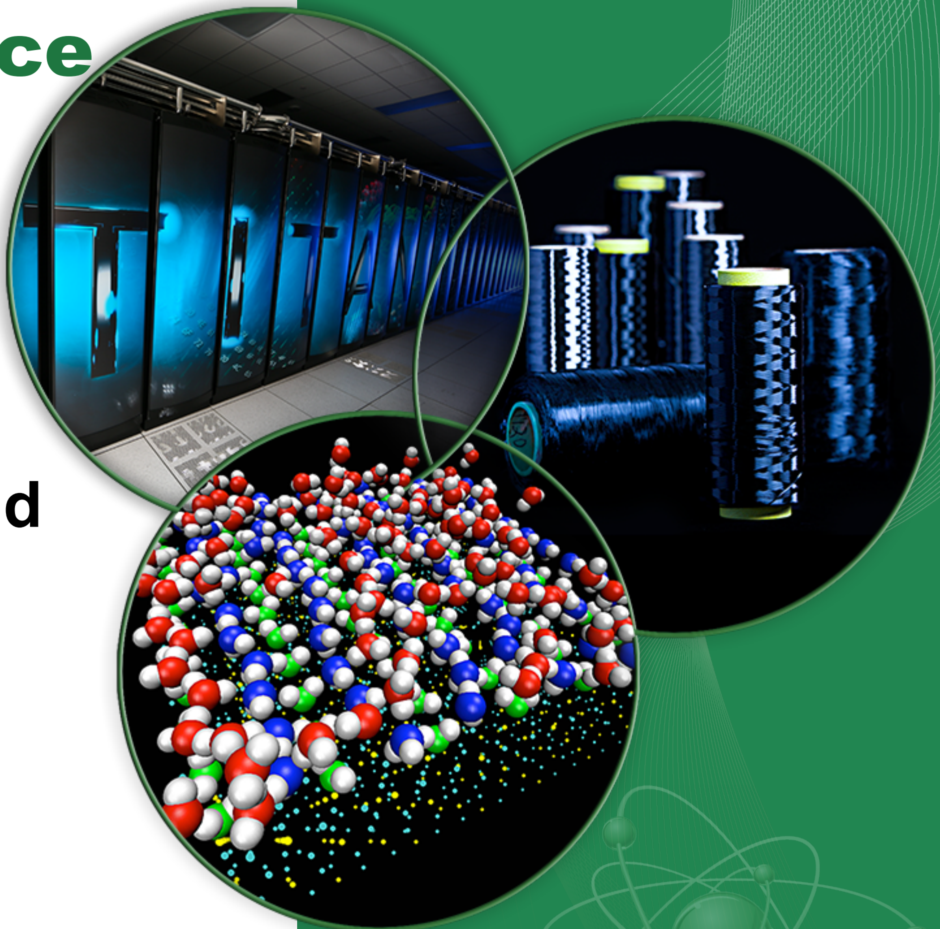
Uncertainty Quantification in (α,n) Neutron Source Calculations in an Oxide Matrix



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Oak Ridge, TN

R-Matrix Code Meeting
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Outline

- Importance of α-particle interactions
- Current status of (α,n) cross sections
- Methodology to generate
  - ^{17,18}O(α,n) cross section covariances
  - U and O stopping power cross section covariances
- Results
- Summary and conclusions
- References

Motivation

- Within a project with LLNL and LANL to quantitatively assess nuclear data uncertainties for safeguards and nonproliferation, ORNL focused on cross section covariance data for light nuclei used for neutron source calculations
- SOURCES4C[1,2] code is widely used in nuclear safeguards, spent nuclear fuel characterization, and for neutron source description due to spontaneous fission and (α,n) reactions
- Currently, in SOURCES4C libraries there is no information on covariance data. Therefore, no uncertainty is provided in the estimated neutron source intensities or emission spectra
- Nuclear data describing α-particle interactions on light nuclei, e.g., fluorine and oxygen, are essential for calculating neutron emission via (α,n) processes

(1) W. B. Wilson et al., Report LA-UR-02-1839, LANL (2002)
(2) R. T. Perry et al., Report LA-8869-MS, LANL (1981)

Goal

- Develop a methodology to propagate nuclear data covariance information in neutron source calculations from (α,n) reactions
- The approach is applied to estimate the uncertainty in the neutron generation rates for uranium oxide fuel types due to uncertainties on
  - $^{17,18}O(\alpha,n)$ reaction cross-sections
  - uranium and oxygen stopping power cross sections.

$$R \propto \sum_i \int_0^{E_\alpha} \sigma_i(E)\epsilon(E)^{-1} dE$$

Introduction
(Current status (α,n) cross sections in SOURCES4C code)

- In SOURCES4C code $^{17,18}O(\alpha,n)$ cross sections are based on Bair's thin-, thick-target measurements for lower energy

Nucleus	Target	E_α(MeV)	Unc.(%)	Norm.	Unc.(%)[a]
^{17}O	thin	0.9--5.3	25	1.35	7
^{18}O	thin	1.0--2.5	25	1.35	7
^{18}O	thin	2.4--5.1	25	1.35	7
^{nat}O	thick	3.8--10.0	7	1.00	7

- For incident α-energy >5MeV SOURCES4C code uses Hansen data properly normalized.

