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OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE US DEPARTMENT OF ENERGY

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Reactor and Nuclear Systems Division

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ABBREVIATED TERMS

API	application programming interface
CSEWG	Cross Section Evaluation Working Group
ENS	Evaluated Nuclear Structure
FLiBe	lithium-beryllium-fluoride
GNDS	General Nuclear Database Structure
IAEA	International Atomic Energy Agency
JAEA	The Japan Atomic Energy Agency
LANL	Los Alamos National Laboratory
LLNL	Lawrence Livermore National Laboratory
NDA	nondestructive assay
NNDC	National Nuclear Data Center
NNL	Naval Nuclear Laboratory
PNEM	Passive Neutron Enrichment Meter
RRR	resolved resonance region
RSICC	Radiation Safety Information Computational Center
SNM	special nuclear material
TNYA	total neutron yield per alpha particle emitted

ABSTRACT

Neutrons from the (α,n) reaction are an important component of nondestructive assay techniques to determine enriched uranium and other actinide inventories in a variety of critical points in the nuclear fuel cycle. However, uncertainties in the cross section, total neutron yield and neutron spectrum, and gamma emissions from these reactions, such as $^{19}F(\alpha,n)$ and $^{17,18}O(\alpha,n)$, introduce large uncertainties in the determination of mass of actinides of interest and can represent several significant quantities in unaccounted material in certain facility processes. Calculations and measurements depend on accurate nuclear data; however, much of the relevant data in use today was measured in the 1980s and earlier and has not been updated. Thus, the current uncertainties in the cross sections and neutron emission spectra are unacceptably large. This report documents the results of a scoping study of (α,n) reaction data that considered the current state of the data and recommends areas of improvement. It also addresses the codes use to calculate the (α,n) neutron and gamma source terms and recommends code improvements to support required analysis.

1. INTRODUCTION

The alpha particle decay of heavy nuclei in compounds results in neutron and gamma emissions through the (α,n) reaction. The neutrons and gammas emitted contribute to reactor flux and the dose from materials processing. They also provide information about the material being measured during nondestructive analyses. For most applications, the cross section of (α,n) reactions is needed up to 6.5 MeV, which is above the maximum energy of alpha emission for most alpha emitters. However, the decay chains of uranium, thorium, and actinium include very short-lived isotopes of polonium, astatine, and others that decay with alpha particles with energies at nearly 9 MeV. They are important for neutron background calculations for low-background experiments. For isotope production, (α,n) cross sections up to 30 MeV are required for a few specific isotopes.

The neutron source term can be calculated with transport codes such as MCNP [Werner 2018] and Geant4 [Agostinelli 2003], which transport the alpha particles, tabulate interactions, and create neutrons on an event-by-event basis. These codes are, however, computationally intensive. Another popular solution is a source term calculator such as SOURCES [Wilson 2002], which creates a neutron source term based on dimensions and isotopic concentrations. The transport is then calculated using stopping powers and codes such as SRIM [Ziegler 2017]. While gamma rays are not typically addressed, they can be useful for specific applications.

These calculations and measurements depend on accurate nuclear data to support the analysis. However, much of the relevant data still in use today was measured in the 1980s and earlier and has not been updated. The current uncertainties in the cross sections and neutron emission spectra are unacceptably large. Those uncertainties, coupled with the computational power now available to utilize any improved data, prompted a scoping study of (α,n) reaction data. This report describes the study and its outcomes, including the state of the current data, the needs for enhanced data for specific applications, and the code enhancements required to optimize the use of the data.

1.1 MOTIVATION: OUTCOMES OF THE NUCLEAR DATA ROADMAPPING AND ENHANCEMENT WORKSHOP

This scoping study was prompted by the outcome of the Nuclear Data Roadmapping and Enhancement Workshop [Romano, 2018], which afforded the nuclear data and user communities an opportunity to discuss (α,n) nuclear data needs and to recommend efforts to improve the data that impact nonproliferation applications. The participants discussed issues identified in the outgoing neutron

spectrum, recent measurements of ^{17,18}O(α,n) and ^{19}F(α,n), and evaluation methods that include covariances.

For nonproliferation applications, the priorities are uranium and plutonium isotopes in oxide and fluoride compounds. For oxygen, fluorine, and other elements, differential data are needed over the energy range of < 1 MeV to 9 MeV. There are existing fluorine measurement data that have not been analyzed or evaluated. There are also measured but unanalyzed neutron angular distribution data that would be important for the evaluations.

Many of the differential measurements in the literature use ^{13}C(α,n) as a reference reaction, and improved carbon data are therefore important. It is also important to measure or reevaluate (α,n) reactions on many of the stable, light elements that are relevant to nonproliferation, such as those found in structural materials and those mixed with alpha emitters in reactor fuels. Measurements of the gamma-ray energies and neutron energy spectra are required for many applications, and the current uncertainties are large. For evaluations, improvements are needed in the codes to support the work. New evaluations involve a relatively low-cost, high-impact effort that would directly inform the need for improved data and can provide covariances required for uncertainty quantification. Application-specific benchmarks are also needed, as are thick target measurements to validate both differential data and simulations. Improvement or replacement of the widely used neutron source term code SOURCES 4C is needed. Furthermore, stopping powers and energy loss in mixed materials need to be better understood because variations in neutron rates have been observed from different material forms.

2. ENDF LIBRARIES

JENDL/AN-2005 [Shibata 2011] is the only complete, transport-ready, evaluated (combining theory with experiment) generally available library for alpha particles in ENDF-6 format. It includes data for ^{6,7}Li, ^{9}Be, ^{10,11}B, ^{12,13}C, ^{14,15}N, ^{17,18}O, ^{19}F, ^{23}Na, ^{27}Al, and ^{28,29,30}Si. The TENDL (α,n) libraries [Koning 2019] are also in ENDF format, but they are purely theoretical and are calculated using the TALYS reaction code [Koning 2013]. There are two different sub-versions of TENDL. In one of them, all the nonelastic reactions are grouped together into a single channel. A total nonelastic cross section is provided together with the particle yield and energy-angular distribution of every secondary particle. The other sub-version contains explicit cross sections up to 30 MeV. Plots of the TENDL-2019 and JENDL-2005 library cross sections as compared to experimental data can be found in Appendix A.

2.1 THE CURRENT STATE OF THE DATABASES

2.1.1 IBANDL (2010) and PIGE (2015) Databases

The International Atomic Energy Agency (IAEA) IBANDL and PIGE databases are careful compilations of reaction data for specific channels and (in the case of IBANDL) R-matrix evaluations of cross sections. In both cases, the evaluations are very well done but are incomplete and cannot be used for particle transport. The TENDL project is considering whether to extend both databases with TENDL evaluations. IBANDL is detailed in IAEA-TECDOC-1780 [IAEA 2015]; PIGE is detailed in IAEA-TECDOC-1822 [IAEA 2017].

2.1.2 JENDL (α,n) Reaction Data File 2003 (JENDL/AN-2003)

The Japan Atomic Energy Agency (JAEA) released JENDL (α,n) Reaction Data File 2003 (JENDL/AN-2003) 2005. Neutron emission data for the (α,xn) reactions were evaluated in the incident α-particle energy region below 15 MeV for nuclides important mainly in nuclear fuel-cycle applications, namely, ^{6,7}Li, ^{9}Be, ^{10,11}B, ^{12,13}C, ^{14,15}N, and ^{17,18}O [Murata 2002] and ^{19}F and ^{23}Na [Matsunobu 2002]. Each of the

nuclei that were considered exhibit significant fluctuations, so the evaluation consisted of R-matrix fits to cross sections, supplemented by Kalbach-Mann systematics and a pre-equilibrium model for outgoing particle distributions (mEXIFON).

2.1.3 JENDL (α,n) Reaction Data File 2005 (JENDL/AN-2005)

The Japan Atomic Energy Agency (JAEA) released JENDL (α,n) Reaction Data File 2005 (JENDL/AN-2005) in 2005. Neutron emission data for the (α,xn) reactions were evaluated in the incident α-particle energy region below 15 MeV for nuclides important mainly in nuclear fuel-cycle applications, namely, ^{6,7}Li, ^{9}Be, ^{10,11}B, ^{12,13}C, ^{14,15}N, ^{17,18}O, ^{19}F, ^{23}Na, ^{27}Al and ^{28,29,30}Si. [Murata 2006]. This revision added Al and Si but otherwise was a refinement of JENDL/AN-2003, using a different pre-equilibrium code (EGNASH-2).

2.1.4 TENDL (α,n) sublibrary

The TALYS Evaluated Nuclear Data Library has had 10 releases since its inception in 2008 (TENDL-2008, 2009, 2010, 2011, 2012, 2013, 2014, 2015, 2017, and 2019). Only TENDL-2015 and later versions are currently available; previous versions are outdated. TENDL contains α-induced reaction calculations for all isotopes living longer than 1 s (about 2,800 isotopes), up to 200 MeV, with covariances [Koning 2019]. The cross sections in TENDL are purely theoretical calculations using the Hauser-Feshbach theory coupled with the Avrigeanu optical model [Avrigeanu 2014] to produce smooth average cross sections and angular distributions over a coarse (~1 MeV) energy grid. As such, TENDL cross sections cannot represent either collisional systems with strong resonances or sizeable fluctuations. Furthermore, the Avrigeanu optical model is a spherical potential and therefore poorly reproduces features of reactions on deformed nuclei such as uranium. Nevertheless, TENDL's completeness can prove useful in filling in gaps in other evaluations.

2.1.5 ENDF/B-VIII.0 (α,n) sublibrary

An alpha reaction sublibrary was added in ENDF/B-VIII.0 providing a location for alpha-induced reaction data. At the time, only one evaluation (α + α) was added. That evaluation was taken from the ECPL library [Perkins 1981] [White 1991]. At the time, it was expected that more evaluations would be added, after the initial release of ENDF/B-VIII.0.

2.2 NEW EVALUATIONS READY FOR THE NEXT ENDF/B RELEASE

The Naval Nuclear Laboratory (NNL) recently tested the JENDL/AN-2005 (α,n) evaluations in its MC21 nuclear transport code [Griesheimer 2015]. Calculated neutron yields and energy/angle spectra were compared to available experimentally measured data from public literature. The MC21 neutron yield results compared very well with most experimental data. For neutron energy spectra, MC21 results were noted to deviate significantly from published experimental data for $^{nat}UO_2$, $^{238}PuO_2$, and Am-Be sources. The deviations were determined to be caused by physics deficiencies in the JENDL/AN-2005 evaluations for ^{17}O, ^{18}O, and ^{9}Be. NNL modified the evaluations to address the identified deficiencies and validated the new versions against experimental data. NNL submitted the modified special-purpose (α,n) evaluations for inclusion in the next ENDF/B release.

As of the time of writing, ENDF has five evaluations in the alpha sublibrary: ^{4}He, ^{7}Li, ^{9}Be, ^{17}O, and ^{18}O. Los Alamos National Laboratory (LANL) and Lawrence Livermore National Laboratory (LLNL) prepared ^{4}He and ^{7}Li, respectively; NNL (with IAEA) prepared ^{9}Be and $^{17-18}$O.

2.3 RECOMMENDATIONS FOR NEW LIBRARIES

The new data and capabilities are strongly constrained by the ENDF-6 format. The ENDF-6 format is the currently accepted form for data transfer between laboratories in the Cross Section Evaluation Working Group (CSEWG) collaboration, and all CSEWG processing and transport codes understand it; however, ENDF-6 was designed for storing neutron-induced transport data for use in 1960s-era computers. Although it is "punchcard ready," ENDF-6 forces all alpha-induced reactions into a few slots: MF=3 data for pointwise cross-section data, MF=6 data for outgoing particle energy-angle distributions, and MF=33 data for covariance on the MF=3 data. These formats are very inefficient and, more crucially, they give no provision for covariance data for the energy-angle distributions.

Many of the (α,x) reaction channels are two-body channels (in that there are only two active degrees of freedom, the target and projectile/ejectile), and therefore they are completely specifiable using the R-matrix theory used for the original evaluation. R-matrix fits provide a collection of resonance parameters that completely specify all cross sections, outgoing distributions, and other parameters, using a much smaller set of resonance parameters. Were the incident particle a neutron, the ENDF MF=2 format and accompanying MF=32 covariance format would be used to encode all the required resonance parameter data.

Extending the ENDF format to handle charged-particle incident reactions in MF=2 is possible and will likely be pursued. However, CSEWG and the rest of the nuclear data community are engaged in a major modernization effort, beginning with the format used to store the nuclear data and extending into the application programming interfaces (APIs) that interface with the data and the processing codes that transform the data into a form usable by application codes. It is wise to prepare for these future changes and maximize the return on the investment by developing a General Nuclear Database Structure (GNDS) format and supporting tools to allow for the inclusion of resonance parameter data for (α,x) reactions.

To prepare for the modernization of the database infrastructure (α,n+X) data, it is recommended that (a) the GNDS tools to charged particle reactions, including creating a GNDS format for charged particle incident resolved resonance data be extended and that (b) APIs be developed for common codes used at the national laboratories (such as the FUDGE and AMPX processing codes and the SAMMY evaluation code).

2.3.1 Benchmark Data

Once a robust suite of alpha data evaluations and the process for creating them has been developed, an equally robust series of tests will be needed to verify that the evaluations meet user needs. However, there is no standard process for evaluating averaged quantities such as the total neutron yield or integral measurements, and there are no data files for their storage and dissemination. Rather, that information tends to reside in publications, laboratory reports, and codes used by researchers in safeguards and related areas (e.g., SOURCES 4C). Creation of a repository for benchmark data where all users have ready access to benchmark results should be investigated.

2.4 BENEFIT TO USERS

Creating a new (α,n+X) library with a modern format and covariance data and a benchmark data repository will provide the user community access to complete nuclear data with the required covariances needed by a wide range of applications. The creation of a library with the best available evaluations will enable regular updates as new data are measured and evaluated.

3. EXPERIMENTAL METHODS

The use of best practices in experimental methods will provide the best possible data sets for each measured isotope. Only with a complete set of measurements can an evaluator ensure an accurate assessment of the data. In this section, we focus on the $^{19}F(\alpha,n)$ reaction, but the techniques described for new measurements are representative of those needed for other isotopes. Experimental methods of previous experiments are examined, sources of uncertainty and discrepancies between experimental results are identified, and a set of recommendations for new experiments is given.

