Optimised of the neutron yield calculation from (α,n) reactions with modified SOURCES4 code

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Outline

- Introduction: neutron production in (α,n) reactions in SOURCES4
- 'Optimised' cross-sections and comparison with data.
  - Cross-checks with alpha beam data.
- Neutron yields and spectra from 'optimised' SOURCES4 versus data for radioactive decay chains.
- Conclusions

See my talk at LRT2024 (and contribution to the proceedings) for previous update.

Preprint available: Parvu et al., arXiv:2408.10910 (2024)

SOURCES4

- SOURCES4A/4C: W.B. Wilson, et al., SOURCES4A: a code for calculating (α,n), spontaneous fission, and delayed neutron sources and spectra, Technical Report LA-13639-MS, Los Alamos, 1999;
- Working historically with SOURCES4A; no noticeable difference for our goals.
- The probability for an alpha particle to produce a neutron by interacting with a nuclide i (N_i is the number density of atoms of nuclide i):

$$P\left(E_\alpha\right) = \int_0^{E_\alpha} \frac{N_i \sigma_i \left(E\right)}{\left(-\frac{dE}{dx}\right)} dE$$

- Stopping power cross-sections from tables compiled by Ziegler.
- Approximation of thick target.

Recent modifications to SOURCES4A

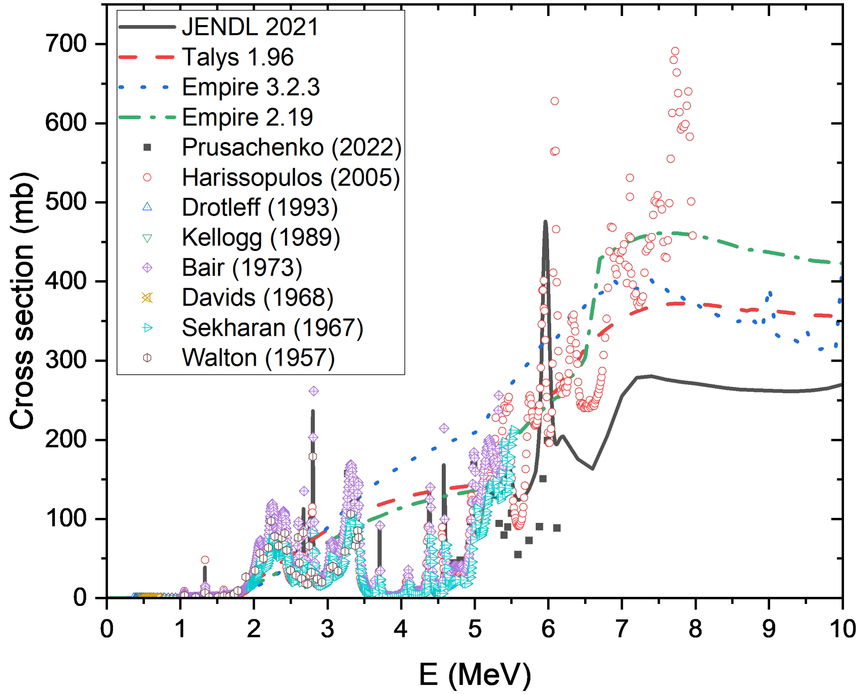
- The **maximum number of discrete nuclear levels** for the product nuclides was increased from 100 to **500**.
- The **maximum number of target elements** was increased from 20 to **110**.
- The cross-sections for (α, n) reactions in SOURCES4 have been taken from reliable experimental data (including some recent ones) where possible and complemented by the calculations with TALYS1.96, EMPIRE2.19/3.2.3, and JENDL-5 where the data were scarce or unavailable.
- Various sets of cross-sections are available in the library → **we just recommend the most reliable in our opinion.**
- The code was modified so **the user does not need to change the order of the cross-section or branching ratios anymore**, but only to indicate which one to use (the order that the cross-section appears in the library) → **data selection option in tape1**.

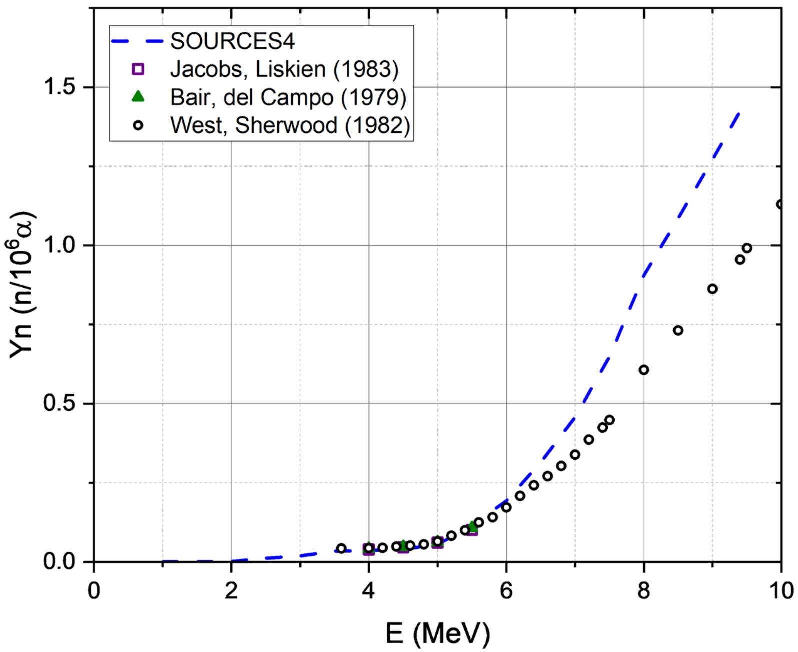
Previous modifications: Carson et al. Astropart. Phys., 21 (2004) 667; Lemrani et al. NIMA 560 (2006) 454; Tomasello et al. NIMA, 595 (2008) 431; Tomasello et al. Astropart. Phys., 34 (2010) 70, Kudryavtsev et al. NIMA 972 (2020) 164095; Kudryavtsev et al. AIP Conf. Proc. 2908 (2023) 1, 100003.

