Recommendations from an (α,n) Nuclear Data Scoping Study

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Project Goal was to provide an actionable plan to resolve (α,n) nuclear data issues that impact applications

- Determine the state of the libraries and modeling capabilities
  - JENDL, TENDL, ENDF
  - SOURCES4C, GEANT, MCNP
- Determine application needs
  - UF_6 enrichment
  - Spent fuel safeguards
  - Passive NDA measurements – neutron/gamma
  - Neutron source characterization – PuBe, AmBe, etc…
  - Natural neutron background calculations
- Create a set of recommended tasks to resolve priority nuclear data and simulation needs.
- Contributors
  - David Brown (BNL)
  - Andrea Favalli (LANL)
  - Marco Pigni (ORNL)
  - Michael S. Smith (ORNL)
  - Michael Zerkle (NNL)
  - Stephen Croft (ORNL)
  - Les Nakae (LLNL)
  - Steve Skutnik (ORNL)
  - Will Wieselquist (ORNL)

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State of the (α,n) Data and Codes

- ENDF library is mostly theoretical (TENDL) or based on JENDL (Japanese library)
  - GNDS format is required to accommodate enhanced data
- Many cross sections for priority isotopes have large uncertainties and are based on old data
- Neutron energy spectra have large uncertainties
  - Require more information from experiments to improve evaluation of spectrum
- Neutron angular distributions not well known
  - Impact differential nuclear data measurements using beams
- Gamma emission energies should be measured
  - Provide information on excitation states populated for improved neutron emission spectra calculations
  - May be useful for NDA measurements
- Stopping powers not well understood
  - Application specific measurements are required
- Evaluation methods require code modifications
  - Experiments that capture the population of excited states using gamma measurement data needed to allow theoretical models to correctly predict the neutron spectrum
- SOURCES4C is unsupported and outdated

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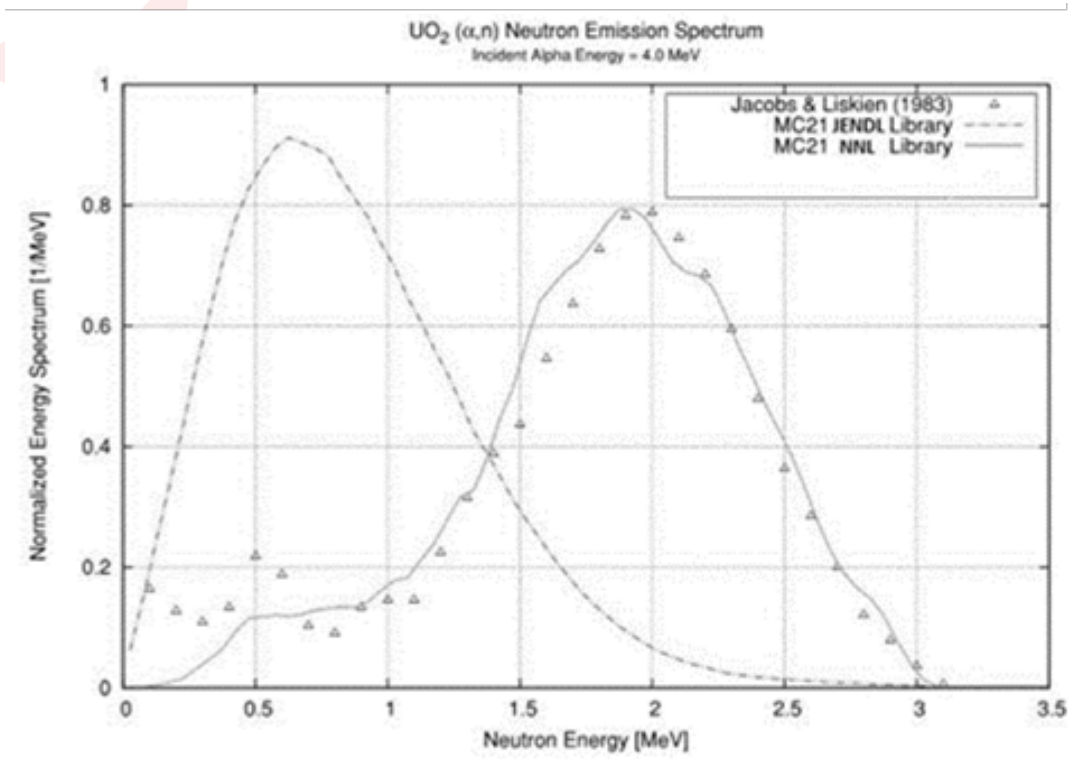
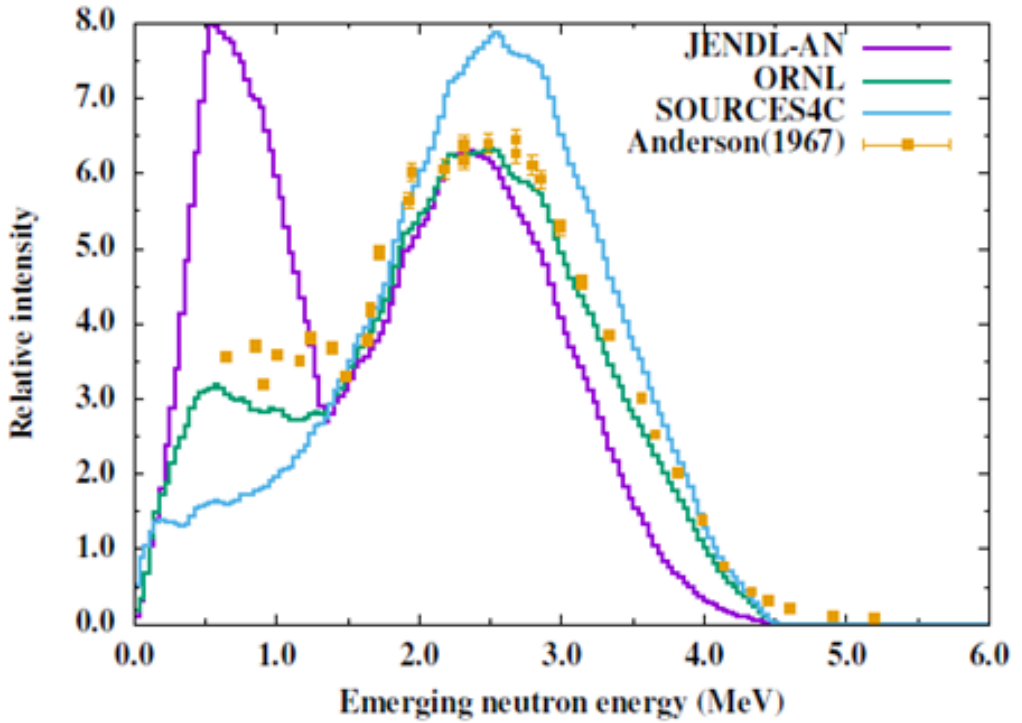
Neutron Spectrum is not well known even if the cross section has relatively low uncertainty