3.1 PREVIOUS EXPERIMENTS

There have been numerous measurements of the $^{19}F(\alpha,n)^{22}Na$ reaction with a variety of target compounds and thicknesses. Three studies used thin targets: Balakrishnan et al. [Balakrishnan 1978], covering an alpha energy range of 2.6 to 5.1 MeV with uncertainties of approximately 15%; Wrean and Kavanagh [Wrean 2000], covering an alpha energy range of 2.3 to 3.1 MeV with uncertainties of 8%; and Peters et al. [Peters 2016b], covering 3.9 to 6.7 MeV at 135 energies with an average absolute cross-section uncertainty of 7.5%. All three thin target energy differential cross-section measurements show numerous resonances in the reaction cross section.

A number of thick-target cross-section measurements have also been made, including Bair and Gomez del Campo [Bair 1979], covering 4 to 8 MeV and using thick PbF_2 and ZnF_2 targets inside a 1.5 m diameter sphere of reactor-grade graphite with eight ^{10}B-enriched BF_3 counters embedded near the surface; Jacobs and Liskien [Jacobs 1983], covering 4.0 to 5.5 MeV at four energies using thick CaF_2 targets and a 5 cm diameter liquid scintillator detector with integrated yield uncertainties of approximately 25%; and Norman et al. [Norman 1984], covering 3.6 to 9.9 MeV using thick PbF_2 targets surrounded by a 1.5 m diameter graphite moderator and an array of 10 ^3He proportional counters. This last measurement, often cited in safeguard studies, determined an average cross section over broad 250 keV energy bins to a precision of approximately 10% to 12%, not including the uncertainty of the alpha stopping powers in the target. In a recent publication, Norman et al. [Norman 2015] derived the neutron yields per 10^6 incident alpha particles based on the data from their 1984 measurement with an uncertainty of 5%. However, their values differ from those of Bair and Gomez del Campo [Bair 1979] by up to 54%. [Simakov 2017] concluded that a recent update to the ^{19}F(alpha,xn) cross section resulted in a 10% to 40% underestimation of the known measured neutron thick target yields in fluorine compounds. This disagreement typifies the experimental situation; measurements with different target compounds (with different stopping powers), thicknesses, and techniques have a scatter of 40% [Bauer 1998, Allmond 2015].

A number of challenges contribute to the total uncertainty of the results for the thick-target measurements. First, the detector system must completely surround the sample because the total yield over all angles is needed; however, most detectors are not thick enough to capture neutrons of all emission energies and at all angles, and a hole is required in the detector for the beam to enter and bombard the sample. Therefore, some neutrons pass through all such detectors without being detected. The loss can be mitigated by modifying the detector, for example, by making the beam entrance as small as possible and increasing the detector's total length and diameter. Second, to estimate the number of neutrons that are not being counted, simulations are needed that require an input of the energy and angular dependence of neutron yields, the possible branching ratios of reactions to various excited states rather than ground states, and the energy dependence of the detector efficiency. Third, stopping powers also play a role in the interpretation of thick-target measurements. Fluorinated actinide samples that are assayed (e.g., UF_6) are not typically bombarded for thick-sample yield measurements because they would contribute a "beam off" neutron background (from alphas generated from actinide decay) that competes with the "beam on" neutron yield arising from alpha bombardment. Also, because the stopping power of alphas in the

bombardment sample (e.g., CaF_2) differs from the stopping power of alphas in the assayed sample (e.g., UF_6), two stopping-power corrections must be made to interpret nondestructive assay (NDA) detector yield determinations—one for the bombardment sample and the other for the actinide assay sample. One can mitigate this issue as much as possible by making a series of self-consistent thick-target measurements with a variety of fluorinated compounds covering a range of atomic numbers, including some fluorinated actinides, to identify and (to some extent) correct for the dependence on stopping powers.

Thick-target yields from fluorine compounds can be considered a fundamental quantity and are inherently easier to measure to better accuracy than differential cross sections. A cross section for comparison with previously measured differential cross-section data can be extracted by averaging thick-target yield measurements over small energy steps and correcting with the best available stopping power values. The combination of thick-target yields and energy-averaged cross sections with neutron spectrum measurements and with thin-target differential cross section (from [Peters 2016b]) can provide stopping power corrections. Furthermore, measurements of neutron yields from different fluorinated compounds provide a direct empirical test of the validity of the additivity of stopping powers in the Bragg-Kleeman rule [Bragg 2005], where sparse experimental data exist for use in determining the role of chemical bonding in stopping powers. Experiments of this type are recommended to verify stopping powers in the compounds required for the application of interest.

3.2 NEUTRON ENERGY AND ANGLE SPECTRA

In an experimental setup, for a given alpha bombarding energy, the neutrons emitted at a particular angle have a range of energies. A determination of the neutron energy spectrum as a function of angle and bombarding energy is needed to improve the precision and accuracy of NDA studies for the following reasons.

1. If the differential cross section is combined with the stopping power to determine the total neutron yield, that cross section must be integrated over all angles, and the assumption of an isotropic cross section was shown to be incorrect in several studies. For example, thin-target yield variations of a factor of three as a function of angle were seen at some alpha-bombarding energies [Peters 2016b]. Angular information on thin-target neutron yields are therefore critical to properly correct such measurements.

2. Detectors in both thick- and thin-target measurements have efficiencies that depend on energy, and the neutron spectra are needed to correct the measured yields appropriately. This effect is most serious for neutrons produced in nuclear reactions with multiple reaction channels open and for studies using moderated neutron detectors. Regarding the reaction channels, the neutron energies released by the $^{19}F(\alpha,n)^{22}Na$ reaction depend on the possible energy levels that can be populated in ^{22}Na for a given incoming alpha energy. For example, if we bombard a sample with alpha particles with 4.78 MeV energy (a critical energy for alpha decay of ^{234}U), neutrons of energies near 2 MeV will be produced via $^{19}F(\alpha,n)$ reactions proceeding to the ^{22}Na ground state (the "ground state reaction channel") as well as neutrons with significantly less energy via $^{19}F(\alpha,n)$ reactions proceeding to one of seven excited states in ^{22}Na. It is critical to understand the energy range of the neutrons because many moderated neutron detectors have a very different efficiency for neutrons over this broad energy range (up to a factor of two for small detectors, but less than 10% for large detectors). Moderated detectors use material such as paraffin to slow the neutrons down before a capture generates the detection signal, making it impossible to know the energies of the neutrons entering the detector (exiting the container being assayed). It is therefore critical to determine the relative neutron intensity at different energies to correct NDA detector signals with their energy-dependent efficiency and thereby determine the true neutron yield from the UF_6 in a container. Simulations such as MCNP

are invaluable for determining this yield correction for any particular combination of NDA detector and container once the neutron energy spectrum from the $^{19}F(\alpha,n)^{22}Na$ reaction is known.

3. The angular and energy dependence of neutrons emitted from $^{19}F(\alpha,n)^{22}Na$ play a critical role in any nuclear data evaluation of the differential reaction cross section using R-matrix theory, which properly combines past measurements of $^{19}F + \alpha$ measurements (with neutrons, alphas, gammas, or other particles in the exit channel) to get a best, recommended value of the cross section (i.e., a value that can then be combined with stopping powers to determine the total neutron yield per alpha particle emitted [TNYA]).

4. The design of next-generation neutron detectors, such as pulse-shape discrimination plastics [Woolf 2015], for the international safeguards, nonproliferation, criticality safety, and security communities may be able to exploit certain features (peaks) in the neutron emission energy spectra to improve the signal-to-noise ratio of their signals and hence their overall precision, and perhaps to increase their fidelity to enable distinguishing the components of mixed-actinide samples. A knowledge of the energy spectrum is needed as input for transport calculations that model such detector systems.

Experiments that provide information on the reaction channels and gamma-ray emissions are critical. Such experiments provide the required information to the evaluator, and for some applications, they generate neutrons in coincidence with gamma rays. Figure 1 illustrates two sub-versions of the $^{13}C(\alpha,xn)$ cross section in TENDL-2017; one version accounts for the reaction channels (dashed line) and the other sums them (solid line). This is also a problem with the data for ^{11}B, $^{14,15}N$, and $^{29,30}Si$ [Mendoza 2019].

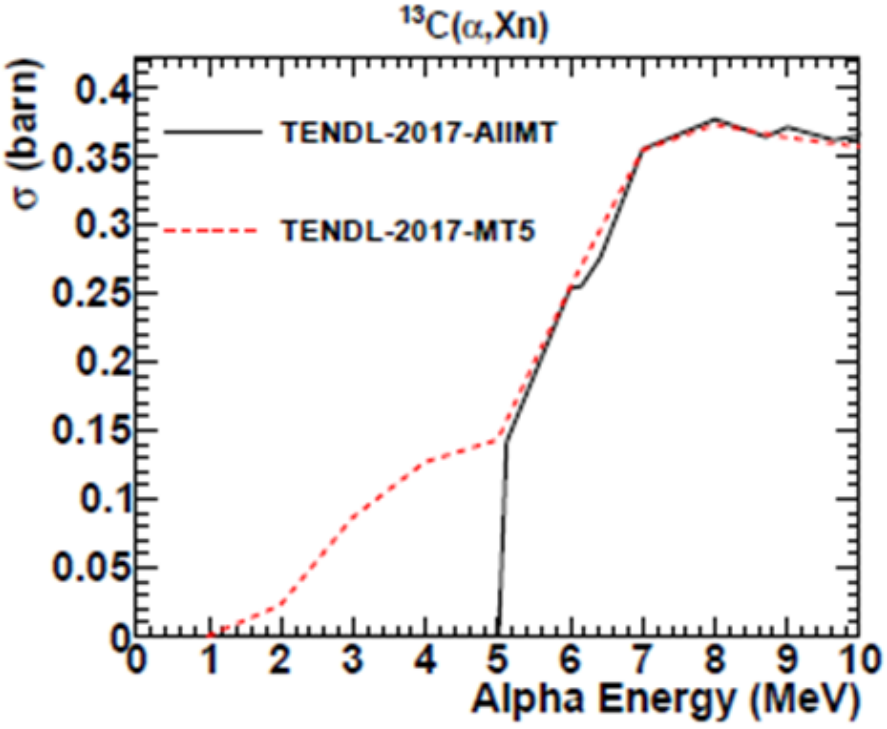

Figure 1. Neutron production cross sections of ^{13}C in the two sub-versions of the TENDL-2017 library, one including all the explicit reaction channels up to 30 MeV (TENDL-2017-AllMT) and the other with all the nonelastic channels grouped together (TENDL-2017-MT5) [Mendoza 2019].

From the study by Mendoza et al. [Mendoza 2019], the effect of accounting for the excitation states and the anisotropic neutron emission spectrum is demonstrated in Figure 2, where the neutron emission

spectrum is calculated in Geant4 using the TENDL-2017 calculation, where all exit channels (α,$n_{0,1,2,\ldots}$) are combined. The calculation uses JENDL/AN-2005 data where each exit channel is accounted for and a modified version of JENDL (JENDL-XS), which uses the JENDL values but assumes an isotropic distribution for the neutron emission. The exit channels and neutron emission spectra need to be better understood through new experiments.

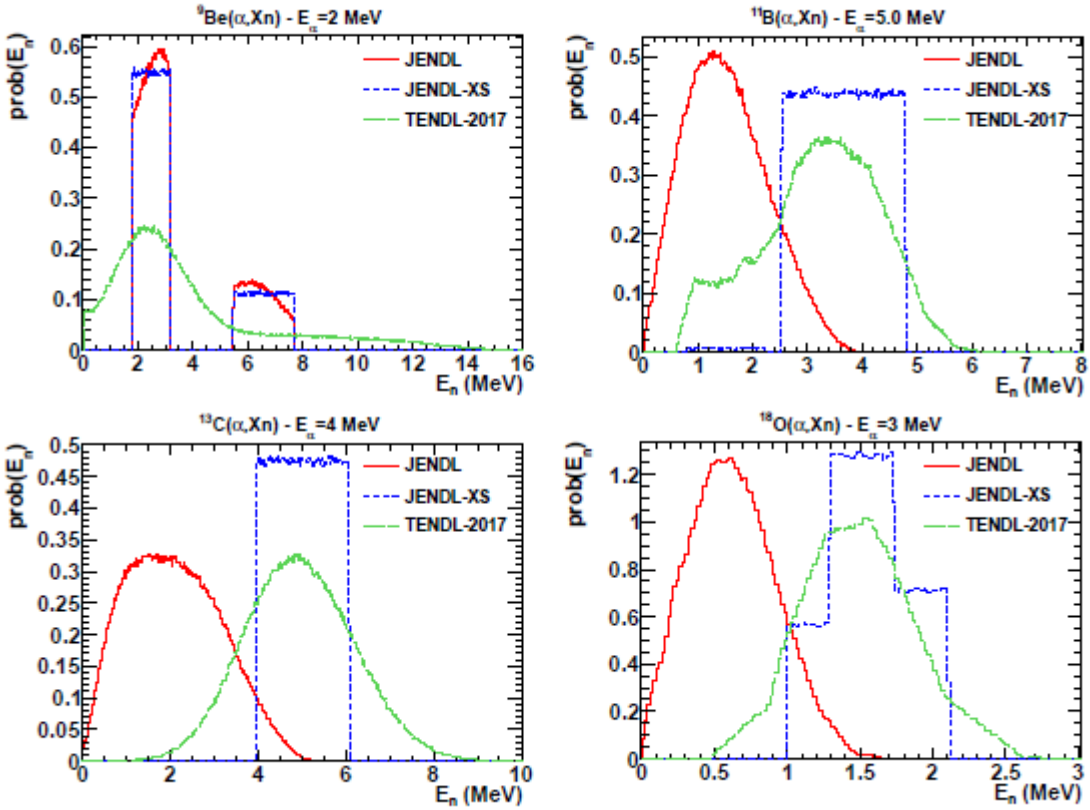

Figure 2. Comparison of neutron spectra when using the JENDL/AN-2005 library, the TENDL-2017 library, and a modified JENDL-XS using the partial (α,n_i) cross sections and assuming isotropic neutron emission [Mendoza 2019].

