

Evaluation and Validation of the $\alpha+^{17,18}\text{O}$ Cross Sections

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Nuclear Data and Criticality Safety

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Recent (α,n) ORNL/Notre Dame Measurements

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Motivations and Background

- Nuclear data impact every aspect of nuclear nonproliferation modeling and simulation, from the design of nuclear detection instruments used in nondestructive technologies to the interpretation of measurement data
- Nuclear data describing α -particle interactions on light nuclei, e.g. fluorine and oxygen, are essential for calculating neutron emission via (α ,n) processes
 - Codes such as SOURCES4C¹ or ORIGEN , widely used in nuclear safeguards and spent nuclear fuel characterization, rely on nuclear data to estimate neutron source intensities for α -particle reactions
 - Currently, evaluated data libraries contain no information on covariance data. Therefore, uncertainties in the estimated neutron source intensities or emission spectra cannot be assessed
- US ENDF/B-VIII.0 library includes a very limited sub-library of evaluated data, such as resonance parameters or cross sections, for induced α -particle reactions
- Within a previous project (2016) 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
 - A novel methodology to estimate the uncertainty in the neutron generation rates has been demonstrated for uranium oxide fuel types (*Prog. Nucl. Energy*, **91** 147, 2016)
- This presentation summarizes the formal $\alpha+^{17,18}\text{O}$ evaluations and related validation (*Prog. Nucl. Energy*, **118** 103130, 2020)

