Calculation of the neutron production in (α,n) reactions with SOURCES4

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Outline

- Introduction: neutron production in (α,n) reactions in SOURCES4
- Cross-sections and branching ratios in different models versus experimental data
- Neutron yields and spectra from different models and comparison with data
- Conclusions

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.

Modifications to SOURCES4A

- Original versions of SOURCES4A/4C are limited to 6.5 MeV α energies.
- Modifications to SOURCES4A:
  - α energies up to 9.8 MeV.
  - More cross-sections and excitation functions added in 2004-2010, mainly from EMPIRE2.19 (until 2009); M. Herman et al. Nucl. Data Sheets 108 (2007) 2655.
  - Modifications described in 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.
  - Recent cross-sections from TALYS1.9 (TENDL-2017) and EMPIRE3.2.3 have been added together with new data for some isotopes in 2019-2021.
- The code (modified versions) is used by several experiments in dark matter search and neutrino physics.

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.

Other tools

- USD web-based tool: http://neutronyield.usd.edu; Mei et al. NIMA 606 (2009) 651. Cross-sections from TENDL libraries (TALYS code).
- Comparison: SOURCES4A vs USD tool: J. Cooley et al. NIMA 888 (2018) 110-118.
- NeuCBOT: S. Westerdale and P.D. Meyers. NIMA 875 (2017) 57–64. Neutron spectra from TENDL libraries. Also comparison with SOURCES4. New version is coming.
- Comparison of NeuCBOT and SOURCES4 (just a few isotopes): Kudryavtsev et al., Talk at IDM2018. Results from the NeuCBOT are different from SOURCES4A even if cross-sections from TALYS are used, possibly due to different versions of TALYS/TENDL, different inputs/models.
- NEDIS code: Vlaskin et al. Atomic Energy, Vol. 117, No. 5, March 2015.
- Comparison between codes and data: Fernandes et al. EPJ Web of Conferences 153, 07021 (2017).
- GEANT4.10.6 - (α,n) reactions included with TENDL of JENDL cross-sections; E. Mendoza et al. Nucl. Instrum. & Meth. in Phys. Res. A 960 (2020) 163659; arXiv:1906.03903. Also comparison with other codes. SaG4n – code based on GEANT4.

Inputs to SOURCES4

- Q-value of the reaction and the energy threshold in the laboratory system (projectile: α particle) – included in the code library with cross-sections.
- Cross-section as a function of energy – included in the code library but can be added/changed by a user.
- Branching ratios for transitions to excited states (code library).
- α lines and intensities (code library).
- Stopping power or energy losses of α's (code library): from Ziegler's tables.
- Spontaneous fission spectra (code library).
- Number of atoms of a radioisotope(s) or number density (input from a user).
- Material composition (input from a user).
- Repository: http://www.oecd-nea.org/tools/abstract/detail/ccc-0661/

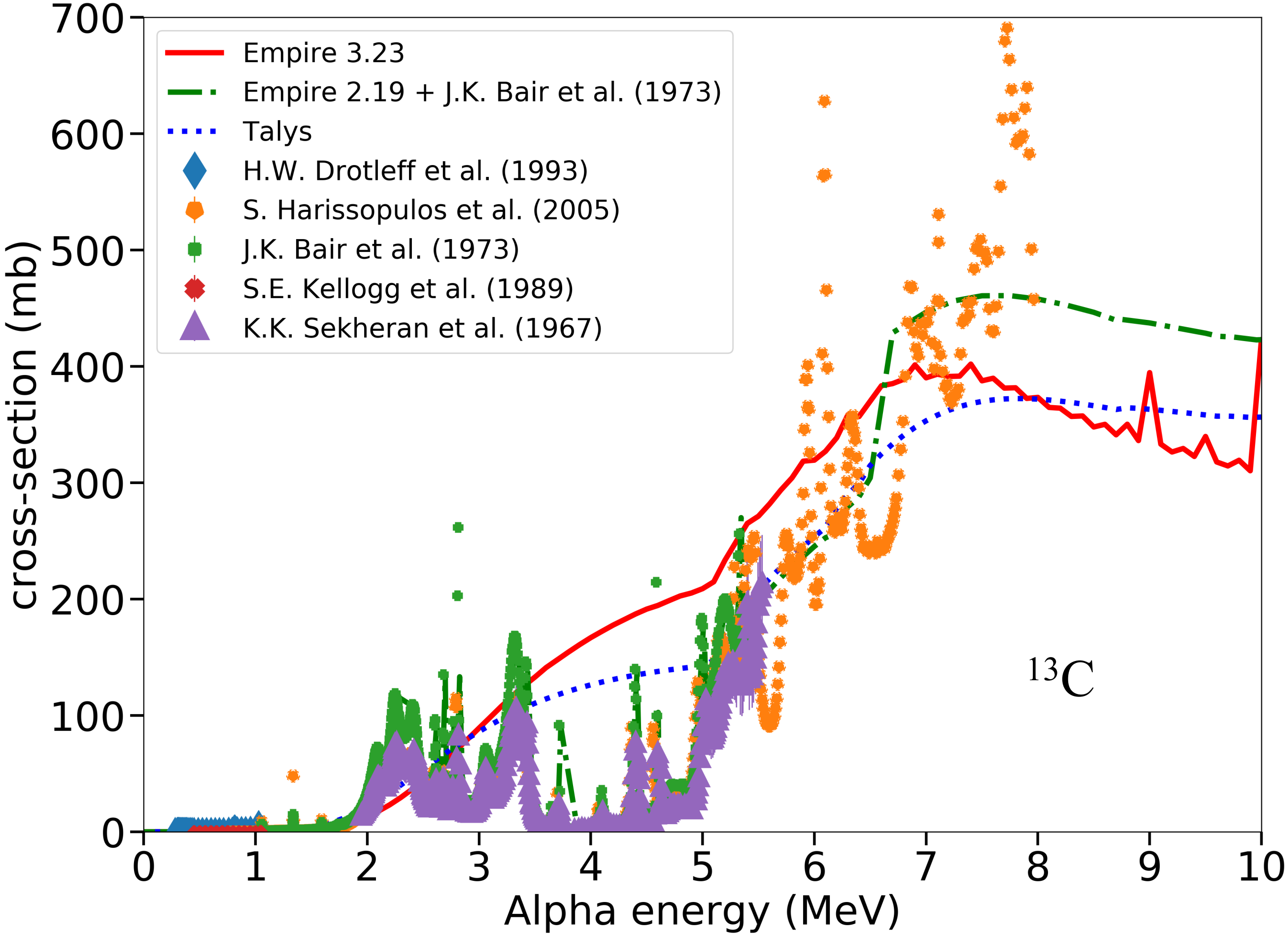
(This is not our code; we have only modified it and support the modified version.)