Advantages and disadvantages

- Advantages
  - Flexible libraries of cross-sections and branching ratios
  - Fast calculation
  - Total neutron spectra; spectra from interactions on individual isotopes and from the variety of radioisotopes in a single calculation; spectra from the ground state and different excited states.
- Disadvantages:
  - Written long time ago (but cross-sections can be added/replaced)
  - Written in Fortran (but no need to intervene if the code works)
  - No gammas generated from de-excitation of final state nuclei (same for other codes)
  - Cannot read ENDF format (but if you know ENDF format, converting the cross-section data into the SOURCES4 format is not a big deal).
  - Cannot deal with 'surface' contaminations/problems.

Cross-sections for ^{13}C (low-A target)

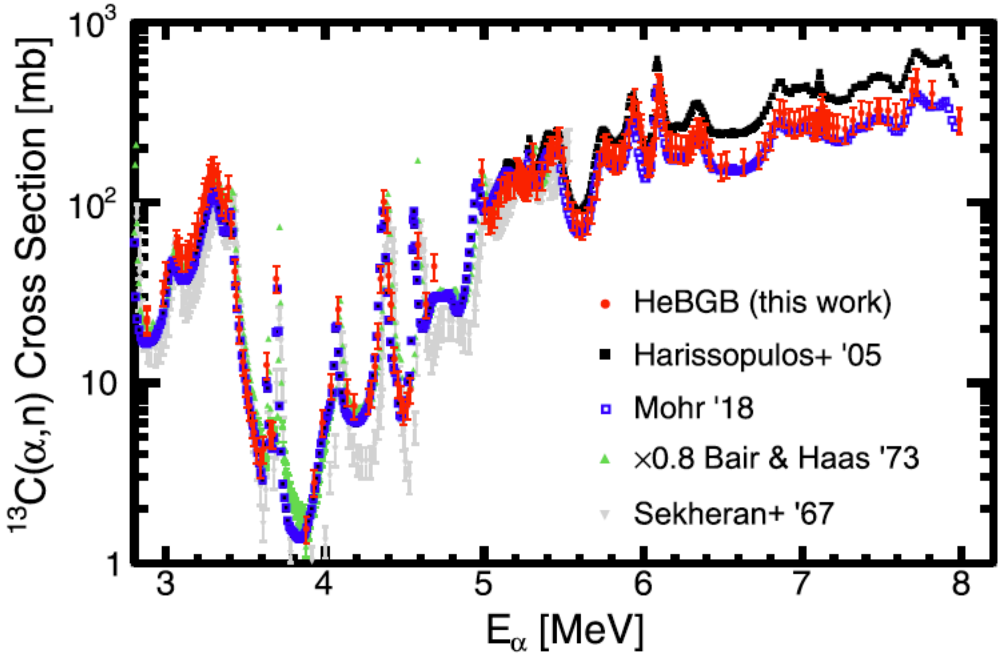


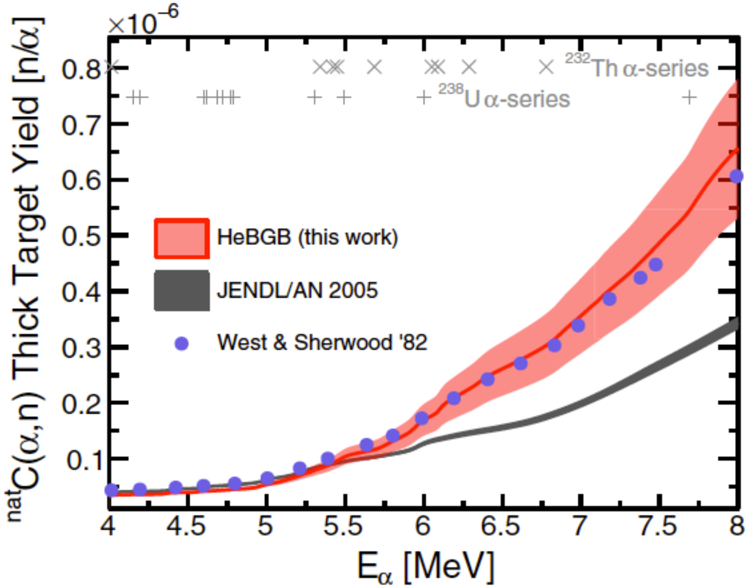


- Codes (based on statistical models) do not predict resonance structure of the cross-sections for light elements.
- Only ^{13}C contributes to the neutron yield on carbon (fraction: 1.07%).
- Harissopulos 2005 + TALYS 1.96 calculations – 'default' in SOURCES4

  → leads to higher neutron yields in materials containing carbon.

New measurements for ^{13}C

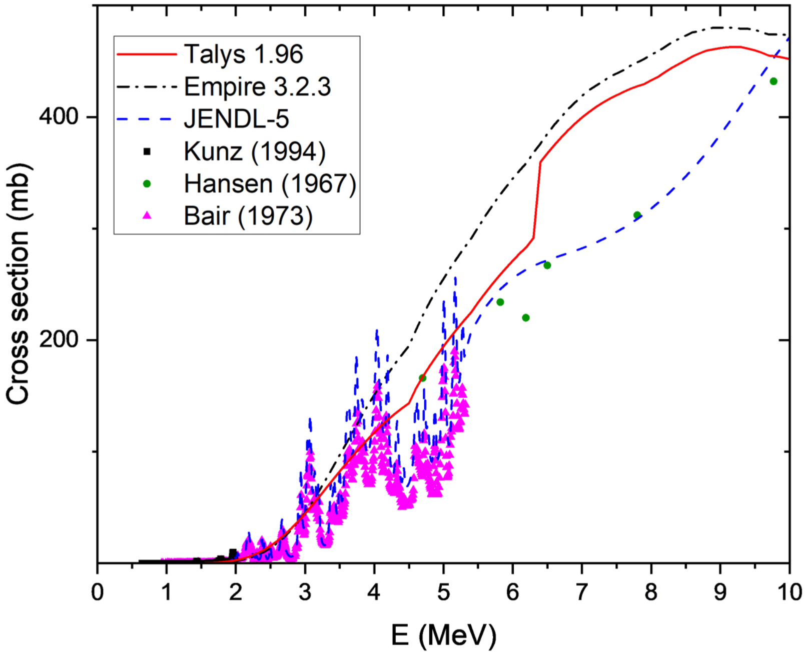




- New measurements from Brandenburg et al, PRC, 108 (2023) L061601.
- Not yet in SOURCES4 library.