(a) Uncertainty after normalization

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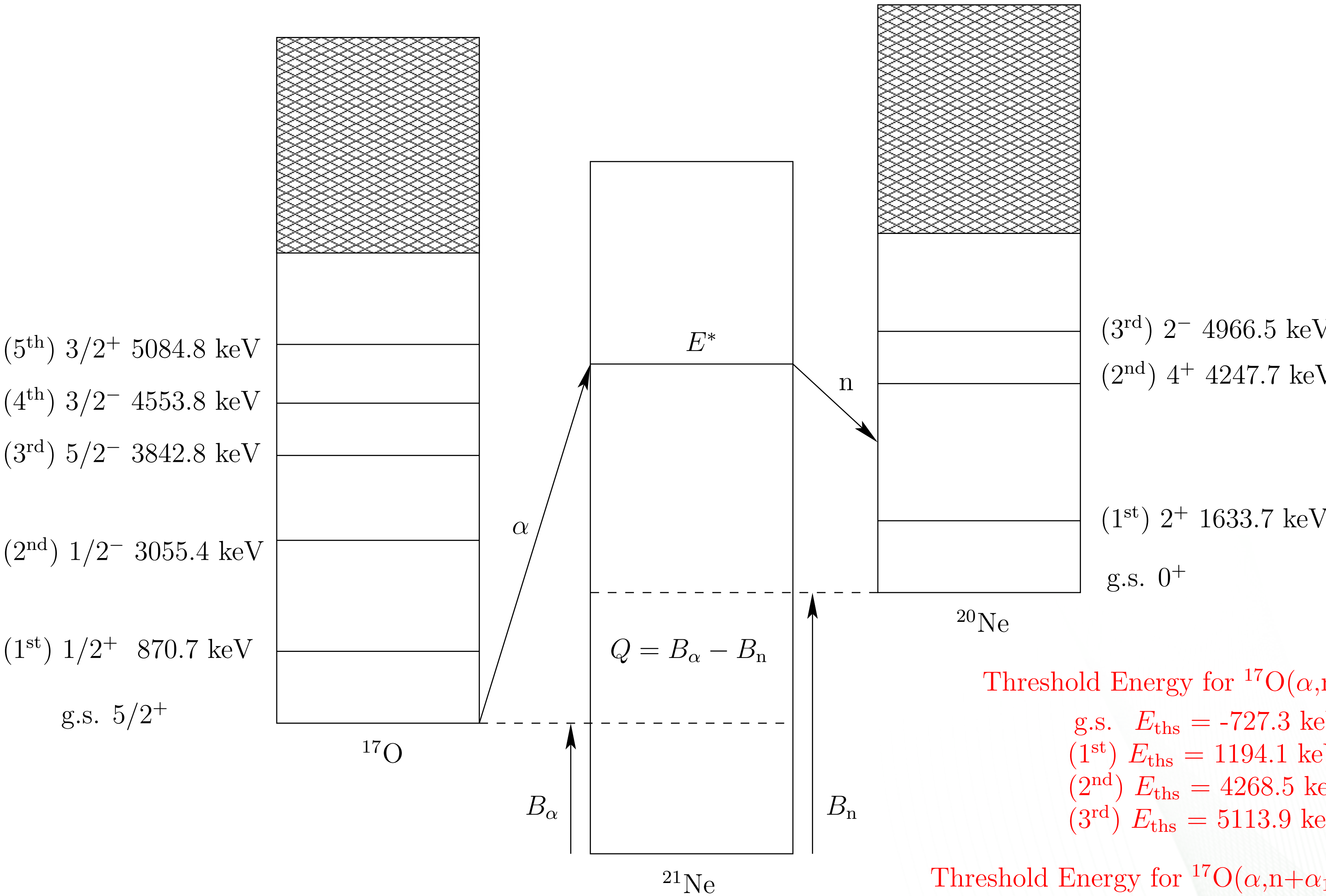
(α,n) reaction scheme for ^{17}O isotope