- Most recent version – SOURCES4C; no difference compared to SOURCES4A (as I am aware of) apart from ^{252}Cf spectrum from SF.

Outputs

- Neutron yields.
- Neutron spectra from SF and (α,n) reactions.
- Spectra for individual radioactive isotopes and for individual isotopes in the target material composition.
- Spectra for transitions to the ground and different excited states of a final state nucleus (excited states are as given in the code library).

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

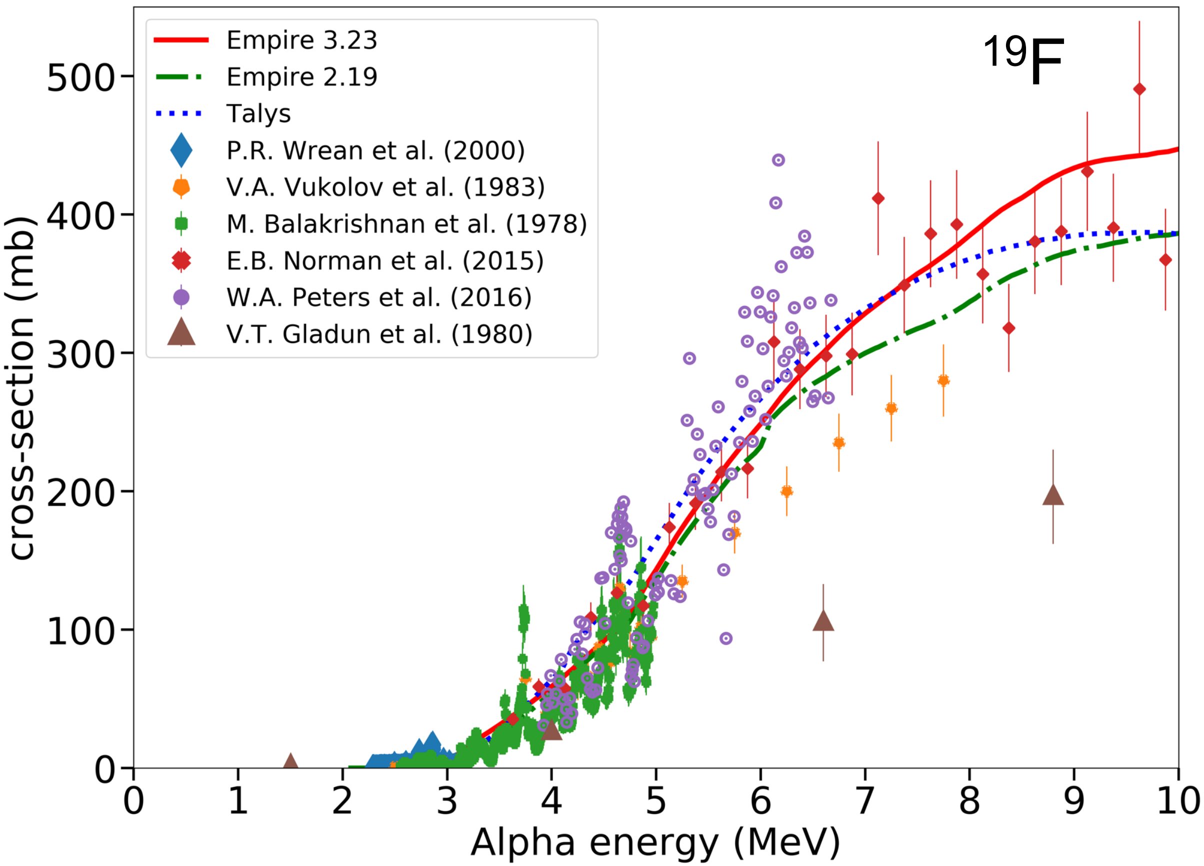


Kudryavtsev, Zakhary and Easeman. Nucl. Intrum. and Meth. A, 972 (2020) 164095.

- Codes (based on statistical models) do not predict resonance structure of the cross-sections for light elements.

- ^{12}C does not contribute (high threshold). Only ^{13}C contributes to the neutron yield (but small abundance).

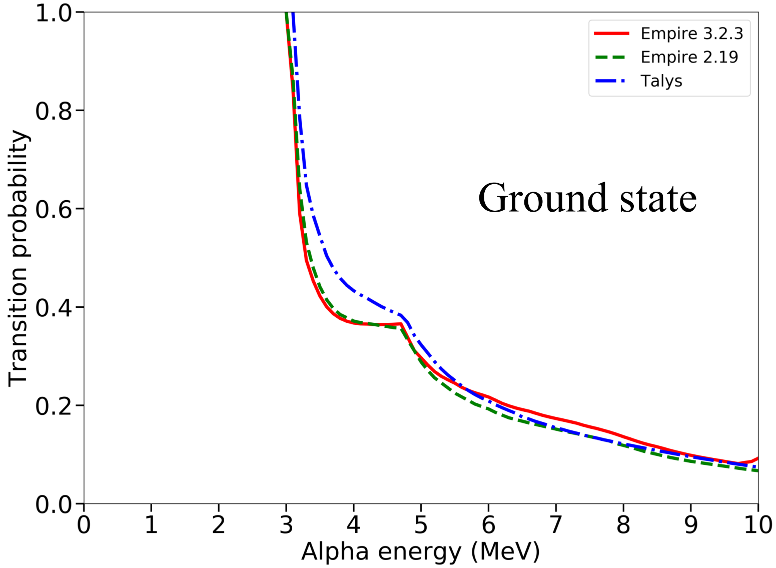
Cross-sections for ^{19}F (low-A target)