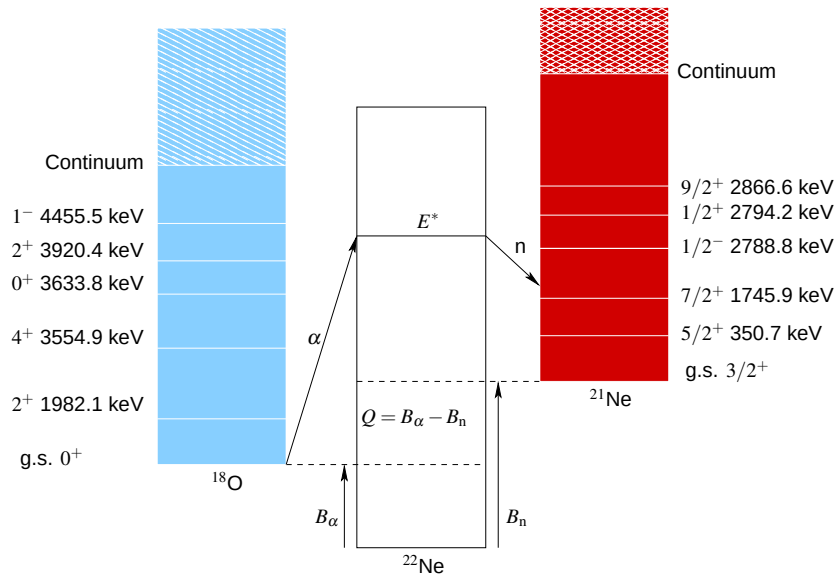
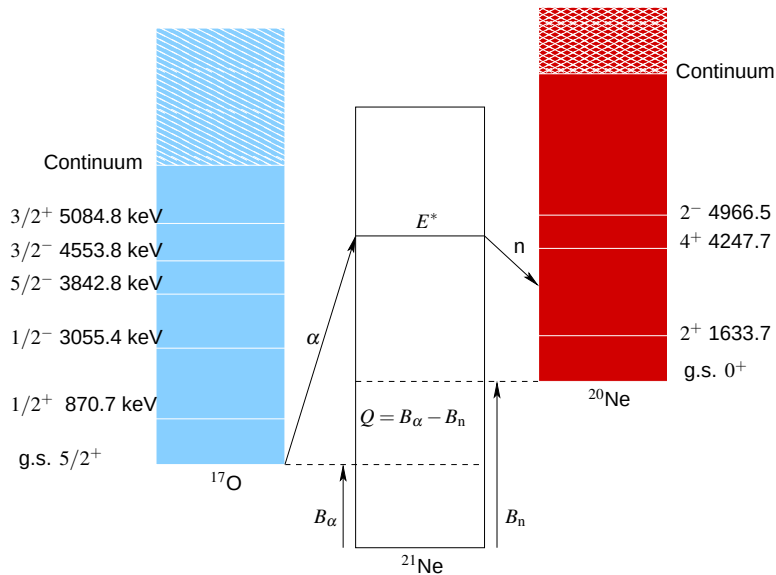
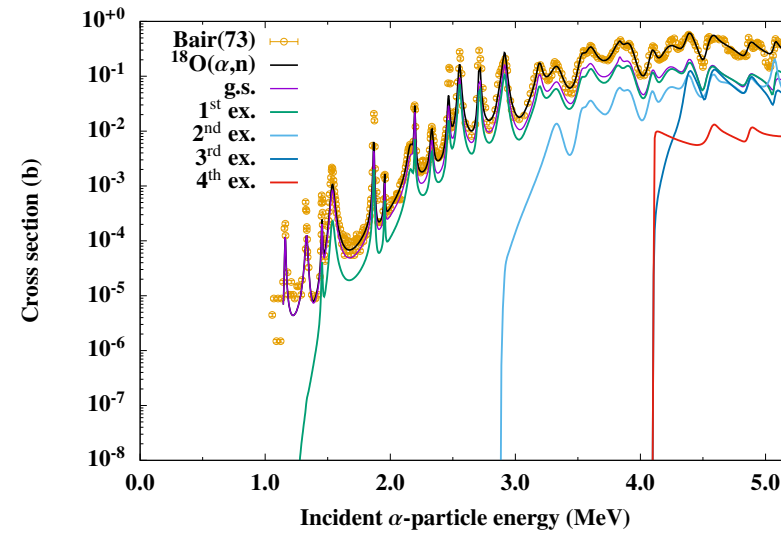
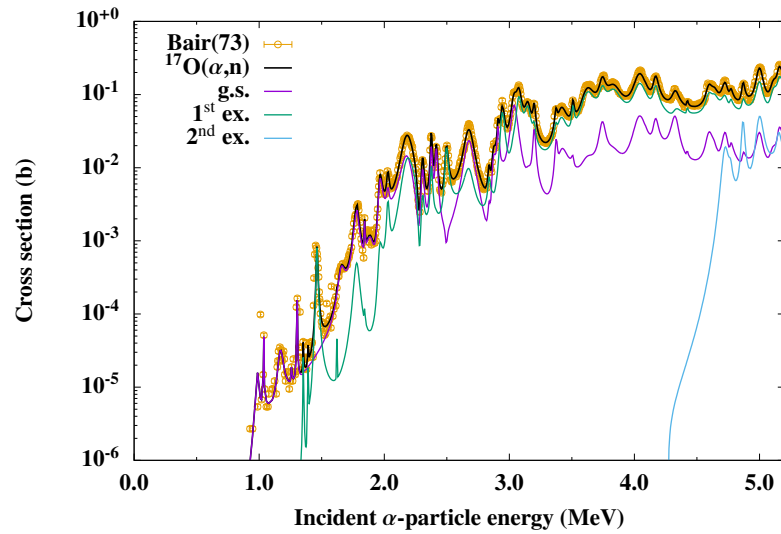
¹W. B. Wilson et al., Report LA-UR-02-1839, LANL (2002)

Introduction

Updates on the $\alpha+^{17,18}\text{O}$ Evaluations

- R-matrix analyses for (α,n) reaction channels up to about 5 MeV was based on available experimental data sets
 - (α,n) experimental data are based on Bair's measurements for both nuclei. However, Bair's data are convoluted and it is impossible to quantify accurately the partial cross sections related each excited states
 - In JENDL-AN (2002) the partial cross sections were based on Hauser-Fashbach (HF) calculations
 - In Sources4C the calculations of the partial cross section related to each excited states are also based on HF calculations performed by the GNASH code
- For ^{18}O Elastic channel, the spin assignment was based on measurement and the resonance analysis of Goldberg (Phys. Rev. C **69**, 24602, 2004) for available excitation energies
 - Goldberg's elastic angular excitation functions and angular distributions not possible to fit together with (α,n) data sets
- Some information on the proper distribution of the excited states may be obtained from integral measurements
- A suite of integral measurements (neutron yields and energy neutron distributions) available for oxide compounds was generated to validate the oxygen cross sections