3.3 STOPPING POWERS

Alpha decay strength functions are well known from extensive studies of actinide decays [ENS 2020]; however the energy loss per unit path length (the slowing down) of alphas as they traverse and interact with a medium is a critical source of uncertainty. The "stopping powers" [Ziegler 1977, Ziegler 1985] depend on the particle type and energy as well as the medium composition. Unfortunately, stopping powers have not been measured for all combinations of alpha energies and sample types, and there are significant disagreements between different tabulations. Further, stopping powers for compound samples (e.g., CaF_2) are often determined by empirically combining those measured separately for the elements (e.g., the Bragg- Kleeman rule [Bragg 1905]), introducing additional inaccuracies. Parameterizations (e.g., Bragg-Kleeman) and codes such as TRIM-89 [Biersack 1989] or SRIM-2013 [Ziegler 2013] are therefore necessary in many studies to estimate the relevant stopping powers, but uncertainties vary substantially for different materials. While some cases have uncertainties of 1% to 3%, many cases have 50% or more, and in some cases disagreements with models by a factor of two are reported [Montanari 2017, Ziegler 2017].

A recent paper by Simakov, et al. [Simakov 2017] indicated that, in uranium compounds, the stopping powers introduce large uncertainties. Specifically, there are differences of 4% to 5% in the neutron yield calculated using SRIM-2013 and using ASTAR alpha-particle stopping powers. For example, in thick target measurements at 5.5 MeV, the yield is sufficiently strong so excellent precision can be achieved in a short time, reducing the influence of impurities and especially surface contaminants and surface stoichiometry.

A better understanding of the stopping powers in an integral sense can be obtained by comparing the thick-target yield integrated over the angle between compounds. Specifically, the stopping power defines the magnitude and shape of the slowing down and stopping of the alphas as well as the mixing rule for how elements in a compound jointly contribute. For some materials such as UF_6, there are no direct accelerator yield measurements, but demonstrating directly how CaF_2, LaF_3, PbF_2, and other compounds scale is very important because different thick target measurements use different targets, and they can be scaled and compared accordingly. This information will be invaluable in the interpretation of UF_6 measurements. By measuring different targets in the same experimental campaign, many potential sources of bias (*e.g.*, systematic uncertainties) should cancel out. The method is, however, not direct because the stopping power cannot be backed out. Preparing targets with layers that are uniform and of known chemical structure is critical for the success of these studies.

3.4 GAMMA RAYS

Gamma multiplicities and energies should be measured during cross-section measurements to provide information on the ratio of excited states populated by the compound nucleus. Although that kind of information is not usually measured directly in nuclear security applications, it is necessary for evaluators to accurately calculate the neutron emission spectrum.

3.5 RECOMMENDATIONS FOR HIGH-RESOLUTION EXPERIMENTS

A series of experiments is recommended to enable neutron detection to be used for precise and accurate NDAs of systems of actinides and light elements. The goal is to reach a combined accuracy of 5% or better on the measured thick-target total neutron yield over the most critical range of energies needed for NDAs. The measurements should address the crucial issues in determining the neutron yield, energy spectrum, gamma emission, cross section uncertainties, and overall normalization. The following experiments are recommended:

1. Address the total neutron yield by bombarding a thick (stopping) target with alpha particles and detecting the emitted neutrons in a 4π-moderated counter that has a detection efficiency that is nearly independent of neutron energy (a “flat-response” detector). Measure a series of compounds to define the role that the stopping power plays in the total yield as appropriate for the application of interest.

2. When appropriate, as it is for $^{19}F(\alpha,n)^{22}Na$ reaction, for example, conduct a second, complementary experiment using the same thick targets as in experiment 1 wherein the decay of the radioactive ^{22}Na nuclei produced in thick samples under alpha bombardment is precisely measured (an “activation” measurement) with no reliance on simulations and without any energy-dependent efficiencies that would add systematic uncertainties.

3. Address the neutron spectrum and angular distribution by bombarding a thick target with alpha particles and measuring the neutrons in an array of liquid scintillator detectors over a selected set of energies. Use of a pulsed beam will enable the use of time-of-flight to determine neutron energy in addition to the detector response.

4. Address overall normalization issues, literature discrepancies, and the neutron spectrum by measuring both thin- and thicker-target energy-differential cross sections with the same array of liquid scintillator detectors in fine energy steps over the complete range needed by NDA studies (0–9 MeV). The neutron spectrum measurements and the compound nucleus excitation function measurements are essential to producing a precision evaluation of (α,n) reactions and neutron emission spectra that can be utilized in a wide variety of nuclear science applications.

5. Incorporate gamma detection to give critical information on the branching ratios to excited states, to verify the yield of low-energy neutrons near and below the neutron detection threshold, and to provide data for applications that make use of the gamma emission data.

6. Implement measurements of multiple target compounds to reduce yield dependencies on stopping powers, low-threshold high-efficiency detectors, low-contamination targets, multiple facilities, and redundant cross checks.

All data must be submitted to the National Nuclear Data Center (NNDC) for incorporation into the EXFOR database. With this goal in mind, it is always good practice to include a nuclear data evaluation expert on the measurement team. A detailed uncertainty analysis should be documented and provided with the data to NNDC for incorporation into EXFOR and to the evaluator.

3.6 IMPACT OF IMPROVED MEASUREMENTS

The use of best practices in experimental methods will provide the best possible data sets for each measured isotope. Only with a complete set of measurements can the evaluator ensure an accurate assessment of the data.

4. (α,N) EVALUATIONS

4.1 CURRENT CAPABILITIES

The goal of obtaining evaluated nuclear data is to provide a data set in agreement with available (differential) measurements and to enable reliable predictions on integral quantities such as independent measurements and sample configurations of the total neutron yields and energy spectra from (α,n) interactions. A reliance on nuclear reaction models defined within the R-matrix theory or the statistical Hauser-Feshbach formalism is the basic and fundamental assumption in generating modern evaluated nuclear data, both for α-induced reactions and for the well-tested case of the neutron sub-library. Such an approach implicitly accounts for all energetically possible reaction channels. Due to the paucity and sometimes discrepancy of differential as well as integral measured data, the use of nuclear reaction models can be seen as an additional constraint on the fit of measured data because it takes into account the correlation among different reaction channels. A simplistic approach such as the adjustment of the (α,n) cross sections performed independently from any reaction models would be incomplete because it will be biased toward particular integral measurements with little clue of the agreement with measured differential data. Furthermore, the covariance matrix of the nuclear model parameters obtained by the fit of the measured data can be used for uncertainty quantification purposes.

Low-mass nuclei have a small number of resonances, making them ideal to analyze their reaction data with R-matrix theory. Although this approach is suitable to evaluate excitation functions over most of the relevant energy range (up to about 5 MeV), the use of other R-matrix approximations such as the reduced R-matrix formalism [Lane 1958] or other models, such as the statistical Hauser-Feshbach model [Herman 2006], are needed to evaluate cross sections for breakup reaction channels or emission of particles such as

deuteron and tritium. For instance, for fluorine and the oxygen isotopes, the energy boundary between the two model approaches is between 5 and 7 MeV, whereas for beryllium it is at about 2.3 MeV.

Furthermore, the neutron and gamma emission spectra have become important sources of nuclear data for applications. The evaluation accounts for the partial cross sections of each excitation state of a compound nucleus after absorption of an alpha particle. These excitation states are required to accurately predict the emitted neutron and gamma energy spectra.

4.2 RECOMMENDATIONS

A new evaluation of the (α,n) cross sections from about 1 to 9 MeV and secondary particles, is recommended for the priority isotopes. The evaluation should include reaction channels such as elastic scattering, inelastic scattering, and other energetically possible reaction channels, including their covariance information. The evaluation methodology will use the SAMMY R-matrix code [Larson 2008] to fit experimental data in the resolved resonance region (RRR). The upper energy range for the RRR usually depends on the available measured data for each reaction channel and the experimental resolution. Above the RRR upper energy limit, the number of energetically possible reaction channels is larger; statistical models within the Hauser-Feshbach theory–implemented in reaction codes such as EMPIRE [Herman 2007] or TALYS [Koning 2019] will be used to evaluate the cross sections in the unresolved and fast energy range. The compilation of the available experimental data will include existing and newly measured data. Among them, the partial cross sections for ^{17,18}O(a,n$_{0,1,2}$,..) recently measured at Notre Dame are seen as a priority to update the current RRR evaluation up to 5 MeV along with the evaluated data for both oxygen isotopes in the energy range above 5 MeV. The existing measured data on ^{19}F(α,n) are also relevant, and the existing experimental information can be used to start the evaluation work that was never initiated for that isotope. The incompatibility between current differential and integral measurements needs immediate investigation. When available, future experimental information on fluorine will be used to update the preliminary evaluation work that, in reverse, will be used also as a parametric tool to validate the newly measured experimental information. Current (α,n) cross-section deficiencies that should be addressed in the future cross-section evaluation work include addressing the disagreement between calculated and measured data and the inconsistency between differential and integral data.

Code development on the R-matrix algorithm is also needed. In the framework of R-matrix theory, one of the major challenges of the evaluation work is the inclusion of reaction channels that are different from (α,n), such as breakup reaction channels for which the two-body formalism is not valid. For nuclei such as oxygen isotopes and fluorine, the breakup channels are in the energy range above 5 MeV, but for lighter nuclei such as ^{9}Be the reaction channels appear at 2.3 MeV. To account for breakup reaction channels in the RRR, the definitions of new penetrability factors or the reduced R-matrix formalism [Lane 1958] are required. These capabilities can be added to the SAMMY evaluation code development to implement reduced R-matrix formalism or penetrability factors needed to describe the breakup reaction channel.

5. PRIORITY ISOTOPES

Table 1 contains a list of isotopes of interest for nonproliferation and nuclear energy applications. The table indicates the threshold energy of the (α,n) reaction, some examples of the applications that require the data, the state of the data, and recommendations. Additional information is also provided for fluorine, oxygen, carbon and beryllium.

Table 1. Priority isotopes for improved (α,n) nuclear data.[a]

Isotope	Current state of data	Reaction threshold	Applications	Recommendation
Highest priority				
^{13}C	Recent data (4–6.5 MeV) to be published, new evaluation needed.	450–800 keV	NDA/ER, safeguards, advanced reactors, background simulations	Low-energy measurement needed < 1–4 MeV. Recent Notre Dame measurement from 4 to 6.5 MeV. Discrepancies in past data are being examined and new evaluation once data issues are resolved.
^{17}O	Recent evaluation available for next ENDF release—discrepancies in neutron spectrum remain	800 keV–1.2 MeV	Safeguards, reactors, NDA/ER, background simulations	Recent Notre Dame measurement of $^{17}O(a,n_{0,1,2})^{20}Ne$ from 0.8 to 7 MeV will provide data to resolve discrepancies in neutron spectrum. Updated evaluation needed.
^{18}O	Recent evaluation available for next ENDF release—discrepancies in neutron spectrum remain	850 keV–1.4 MeV	Safeguards, reactors, NDA/ER, Background simulations	Notre Dame measurement of $^{18}O(a,n_{0,1,2,3,4})^{21}Ne$ from 2 to 8 MeV will provide data to resolve discrepancies in neutron spectrum. Updated evaluation needed.
^{19}F	No evaluation of recent data— benchmark and neutron spectrum data needed	2.3 MeV	Safeguards UF_6, FLiBe reactors, fuel cycle and waste management applications	2016 measurement provided fine structure for 3 to 6.5 MeV alphas.[b] Still need total neutron, neutron spectrum, gamma emission with lower uncertainty. A combination of improved total cross section, angular distributions, neutron energy spectra, activation, and thick target integral measurements are needed to reduce uncertainties.
High priority				
^{7}Li	Adjusted evaluation based on JENDL data available for next ENDF release, large discrepancies in data	3–4 MeV	FLiBe reactors, NDA/ER, safeguards Important for characterization of actinide-Li neutron sources	New experiments with ability to resolve excitation states and reaction channels and new evaluations are needed to accurately model neutron sources.
^{9}Be	Neutron spectrum has been validated and new evaluation available for next ENDF release	200 KeV	FLiBe reactors, NDA/ER, safeguards Important for characterization of actinide-Li neutron sources	New data are required specifically above 5 MeV to address the multiple breakup channels in the cross section and to collect neutron energy and angular distributions. Evaluations based on new data are recommended that address the reaction channels.
^{11}B	No evaluation	600 keV	Reactors, safeguards	Only measured up to 2.5 MeV.
^{10}B	Recent Experiments up to 4.5 MeV	1 MeV	Reactors, safeguards	Recent $^{10}B(a,n_0)^{13}N$ experiments performed at 2.2 to 4.9 MeV and at 1.5 to 4.5 MeV. Data above 4.9 MeV are needed along with a new evaluation.
^{27}Al	Discrepancies in data from 1970s. No recent evaluation	3 MeV	NDA/ER, advanced reactor fuel	New experiments and evaluations with resolved excitation states and neutron spectrum and evaluations are needed.

Table 1 (continued)[a]

Isotope	Current state of data	Reaction threshold	Applications	Recommendation
Lower priority				
^{23}Na	Need new experiments and evaluation	3.5 MeV	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	3 MeV	MgCl molten salt, fuel cladding, astrophysics	Recent measurement from 1.5 to 3.5 MeV. Experiments up to 6.5 MeV needed for advanced reactors. Evaluation needed.
^{26}Mg	Need new experiments and evaluation	3 MeV	MgCl molten salt, fuel cladding, astrophysics	Planned measurement from 1.5 to 3.5 MeV. Experiments up to 6.5 MeV needed for advanced reactors. Evaluation needed.
^{29}Si	Need new experiments and evaluation	2 MeV	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	4 MeV	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	1 MeV	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 and fuel cycle processes.
^{41}K	Need new experiments and evaluation	3.5 MeV	Pyroprocessing, molten salt fuel (FLiNaK)	New experiments with ability to resolve excitation states and evaluations are needed for advanced reactor fuels and fuel cycle processes.

[a] The isotope priorities are based on nonproliferation applications. Advanced reactors, fuel cycle processes and advanced reactor safeguards may have different priorities. Additionally, low-background measurement calculations require data up to 9.5 MeV.

[b] W. A. Peters et al., *A Kinematically Complete, Interdisciplinary, and Co-Institutional Measurement of the $^{19}F(\alpha,n)^{22}Na$ Cross Section for Nuclear Safeguards Science*, INL/EXT-16-38791, OSTI 1263500, https://www.osti.gov/scitech/biblio/1263500 (2016).