Calculations of neutron spectrum using MC21 with the JENDL O(α,n) library and the Naval Nuclear Laboratory (α,n) library compared to experimental data (left) and theoretical calculations of the neutron spectrum for O(α,n) reaction compared to the JENDLE library, SOURCES4C, and experimental data (right) [Pigni, 2016].

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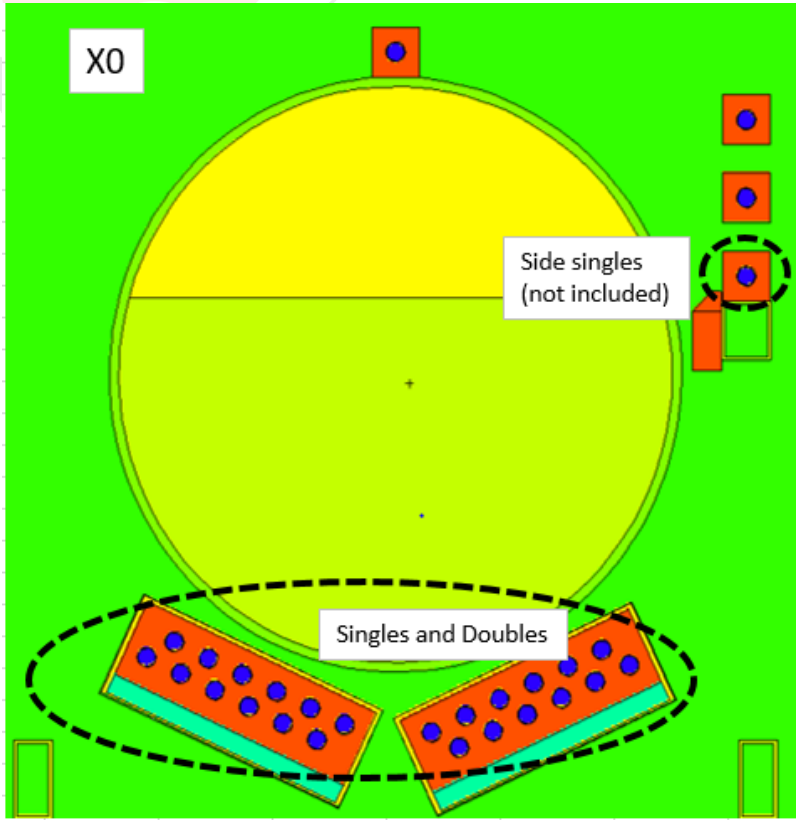
Uncertainty study of UF_6 cylinders at LANL