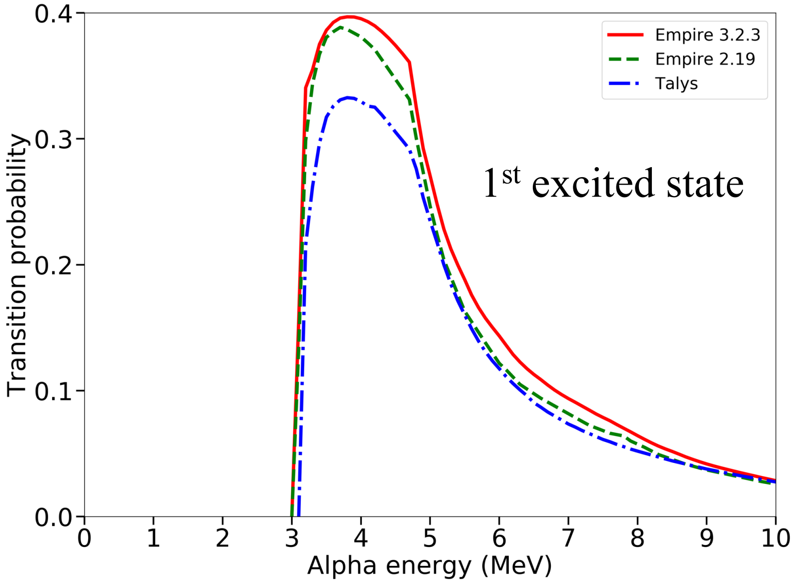


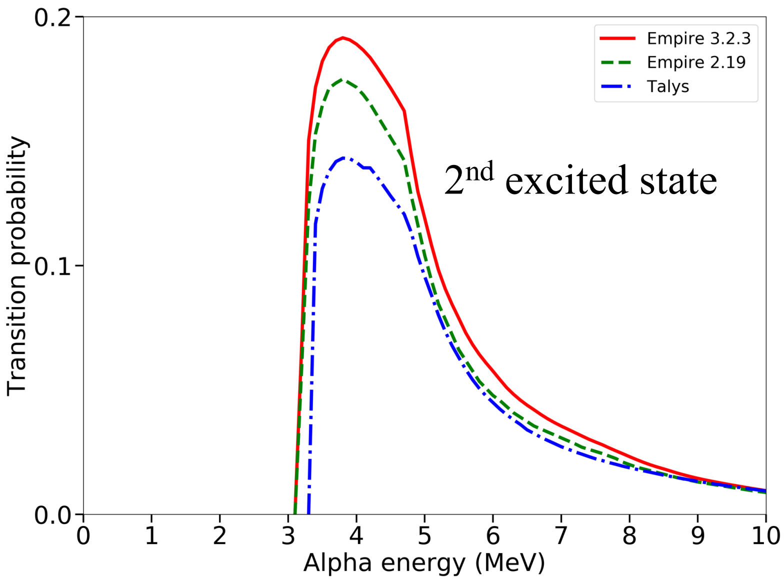
- Kudryavtsev, Zakhary and Easeman. Nucl. Intrum. and Meth. A, 972 (2020) 164095.
- Codes (based on statistical models) do not predict resonance structure of the cross-sections for light elements.
- Measurements of cross-section do not always agree with each other.
- Data for branching ratios are scarce and cover only low-energy states.

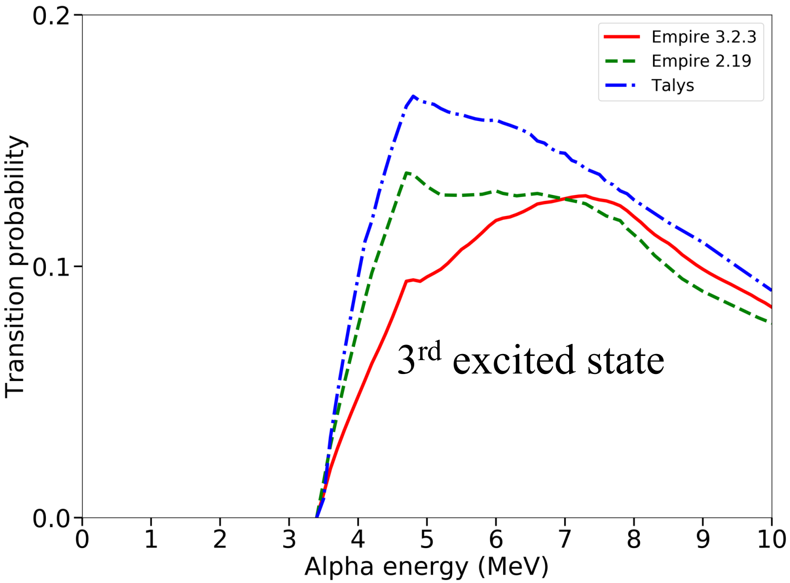
- Data do not allow us to choose the optimum model for ^{19}F.
- Data can be used in SOURCES4A (possibly in combination with another model). The results are quite different depending on a specific measurement.