5.1 FLUORINE

None of the previous evaluations and assessments of the $^{19}F(\alpha,n)^{22}Na$ differential cross section have presented a thorough treatment of that reaction. Vukulov et al. [Vukulov 1983] covered the energy range 2.5 to 7.75 MeV and assigned an uncertainty of up to 10% to the cross section. Murata et al. [Murata 2006] used the EG-GNASH code [Yamamuro 1990] to calculate a cross section based on the data of Norman and Bair [Norman 1984, [Bair 1979], but assigned no uncertainty, exhibited no fine (resonant) structure (such as seen in [Balakrishnan 1978] and [Peters 2016b]), and did not discuss the significant spread of the measurements. The TALYS reaction code [Koning 2013] was used to calculate the cross section in the TENDL 2012 library [Koning 2012] and in the TENDL-2015 library [Koning 2015]. The calculation, however, exhibited no fine structure, had no uncertainty, was made on a grid with 1 MeV energy spacing, and was not normalized to any of the measurements. In the most recent assessment, Simakov and van den Berg [Simakov 2017] discussed only four of the data sets and did not adequately address their discrepancies or the differences with other data sets. In Peters et al. [Peters 2016b], comparisons are given between their thin target measurement and that of Balakrishnan et al. [Balakrishnan 1978] as well as with the assessments of Vukulov et al. [Vukulov 1983], Murata et al. [Murata 2006], TENDL [Koning 2019], and van den Berg and Simakov [Simakov 2017]. They also average their cross section into 250 keV alpha energy bins to compare it with the Norman et al. [Norman 1984] result. The differential measurement of Peters et al. [Peters 2016b] has not yet been evaluated.

A new, thorough data evaluation is needed with additional experimental input for the $^{19}F(\alpha,n)^{22}Na$ cross section so that the safeguards and related communities can all use the same "best" value of that critical quantity. The cross section furthermore needs to be reconciled with previous determinations of thick-target yields and the total neutron yield used to interpret NDA measurements. Differently from the oxygen isotopes, the R-matrix evaluation work on the fluorine cross sections for α-particle–induced reactions was never initiated. Basic work to check the consistency of the microscopic (α,n) cross sections in thick-target neutron yield calculations was shown by Simakov [Simakov 2017]. The main outcome of Simakov's work showed that microscopic cross sections measured by Wrean in 2000 and Balakrishnan in 1978 are incompatible with the measured thick-target yield data sets. The update of the cross section corresponding to Wrean and Balakrishnan data sets leads to a 30% to 40% underestimation of the known measured neutron thick-target yields in fluoride compounds. The upper energy limit for which emission of deuteron, tritium, and other particles generated in breakup reactions are energetically possible is approximately 8 MeV. The quantification of the (α,2α) reaction channel is also needed because it has a positive Q-value and is always energetically possible. In view of the large discrepancy between differential and integral values, new measurements and a full evaluation of the (α,nx) cross sections are needed and are given the highest priority. Figure 3 shows the available set of experimental data for the (α,n) reaction (on the left) and other reaction channels (on the right). The discrepancy between the Wrean (2005) and Balakrishnan (1978) data sets is clearly evident below 3 MeV. Both data sets have a slightly lower energy spacing than very recently measured data [Peters 2016]. The data sets display systematic disagreement in the overlapping energy range.

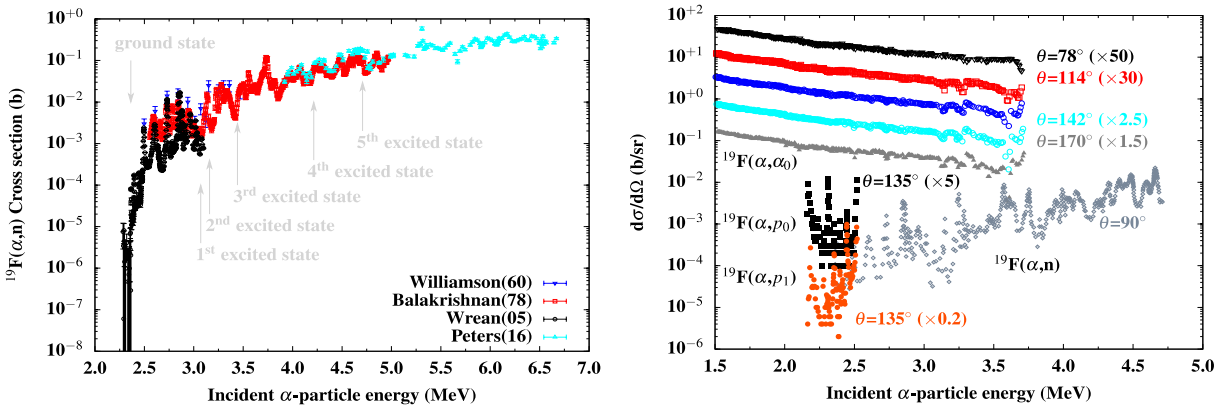

Figure 3. Comparison of $^{19}F(\alpha,n)^{22}Na$ data (left), and the cross section as a function of excitation states (right).

Any new experiments may take several years; therefore, an immediate evaluation of fluorine to support users until new data that include new information on reaction channels, neutron energy and angular distributions become available. SAMMY code development to implement reduced R-matrix formalism or penetrability factors to describe the breakup reaction channel will be necessary to support the evaluation. Evaluated data should be provided in both ENDF and GNDS format to NNDC for inclusion in the next version of ENDF.

5.2 OXYGEN

Previous R-matrix analyses [Pigni 2016, Pigni 2020] showed the need for new experimental data to measure the energy-dependent partial contributions of the (α,n) cross sections to each excited state of the compound nucleus. This information proved to be important in evaluating the shape and magnitude of the calculated neutron energy spectra. This type of measurement for both isotopes were recently initiated at the Notre Dame facility and the goal is to include them in the previous R-matrix analyses. Similar to the work performed in [Pigni 2020], other independent analyses [Simakov 2017] showed the use of energy-differential experimental nuclear data to predict thick-target neutron yields. As shown in Figure 8 of Simakov's paper, below 5.5 MeV, the discrepancy between calculated and measured neutron thick-target yields can be up to 15% in the mean values and up to 30% in the uncertainty limits, although they all seem to agree for monoenergetic incident α-particle energy at 5 MeV. In particular, the measured neutron thick-target yield data obtained by Jacobs and Lisken [Jacobs 1983] do not show full consistency with the data sets by Bair [Bair 1979] and West [West 1982] in the overlapping incident α-particle energy region. This is particularly important because Jacobs' experimental campaign included measurements of neutron energy distributions that are very relevant to validating the underlying microscopic cross sections.

Full consistency between neutron energy distributions and thick-target yields should be achieved by the evaluated cross-section data. Figure 4 shows the partial contributions calculated from the partial $^{17,18}O(\alpha,n)$ cross sections to the energy neutron spectrum distribution of a plutonium oxide configuration. This is an example how the total neutron energy spectrum is shaped by the partial contributions directly dependent from the partial cross section (α,n_0), (α,n_1), ..., (α,n_x). Another issue associated with the (α,n) cross sections and purely related to the cross-section evaluation work is the incompatibility of the $^{18}O(\alpha,\alpha)$ data in the simultaneous fit of the (α,n) data in the energy region above 3 MeV. That issue represents an open problem for the ^{18}O isotope.

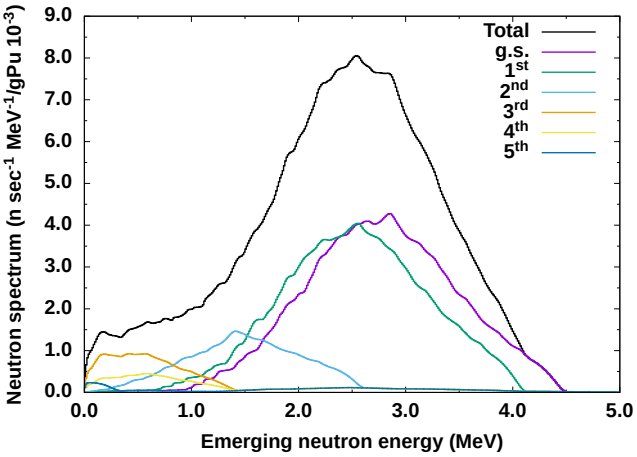

Figure 4. The partial contributions of the ^{17,18}O(α,n) cross section to the total neutron energy spectrum.

5.2.1 Recent Efforts

A recent evaluation of ^{17}O by NNL is based on the original JENDL/AN-2005 version with specific modifications to the neutron energy/angle distribution treatment for different reactions. No modifications were made to cross-section data [Griesheimer 2017]. The total neutron production file (MT=4) was removed and replaced with the cross sections for neutron emission leaving the compound nucleus in the ground state or an excited state (MT=50-53,91). The NNL ^{18}O evaluation is based on the original JENDL/AN-2005 version with specific modifications to the neutron energy/angle distribution treatment for different reactions. No modifications were made to cross section data [Griesheimer 2017]. The modified NNL versions for ^{17}O and ^{18}O were retested with MC21 [Griesheimer 2017]. The resulting neutron energy spectra for thick $^{238}UO_2$ and $^{238}PuO_2$ compared well to previous results [Jacobs 1983, Herold 1968] [Anderson 1967, Anderson 1980]. These adjusted data sets are available for the next ENDF/B release.

New, higher-resolution data sets will be available from Notre Dame with information to verify partial cross sections that will reduce uncertainties at lower energies in the previous evaluations. A new evaluation that is recommended includes new data that provide information on the multiple reaction channels along with neutron energy and angle distributions. Data up to 6.5 MeV are needed for NDA analysis and reactor calculations, and data up to 9 MeV are needed to support neutron background calculations. The resulting data sets should be submitted to NNDC in both ENDF and GNDS format. Evaluation code development to implement the multiple possible breakup channels is required.

5.3 CARBON

Carbon-13 data are used as a calibration benchmark standard for the evaluation of other isotopes, which requires low uncertainty. The evaluated ^{16}O(n,α)^{13}C cross section in the ENDF/B-VII.1 nuclear data library is still uncertain because of systematic discrepancies between measured data. The evaluation heavily relies on cross sections for the ^{13}C(α,n)^{16}O inverse reaction reported by Harissopulos [Harissopulos 2005], which are systematically lower than previous values reported by Bair [Bair 1979]. Previous works [Pigni 2016b] briefly describe two sets of measured cross sections and show on the basis of the experimental information that they can be made more consistent. However, a dedicated experiment to confirm such a hypothesis is still needed for thin- and thick-target samples. Figure 5 shows the discrepancy up to 30% between measured cross section of Bair and Harissopulos. Above 5 MeV, where the ^{13}C(α,n$_1$)^{16}O channel opens up, it is well known (e.g., [Peters 2017], [Febbraro 2020]) that the

Harissopulos result is incorrect by up to a factor of two because that experiment could not distinguish between neutrons from the (α,n_0) and (α,n_1) channels, which have a very different detection efficiency.

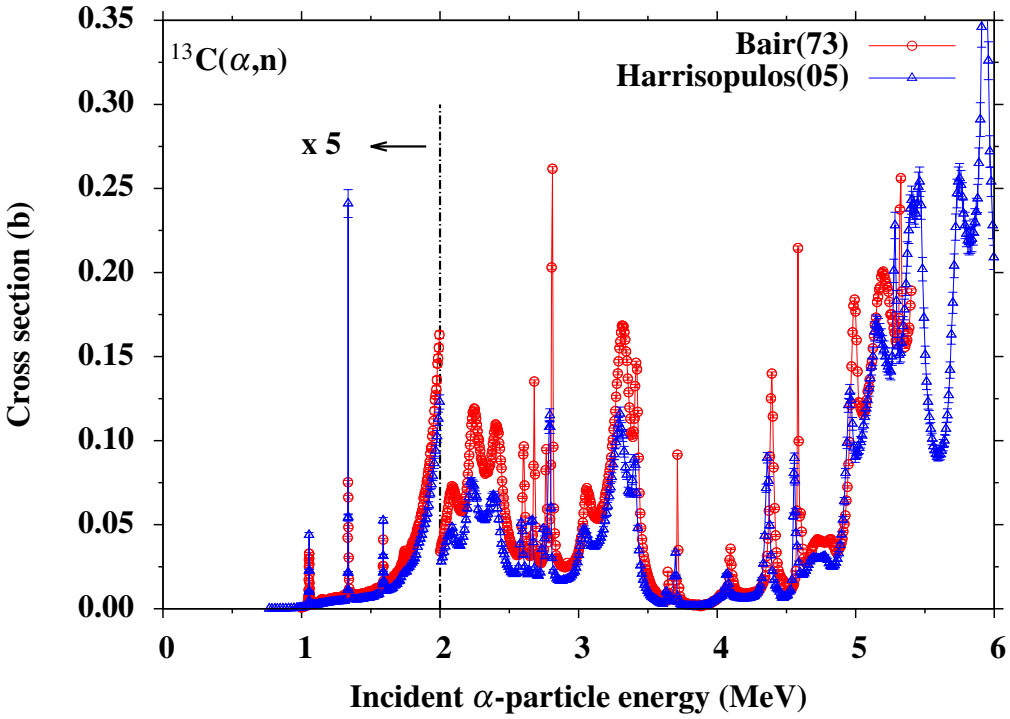

Figure 5. The discrepancy of up to 30% between measured cross section of Bair and Harrissopulos [Bair 1979] [Harissopulos 2005].

5.4 BERYLLIUM

There have been R-matrix analyses [Wrean1994, Freer 2011] for α-particle induced reactions on ^{9}Be. However, their resonance parameters were not reported in an official nuclear evaluated data library. One of the major challenges in evaluating the ^{9}Be resonance parameters and cross sections is the number of breakup reaction channels such as (α,nα) and (α,n2α) as well as other reaction channels, such as (α,d) and (α,t), opening at 2.3 MeV. Because those reaction channels cannot be analyzed within a conventional R-matrix analysis, the reduced R-matrix formalism could be used to evaluate the lumped contribution of the reaction channels defined by a set of absorption widths. In this regard, the Reich-Moore approximation [Reich 1958] can be seen as a special case of the reduced R-matrix shown in Figure 6.

This approach might be useful also to evaluate the ^{9}Be(α,n) cross sections in the energy range above 5 MeV. In fact, as shown in Figure 6, the (α,n) cross sections are generally in good agreement with experimental data, but large discrepancies are clearly evident above 5 MeV. Some work is needed to understand if either the measured data might be contaminated by reaction channels different from (α,n) or if the evaluated data need to be updated above 5 MeV.