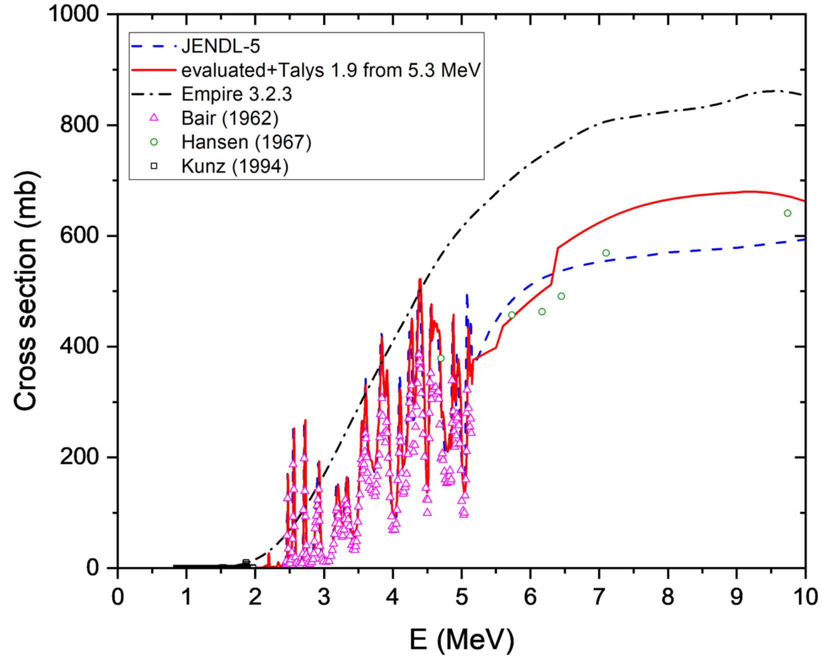
Oxygen

Oxygen-17 (0.038%)



Kunz up to 1 MeV + Bair + JENDL-2021 above 5.3 MeV

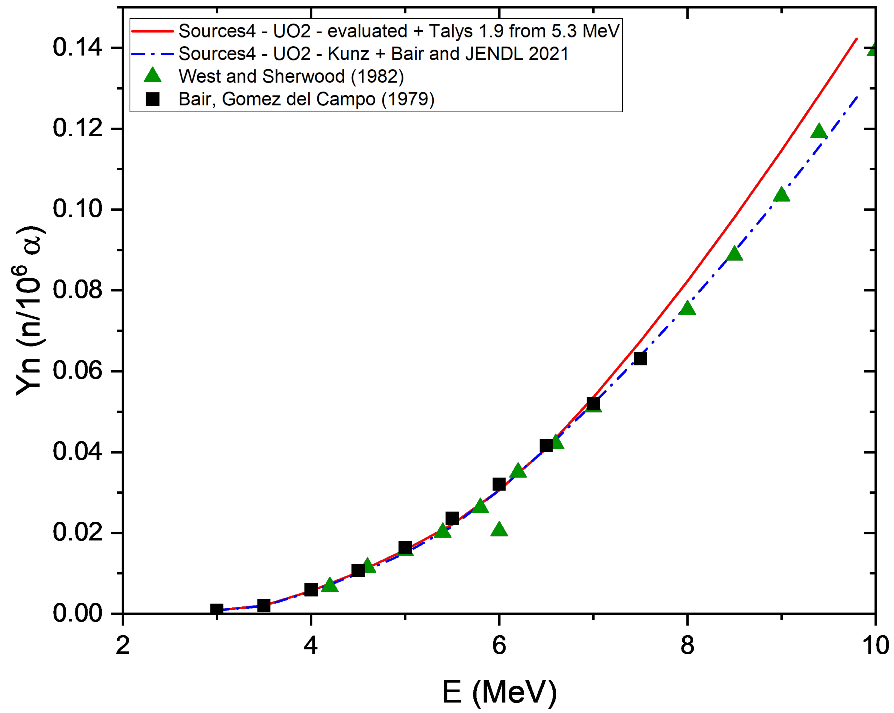
Oxygen-18 (0.205%)



JENDL-2021 (overlapping with data)

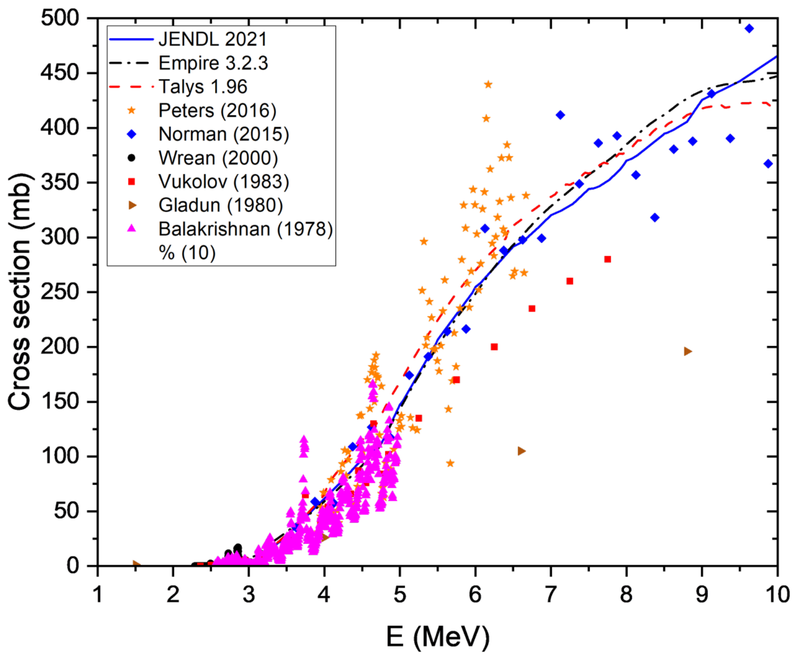
- The choice of cross-section above 5.3 MeV is not obvious.
- JENDL follows experimental data below 5.3 MeV and can be the best option to use above this energy.