$\alpha + {}^{17,18}\text{O}$ Partial Cross Sections



Neutron Energy Distribution (Isotropic Approximation)

The fraction of the total neutron distribution can be defined (E, laboratory)

$$dq_i^m = \int_{E_m}^{E_\alpha} \frac{d\sigma_i^m}{d\Omega} \varepsilon(E)^{-1} dE d\Omega. \quad (1)$$

The differential cross sections written in terms of Legendre polynomials ($d\Omega = 2\pi d\mu$ with $\mu \equiv \cos \theta$, center of mass)

$$\frac{d\sigma_i^m}{d\Omega}(E, \mu) = \frac{\sigma_i^m(E)}{4\pi} \left[1 + \sum_{l \geq 1} b_l P_l(\mu) \right], \quad (2)$$

In isotropic approximation ($b_l=0$)² and from the conservation of the interaction probability between center of mass and laboratory frame systems,

$$\text{pdf}(\mu) d\mu = \text{pdf}(E_n) dE_n, \quad (3)$$

and the outgoing neutron energy (in the laboratory system)

$$E_n^m(E, \mu) = g_m(E) + f_m(E)\mu, \quad (4)$$

the probability density function $\text{pdf}(E_n)$ for E_n is also constant being the neutron distribution in angle given by

$$dE_n/d\mu = f_m(E)/2\pi. \quad (5)$$

²The probability to scatter in any direction is constant and the probability density function for μ is $\text{pdf}(\mu) = 1/2$ with $\mu \in [-1:1]$.

Suite of Neutron Yield Source Calculations

Validation Procedure

Table 2

Calculated and measured values of total neutron yields for a selected suite of neutron sources.

Source	$\langle E_\alpha \rangle$ (MeV)	$p \pm \Delta p$	Units	SOURCES4C	JENDL	This work	Reference
PuO ₂ (801)	5.3	157.8 ± 9.8	n/s/sample ^a	144.4	145.8	146.8	(Kimura et al., 1986) ^b
PuO ₂ (802)	5.2	51.3 ± 2.5	n/s/sample ^a	54.1	54.5	55.1	(Kimura et al., 1986) ^b
²⁴¹ AmO ₂	5.5	236.9 ± 5.9	n/s/sample ^a	256.1	259.8	260.3	(Kimura et al., 1986) ^b
²¹⁰ Po-O ^{nat}	5.3	0.068 ± 0.011	n/10 ⁶ α	0.063	0.063	0.064	Vukolov (1983)
²³⁸ PuO ₂	5.5	2.0 ± 0.2	n/10 ⁸ α	2.189	2.221	2.225	Vukolov (1983)
²³⁸ PuO ₂	5.5	2.204 ± 0.033	n/10 ⁸ α	2.189	2.221	2.225	Vukolov (1983)
²³⁸ PuO ₂	5.5	2.31 ± 0.14	n/10 ⁸ α	2.189	2.221	2.225	Vukolov (1983)
²³⁸ PuO ₂	5.5	2.291 ± 0.027	n/10 ⁸ α	2.189	2.221	2.225	Vukolov (1983)
²³⁸ PuO ₂	5.5	2.25 ± 0.11	n/10 ⁸ α	2.189	2.221	2.225	Vukolov (1983)
²⁴¹ AmO ₂	5.5	2.2 ± 0.3	n/10 ⁸ α	2.189	2.221	2.225	Vukolov (1983)
²⁴¹ AmO ₂	5.5	2.78 ± 0.41	10 ³ n/g ²⁴¹ Am	2.75	2.78	2.79	Vukolov and Chukreev (1987)
UO ₂	4.0 ^c	5.1 ± 0.3	n/10 ⁹ α	5.6	5.6	5.7	Jacobs and Liskien (1983)
UO ₂	4.5 ^c	9.1 ± 0.5	n/10 ⁹ α	10.1	10.1	10.3	Jacobs and Liskien (1983)
UO ₂	5.0 ^c	16.0 ± 0.9	n/10 ⁹ α	15.5	15.3	15.8	Jacobs and Liskien (1983)
UO ₂	5.5 ^c	19.9 ± 1.0	n/10 ⁹ α	21.7	22.0	22.0	Jacobs and Liskien (1983)
UO ₂	4.0 ^c	4.9 ± 0.1	n/10 ⁹ α	5.6	5.6	5.7	West and Sherwood (1982)
UO ₂	4.5 ^c	10.3 ± 0.1	n/10 ⁹ α	10.1	10.1	10.3	West and Sherwood (1982)
UO ₂	5.0 ^c	15.7 ± 0.2	n/10 ⁹ α	15.5	15.3	15.8	West and Sherwood (1982)
UO ₂	5.5 ^c	22.1 ± 0.3	n/10 ⁹ α	21.7	22.0	22.0	West and Sherwood (1982)
UO ₂	4.0 ^c	5.9 ± 0.6	n/10 ⁹ α	5.6	5.6	5.7	Bair as in (Jacobs and Liskien, 1983)
UO ₂	4.5 ^c	10.7 ± 1.1	n/10 ⁹ α	10.1	10.1	10.3	Bair as in (Jacobs and Liskien, 1983)
UO ₂	5.0 ^c	16.4 ± 1.6	n/10 ⁹ α	15.5	15.3	15.8	Bair as in (Jacobs and Liskien, 1983)
UO ₂	5.5 ^c	23.6 ± 2.4	n/10 ⁹ α	21.7	22.0	22.0	Bair as in (Jacobs and Liskien, 1983)