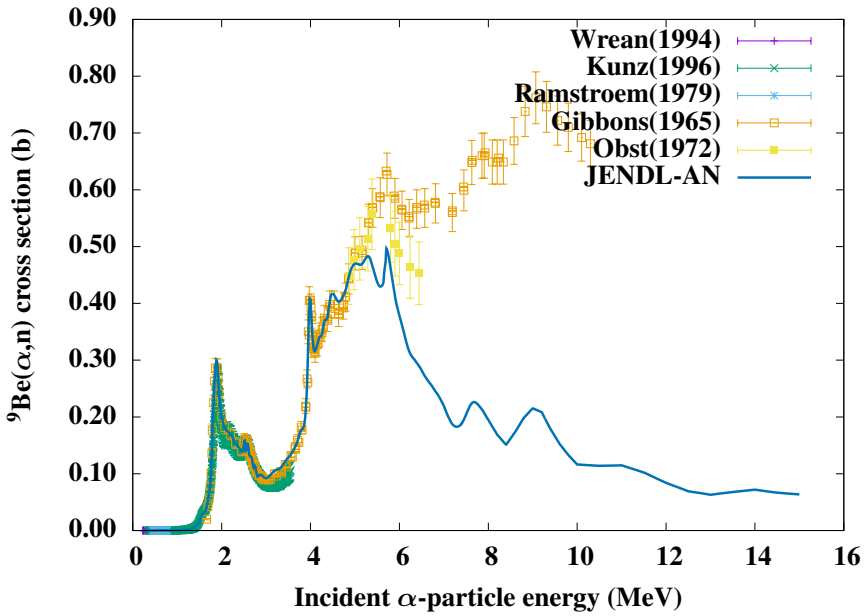

Figure 6. Comparison of ^{9}Be(α,n) cross-section data.

Recently, NNL submitted new evaluation for inclusion in ENDF based on the JENDL/AN-2005 version (NMOD=2) with specific modifications made to the cross-section data for different reactions. No modifications were made to neutron energy/angle distribution data. Within the alpha energy range 4.64 to 7.90 MeV (the maximum energy study reported in [Geiger 1976]), total neutron emission file (MT=4) and (alpha,n+alpha) (MT=22) were adjusted to match the experimentally observed MT=22/MT=4 ratios from [Geiger 1976]. The total cross sections were unchanged, and the sum of MT=4 + MT=22 remains equal to the original total cross sections. The sum of the cross section the MT=50–52,91 excitation states were rescaled to be consistent with the modified total neutron production cross section (MT=4). The original energy grids were retained in all cases.

The new data were retested with MC21. The resulting Am-Be neutron energy spectrum agreed well with Marsh's spectrum [Marsh 1995] and showed marked improvement in the low-energy region below 1.5 MeV. The MC21 results and improvement in the predicted neutron energy spectrum are consistent with those of Shores et al. [Shores 2003], where the SOURCES code is specifically modified, based on the findings of Geiger [Geiger 1976], to better account for the ^{9}Be breakup reaction cross sections. Neutrons emitted in the continuum (MT=91) uses the same Kalbach-Mann systematics neutron distribution data previously used for the total neutron emission MT=4 [Griesheimer 2017].

New data are especially needed above 5 MeV to address the multiple breakup channels in the cross section and to determine neutron energy and angular distributions. Evaluations that are based on new data and that address all reaction channels are recommended.

6. MODELING METHODS

Any improvement in nuclear data impacts the user only if the simulation codes have access to the data and include the advanced physics capabilities to use the data. Therefore, a discussion of the codes used to model the (α,n) source term and recommendations for improvements is provided here.

6.1 COMMON CODES AND CAPABILITIES

There are several capabilities incorporated into current codes to calculate the neutron source terms from (α,n) reactions. Monte Carlo transport calculations (e.g., MCNP, Geant4) suffer from the fact that charged particle transport is computationally intensive, whereas SOURCES 4C is very fast but has limited capabilities. Users typically utilize both types of codes by creating a neutron source term using SOURCES4C and manually transferring that source term into the input of a Monte Carlo calculation.

6.1.1 SOURCES 4C

SOURCES 4C is the most widely used code to calculate the neutron source term from (α,n) reactions and is considered the standard for work in nonproliferation, safeguards, and a variety of other applications. The SOURCES 4C code [Brown 2000, Wilson 2002] is used to define (α,n)-based neutron sources for Monte Carlo modeling and for the optimization of NDA detector systems. It is both a convenient repository of data and a codification of the underlying physics necessary to estimate the strength and spectrum of neutrons resulting from (α,n) reactions for any homogeneous material containing α-emitting and (α,n) target elements. The code includes (α,n) cross sections for target nuclei taken from the EMPIRE code [Herman 2007] and in some cases from experimental information [Cooley 2017]. It also includes α-emission energy spectra, elemental stopping powers, and branching ratios to product-nuclide energy levels. The origin of some of the data coded into SOURCES4C is uncertain, and it does not actively retrieve results from standardized databases or other codes. Using the cross sections and stopping powers, the code first calculates the neutron yield value resulting from (α,n) reactions, with alpha particles over a discrete energy range, caused by all α-emitting nuclides in the material and summed over all target nuclides. The total neutron yield is then determined by multiplying the total alpha source strength for each alpha energy range times the corresponding (α,n) yield value [Gauld 2002]. Because of the utility of SOURCES 4C for a variety of applications, it is important that the calculated neutron source strengths be based on the best cross-section information.

SOURCES 4C is incorporated into ORIGEN, a depletion code relied on worldwide for calculation of isotopics in spent fuel and for post-detonation forensics decay. SOURCES 4C is also used as the source term input for MCNP neutronics calculations, and the calculations are used to interpret NDA measurements where alpha-emitting actinides are present.

6.1.2 MCNP

For alpha particle transport, MCNP can either utilize data tables or model physics to simulate the alpha interactions and secondary particle production. The most recent published work on both alpha production and transport in MCNP is detailed by McMath and McKinney [McMath 2015], where SOURCES 4C and MCNP are compared using some TENDL2012 cross-section tables. Stopping powers have not been updated in MCNP for many years and require modernization; however, the low-energy stopping powers performed reasonably well. A limited set of (α,n) cross sections is available in MCNP, but more formatted data tables can be used if included in the usual manner (i.e., xsdir modifications).

6.1.3 MC21

NNL recently implemented an in-line (α,n) source sampling methodology in MC21 for Monte Carlo radiation transport simulations. The LLNL COG Monte Carlo code uses a similar approach, but only the ability to calculate neutron yields has been studied.

6.1.4 Geant4

Geant4 now includes a module called ParticleHP, which allows Geant4 to use nuclear data libraries for charged particles available in ENDF format. This capability was developed to support the calculation of the neutron source from uranium and thorium decay chains for low-background measurements. Geant4 performs the alpha particle transport and generates neutrons and gamma-rays on an event-by-event basis. Although this capability has shown to be robust [Mendoza 2019], it is dependent on the nuclear data used, specifically on the problematic (α,n) evaluations included in ENDF/B-VIII, such as the coarsely gridded TALYS statistical model calculation used for $^{19}F(\alpha,n)$.

MCNP and Geant4 simulations of experimental detector response gave results that differed on average by 4% for the $^{19}F(\alpha,n)$ measurements by Peters et al. [Peters 2016b]. This uncertainty was added into the experimental uncertainty budget and was the dominant term for many energies. Enabling a validated SOURCES 4C code to be used with MCNP and Geant4 may reduce some of these sources of uncertainty.

6.2 RECOMMENDED CODE IMPROVEMENTS

SOURCES 4C is the most broadly used source term code and can be used to provide an input source file to common transport codes. Therefore, the recommendation is to update and validate the SOURCES 4C code.

6.2.1 Issues and Recommended Improvements to SOURCES 4C

SOURCES was originally developed in 1982 and is written in Fortran 77. SOURCES 4C represents the last distributed code update as of 2002, when the current user manual was written. Since then, it has not been continuously maintained or validated in a controlled manner to ensure its continued utility. The code and data infrastructure within SOURCES 4C are both inflexible and outdated, limiting opportunities for direct integration with codes like MCNP and Origen. Modernization of SOURCES to take advantage of modern computational methods and providing for more flexible means of updating nuclear data are required to ensure its future usability for these codes as well as for inputs into other NDA tools for nonproliferation and safeguards.

SOURCES 4C uses an internal nuclear data reaction file that individuals have updated for their own use (see Table 2), but the updated data have not been included in the distributed version of the code through Radiation Safety Information Computational Center . Additionally, the SOURCES 4C nuclear data files do not correspond to any standardized nuclear data format (e.g., ENDF6, AMPX, NJOY) and as a result, updating data to reflect current developments is time-consuming and labor-intensive. The (α,n) interaction cross sections and yields in SOURCES 4C are largely based on calculations from GNASH [Young 1992]. There are no gamma emission data, and no capability to use covariance data.

Because SOURCES 4C uses its own internal data libraries, any improvements in the ENDF libraries would not be realized within SOURCES 4C. The code's alpha decay library tracks 89 nuclide decay alpha spectra with 24 sets of recoil nucleus-branching fractions, however data for most target nuclei are limited to a maximum energy of 6.5 MeV. While this is sufficient for most cases, there are 21 nuclides in the natural decay chains with one or more alphas above this threshold required for calculation of low-background measurements.

Covariance information needed for performing sensitivity and uncertainty analyses, which are especially vital for the design of precision measurement systems that take advantage of (α,n) reactions to infer information about actinide content.

SOURCES 4C uses the user-supplied information for the target medium composition and the alpha sources to calculate a multigroup neutron energy spectrum from (α,n) interactions based upon a user-defined energy grouping. Stopping-power data are tabulated in a master (α,n) cross-section file, which was last updated in 1989. The alpha transport calculations used for calculating branching and stopping power are substantially out of date, based upon work by Ziegler [Ziegler 1977].

Table 2. Isotope cross sections hard-coded into SOURCES 4C and the source of the data.

Isotope	ZAID[a]	Level-branching fraction source data	Cross-section data
^{7}Li	30070	GNASH[b]	Gibbons and Macklin[c]
^{9}Be	40090	Geiger and Van der Zwain[d]	Geiger and Van der Zwain
^{10}B	50010	GNASH	Bair and Gomez del Campo[e]
^{11}B	50110	GNASH	Bair and Gomez del Campo
^{13}C	60130	GNASH	Bair and Haas[f]
^{14}N	70140	N/A	GNASH
^{17}O	80170	Lessor and Schenter[g]	Perry and Wilson[h]
^{18}O	80180	Lessor and Schenter	Perry and Wilson
^{19}F	90190	Lessor and Schenter	Balakrishnan et al.[i]
^{21}Ne	100210	N/A	GNASH
^{22}Ne	100220	N/A	GNASH
^{23}Na	110230	GNASH	GNASH
^{25}Mg	120250	GNASH	GNASH
^{26}Mg	120260	GNASH	GNASH
^{27}Al	130270	GNASH	GNASH
^{29}Si	140290	GNASH	GNASH
^{30}Si	140300	GNASH	GNASH
^{31}P	150310	GNASH	GNASH
^{37}Cl	170370	GNASH	Woosley et al.[j]

[a] ZAID is the MCNP format for material identifier where Z is the atomic number and A is the atomic mass number.
[b] P. G. Young et al., LA-12343-MS, Los Alamos National Laboratory, 1992.
[c] J. H. Gibbons and R. L. Macklin, *Phys. Rev*. 114, 571, 1959.
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6.3 RECOMMENDED SOURCES 4C CODE IMPROVEMENTS

A new version of SOURCES should be created that takes advantage of modern computational capabilities. The creation of a Fortran/C++/Python API allows calculations to be set up and run and for results to be retrieved from other codes such as MCNP or SCALE/ORIGEN.

6.3.1 Data Libraries

It is recommended that SOURCES 4C be modified to directly process ENDF-formatted data libraries (likely through the GNDS format standard). The modification will allow the user to incorporate more recent evaluations and uncertainty estimates, including evaluations such as the new ^{16}O evaluation (which

is presently missing). This recommendation is made in parallel with the recommendation to significantly improve the (α,n) evaluations in ENDF. The addition of sensitivity analysis capabilities for the neutron source rate density with respect to cross sections and elemental stopping powers is also recommended. These capabilities, when combined with covariance information, would provide invaluable information to researchers to determine sensitivity and propagated uncertainties of evaluated systems, greatly aiding in the prioritization of targeted nuclear data measurements.

6.3.2 Alpha Transport Updates

It is recommended that the SOURCES 4C stopping power libraries be updated to use SRIM 2013, which contains a substantially expanded body of experimental data for alpha stopping powers (approximately 6,300 data points). The code should also allow for user input to adapt the physics for specific applications.

6.3.3 Validation Test Suite

SOURCES 4C comes with a limited set of validation data, primarily presented in the context of example problems at the end of the manual to illustrate the use and performance of the SOURCES 4C code. A validation test suite for SOURCES that can easily be executed by the user for predefined validation problems is recommended. The capability to conduct validation tests would establish the accuracy of the SOURCES code and methods and would likewise permit users to directly evaluate the effects of updated or alternative data evaluations.

7. IMPACT OF (α,n) NUCLEAR DATA TO APPLICATIONS

Improving the understanding of and the ability to model the (α,n) source term will facilitate nondestructive measurements for safeguards and nonproliferation and will improve criticality and dose calculations for advanced reactors. The IAEA has expressed a need for high-fidelity $^{19}F(\alpha,n)^{22}Na$ cross sections and secondary particle information for UF_6 cylinder enrichment monitoring and other applications. For emergency response, there is interest in the gamma/neutron ratio for material identification. For advanced reactors, actinides in an oxide, nitride, or oxycarbide matrix can be measured using the (α,n) neutrons and gamma rays, and the (α,n) contribution must be known to interpret spent fuel measurements. For advanced reactors, it is expected that the (α,n) source term in molten salts will be large due to the buildup of minor actinides over time and thus will require knowledge of the K, Mg, F, Li, Be, Cl (α,n) cross sections and neutron emission spectra.