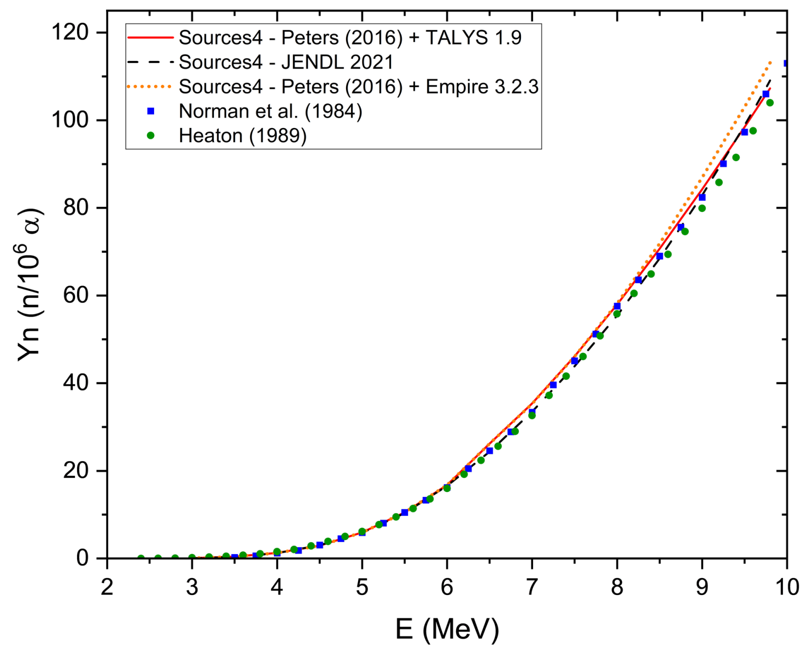
Uranium oxide



- JENDL-2021 above 5.3 MeV provides a better agreement with data from alpha beams than TALYS1.96.

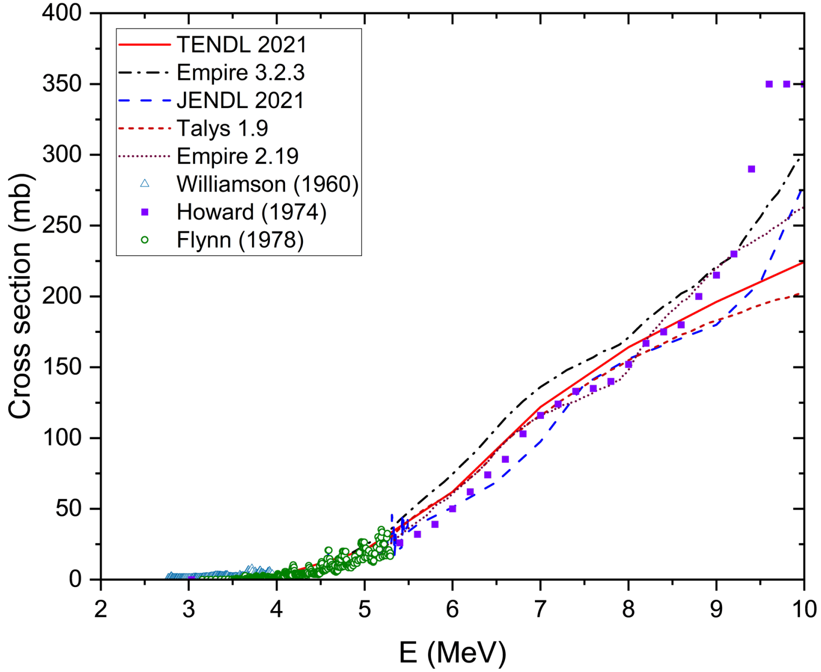
^{19}F

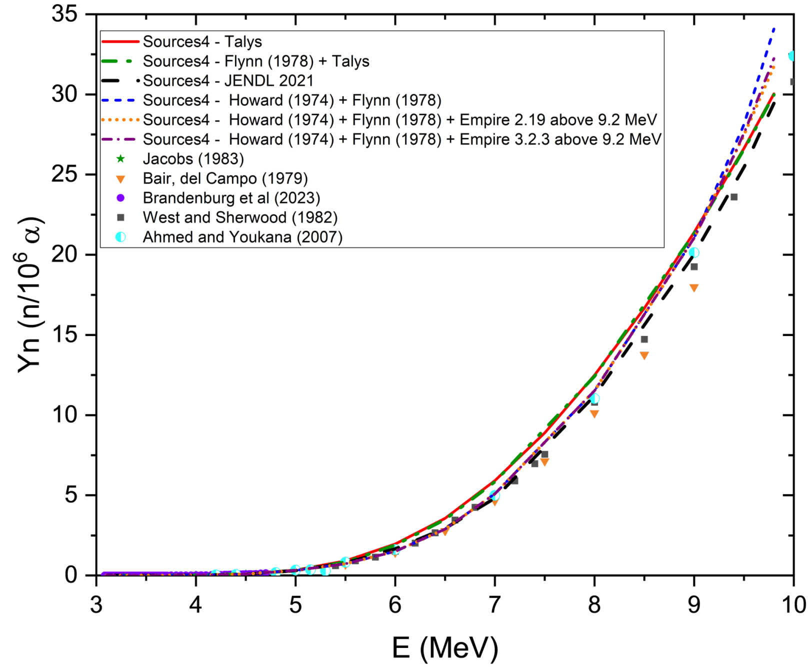




- Sometimes data do not allow us to choose the optimum cross-section, as for ^{19}F.
- The cross-sections measured by Peters (2016) complemented with those from TALYS1.96 above 7 MeV.
- The choice of the code above 7 MeV is not critical but the choice of the data below 7-8 MeV is important.