^a (α,n) component only.

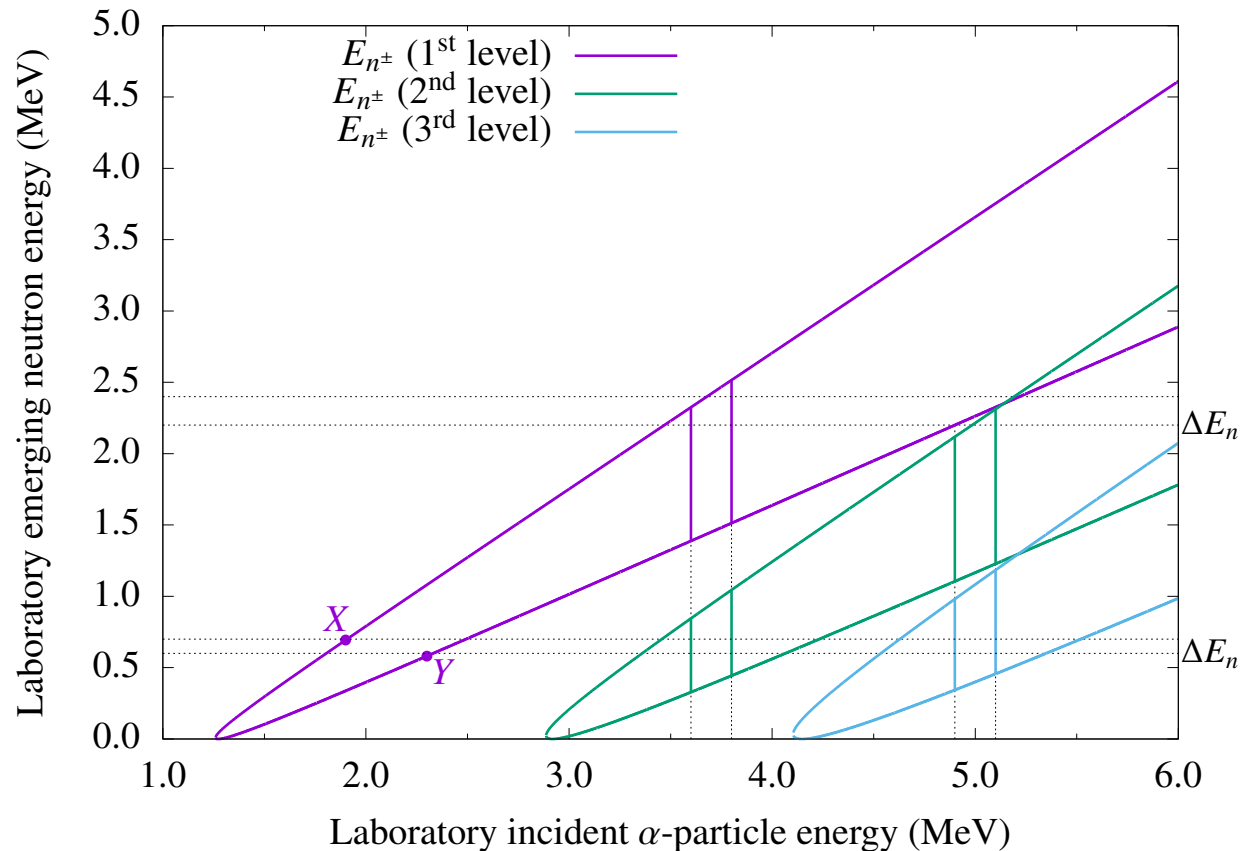
^b A re-analysis of the experimental setup suggested the energy dependence of the BF₃ array could lead to statistically significant corrections and was marginally taken into consideration in the measurements. Since such a correction can reduce the measured data by at least 10%, Kimura's measured data cannot be considered as quality benchmarks.