7.1 IMPORTANCE OF (α,n) TO NONDESTRUCTIVE ASSAYS

NDAs are a fundamental technique for safeguards verification and special nuclear material (SNM) inventories. Safeguards assessments are based on physical measurements, and NDAs are the only viable approach to quickly and cost-effectively determine SNM amounts and flows in situ or in toto for bulk materials. NDAs are critically needed to match the physical amounts of SNMs in a facility with the amounts listed in their official inventory. The accuracy of NDA techniques determines the number of measurements required by safeguards inspectors as well as the overall quality of the verification. Neutron-based NDAs are techniques used the most often for SNM mass determinations because neutrons are very penetrating and provide information on the entire mass of most items. This is in contrast to gamma-ray-based NDAs; gamma-rays are less penetrating than neutrons, and bulk samples such as UF_6 cylinders are many times larger than the mean free path of their gamma emissions. In addition to safeguards verification, NDA methods are also extensively used in waste measurement and decommissioning work, especially for their necessary criticality control measurements.

7.1.1 Total Neutron Yields

A precision determination of the differential $^{19}F(\alpha,n)^{22}Na$ cross section is, by itself, not sufficient to translate a passive measurement of neutrons emitted from a UF_6 container to the amount of ^{235}U in the container. Rather, researchers often rely on the total yield of neutrons from $^{19}F(\alpha,n)^{22}Na$ reaction per incident alpha particle, integrated over all angles and all alpha energies i.e., the TNYA). Some studies in the literature (e.g., [Norman 2015], [Reilly 1991]) quote the total neutron yield per million alpha particles. For a number of reasons, the TNYA is the fundamental quantity of interest needed to interpret the results of NDAs in specific chemical compounds. First, the materials to be measured are typically much larger than the range of the alpha particles, and so emitted neutrons represent an average total production per alpha particle as the alpha particles slow down (losing energy) and stop in the material. Second, because the alpha emitters (e.g., U in UF_6) are homogeneously distributed throughout the material, and alpha particles are isotopically emitted (no preferred direction), the emitted neutrons represent an average over all emission angles.

There are two primary methods to determine the TNYA, one starting from a differential cross section and the other starting from a thick-target measurement. For the first, the conversion of the differential cross section into the TNYA requires one additional property (the stopping power of alpha particles in the fluorinated actinide medium). The first step in the conversion process is to calculate the neutron yield over all emission angles resulting from alpha particles of a specific energy as they slow down (through interactions with the sample) and undergo (α,n) reactions. That quantity is then multiplied by the normalized number of alpha particles emitted from the actinide decay at each energy (the alpha decay strength function) and summed over all alpha energies to get the TNYA. Some studies use a related quantity to connect detected neutrons with the assayed material (the emitted neutrons per second per gram of actinide), which is related to the TNYA by multiplying by the number of decay alphas per second, and dividing by the atomic mass, for the actinide being assayed. A serious issue with calculating the TNYA from a differential cross section is the insufficient precision of both the differential cross section (as a function of alpha angle and energy) and the stopping power of alphas in the material (as a function of alpha energy and medium composition).

For the second method, the neutron emission resulting from bombarding thick (stopping) sample with alpha particles is already integrated over energy and angle, and so it is closely related to the TNYA. However, corrections need to be made because the stopping power in the sample (e.g., CaF_2, LaF_3) differs from the stopping power in the UF_x sample of interest. If a moderated neutron detector is used for the laboratory experiment, then branching ratios (from separate experiments or from theory) need to be used to correct for the different efficiencies of neutron detection from different excited states. Also, in some cases, there may be geometry effects (e.g., surface area, edges, placement of detectors) that need correction.

7.2 MOLTEN SALT REACTORS

Several advanced molten salt reactor concepts are being explored for future nuclear power generation [Kamei 2013]. Some designs incorporate nuclear fuel into the flowing molten salt [Dewan 2016, Holcolmb 2011, Forsberg 2003]. According to Dewan, the vast majority of past work has used lithium-beryllium-fluoride (FLiBe) salt, but other inorganic salts are now of interest, including NaF-ZrF_4, ^{7}LiF-(LEU)F_4, ^{7}LiF/NaF-(TRU)F_3, and chloride salts, depending on the objectives of reactor operation and its design (e.g., thermal or fast spectrum, actinide solubility, salt processing options). In all cases, however, (α,n) production can be anticipated as being an important additional source of neutron radiation, particularly from the irradiated salt ex-core. Minor actinides produce high-energy alpha particles, and the flux and reactivity predictions of molten salts will be improved by the cross section and neutron emission

energy spectra. Stopping power will affect the TNYA, and alpha particle transport is mostly unknown in molten salts.

Additionally, the LiCl-KCl eutectic employed for electrochemical separations likewise is expected to exhibit a comparatively significant (α,n) component to the total neutron yield within the electrorefiner vessel. For this application, the total neutron yield is primarily attributable to spontaneous fission of ^{244}Cm and (α,n) neutrons produced by the decay of ^{238}Pu and ^{244}Cm [Gilliam 2018]. The difference in neutron yield is especially prominent in the region of 0 to 300 keV, wherein the (α,n) component will tend to dominate total neutron emissions up to relatively high (> 40 GWd/MTU) fuel discharge burnups, at which point it is still expected to contribute up to half of the total neutron source within this region [Gilliam 2018], as illustrated in Figure 7. The primary targets of interest are ^{7}Li and ^{37}Cl. Of the (α,n) source term, ^{238}Pu is estimated to provide roughly 30% to 50% of the total (α,n) component (Figure 8) [Gilliam 2019]; however, further refinement and investigation of this method as a viable alternative modality for in situ material accountancy measurements would require significant improvements to the relative uncertainty associated with alpha-induced neutron yields for ^{7}Li and ^{37}Cl, two isotopes identified as high priority for study within this scope.

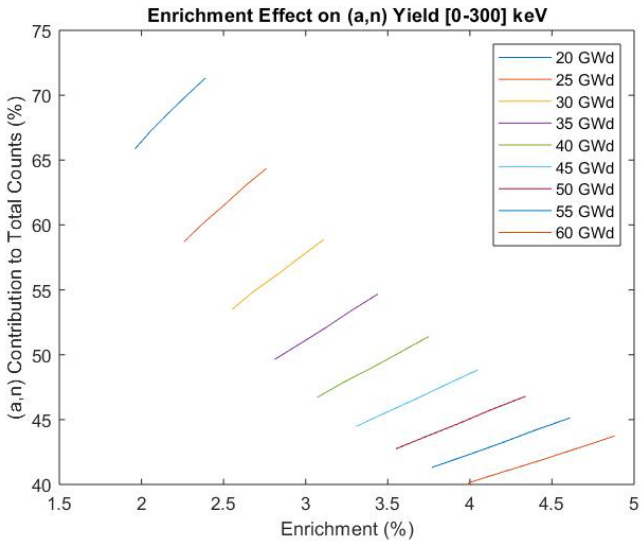

Figure 7. Relative contribution of the (α,n) source term to the total neutron source from 0 to 300 keV for representative used nuclear fuel compositions dissolved in LiCl-KCl eutectic salt [Gilliam 2019].

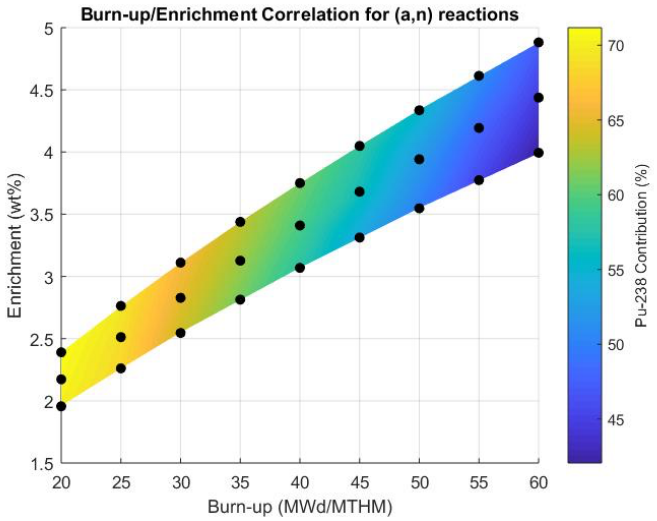

Figure 8. Correlation of ^{238}Pu contribution to (α,n) reactions as a function of discharged fuel burnup and enrichment at 5 years cooling time, dissolved in a LiCl-KCl eutectic salt [Gilliam 2018].

7.3 BACKGROUND CALCULATIONS FOR LOW-BACKGROUND MEASUREMENTS

Low-background experiments such as those that utilize underground detection systems require high-sensitivity measurements and so require accurate determinations of natural background. One source of neutrons is due to the (α,n) reactions caused by alpha emission of naturally occurring isotopes of the uranium and actinium decay series, including $^{211, 212, 213, 214, 215, 216}$Po, $^{215, 217, 218}$At, $^{218, 219, 222}$Rn, and ^{211}Bi. Those isotopes, which collectively contribute 70 alpha emission energies above 6.5 MeV (the current upper limit included in SOURCES 4C). The ^{13}C(a,n) reaction is one example of a critical background in neutrino experiments (e.g., [Febbraro 2020] and references therein). In addition to the total neutron yield, the neutron energy spectra are also important for such low-background experiments because they can result in an energy-dependent background that can introduce significant difficulties on the interpretation of the experimental results.

7.4 (α,n) NEUTRON AND GAMMA EMISSION RATIOS FOR NDA

It has been shown in previous work that the gamma/neutron ratios emitted from compounds can be considered constant when the compounds are produced in an identical manner. The energy of alpha interaction can change the populated excitation states and thus change the gamma emission energy distribution. The ratio of the passive gamma emission from the heavy nucleus can also be used as a predictor of the number and energy of (α,nγ) interactions. This ratio is also influenced by the chemistry of the compound and particle size, and the influence of particle type and size on stopping powers is not well understood. This information may be useful for some types of NDAs and should be investigated for specific applications [Croft 2004].

The production of gamma rays accompanying F(α,n) production in UF_6 cylinders has been looked at computationally [Croft 2013]. For this application, the gamma rays are well shielded and are not considered as useful as neutrons for assays. In addition to F(α,n) neutrons, α-induced reaction gamma rays are generated, notably at 110, 197, 582, 891, 1,236, and 1,275 keV. If one could observe ^{19}F(α,xγ) gamma lines in the HRGS spectra, the α-activity could be estimated directly, and in turn the ^{234}U abundance could be obtained, for example, by utilizing the ratio of the detected 197 keV to 186 keV full energy peaks. However, until now there has been no readily available estimate of the expected strength of the reaction gamma rays an no serious consideration as to whether they might be diagnostic or not.

Croft et al. used published thin-target data to calculate the thick-target yields of the chief reaction gamma-rays in UF_6 [Croft 2013]. Comparisons were made to the neutron production rates to obtain γ/n estimates and to the ^{235}U decay line at 186 keV, which was taken as a fiducial line. It is shown that the reaction gamma rays are produced but suffer from strong attenuation and are not useful safeguards purposes.

Now that the underlying numerical data are readily available, however, it can be used to support neutron and gamma production calculations in other fluorine compounds, such as impure plutonium reference materials, where fluorine may be present only at the parts per million by weight level still present a serious background.

7.4.1 F(α,n) for UF_6 cylinders

Fluorine is an especially important element in safeguards, particularly for uranium enrichment plants, which have holdups in the form of UO_2F_2 and UF_4. In fact, fluorine compounds of U (e.g., UF_4 and UF_6) and Pu (e.g., PuF_6) appear throughout the nuclear fuel cycle. The alpha particles emitted from spontaneous decay of the actinides in those compounds interact with F to copiously produce neutrons via the $^{19}F(\alpha,n)^{22}Na$ reaction. The $^{19}F(\alpha,n)^{22}Na$ reaction is the physics foundation for a "self-interrogating" NDA technique wherein the grams of actinides can be determined from a passive measurement of neutrons emitted from the material (Figure 9). There are two source terms for the total neutron emission from a fluorinated actinide sample: spontaneous fission and (α,n) reactions. The former is determined by the actinide isotopic composition (i.e., the enrichment); the latter depends on the chemical compound as well as the yield of neutrons from the $^{19}F(\alpha,n)^{22}Na$ reaction. In enrichments plants,

Figure 9. Passive neutron measurement of: (left) a large UF_6 cylinder with the UCAS at Rokkasho Enrichment Plant in Japan; (center) a small UF_6 cylinder with the Mini-ENMC [LaF_{11}] at LANL; (right) passive, self-interrogating NDA setup on a Model 30B UF_6 container at Rokkasho enrichment plant [Miller 2012].

^{234}U gets enriched along with ^{235}U, and alphas from ^{234}U decay interacting with fluorine in those compounds are the dominant source of neutrons. For example, one can calculate from the relative half-lives that more than 90% of the neutrons emitted from a cylinder of UF_6 with a 4% ^{235}U enrichment are from $^{19}F(\alpha,n)$ reactions, primarily from α particles produced via ^{234}U decay, and a 5% enrichment has a 20:1 ratio of (α,n) to fission neutrons. By comparison, the neutrons from fission and (α,n) are approximately equal in plutonium oxide. Given the dominant 4.7 MeV alpha energy from ^{234}U decay and the 2.36 MeV threshold for the (α,n) reaction, the emitted neutron spectrum is fairly soft and results in little induced fission of the actinides in the sample.

A detailed understanding of the yield of neutrons from $^{19}F(\alpha,n)^{22}Na$ can therefore be combined with the $^{235}U/^{234}U$ enrichment ratio to assay the sample for ^{235}U. It is therefore critical to understand the neutron yield of $^{19}F(\alpha,n)^{22}Na$ to interpret NDA measurements. Total neutron counting is often the favored means to locate and quantify U deposits in enrichment plants and prevent criticality incidents. Understanding the

yield of neutrons from $^{19}F(\alpha,n)^{22}Na$ can also help designs of next-generation instruments needed to meet the ever more stringent needs of the international safeguards, nonproliferation, criticality safety, and security communities [Bernstein 2015].

A study of the Passive Neutron Enrichment Meter System is provided in Appendix B.