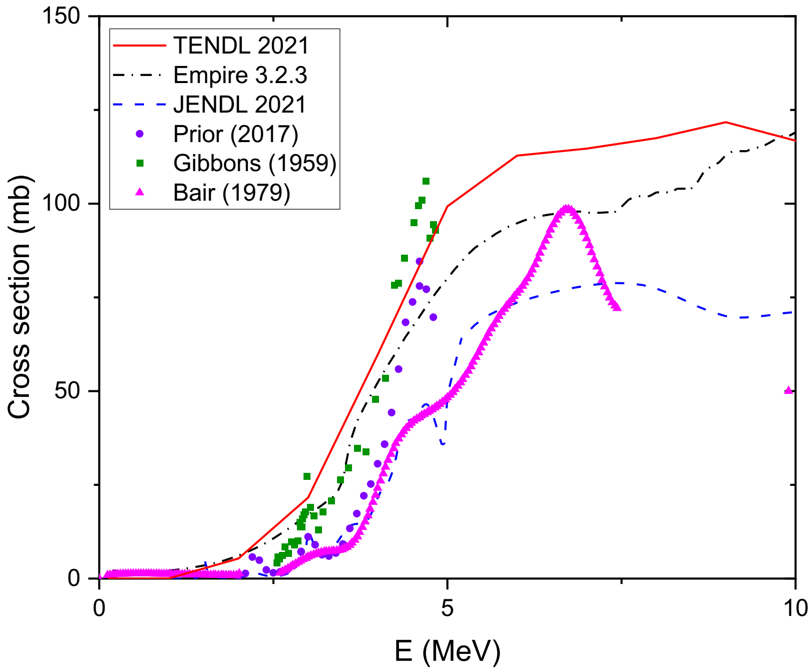
^{27}Al

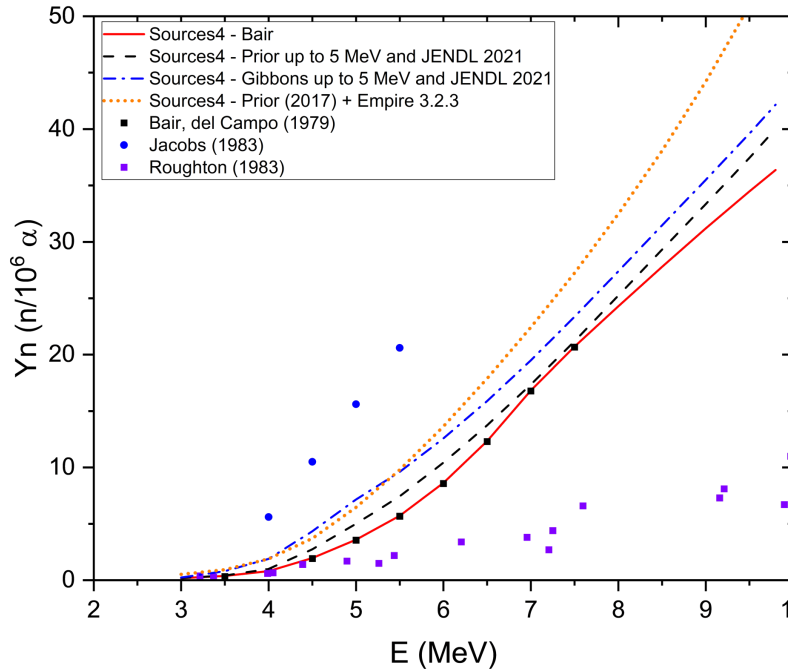




- The cross-sections measured by Flynn and Howard were used complemented with those calculated with EMPIRE 3.2.3 above 9.2 MeV.
- Not a big difference compared to models.

$^{10}B(\alpha,n)^{13}N$ (19.9%)