Threshold Energy for ^{17}O(α,n) reactions

g.s. E_{ths} = -727.3 keV

(1$^{\text{st}}$) E_{ths} = 1194.1 keV

(2$^{\text{nd}}$) E_{ths} = 4268.5 keV

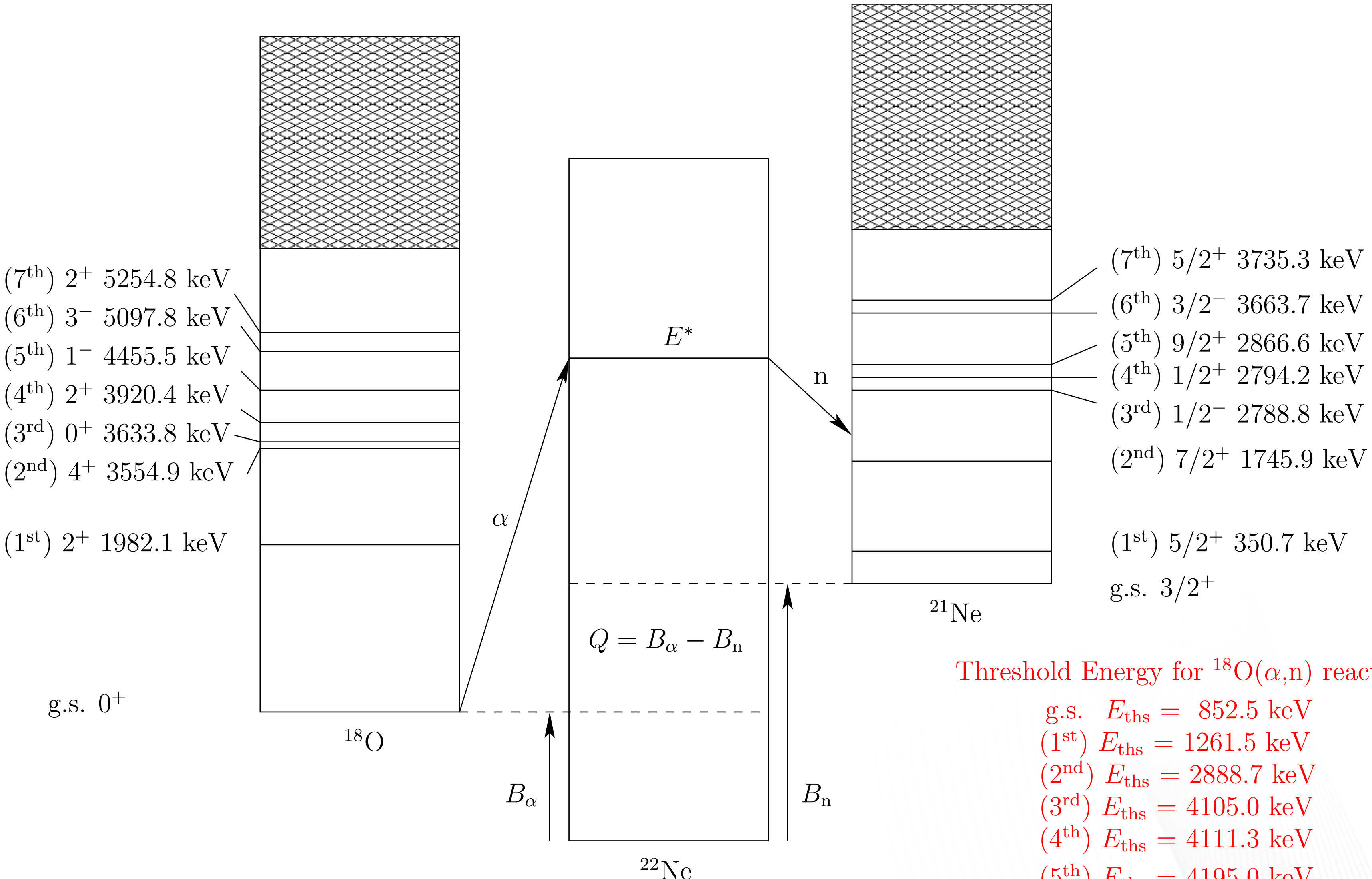
(3$^{\text{rd}}$) E_{ths} = 5113.9 keV

Threshold Energy for ^{17}O(α,n+α_1) reactions

(1$^{\text{st}}$) E_{ths} = 5118.6 keV

Schematic of the ^{17}O(α,n)^{20}Ne nuclear reaction process

(α,n) reaction scheme for ^{18}O isotope



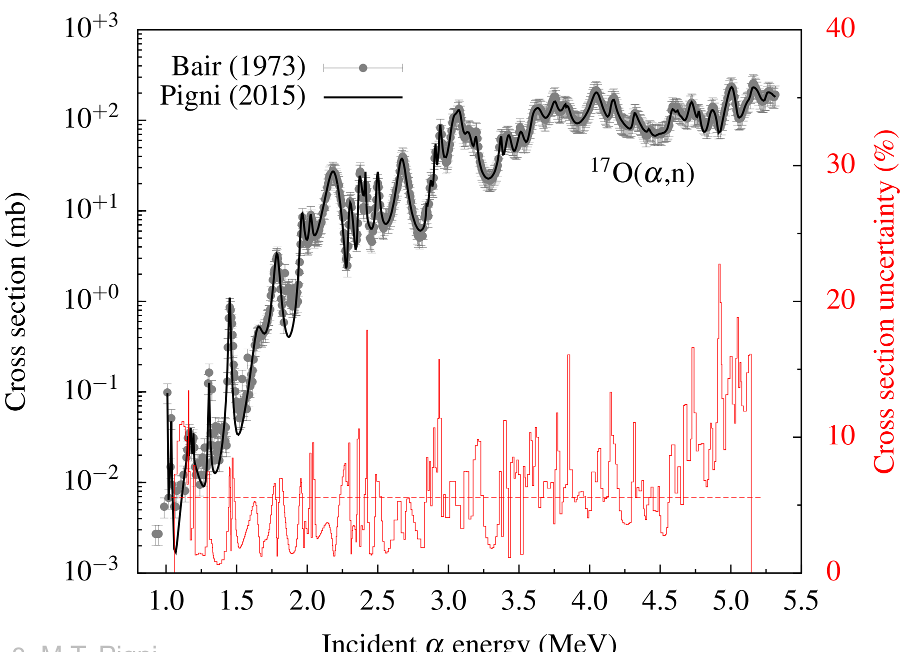
Threshold Energy for ^{18}O(α,n) reactions

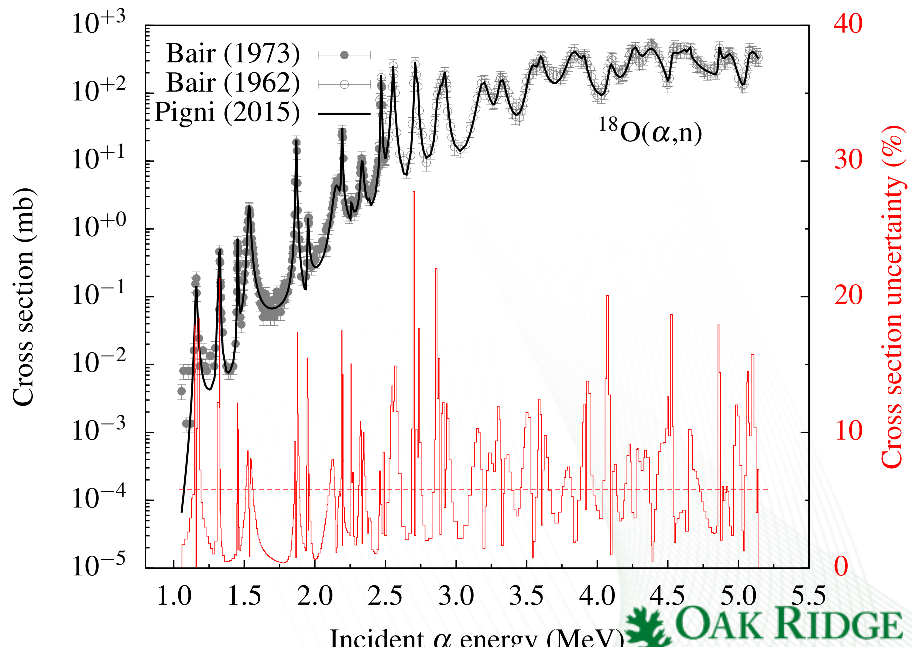
g.s. $E_{\text{ths}} = 852.5$ keV
(1st) $E_{\text{ths}} = 1261.5$ keV
(2nd) $E_{\text{ths}} = 2888.7$ keV
(3rd) $E_{\text{ths}} = 4105.0$ keV
(4th) $E_{\text{ths}} = 4111.3$ keV
(5th) $E_{\text{ths}} = 4195.0$ keV
(6th) $E_{\text{ths}} = 6826.9$ keV
(7th) $E_{\text{ths}} = 6924.4$ keV

Schematic of the ^{18}O(α,n)^{21}Ne nuclear reaction process

Methodology
(to generate (α,n) cross section covariances up to 5 MeV)

- R-matrix SAMMY code for lower energy range up to about 5 MeV based on Bair's experimental data
  - Reich-Moore parameterization of (α,n) reactions
    - For ^{18}O Elastic channel (as well as spin assignment) based on measurement of Goldberg et al. for available excitation energies
    - For ^{17}O up to about 2 MeV important work of Best et al.