8. CONCLUSIONS

There is a general lack of data for (α,n) cross sections, neutron emission spectra, and associated gamma rays within the ENDF database, save for a few recent contributions, which are required for determination of neutron yields from (α,n) reactions in nuclear security and nuclear energy applications. Some recent measurements have not been evaluated or validated, and no covariance data are available. Creating a database of validated nuclear data with uncertainties available to the user will provide confidence levels for the interpretation of NDA assays. Currently, the ENDF/B-VIII.0 (α,n) sub-library contains only one incident alpha data set for $^{4}He(\alpha,\gamma)$, and four recent evaluations are ready for the next ENDF/B release: ^{7}Li, ^{9}Be, ^{17}O, and ^{18}O. JENDL/AN-2005 [Murata 2006] is the only publicly available ENDF-6 format library for incident alpha particles produced by evaluations of experimental data. It contains neutron production cross sections and secondary neutron energy and angular distributions for the following isotopes: $^{6,7}Li$, ^{9}Be, $^{10,11}B$,$^{12,13}C$, $^{14,15}N$, $^{17,18}O$, ^{19}F, ^{23}Na, ^{27}Al, and $^{28,29,30}Si$. The TENDL database contains (α,n) cross sections produced from calculations of the TALYS code and includes the total yield and an energy-angular distribution of every secondary particle for all relevant isotopes. However, being purely theoretical, there are large uncertainties and important discrepancies in the cross sections and the neutron emission spectra. The TENDL cross sections are also usually given on a coarse (~1 MeV) energy grid with limited values in the 1 to 9 MeV energy range, and therefore they have no capability to represent any reaction fine structure.

There are discrepancies in the cross sections between the measured data sets, and discrepancies between measured and evaluated data sets. The neutron emission spectra have even larger uncertainties. Taken together, they result in serious uncertainties in NDAs and neutron source term calculations. Much of the experimental data are from the 1980s or earlier and do not provide information on all possible reaction channels and excitation states. Stopping powers also add uncertainty to the experiments, and they need to be better understood for many common compounds.

Experiments that provide information on the partial cross section of each excitation state and reaction channel of the compound nuclei are critical to properly calculate the neutron emission spectrum because each excitation state emits a different energy spectrum of neutrons, which directly impacts their detection efficiency. Experiments that provide the partial cross sections of each excited state through the use of new high-resolution neutron detectors over all angles of interest and with correlated gamma energies are needed. Evaluation of the experimental data will enable the creation of a new ENDF/B (α,n) sub-library in a more flexible GNDS format; the sublibrary will include secondary particle information (i.e., neutron spectra and gamma rays) and cross-section covariances.

Modeling and simulation capabilities require modernization in order to optimize the use of the data. SOURCES 4C must be updated as a useful tool to create (α,n) neutron and gamma source terms that can interface readily with commonly used transport codes.

The benefits to the users and their applications will be reduced uncertainty in NDA measurements through the knowledge of the cross section, neutron spectrum, and gamma emission. Inclusion of covariance data will enable uncertainty studies. For reactor applications, uncertainty in the (α,n) contribution to the flux increases the safety margins as well as safeguards measurements. Molten salts are of particular interest due to the mixture of the fuel with light elements. In the safeguards community,

reduced uncertainties in the F(α,n) cross sections and neutron spectra have high impact due to the quantity of material that is accounted for through neutron measurements.

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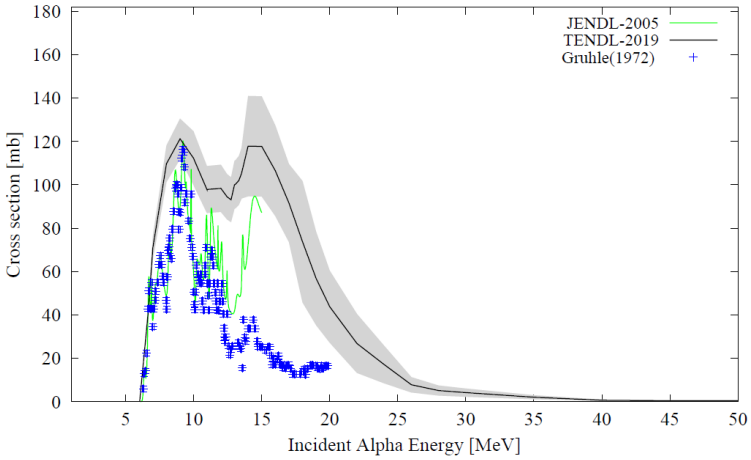
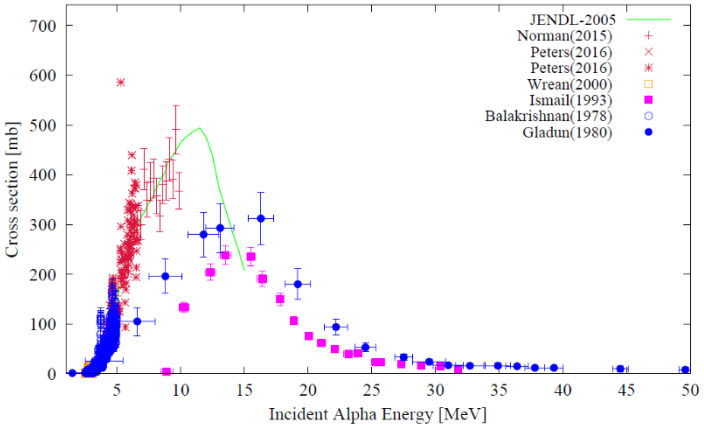
APPENDIX A. PLOTS OF TENDL-2019 AND JENDL-2005 DATA COMPARED TO EXPERIMENTAL DATA

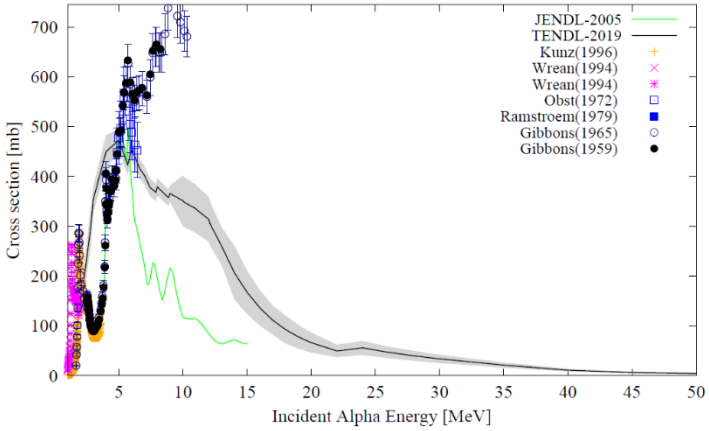
$^{14}N(\alpha,n)^{17}F$



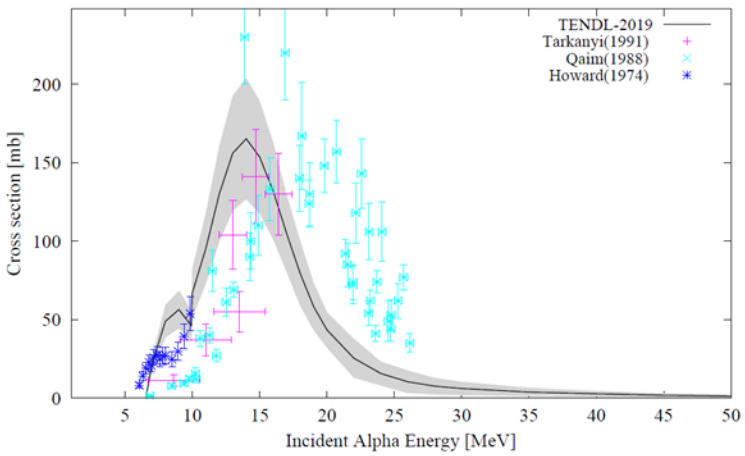
$^{19}F(\alpha,n)^{22}Na$



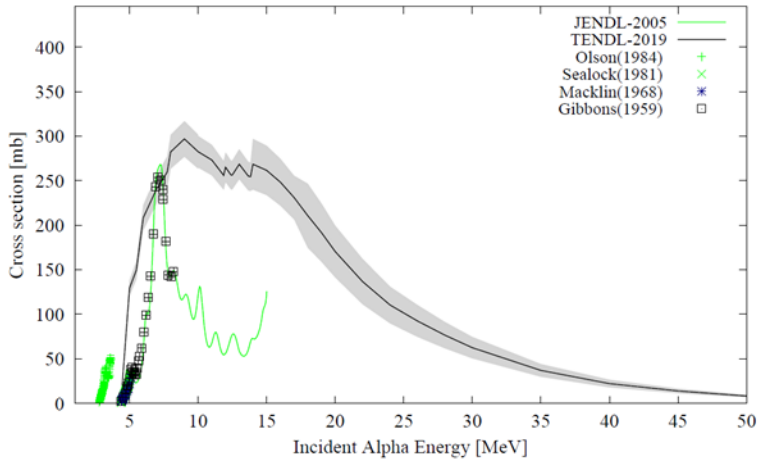
$^{9}Be(\alpha,n)^{12}C$



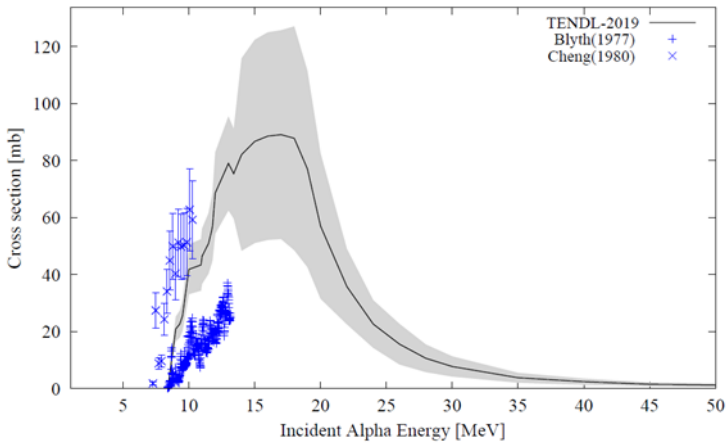
$^{35}Cl(\alpha,n)^{38}K$



$^{7}Li(\alpha,n)^{10}B$



$^{24}Mg(\alpha,n)^{27}Si$



APPENDIX B. THE IMPACT OF (α,n) NUCLEAR DATA UNCERTAINTIES TO UF_6 CYLINDER ASSAY USING THE PASSIVE NEUTRON ENRICHMENT METER

To evaluate sensitivity to energy spectra, the response of the Passive Neutron Enrichment Meter (PNEM) was modeled for 30B cylinders with 2,230 kg of UF_6 (corresponding to 98% of the 2,277 kg fill limit). To include effects of UF_6 distribution the two extreme distributions from the X-factor were included in this study as shown in Figure B-1. The MCNP model that was used in this study corresponds to that used by Broughton and Swinhoe (2019).

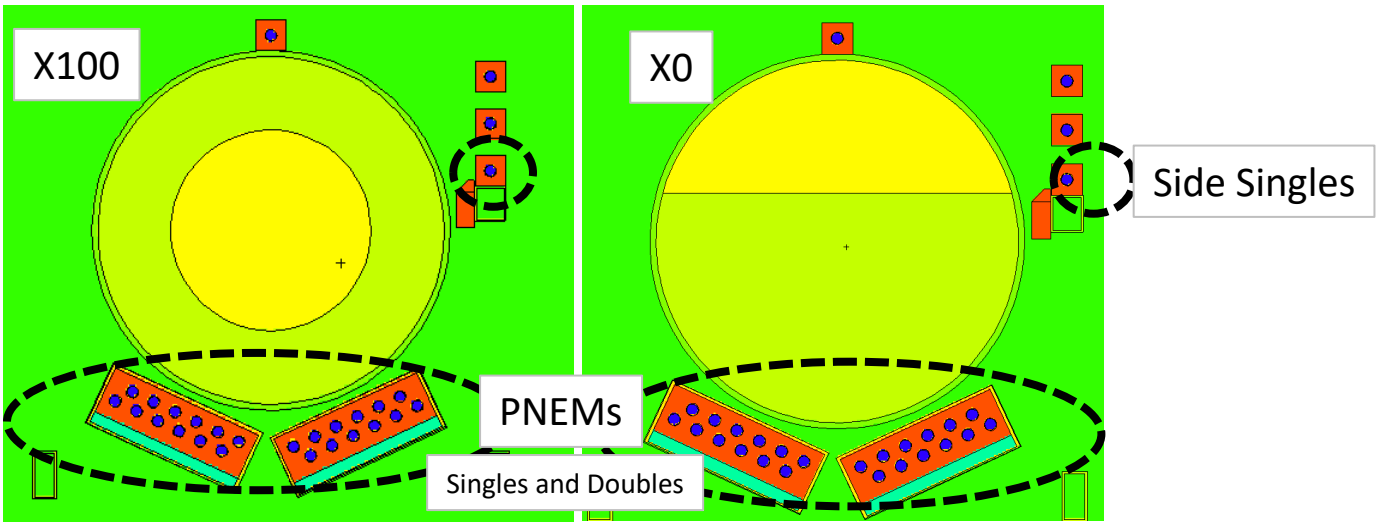


Figure B-1. Simulated geometry showing UF_6 geometry and detector location.

The initial (α,n) spectra used during modeling were taken either from the semiempirical SOURCES 4C (Wilson et al. 2002) spectrum or the experimentally measured spectrum of Jacobs and Liskien (1983). The modified SOURCES4C spectra were generated using a customized version of sources with updated cross sections. The simulated spectra are shown in the Figure B-2, where the spectrum denoted "J&L 'Hard'" corresponds to the experimental Jacobs and Liskien (1983) spectrum after applying a linear extrapolation to 0 from 300 keV. The extrapolation may be appropriate due to the high experimental uncertainty in that range and to the fact that the data become much more representative of the SOURCES 4C (Wilson et al. 2002) data when the extrapolation is used. This work assessing uncertainty due to source is based on the instrument response that is dependent on the simulated spectra used and does not perturb any individual spectrum beyond what is shown in Figure B-2.