Branching ratios for fluorine

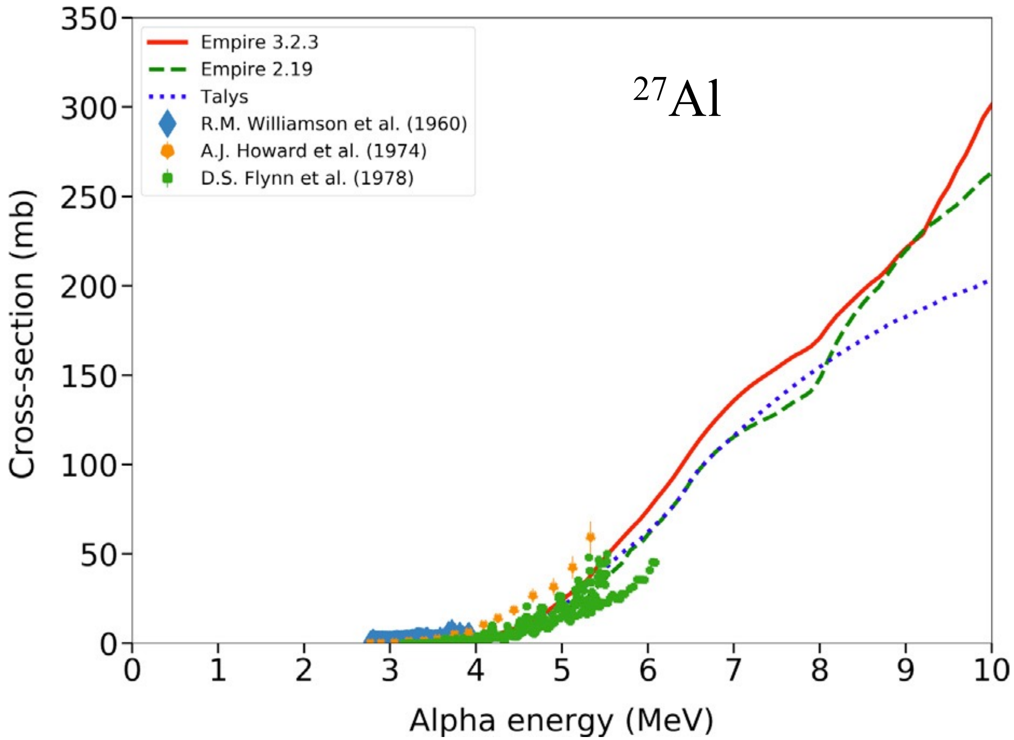








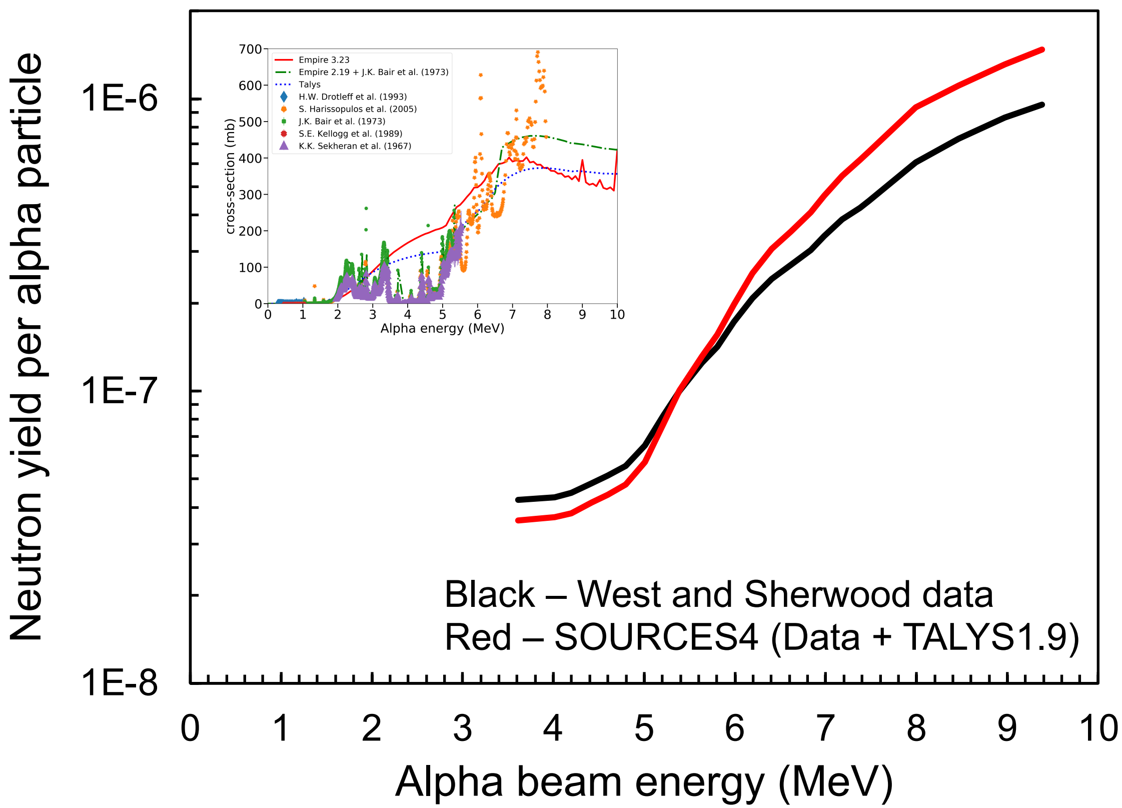
Cross-sections for aluminium



- Data are limited (quite old) and cannot help with the choice of the model. Using measured cross-sections leads to large variation in the neutron yield, depending on a specific measurements.
- We still need codes where there are no measurements.

SOURCES4 vs data: ^{13}C

- Recent work: use experimental data for cross-sections if available and reliable and complement them with a model where there are no data.
- Branching ratio: we still need a model.
- To check calculations, compare the results with alpha beam experiments.



^{13}C: cross-section data from Harissopoulos et al (2005) up to 8 MeV, TALYS1.9 above 8 MeV.

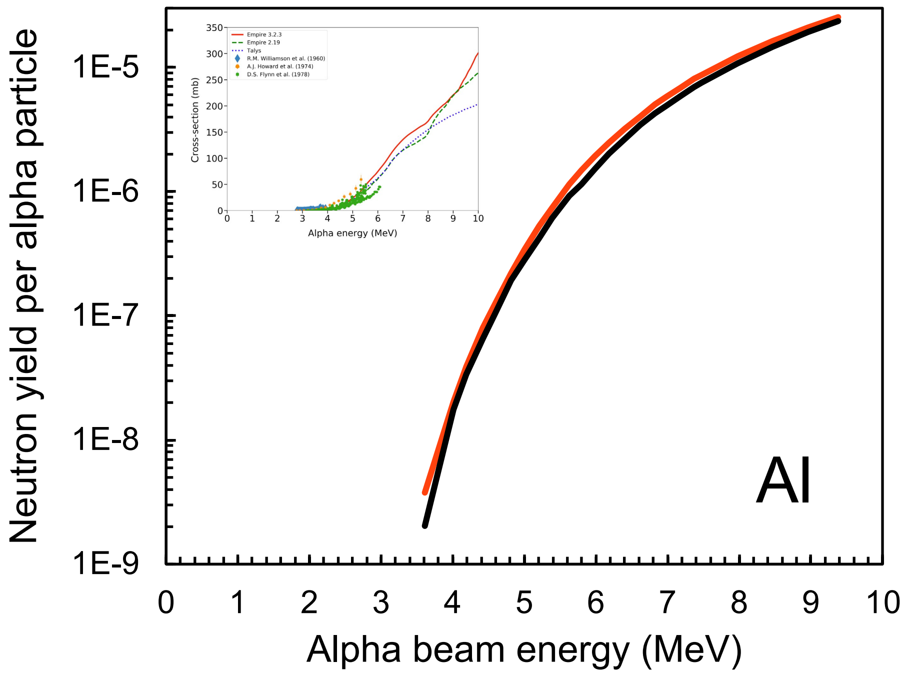
Reasonable agreement between the code and alpha beam data.