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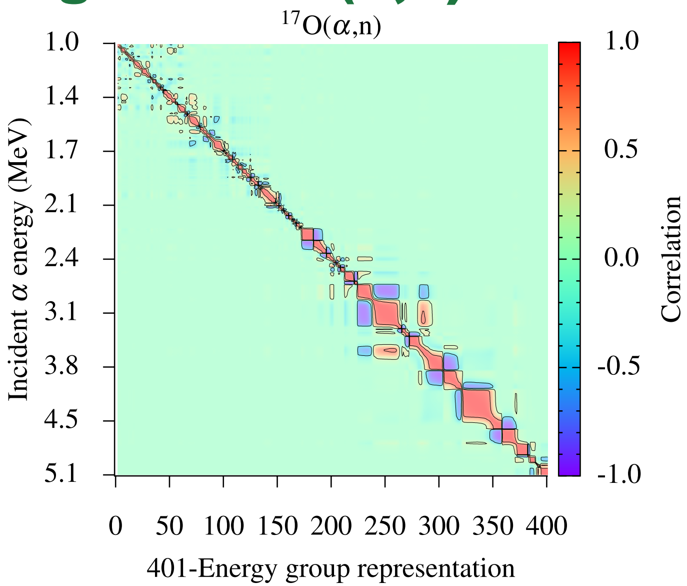
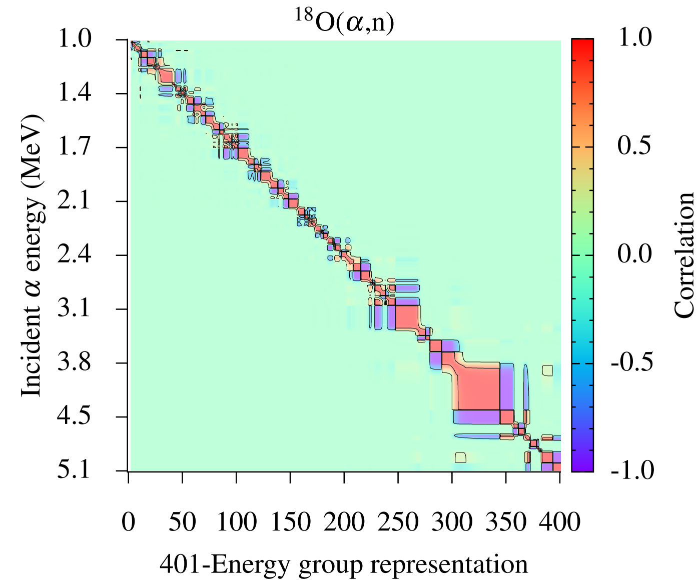
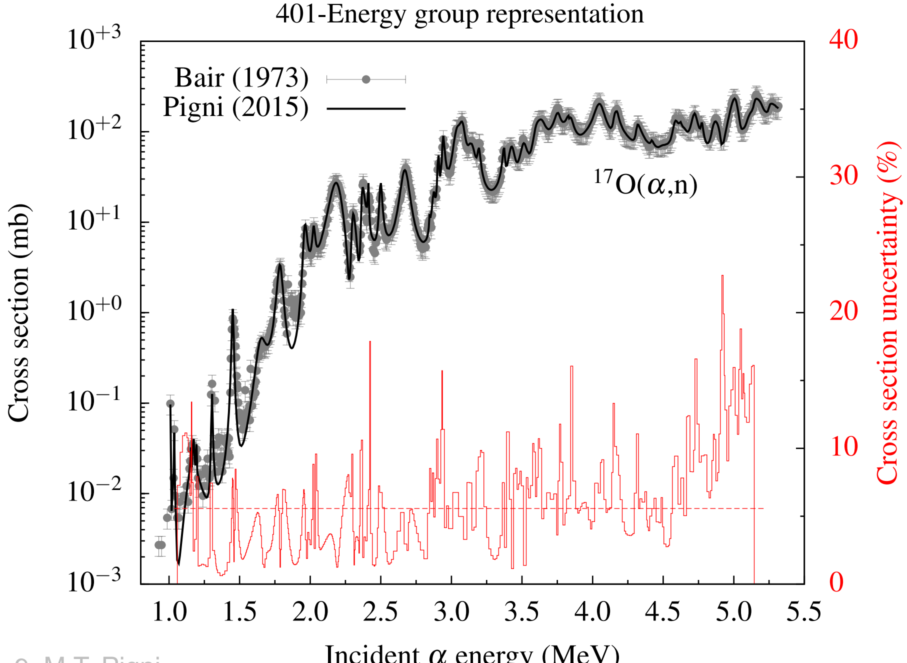
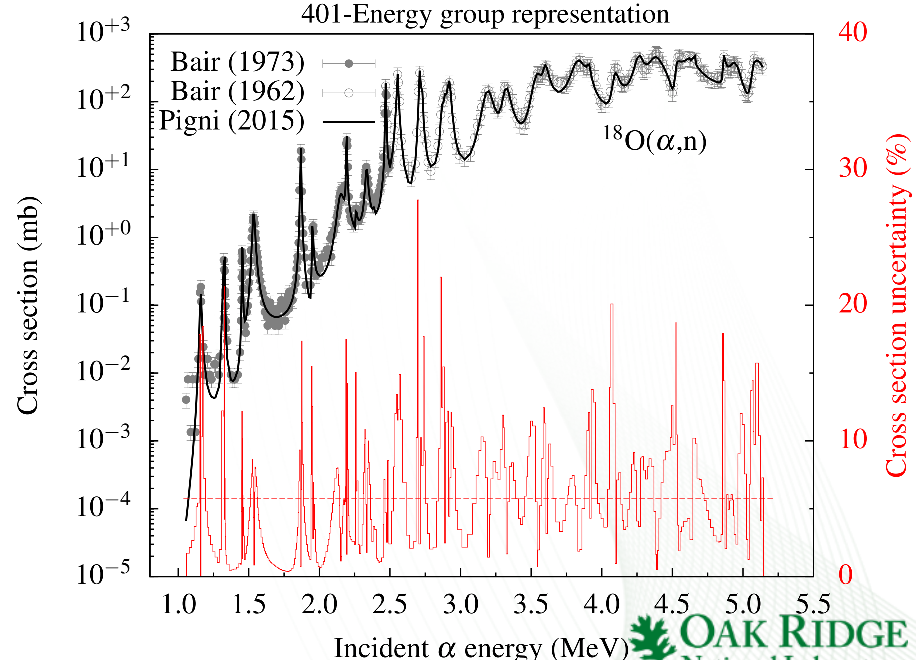
Methodology
(to generate (α,n) cross section covariances up to 5 MeV)