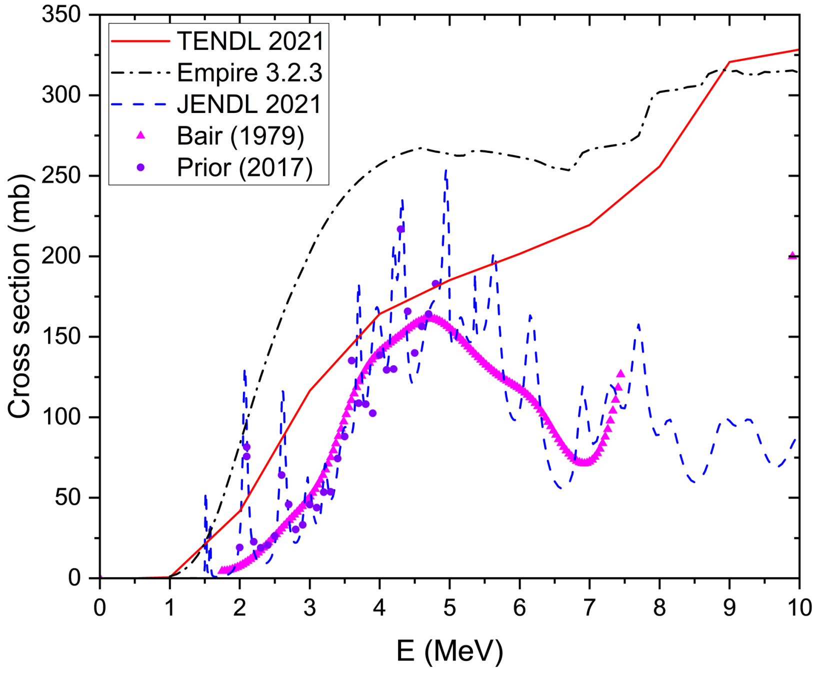


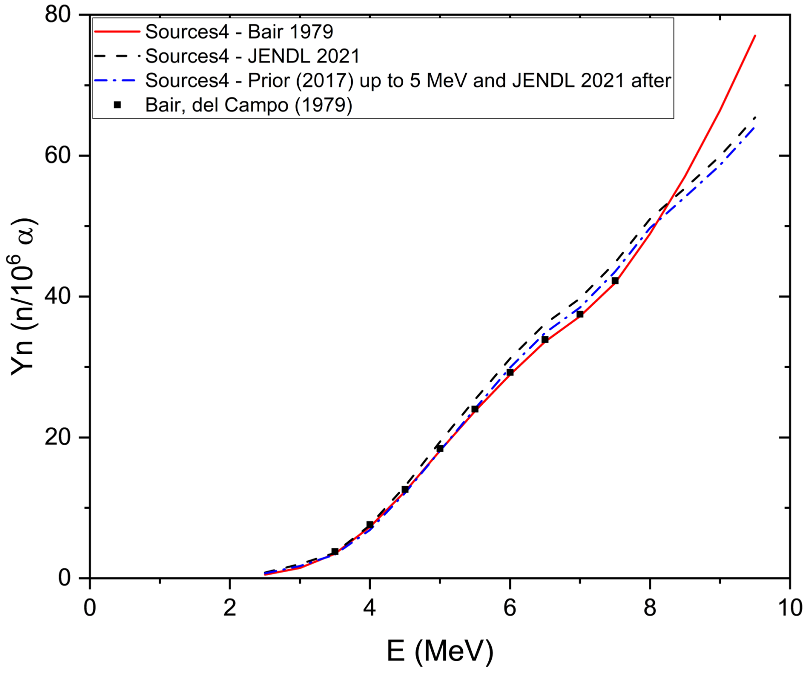


Typical example of multiple data sets not agreeing with each other both for cross-sections and neutron yields from alpha beams.

Prior + JENDL 2021 – default in SOURCES4

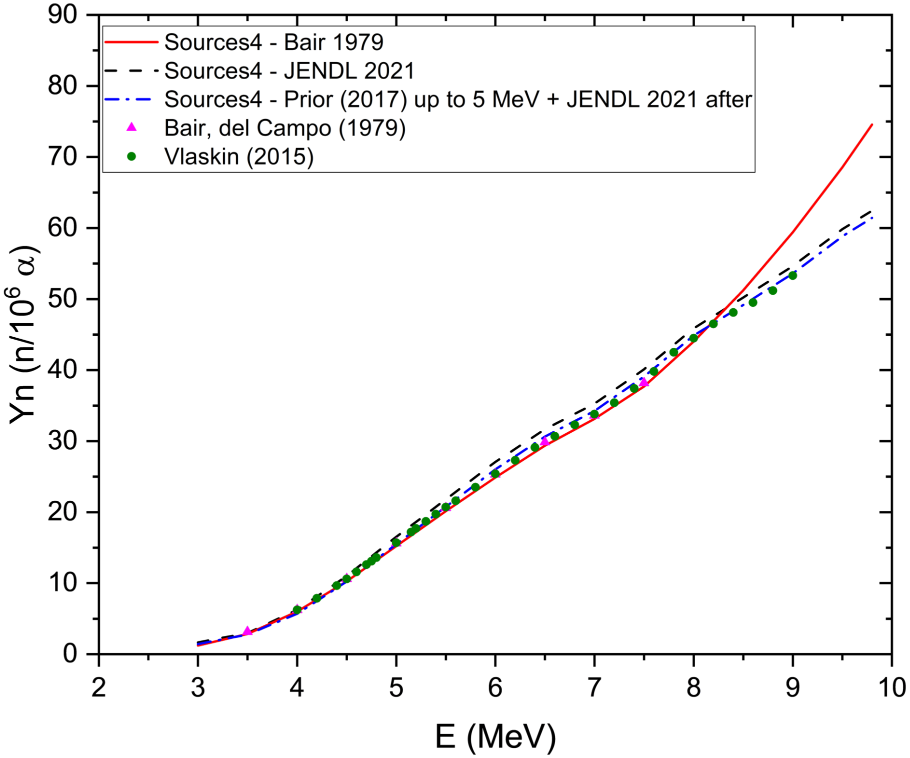
$^{11}B(\alpha,n)^{14}N$ (80.1%)





Prior + JENDL 2021 – default in SOURCES4.
The choice is driven by the agreement observed with alpha beam data.