^c Mono energetic α-particle.

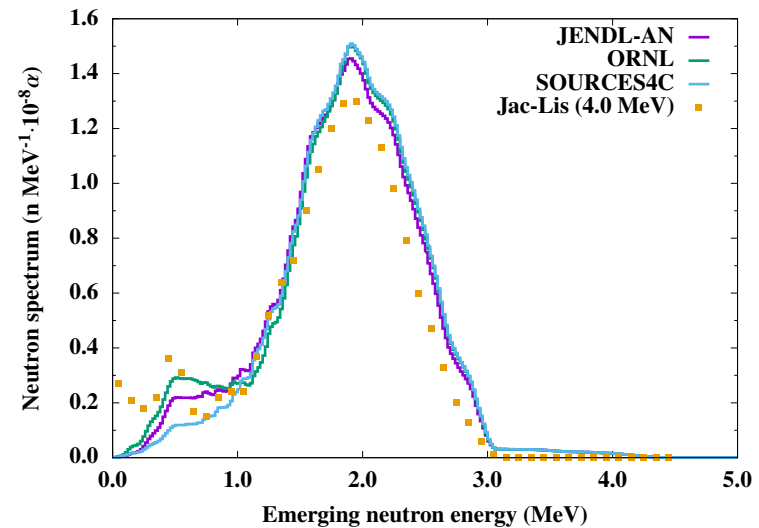
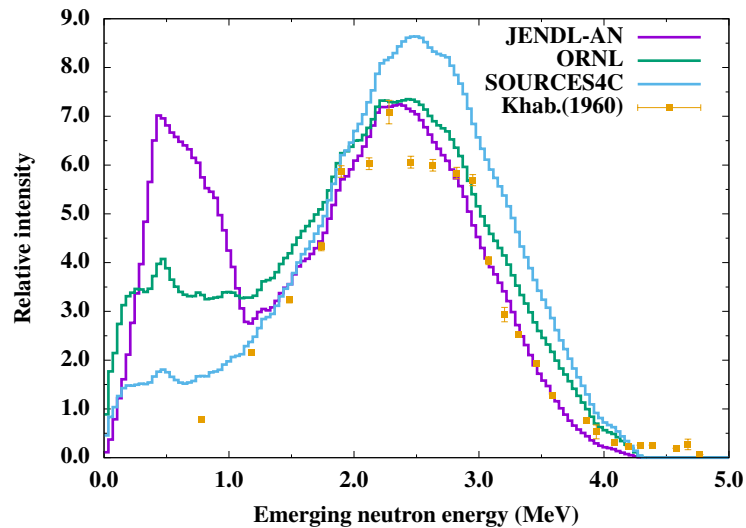
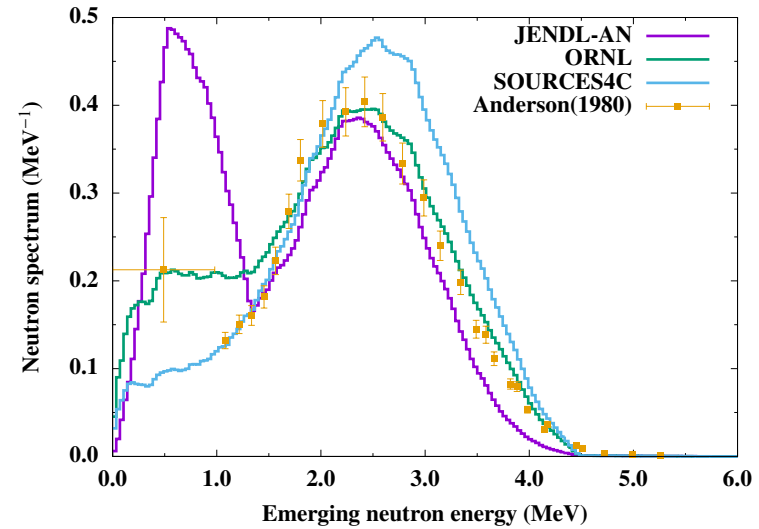
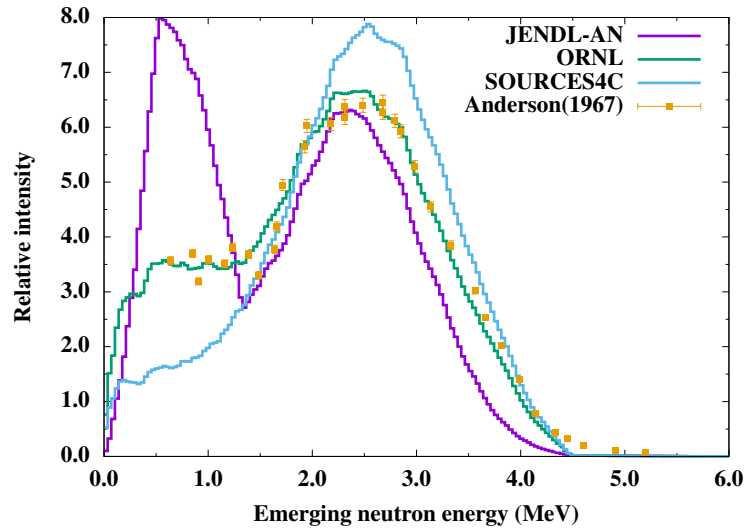
Neutron Energy Distribution (Isotropic Approximation)

The neutron energy distribution for the target nuclide i related to the (α, n) in the approximation of isotropic angular distribution of neutron emitted in the center of mass

$$\frac{dq_i^m}{dE_n} = \int_{X(E_n)}^{Y(E_n)} \frac{\sigma_i^m(E) \varepsilon(E)^{-1}}{(E_{n+}^m - E_{n-}^m)} dE, \quad (6)$$



Neutron Energy Distribution Calculations



Joint Measurement Campaign (ORNL + Notre Dame)