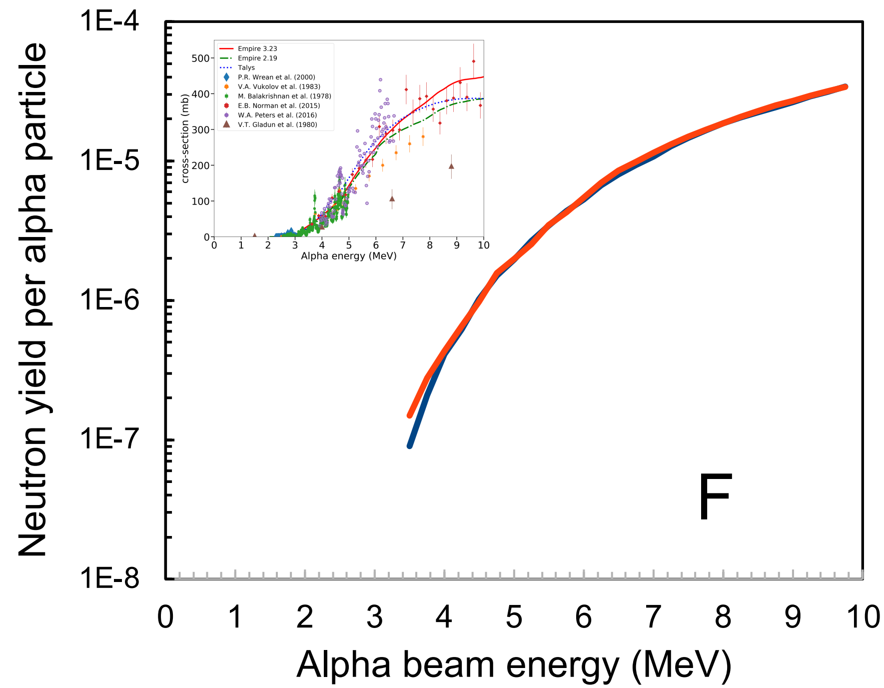
Note the use of two different data sets for each material:

1. Cross-sections as input to SOURCES4.

2. Alpha beam measurements to compare with output of SOURCES4.

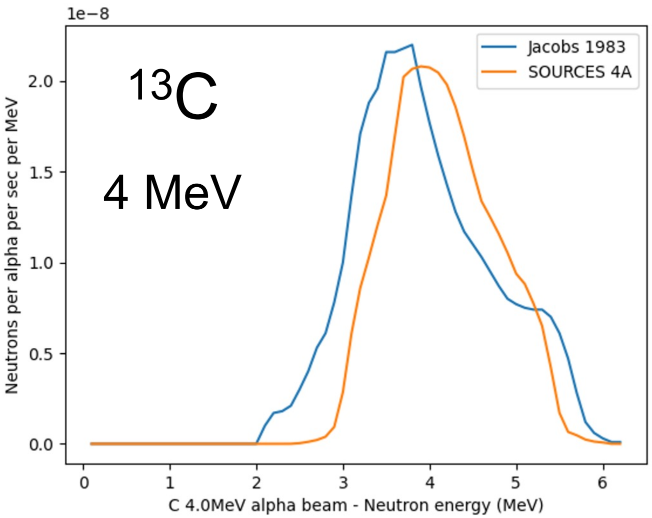
SOURCES4 vs data: aluminium and fluorine

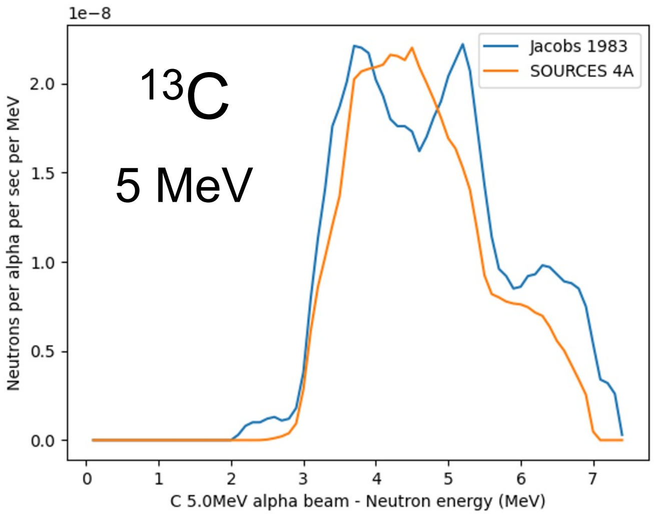


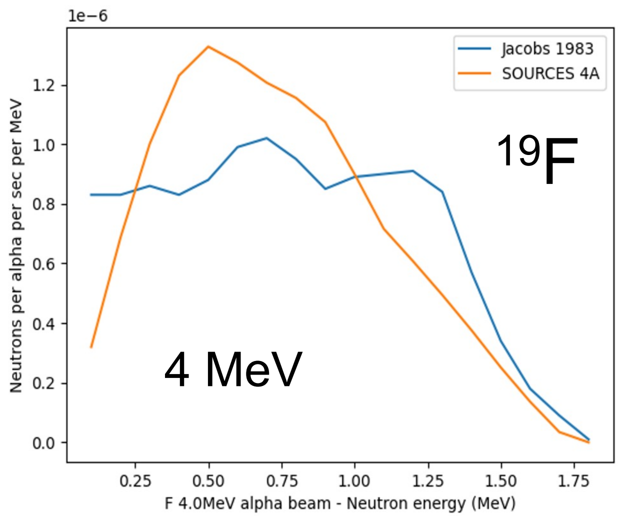


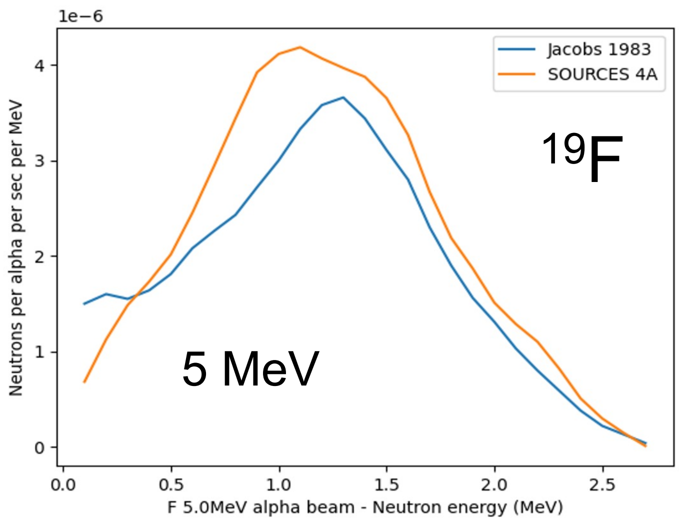
- First result with optimised cross-sections:
  - Al – data are inconclusive, TALYS is used for all energies.
  - F – data from Peters et al (2016) up to 6.7 MeV, TALYS above this energy.
- Tables are on the backup slides.