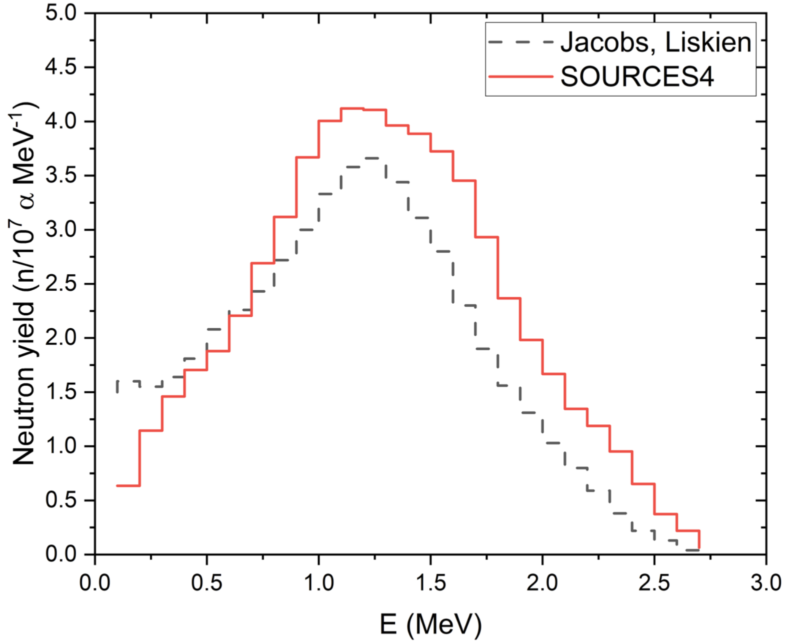
$^{nat}B(\alpha,n)N$



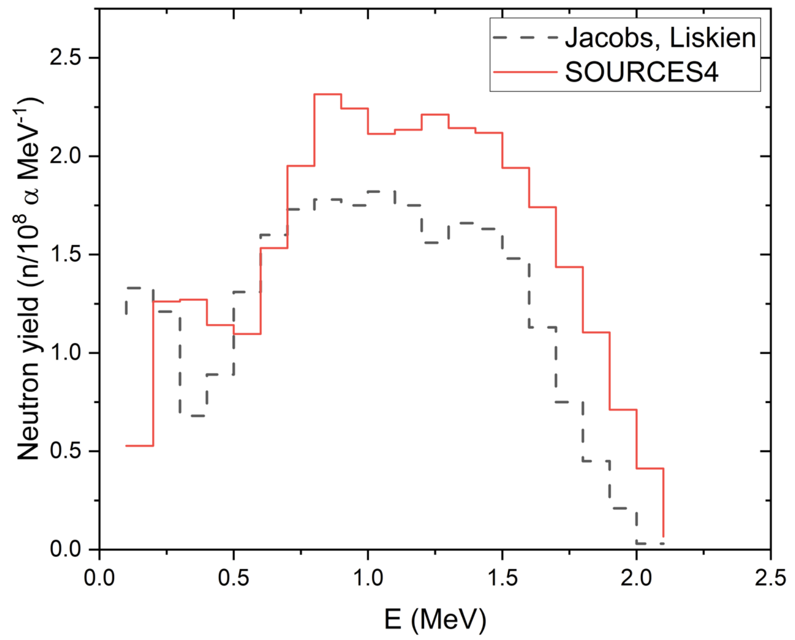
B-11 has 80.1% abundance
→B-10 is not so critical.

* Vlaskin's points are **not** experimental data, but an evaluation of neutron yield based on experimental data.