Recent Experimental Activities

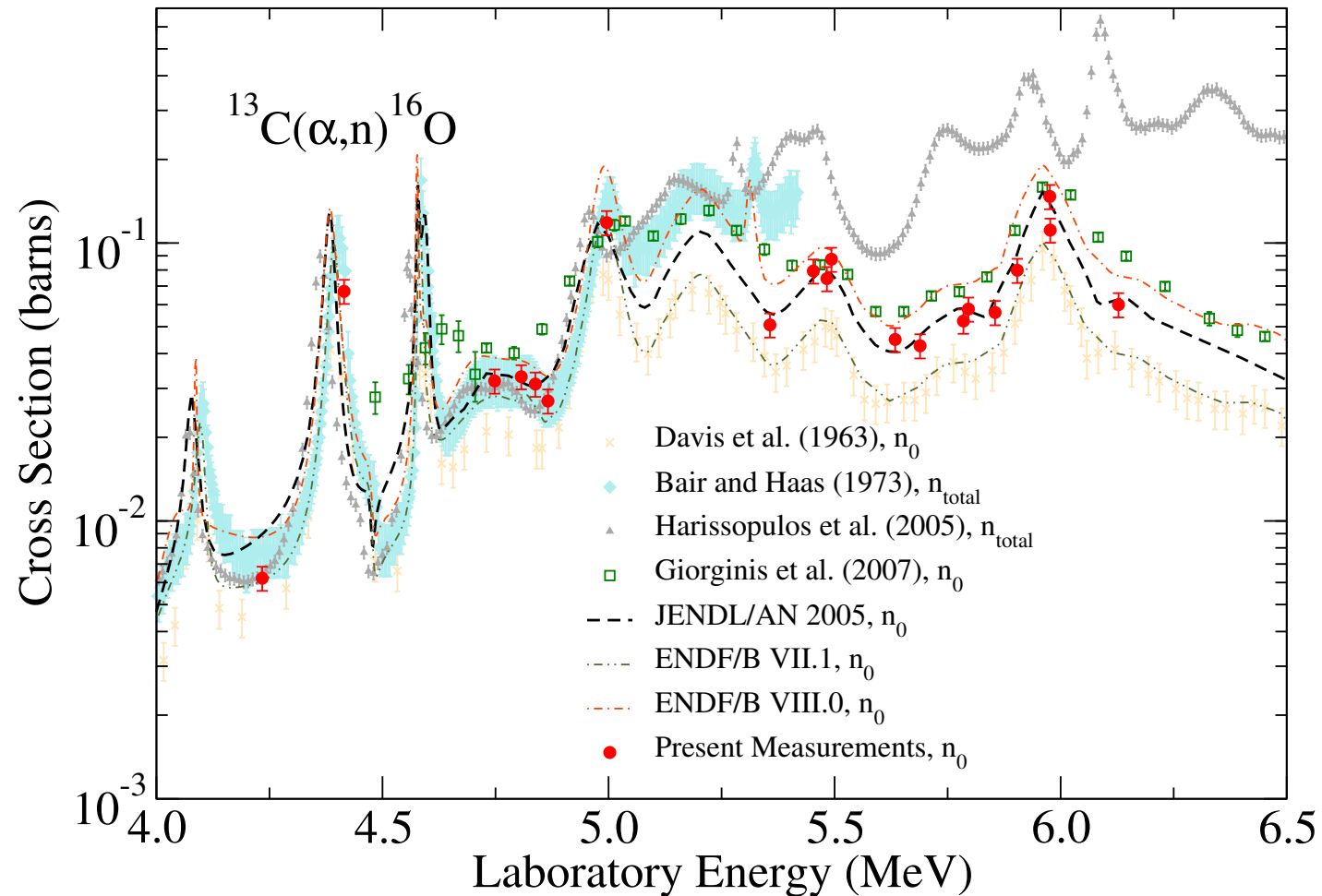
- $^{10}\text{B}(\alpha, n_0)^{13}\text{N}$ ($2.2 < E_\alpha < 4.9$ MeV) performed at National Ignition Facility
- $^{17}\text{O}(\alpha, n_{0,1,2})^{20}\text{Ne}$ performed at Notre Dame facility (2019)
- $^{18}\text{O}(\alpha, n_{0,1,2,3,4})^{21}\text{Ne}$ performed at Notre Dame facility (in progress)
- $^{13}\text{C}(\alpha, n_0)^{16}\text{O}$ performed at Notre Dame to test agreement/disagreement with Harissopulos' measurements

Future Plans

- To propose high-resolution measurement on $^{19}\text{F}(\alpha, n)$ cross sections

Joint Measurement Campaign (ORNL + Notre Dame)

SUBMITTED FOR PUBLICATION



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Thank you!