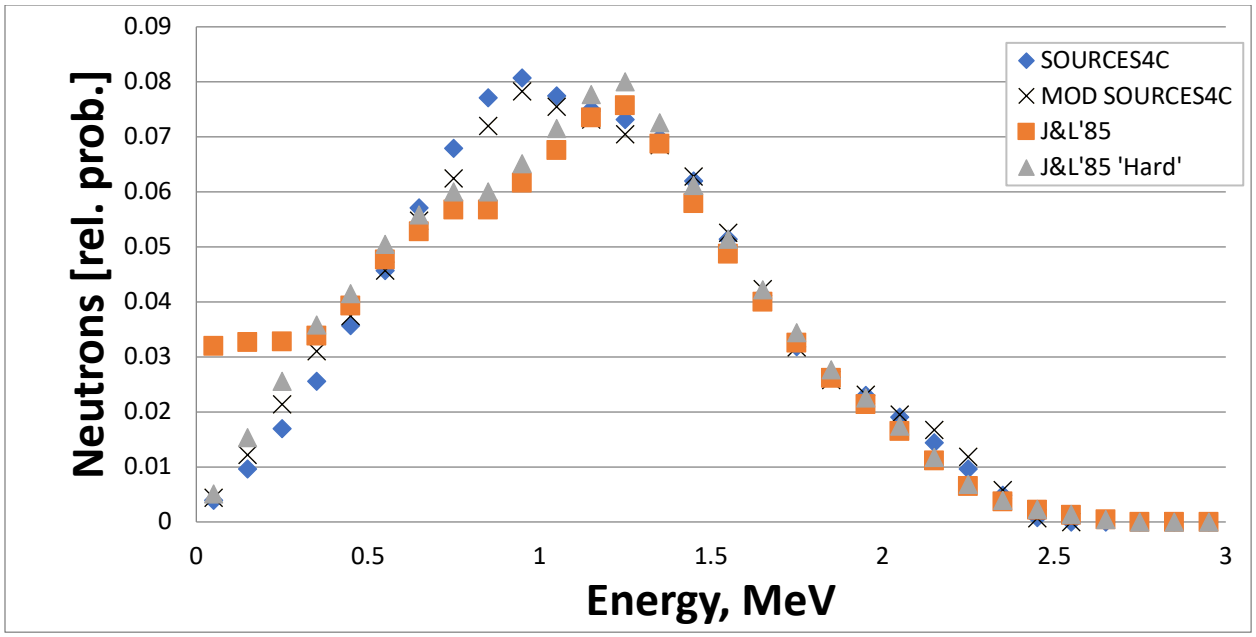


Figure B-2. Simulated $^{19}F(\alpha,n)$ spectra.

To assess energy dependence, 40 monoenergetic simulations were conducted to gauge the overall energy dependence for neutrons ranging from 0 to 4.5 MeV. The Singles (Figure B-3) show relatively little dependence on the initial AmLi spectrum. The Doubles (Figure B-4) show greater dependence potentially due to the thermal albedo being higher for fast neutrons, which is more likely to induce fission nearer the PNEM pod than thermal neutrons, which are likely to induce fission near where they are initially created. Similar to the Singles in the PNEM pod the Side Singles (Figure B-5) show relatively low energy dependence. For all three of the assay signals there is a slight change in energy response due to geometry from self-moderation, but the change in detection efficiency and multiplication are the dominant effects (the X0 responses for Singles and Doubles are considerably greater than those for X100).

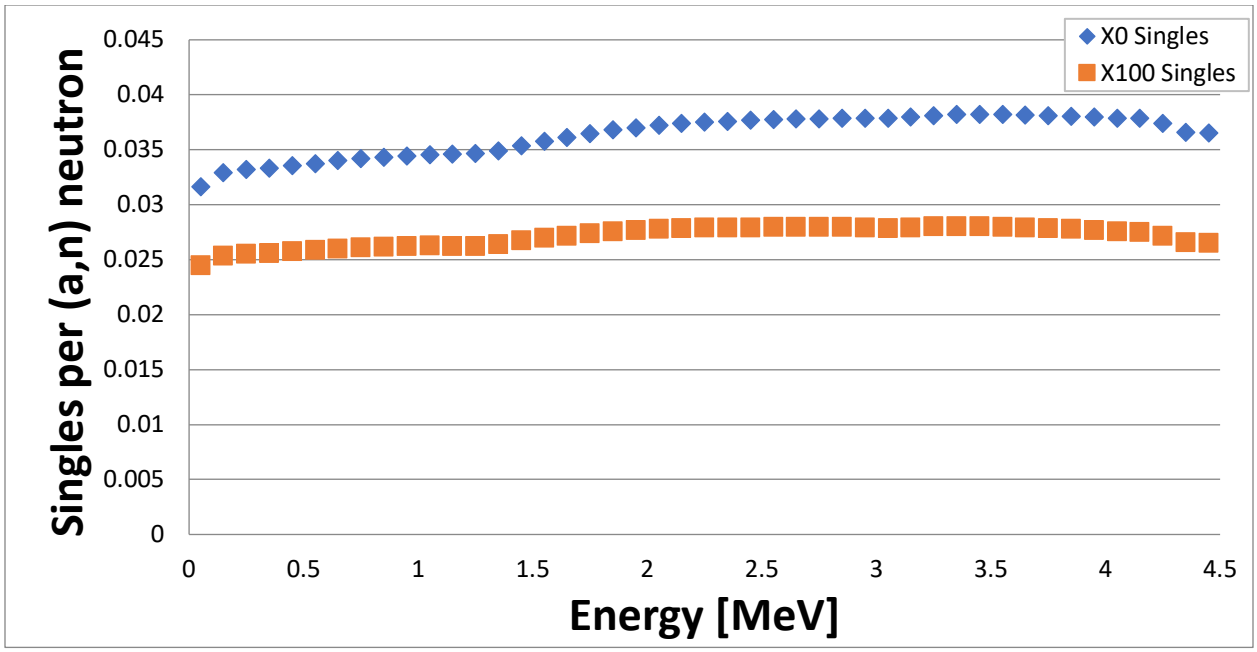


Figure B-3. Simulated singles response as a function of energy for mono-energetic neutrons.

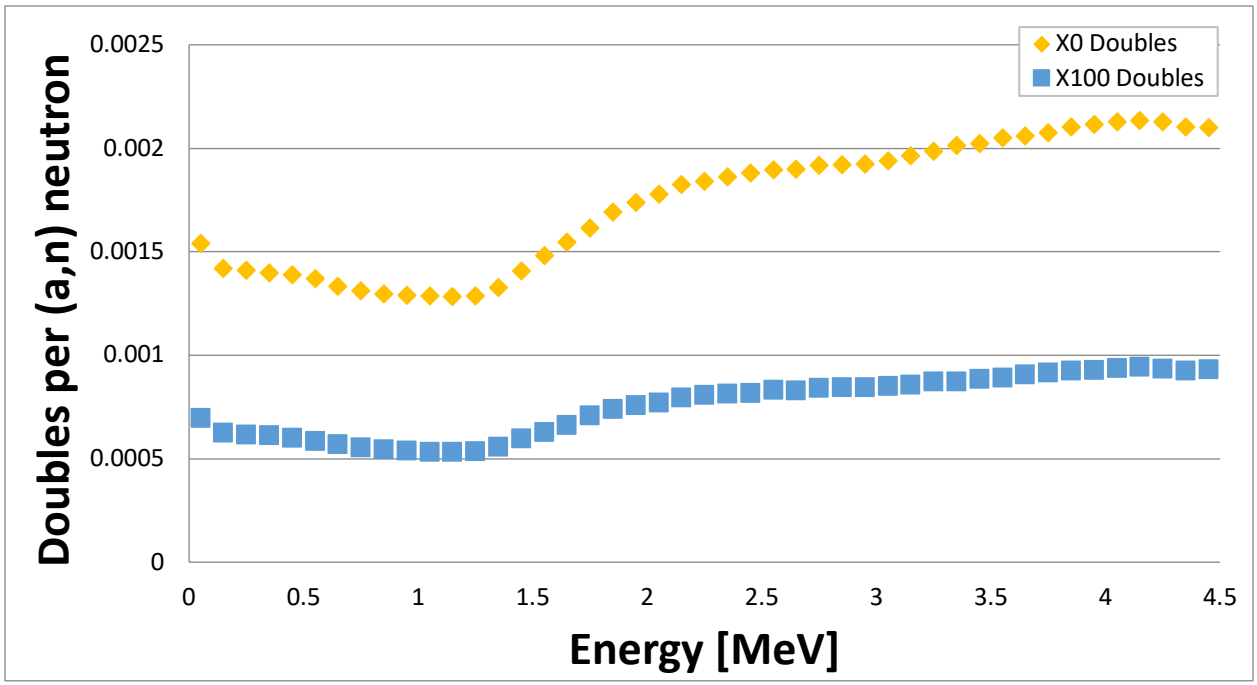


Figure B-4. Simulated doubles response as a function of energy for mono-energetic neutrons.

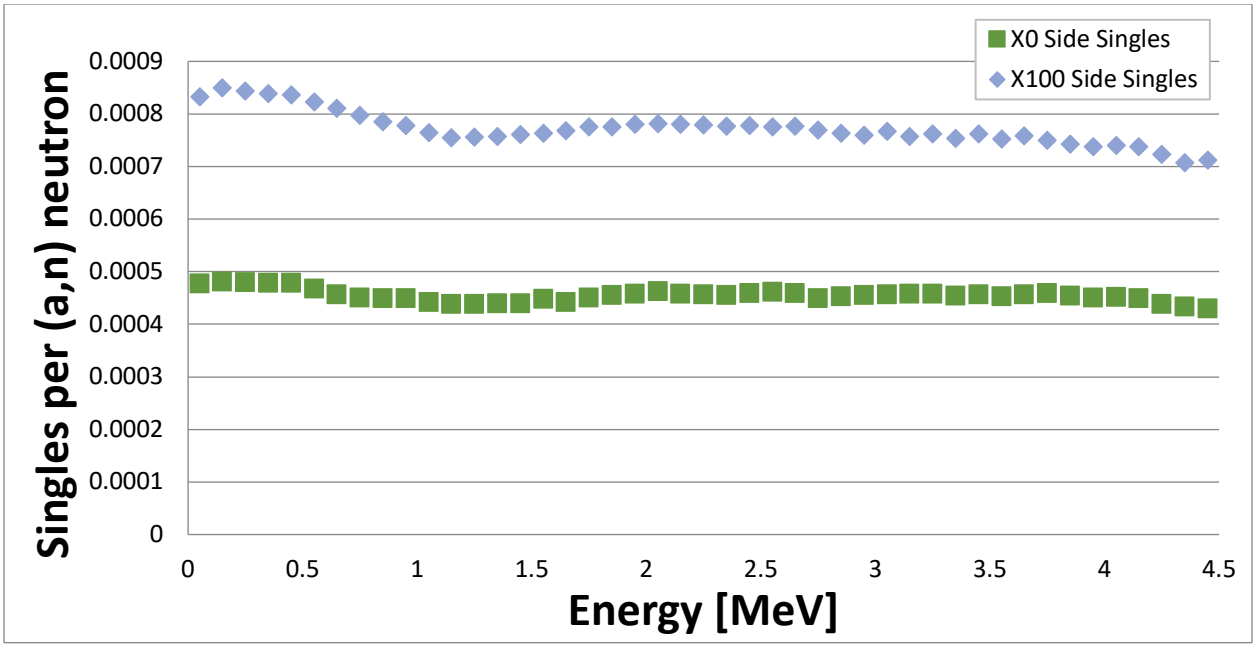


Figure B-5. Simulated side singles response as a function of energy for mono-energetic neutrons.

Each of the four possible $^{19}F(\alpha,n)$ spectra was simulated at four enrichments ranging from 0.2% to 4.947%. The atom percentages used for each simulated enrichment are listed in Table B-1. At each enrichment the associated spontaneous fission contribution was also simulated by using PAR=SF on the SDEF card. Absolute emission rates were calculated using the nuclear data listed in Table B-2. The simulated response is then summarized in Table B-3, where the relative standard deviation is given for each enrichment-distribution set both for the (α,n) contribution and then for the total signal. The effect of the (α,n) spectrum is considerably reduced when looking at the total signal for the depleted uranium (DU) simulations especially as the spontaneous fission of ^{238}U is the main neutron source.

Table B-1. Atom percentages used for each simulated enrichment (%).

Enrichment	DU (0.20%)	1.50%	3.00%	4.95%
U-234	0.000150	0.00143	0.00318	0.00627
U-235	0.0289	0.217	0.434	0.707
U-236	0.00	0.000994	0.00199	0.000149
U-238	14.26	14.07	13.85	13.57
F-19	85.71	85.71	85.71	85.71

Table B-2. Source yields for uranium isotopes in UF_6: (α,n): ^{234}U (Miller et al. 2014), ^{235}U and ^{238}U (Croft 1997), ^{236}U (Reilly et al. 1991), SF: (Reilly et al. 1991)

Isotope	F(α,n) [n/s/g]	Spontaneous Fission [n/s/g]
U-234	474	$5.02 \cdot 10^{-3}$
U-235	0.118	$2.99 \cdot 10^{-4}$
U-236	2.90	$5.49 \cdot 10^{-3}$
U-238	$1.21 \cdot 10^{-2}$	$1.36 \cdot 10^{-2}$

Table B-3. Relative standard deviation (%) for the simulated response when varying the initial (α,n) spectrum for each enrichment-distribution pair

Enrichment	UF_6 Distribution	Singles σ_R (%)		Doubles σ_R (%)		Side Singles σ_R (%)	
		(α,n)	Total (α,n)+(SF)	(α,n)	Total (α,n)+(SF)	(α,n)	Total (α,n)+(SF)
DU (0.20%)	X0	0.68	0.37	2.62	0.20	0.53	0.29
	X100	0.55	0.30	3.37	0.21	0.03	0.02
1.50%	X0	0.38	0.31	0.28	0.14	0.46	0.38
	X100	0.26	0.22	0.53	0.25	0.19	0.16
3.00%	X0	0.31	0.28	0.16	0.12	0.62	0.56
	X100	0.21	0.19	0.64	0.45	0.13	0.12
4.947%	X0	0.24	0.23	0.23	0.20	0.23	0.22
	X100	0.19	0.18	0.39	0.33	0.26	0.25

The energy sensitivity of the PNEM and the secondary-side detector appears relatively small in all cases using the four simulated (α,n) starting spectra. This is likely due to the main differences in the simulated spectra being < 0.3 MeV and in the 0.95 to 1.25 MeV energy range where a change in the initial neutron energy has minimal effect on count rate.

It is believed that rather than being sensitive to energy spectrum, the factor limiting accuracy of simulations is likely to be the thick target (α,n) yield. The uncertainty in the yield is reported as being 576 ± 7.3% (Sampson 1974), 474 ± 4.4% (Miller et al. 2014), 503 ± ~4% (Kulisek et al. 2017; uncertainty estimated by Croft et al. 2020), and 507 ± ~1.1% (Croft et al. 2020).

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