Goal: to determine the nuclear data contribution to the uncertainty budget for UF_6 cylinder measurements

Passive Neutron Enrichment Meter (PNEM)



- Two briefcase size polyethylene pods with twelve ^{3}He tubes at 10 atm.
- Varied enrichments and fraction of UF_6 bound to walls
- (α,n) source terms built on the Jacobs & Liskien data (‘83 and ‘85), modeled with SOURCES4C
- Included updated treatment of the stopping power.

Conclusion: The total neutron yield has biggest impact

- These detectors are relatively insensitive to neutron energy

Neutrons/sec/g ^{234}U	Source
460	(Reilly et al. 1971)
576 ± 7.3% (533-618)	(Sampson 1974)
474 ± 4.4% (453-494)	(Miller et al. 2014)
503 ± ~4% (483-523)	(Kulisek et al. 2017; uncertainty estimated by Croft et al. 2020)
507 ± 1.1% (501-513)	(Croft et al. 2020)
610	SOURCES-4C (Wilson et al. 2002)
604	Modified SOURCES-4C (Wilson et al. 2002; private comm. Croft & Gauld 2020)

D.P. Broughton, S. Croft, C. Romano, A. Favalli, “Sensitivity of the simulations of passive neutron emission from UF_6 cylinder to the uncertainties in both $^{19}F(\alpha,n)$ energy spectrum and thick target yield of ^{234}U in UF_6”, United States. https://doi.org/10.1016/j.nima.2021.165485

Priority Recommendations – 5 Year Plan

Prioritized by cross-cutting impact and program specific need for improved fluorine data

- SOURCES4C code modernization
- Evaluation of fluorine, oxygen and carbon (α,n) data
- Fluorine thick target integral and activation experiments
- ENDF database modernization, GNDS format
- Processing code updates

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Modernization of the SOURCES4C Code

PRIORITY TASKS

- Open-source distribution with software management – requires ownership/maintenance
- Nuclear data read from ENDF/B
  - Pointwise in energy (α,n) cross sections.
  - Prompt neutron emission spectra.
  - Prompt gamma emission spectra.
- Enable user input to supply specialized nuclear data files, stopping powers
- The code output will include reaction rates, neutron and gamma energies
- A Fortran/C++/Python API allows interface with other codes - MCNP or SCALE/ORIGEN

FOLLOW ON TASKS

- Verification and Validation
  - Application specific test suites

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Evaluation of Current Data

- **Fluorine Evaluation**
  - An evaluation of Fluorine in year one will provide useful evaluated data to the users until improved benchmark data can be provided.
    - Improved data requested by IAEA, recent measurement data (Peters 2016) never evaluated
  - Updated evaluation based on new data once experiments are complete.
- **Oxygen Evaluation**
  - New, higher resolution data sets will be available from Notre Dame with information on excitation states that will reduce uncertainties at lower energies in the previous evaluations.
- **Carbon Evaluation**
  - Carbon is the isotope used to calibrate detector systems for (α,n) measurements. There are large discrepancies in the data that are currently being examined by multiple groups. An evaluation of the data is needed once the discrepancies are resolved.
- **Code Development**
  - Reduced R-matrix formalism and penetrability factors to describe the break-up reaction channel and account for both gamma and neutron emission.

Postdoc or early career mentoring recommended

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Experiments on Fluorine to Reduce Uncertainties

PRIORITY TASKS

- **Fluorine Flat-response 4 pi Moderated Neutron Counter Measurements**
  - Measure total yield of neutrons from $^{19}F(\alpha,n)^{22}Na$ per incident alpha particle, integrated over all angles and all alpha energies.
  - Measure a series of fluorinated compounds to define the role that the stopping power plays in the total yield is important.
- **Fluorine Activation Measurements**
  - Measure ^{22}Na decay produced by the $^{19}F(a,n)^{22}Na$ reaction in a thick fluorinated target after bombardment by alpha particles.
- **Fluorine Neutron Spectrum Measurements**
  - Measure the energy spectrum and angular dependence of neutrons emitted from the thick stopping fluorinated sample simultaneously with the activation measurement
  - Use time-of-flight combined with deuterated liquid scintillator detector spectrum unfolding.
- **Thin Fluorine Target Energy-Differential Excitation Function Measurements**
  - 10 keV energy steps including neutron and gamma spectrum benchmarked to integral measurements.