Neutron spectra: 5 MeV alphas

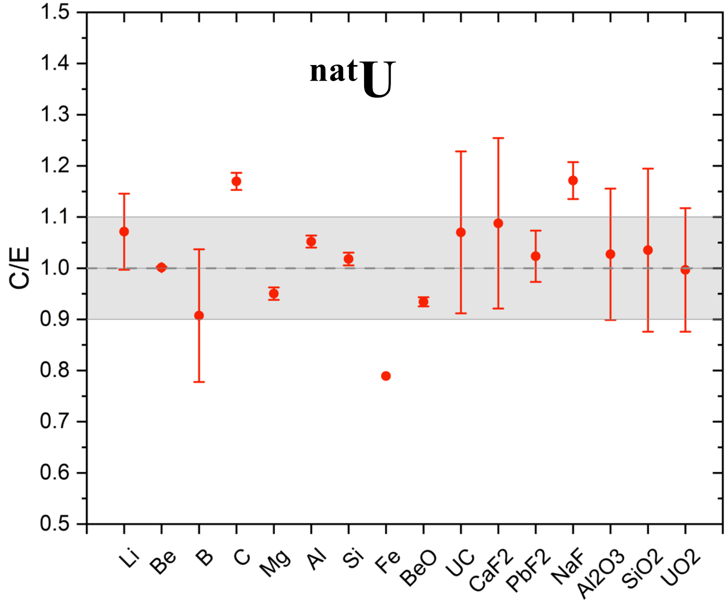


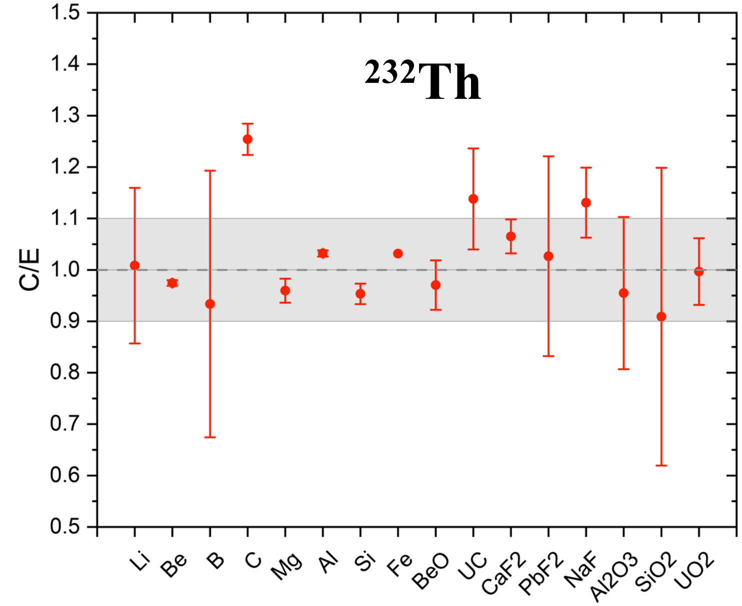
^{19}F



^{27}Al

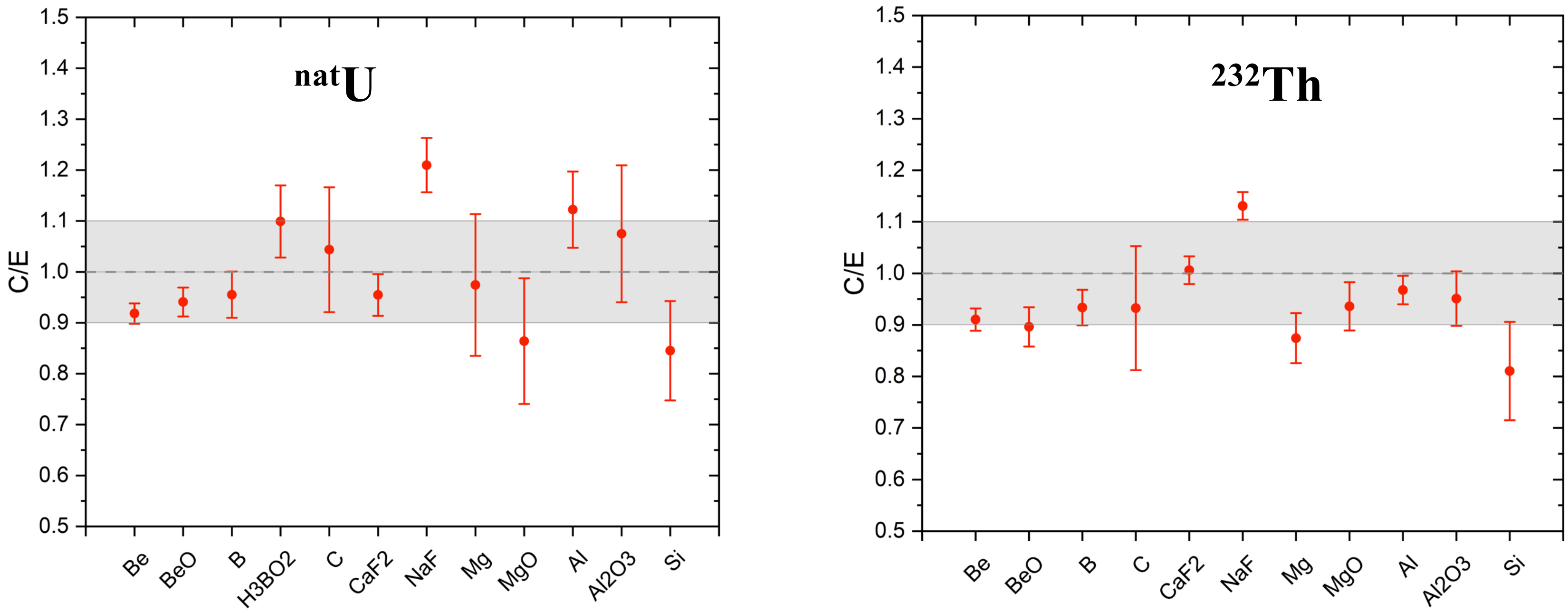
Neutrons from radioactive decay chains





- Fernandes et al. EPJ Web of Conferences, 153 (2017) 07021.
- Not the real measurements but evaluation of neutron yields from radioactive decay chains from alpha beam measurements
  - Systematic uncertainties due to the procedure used may be quite high.
- Higher yield for carbon from SOURCES4 – not a surprise (but see next slide).
- Fe: Lower for U and ok for Th -> uncertainty in 'measurements'?
- The agreement is within 10% for most materials tested.

Neutrons from radioactive decay chains



- G. V. Gorshkov, O. S. Tsvetkov, Soviet Atomic Energy, 14 (1964) 573–577.
- Direct measurements of neutron yields from radioactivity.
- Carbon looks fine but with quite a big uncertainty.
- Strangely, NaF shows higher neutron yield in SOURCES4 whereas CaF_2 better agrees with data? Still some uncertainty in data?
- The agreement is within 10% for most materials tested.

Conclusions

- Statistical models (TALYS1.9 and EMPIRE2.19/3.2.3) are not recommended for use for light isotopes at low energies where the cross-sections show a resonant behaviour (from the authors of the codes).
- An optimised approach:
  - Use data where possible (usually at low alpha energies and no controversy) and a model at higher energies that agrees with data at low energies
  - If no data exist for an isotope, use a model (TALYS or EMPIRE) based on comparison of the neutron yields with alpha beam data (if available)
  - A model for branching ratios.
  - Comparison with alpha beam data show good agreement (^{13}C is still a question).
- Neutron yields from decay chains:
  - Neutron yields with the optimised approach show a good agreement with data
    - within 10% for most materials (mainly light elements)
  - Calculated neutron spectra agree reasonably well with the measured ones
    - differences still exist but the measurements are not easy.

Backup: table with neutron yields

Element	^{nat}U	^{232}Th	Compound	^{nat}U	^{232}Th
Li	7.12×10^{-10}	2.95×10^{-10}	Al_2O_3	8.53×10^{-11}	4.17×10^{-11}
Be	8.38×10^{-9}	2.79×10^{-9}	BeO	3.08×10^{-9}	1.03×10^{-9}
B	1.99×10^{-9}	6.14×10^{-10}	C_2F_4	9.76×10^{-10}	3.90×10^{-10}
C	1.76×10^{-11}	7.04×10^{-12}	$CaCO_3$	7.28×10^{-12}	2.89×10^{-12}
N	5.80×10^{-11}	3.23×10^{-11}	CaF_2	6.63×10^{-10}	2.75×10^{-10}
Na	4.13×10^{-10}	1.93×10^{-10}	CH_2	1.71×10^{-11}	7.04×10^{-12}
Mg	2.03×10^{-10}	7.67×10^{-11}	H_2O	3.98×10^{-12}	1.39×10^{-12}
Al	1.67×10^{-10}	8.25×10^{-11}	H_3BO_3	3.38×10^{-10}	9.64×10^{-11}
Si	2.18×10^{-11}	1.01×10^{-11}	MgO	1.20×10^{-10}	4.56×10^{-11}
P	2.85×10^{-11}	1.94×10^{-11}	Na_2CO_3	2.78×10^{-10}	1.29×10^{-10}
Cl	8.08×10^{-11}	4.36×10^{-11}	NaCl	1.52×10^{-9}	6.07×10^{-10}
Ar	1.52×10^{-10}	9.00×10^{-11}	NaF	8.16×10^{-10}	3.50×10^{-10}
Ca	1.80×10^{-12}	1.22×10^{-12}	PbF_2	4.39×10^{-10}	1.74×10^{-10}
Ti	3.63×10^{-11}	3.18×10^{-11}	SiO_2	1.41×10^{-11}	5.98×10^{-12}
Cr	1.40×10^{-11}	1.45×10^{-11}	Stainless steel Fe(66%), Cr(17%), Ni(12%), Mn(2%), Mo(2%), Si(1%)	7.38×10^{-12}	8.91×10^{-12}
Mn	9.29×10^{-12}	1.04×10^{-11}			
Fe	4.74×10^{-12}	6.68×10^{-12}			
Ni	1.02×10^{-13}	2.63×10^{-13}	UC	2.50×10^{-12}	1.02×10^{-12}
Cu	3.67×10^{-13}	1.07×10^{-12}	UO_2	2.41×10^{-12}	8.37×10^{-13}

Whole U and Th decay chains. Units: n/g/s/ppb.
Additionally, spontaneous fission of ^{238}U gives 1.35×10^{-11} n/g/s/ppb.