Neutron spectra: ^{13}C and ^{19}F

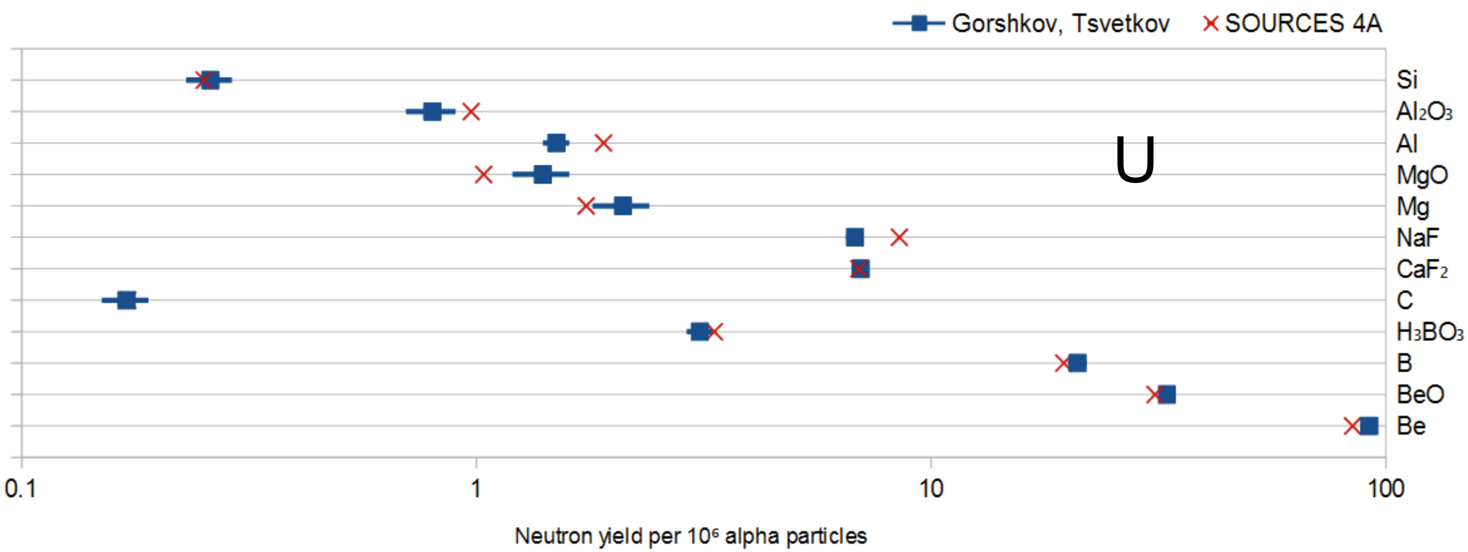


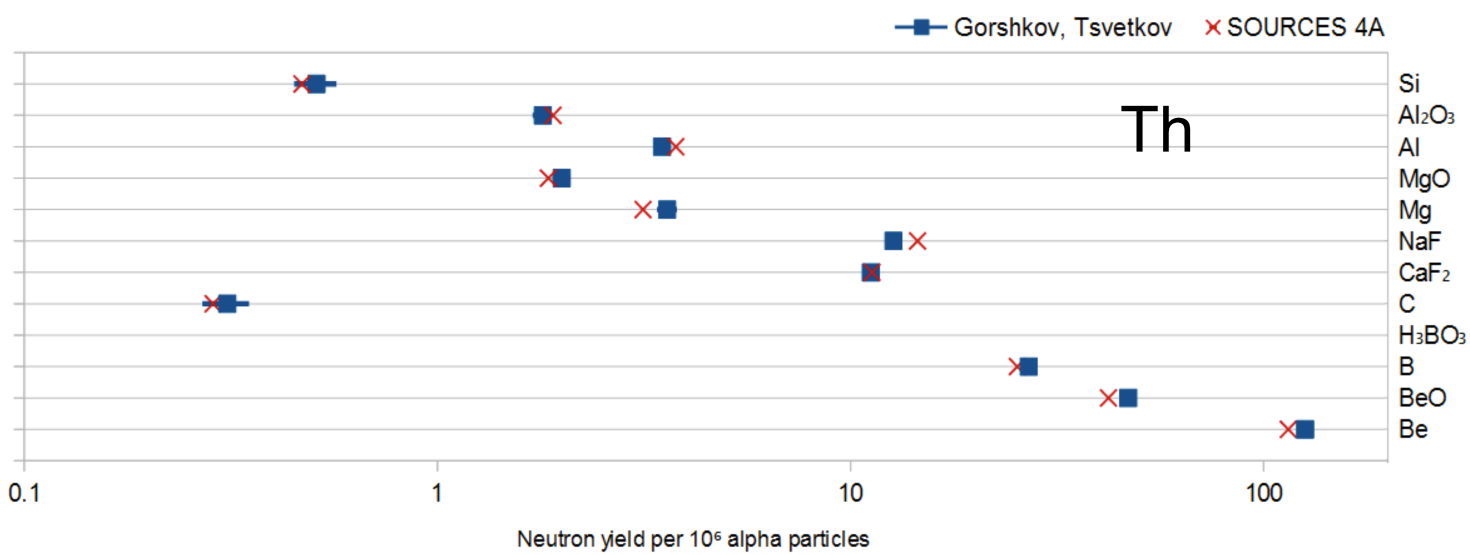






Total neutron yield from U/Th chain:





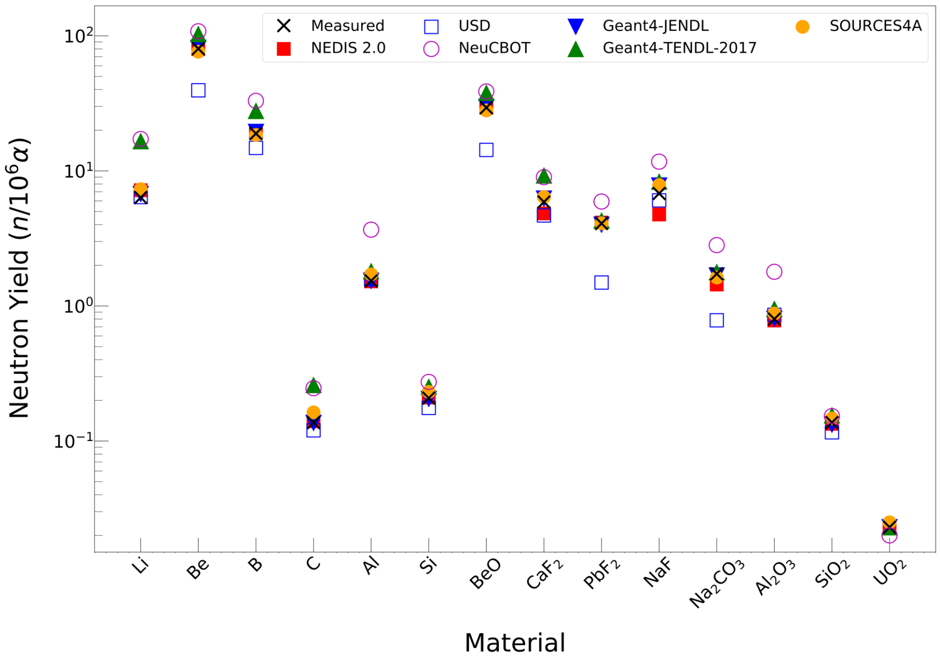
Data from Gorshkov, Zvetkov, Atomnaya Energiya, 14, 550-554,1963.

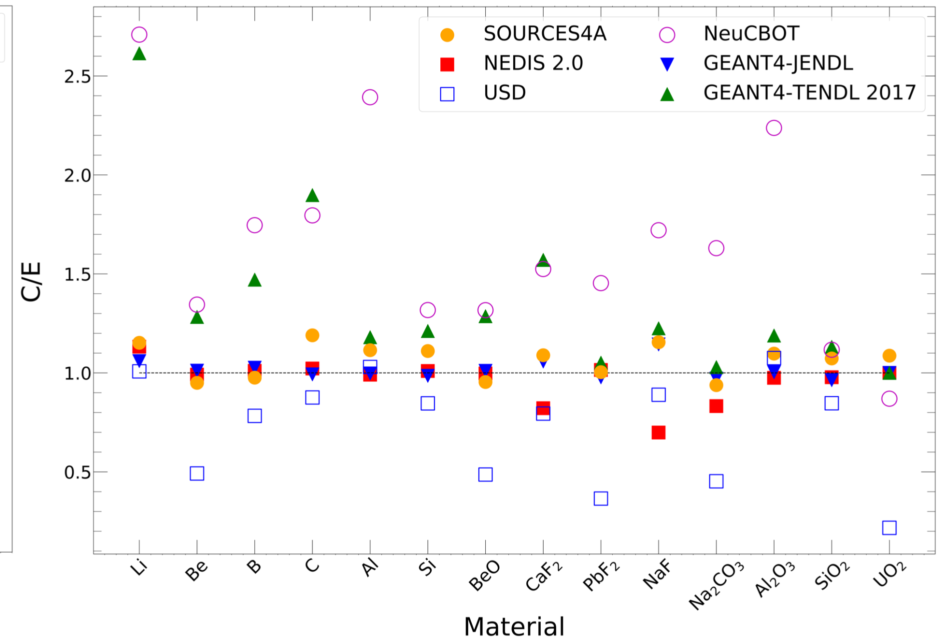
The only known (to me) paper with direct measurements of neutron yields from U/Th series.

Only light materials.

Total neutron yields from U chain

- Graphs are for U chain and a selection of materials used also in other publications. Other calculations and experimental data were taken from Fernandes et al. EPJ Web of Conferences 153, 07021 (2017); E. Mendoza et al. NIMA 960 (2020) 163659.
- 'Experimental data': combination of neutron yields from individual alphas at fixed energies on individual isotopes in a material – not direct measurements of neutrons from the whole U/Th series.





(α,*n*) reaction working group

- Motivated by:
  - Future low-background experiments in dark matter and neutrino physics;
  - Potential effect of neutrons on the future detector sensitivities;
  - Uncertainties in the cross-section and neutron yield measurements and calculations;
- A working group has been formed to:
  - Assess the need of different communities in the knowledge of neutron productions (materials, energies etc);
  - Assess the status of the codes, databases, measurements;
  - Formulate the needs for further work to obtain more data and improve the codes;
  - Write a white paper with the findings.
- Interested to join?
  - Contact Roberto Santorelli: roberto.santorelli@ciemat.es.
  - Or myself: v.kudryavtsev@sheffield.ac.uk.

Conclusions

- Statistical models (TALYS1.9 and EMPIRE2.19/3.2.3) give similar cross-sections (within 20-30%), at least for most critical isotopes and most energies of interest, but are not recommended for use for light isotopes.
- Cross-sections as input to SOURCES4:
  - Data on cross-sections are not always sufficient to choose the best code/model – large variations exist among experimental cross-sections.
  - An optimised approach (started recently):
    - 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.
    - A model for branching ratios.
- Neutron yields from alpha beams and decay chains:
  - Different models for the cross-sections result in 20-25% difference between neutron yields from SOURCES4.
  - Neutron yields with the optimised approach show a good agreement with data.
  - Calculated neutron spectra agree reasonably well with the measured ones; differences still exist but the measurements are not easy.