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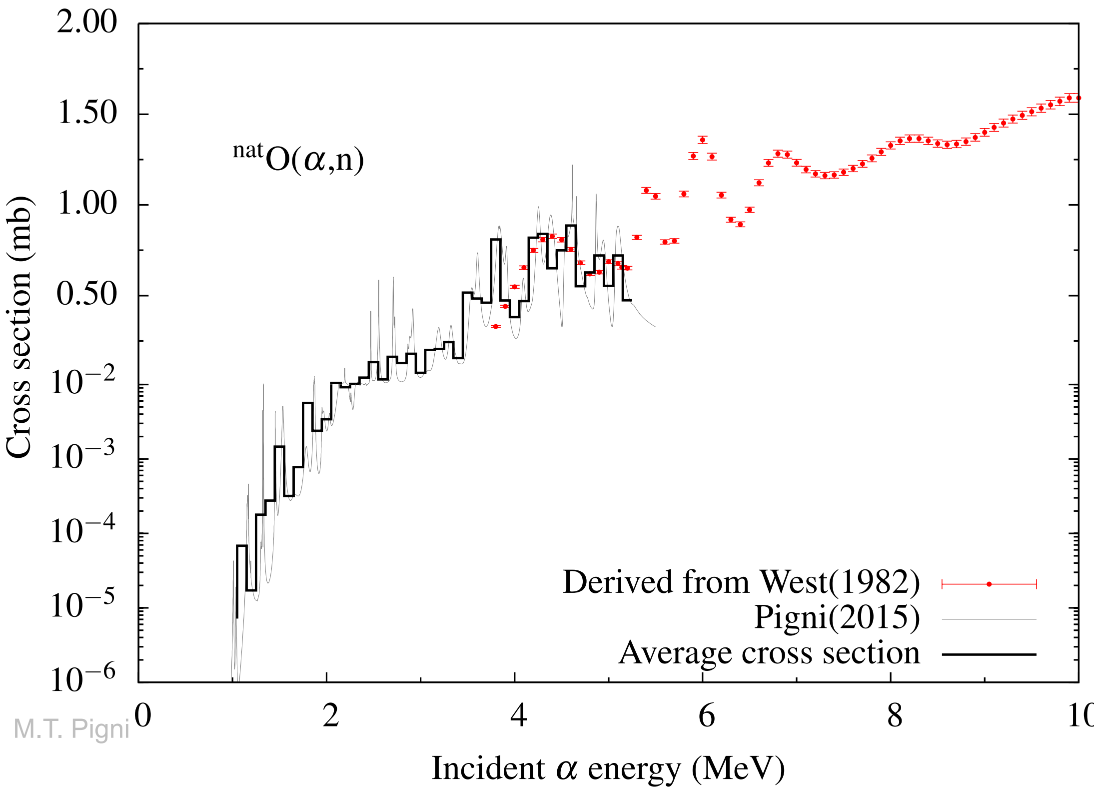
Methodology
(to generate (α,n) cross section covariances above 5 MeV)

