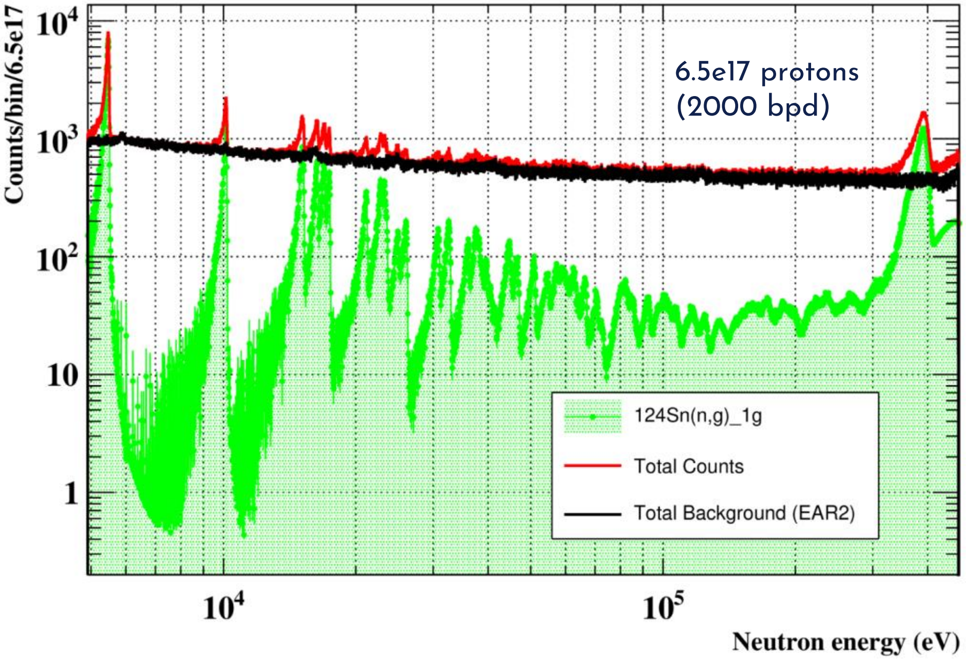
Total Counts = 124Sn(n,g)_0.1/1g + total background

Proton Request [EAR2]

Counts estimation:

- 1.0 g sample, 0.00618 at/b (10 mm-diameter)

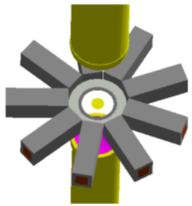
Eff-4.5% (9 sTEDs), JEFF-3.3



Proton Request [EAR2]

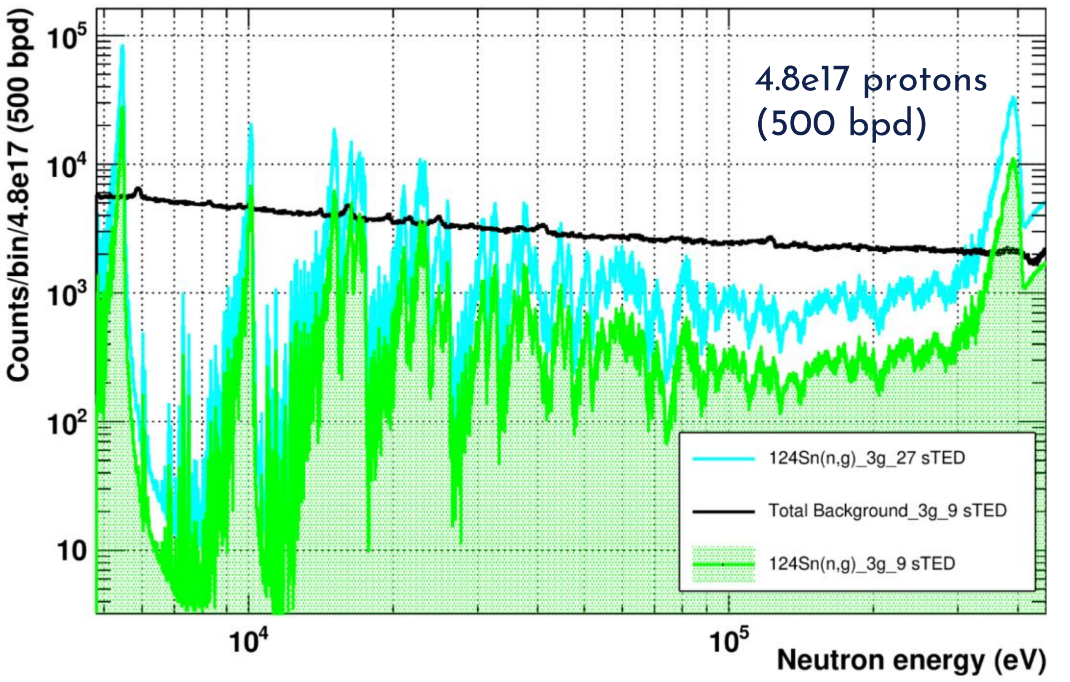
Counts estimation:

- 3.0 g sample, 0.01856 at/b (10 mm-diameter)



Eff-4.5%, JEFF-3.3 [9 sTEDs]

Eff-13.5%, JEFF-3.3 [27 sTEDs]



Proton Request [EAR2]

For ^{124}Sn (1 g sample) -> 6.5×10^{17} protons

For ^{124}Sn (0.1 g sample) -> 1.1×10^{17} protons

For ^{124}Sn (3 g sample) -> 4.8×10^{17} protons

For ^{nat}Sn -> 1.1×10^{17} protons

Ancillary: normalisation (^{197}Au) + background estimation (^{nat}Pb, ^{nat}C, Empty) -> 6.5×10^{17} protons

In total: 2.0×10^{18} protons

Summary

- ❑ Motivation:
- ✔ A: Neutrinoless double β decay searches
- ✔ B: Nuclear fuel depletion calculations: ^{125}Sb problem

- ❑ Status of data:
- ✔ No ToF neutron capture data exist to map out first resonances below 10 keV, questionable resonances…

- ❑ Experiment goals:
- ✔ To provide for the first time reliable, low uncertainty neutron capture ToF data from thermal to 15-20 keV -> resonance parameters for the most intense resonances
- ✔ Possibly average cross section above 20 keV

- ❑ Impact:
  - ✔ To better quantify the neutron-induced background for neutrinoless double β decay ($0\nu\beta\beta$) searches
  - ✔ Optimistically, to at least partially clarify the differences between various evaluations -> improve on ^{125}Sb problem

Proton request	
Sample	**Protons**
^{124}Sn (1 g)	6.5×10^{17}
^{124}Sn (0.1 g)	1.1×10^{17}
^{124}Sn (3.0 g)	4.8×10^{17}
^{nat}Sn	1.1×10^{17}
^{nat}C, ^{nat}Pb, ^{197}Au + Empty	6.5×10^{17}
Total	$\mathbf{2.0\times10^{18}}$

Thanks

Do you have any questions?

https://www.nipne.ro/proiecte/pn3/ntof
/
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