Backup: tables with neutron yields

SOURCES4A: total neutron yields from U/Th chains

Material	Cross-section	$^{238}U + ^{235}U$	^{232}Th
Aluminium	TALYS1.9	1.86×10^{-10}	9.05×10^{-11}
	EMPIRE2.19	1.69×10^{-10}	8.59×10^{-11}
	EMPIRE3.2.3	2.20×10^{-10}	1.07×10^{-10}
Al_2O_3	TALYS1.9	9.48×10^{-11}	4.56×10^{-11}
	EMPIRE2.19	8.59×10^{-11}	4.32×10^{-11}
	EMPIRE3.2.3	11.42×10^{-11}	5.45×10^{-11}
Howard, 1974	EMPIRE3.2.3 +Experiment	13.55×10^{-11}	6.04×10^{-11}
PTFE	TALYS1.9	10.21×10^{-10}	4.03×10^{-10}
	EMPIRE2.19	8.72×10^{-10}	3.50×10^{-10}
	EMPIRE3.2.3	9.39×10^{-10}	3.78×10^{-10}
	EMPIRE3.2.3 +Experiment	9.68×10^{-10}	3.91×10^{-10}

Units: neutrons/g/s/ppb of the parent isotope. Only (α, n) reactions, no SF.

SOURCES4A: total neutron yields from U/Th chains

Material	Cross-section	$^{238}U + ^{235}U$	^{232}Th
SiO_2	TALYS1.9	1.54×10^{-11}	6.75×10^{-12}
	EMPIRE2.19	1.59×10^{-11}	7.03×10^{-12}
	EMPIRE3.2.3	2.07×10^{-11}	8.61×10^{-12}
	EMPIRE3.2.3 +Experiment	1.35×10^{-11}	6.21×10^{-12}
Ti	TALYS1.9	2.80×10^{-11}	2.33×10^{-11}
	EMPIRE2.19	2.55×10^{-11}	2.15×10^{-11}
	EMPIRE3.2.3	3.39×10^{-11}	2.48×10^{-11}
	EMPIRE3.2.3 +Experiment	3.39×10^{-11}	2.46×10^{-11}

Units: neutrons/g/s/ppb of the parent isotope. Only (α, n) reactions, no SF.

Comparison with data

Target	Uranium					Thorium				
	Gorshkov, Tsvetkov		Sources			Gorshkov, Tsvetkov		Sources		
		±	Raw	Converted	Agreement (%)		±	Raw	Converted	Agreement (%)
Be	92	2	8.39E-09	84.5	-8	126	3	2.79E-09	114.7	-9
BeO	33	1	3.08E-09	31.1	-6	47	2	1.03E-09	42.1	-10
B	21	1	1.94E-09	19.6	-7	27	1	6.17E-10	25.3	-6
H3BO3	3.1	0.2	3.31E-10	3.3	8					
C	0.17	0.02	1.70E-11	0.2	1	0.31	0.04	6.97E-12	0.3	-8
CaF2	7	0.3	6.88E-10	6.9	-1	11.2	0.3	2.75E-10	11.3	1
NaF	6.8	0.3	8.45E-10	8.5	25	12.7	0.3	3.54E-10	14.5	14
Mg	2.1	0.3	1.73E-10	1.7	-17	3.6	0.2	7.66E-11	3.1	-13
MgO	1.4	0.2	1.03E-10	1.0	-26	2	0.1	4.52E-11	1.9	-7
Al	1.5	0.1	1.89E-10	1.9	27	3.5	0.1	9.20E-11	3.8	8
Al2O3	0.8	0.1	9.66E-11	1.0	22	1.8	0.1	4.64E-11	1.9	6
Si	0.26	0.03	2.50E-11	0.3	-3	0.51	0.06	1.14E-11	0.5	-8

- Comparison of SOURCES4A with 'partly' optimised (still working on this) cross-sections with data on neutron yields from U/Th series from Gorshkov and Zvetkov (1963).
- The last column for each series show the percentage difference.

Tables with neutron yield

Beam (MeV)	Neutrons per α, aluminium		
	West and Sherwood	SOURCES	%
3.611	2.033E-9	3.77E-09	85
4.013	1.748E-8	2.03E-08	16
4.193	3.382E-8	3.91E-08	16
4.423	6.598E-8	8.02E-08	22
4.598	1.066E-7	1.28E-07	20
4.817	1.925E-7	2.19E-07	14
5.004	2.836E-7	3.42E-07	21
5.211	4.247E-7	5.30E-07	25
5.388	6.133E-7	7.39E-07	20
5.63	9.233E-7	1.14E-06	23
5.799	1.149E-6	1.48E-06	29
5.988	1.520E-6	1.93E-06	27
6.184	2.020E-6	2.47E-06	22
6.403	2.663E-6	3.20E-06	20
6.615	3.456E-6	4.05E-06	17
6.827	4.252E-6	5.03E-06	18
6.985	4.930E-6	5.84E-06	18
7.179	5.903E-6	6.91E-06	17
7.373	6.976E-6	8.08E-06	16
7.474	7.557E-6	8.72E-06	15
7.976	1.081E-5	1.23E-05	14
8.466	1.473E-5	1.64E-05	11
8.963	1.926E-5	2.10E-05	9
9.381	2.361E-5	2.54E-05	8

Beam (MeV)	Neutrons per α, fluorine		
	Norman and Chupp	SOURCES	%
3.50	9.00E-8	1.49E-7	65.6
3.75	2.08E-7	2.79E-7	34.1
4.00	4.11E-7	4.34E-7	5.6
4.25	6.17E-7	6.52E-7	5.7
4.50	1.02E-6	9.78E-7	-4.1
4.75	1.50E-6	1.58E-6	5.3
5.00	1.96E-6	1.98E-6	1.0
5.25	2.66E-6	2.53E-6	-4.9
5.50	3.45E-6	3.46E-6	0.3
5.75	4.37E-6	4.28E-6	-2.1
6.00	5.31E-6	5.50E-6	3.6
6.25	6.68E-6	7.01E-6	4.9
6.50	8.00E-6	8.55E-6	6.9
6.75	9.40E-6	9.92E-6	5.5
7.00	1.08E-5	1.15E-5	6.5
7.25	1.28E-5	1.32E-5	3.1
7.50	1.46E-5	1.50E-5	2.7
7.75	1.66E-5	1.68E-5	1.2
8.00	1.86E-5	1.87E-5	0.5
8.25	2.05E-5	2.08E-5	1.5
8.50	2.23E-5	2.28E-5	2.2
8.75	2.43E-5	2.50E-5	2.9
9.00	2.65E-5	2.71E-5	2.3
9.25	2.90E-5	2.94E-5	1.4
9.50	3.12E-5	3.17E-5	1.6
9.75	3.42E-5	3.40E-5	-0.6

SOURCES4 vs data: left – aluminium, right – fluorine. Last column: percentage difference.