ENDF (α,n) Nuclear Data Sub-Library in GNDS Format

PRIMARY GOAL: Expanded ENDF (α,n) library

- Extend the GNDS tools and format to allow for charged particle reactions
- Develop processing and evaluation codes Fudge, GIDI and AMPX managed by the NEA GNDS Expert Group (EG-GNDS)
  - Extend FUDGE processing code and the open source GIDI API.
    - Used by LLNL codes and, in the future, GEANT4
  - Extend the SAMMY evaluation tool and AMPX processing code.
    - Used by SCALE/ORIGEN

Top Priority isotopes for Nonproliferation

Isotope	Current State of Data	Applications	Recommendation
^{13}C	Recent data (4-6.5 MeV) to be published, No evaluation	NDA/ER, safeguards, advanced reactors, background simulations, alpha therapy	Low energy measurement needed < 1MeV - 4 MeV. Recent Notre Dame measurement from 4-6.5 MeV. Discrepancies in past data are being examined and new evaluation once data issues resolved.
^{17}O	NNL and ORNL evaluation available for next ENDF release - still discrepancies in neutron spectrum	safeguards, reactors, NDA/ER, Background simulations, alpha therapy	Recent Notre Dame measurement from 0.8 - 7 MeV will provide data to resolve discrepancies in neutron spectrum. Updated evaluation needed
^{18}O	NNL and ORNL evaluation available for next ENDF release - still discrepancies in neutron spectrum	safeguards, reactors, NDA/ER, Background simulations, alpha therapy	Recent Notre Dame measurement from 2-8 MeV will provide data to resolve discrepancies in neutron spectrum. Updated evaluation needed
^{19}F	No evaluation on recent data, benchmark needed	safeguards UF6, FLiBe reactors, fuel cycle & waste management applications (fluoride volatility process)	Previous cross section measurements provided improvement. Still need total neutron, neutron spectrum, gamma emission with lower uncertainty per IAEA. A combination of improved total cross section, angular distributions, neutron energy spectra, activation, and thick target integral measurements are needed to reduce uncertainties.

Secondary Priority – Safeguards and Reactors

^{7}Li	Adjusted evaluation based on JENDL data available for next ENDF release, large discrepancies in data require new experiments	FLIBe reactors, NDA/ER, spent fuel safeguards, Actinide-Li neutron sources	New experiments with ability to resolve excitation states and reaction channels and new evaluations are needed in order to accurately model neutron sources.
^{9}Be	NNL evaluation available for next ENDF release	FLIBe reactors, NDA/ER, spent fuel safeguards, Actinide-Be neutron sources	Neutron spectrum been validated by NNL by adjusting JENDL for additional breakup reaction channels. Would benefit from new measurements that provide information on reaction channels
^{11}B	No evaluation	Reactors, safeguards	Only measured up to 2.5 MeV
^{10}B	Recent Experiments up to 4.5 MeV, requires evaluation	Reactors, safeguards	Recent experiments from 1.5-4.5 MeV conducted at Notre Dame. Data above 4.5 MeV and an evaluation are needed
^{27}Al	Discrepancies in data from 1970s No evaluation	NDA/ER, advanced reactor fuel and spent fuel safeguards	New experiments and evaluations with resolved excitation states and neutron spectrum are needed for advanced reactor fuels and emergency response.

Lower priority

^{23}Na	Need new experiments and evaluation	molten salt reactors	New experiments with ability to resolve excitation states and evaluations are needed for advanced reactor fuels
^{25}Mg	Need new experiments and evaluation	MgCl molten salt, fuel cladding, astrophysics	Recent Notre Dame measurement from 1.5-3.5 MeV. Experiments up to 6.5 MeV needed for advanced reactors. Evaluation needed
^{26}Mg	Need new experiments and evaluation	MgCl molten salt, fuel cladding, astrophysics	Planned Notre Dame measurement from 1.5-3.5 MeV. Experiments up to 6.5 MeV needed for advanced reactors. Evaluation needed
^{29}Si	Need new experiments and evaluation	Advanced reactor fuels	New experiments with ability to resolve excitation states and evaluations are needed for advanced reactor fuels
^{30}Si	Need new experiments and evaluation	Advanced reactor fuels	New experiments with ability to resolve excitation states and evaluations are needed for advanced reactor fuels
^{37}Cl	Need new experiments and evaluation	Chloride-based (Na, LiCl, MgCl) molten salt fuel, pyroprocessing	New experiments with ability to resolve excitation states and evaluations are needed for advanced reactor fuels
^{41}K	Need new experiments and evaluation	Pyroprocessing, molten salt fuel (FLiNaK)	New experiments with ability to resolve excitation states and evaluations are needed for advanced reactor fuels

Thank You!

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