- Above 5 MeV cross sections based on West's experimental data on ^{nat}O
  - West's data measured on a thick target (1.5% uncertainty)
- JENDL cross section data for ^{17}O (0.038%) to determine ^{18}O (0.205%) out of ^{nat}O.



Integrating over the energy range 3.8 up to 5 MeV, i.e. the energy range common to both data sets, we found the average cross sections (black line) were 3% lower than the West's cross sections (red dots).

Methodology
(to generate stopping power cross section covariances)

- The stopping power cross sections and the (α,n) reaction cross sections are both and concurrently used to compute the average number of emitted (α,n) neutrons
- Fit of ASTAR data with analytical functions

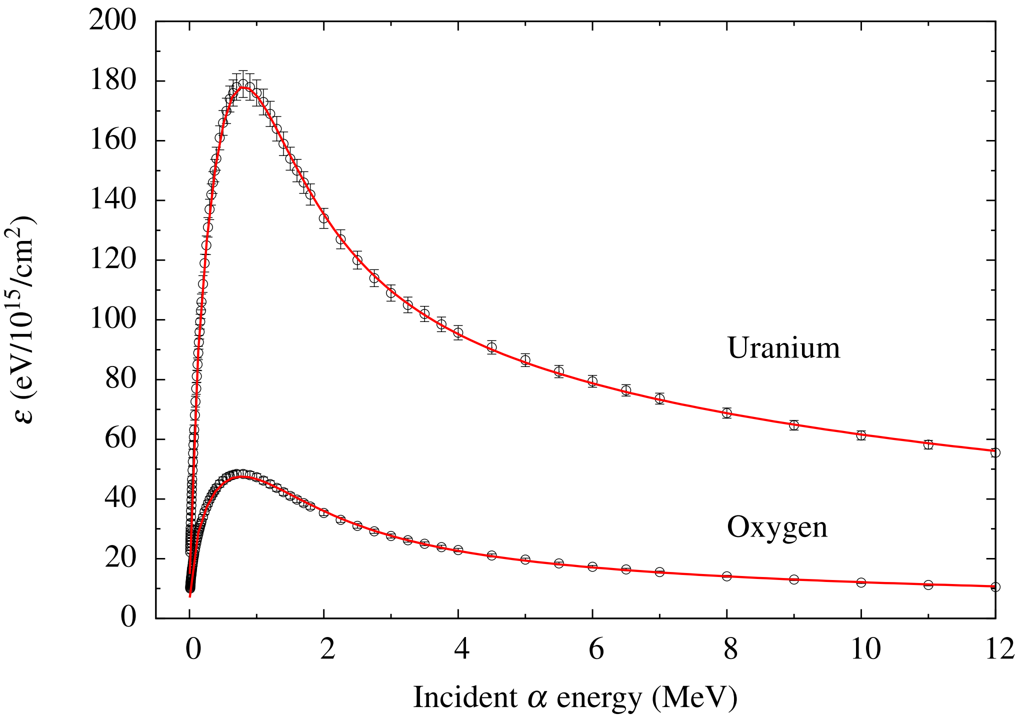
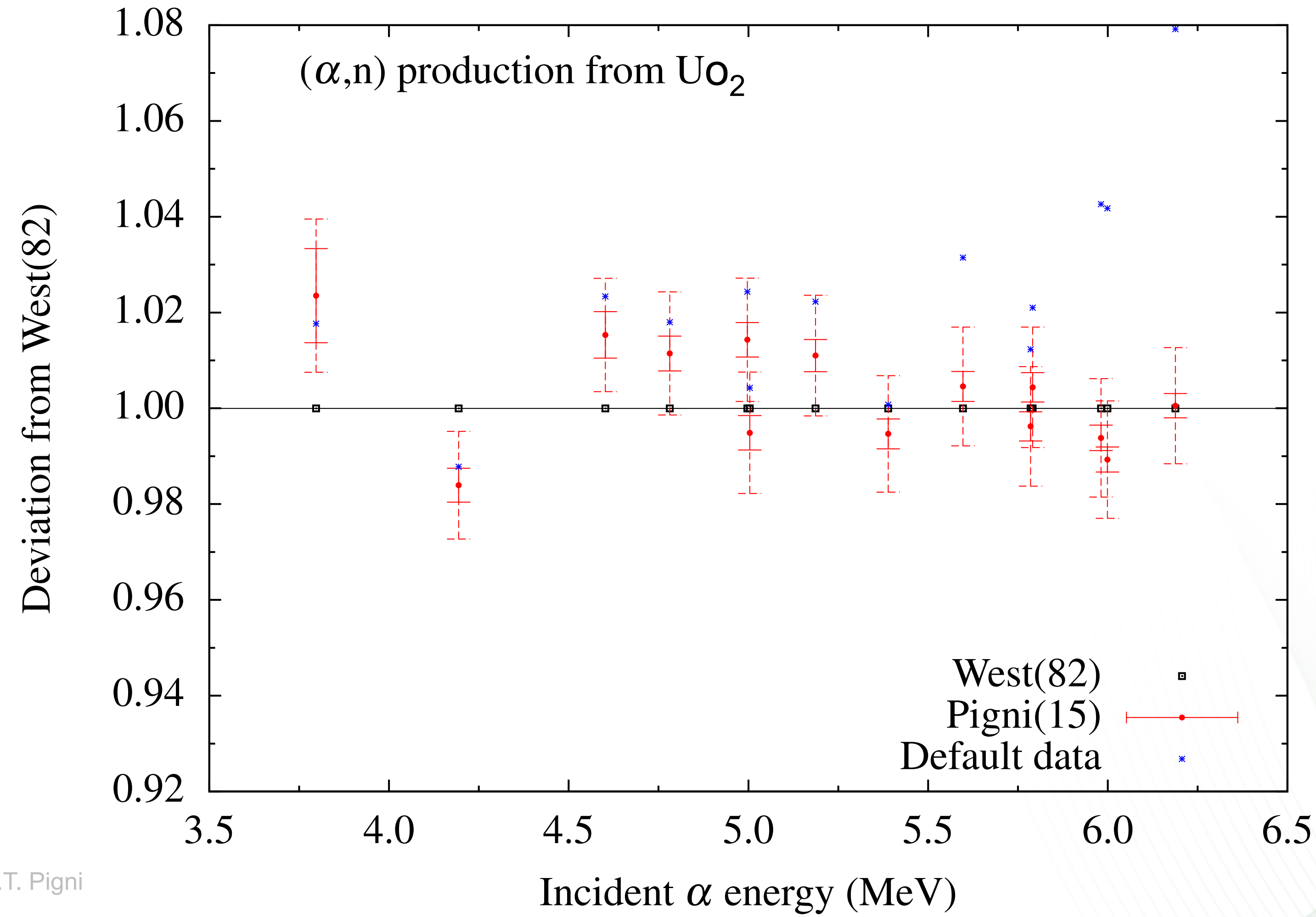


TABLE II. Coefficient factors and related covariance information to compute from analytic functions of Eq. (7) the oxygen and uranium stopping power cross section of Fig. 6.

oxygen							
No.	c_i	$\pm\Delta c_i(\%)$	Correlation matrix				
1	3.439	1.12	1.000				
2	0.438	0.62	-0.967	1.000			
3	45.426	9.18	0.450	-0.546	1.000		
4	17.111	23.21	-0.364	0.449	-0.984	1.000	
5	1.458	26.84	-0.459	0.556	-0.996	0.971	1.000
uranium							
No.	c_i	$\pm\Delta c_i(\%)$	Correlation matrix				
1	6.072	0.42	1.000				
2	0.554	0.18	-0.965	1.000			
3	368.5	2.55	0.302	-0.376	1.000		
4	2.090	3.57	-0.106	0.148	-0.930	1.000	
5	0.472	5.06	-0.306	0.381	-0.995	0.908	1.000

Results

- Uncertainties on (α,n) production from uranium computed from covariance information on reaction cross section of $^{17,18}O(\alpha,n)$ and stopping power cross section on oxygen and uranium. Deviation from West data (in blue and red dots) is also shown.



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Summary and Conclusions

- Developed methodology to generate uncertainty on neutron source emission spectra based on
  - Reaction cross sections: $^{17,18}O(\alpha,n)$
  - Stopping power cross sections: U and O
- High energy data suggest a renormalization (+3%) of Bair's data in the low energy range
- Application of the method was performed on a simple but realistic case for uranium oxide fuel showing the impact of
  - Reaction cross section is about <0.5%
  - Stopping power cross section is about >1%
- **Paper published in *Prog. Nucl. Energy 91, 147 (2016)***

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