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Our Future Nuclear Data Needs

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Abstract

A well-established knowledge of nuclear phenomena including fission, reaction cross sections, and structure/decay properties is critical for applications ranging from the design of new reactors to nonproliferation to the production of radioisotopes for the diagnosis and treatment of illness. However, the lack of a well-quantified, predictive theoretical capability means that most nuclear observables must be measured directly and used to calibrate empirical models, which in turn provide the data needed for these applications. In many cases, either there is a lack of data needed to guide the models or the results of the different measurements are discrepant, leading to the development of evaluation methodologies to provide recommended values and uncertainties. In this review, we describe the nuclear data evaluation process and the international community that carries it out. We then discuss new measurements and improved theory and/or modeling needed to address future challenges in applied nuclear science.

Contents

1. INTRODUCTION

1.1. The Two Faces of Nuclear Data

Low-energy nuclear science (LENS) straddles the fence between curiosity- and application-driven pursuits. On the curiosity-driven side, low-energy nuclear structure offers a unique laboratory to examine the interplay between single-particle and collective behavior and to explore the transition from quantum to continuum behavior in a mesoscopic setting. LENS also plays an important role in nuclear astrophysics, improving models of stellar energy generation and allowing isotopic abundances to be used to inform models of the astrophysical environments where heavy nuclei are formed. On the application-driven side, a well-quantified knowledge of low-lying nuclear structure and reactions is needed to model energy generation and isotope production for medical and industrial uses and for the national security and counterproliferation communities.

A hallmark of LENS is that most theoretical descriptions are descriptive rather than rigorously predictive at the level of accuracy and over the entire range of energies, et cetera, needed for real-world applications. As a result, targeted measurements coupled to a data evaluation process are used to produce databases of recommended values that can be employed by the user communities. The evaluation processes range from close adherence to experimental results in the case of low-energy nuclear structure and decay data to experiment-guided modeling in the case of nuclear reactions. This work is carried out by subject-matter experts who usually come from the field utilizing the data. These individuals spend many years studying under experienced evaluators in order to develop the highly specialized skills required for the evaluation process.

This review begins with a description of the nuclear data evaluation process, including the domestic and international organizations that help organize evaluation efforts. It then describes ways to improve the modeling of (n, x) reactions on nuclides important for applications. These improvements to the modeling include providing a better understanding of fission, improving the modeling of inelastic neutron scattering, and better determining the properties of the highly excited

continuum states that determine which reactions dominate at a given excitation energy. The review then discusses needs for specific applications including national security/nonproliferation, medical isotope production, and nuclear energy. It ends by introducing the recently formed Nuclear Data Interagency Working Group (NDIAWG), which is formulating a national plan to address high-priority needs, and by presenting ideas about how to change the way we educate evaluators to ensure that a well-trained workforce is available to meet these important needs.

1.2. The Nuclear Data Pipeline

Before diving into the data needs, one must understand the nuclear data pipeline that takes experimental or theoretical results and prepares them for use in applications. This process is illustrated in **Figure 1** and can be divided into four steps:

1. compilation,
2. evaluation,
3. processing, and
4. validation.

This process ends with the users and their application, but actually begins with the performance of carefully designed measurements with well-defined uncertainties. Therefore, those funding or performing the original experiment would be wise to understand the entire nuclear data workflow in order to ensure that new results find their way to the intended user. For example, if a new result forces changes to data formats (step 2) or processing (step 3) or to application codes (step 4 and the eventual user), this fact should be considered when developing a new activity.

The first step in the pipeline is compilation, which begins with potentially the most important database: Nuclear Science References (NSR) (1). NSR contains references to publications from major LENS journals as well as internal reports from labs throughout the world and serves as the starting point for the data evaluation process. Descriptive keywords are assigned to these articles by individuals with LENS backgrounds, enabling the use of this information in later portions of the evaluation process.

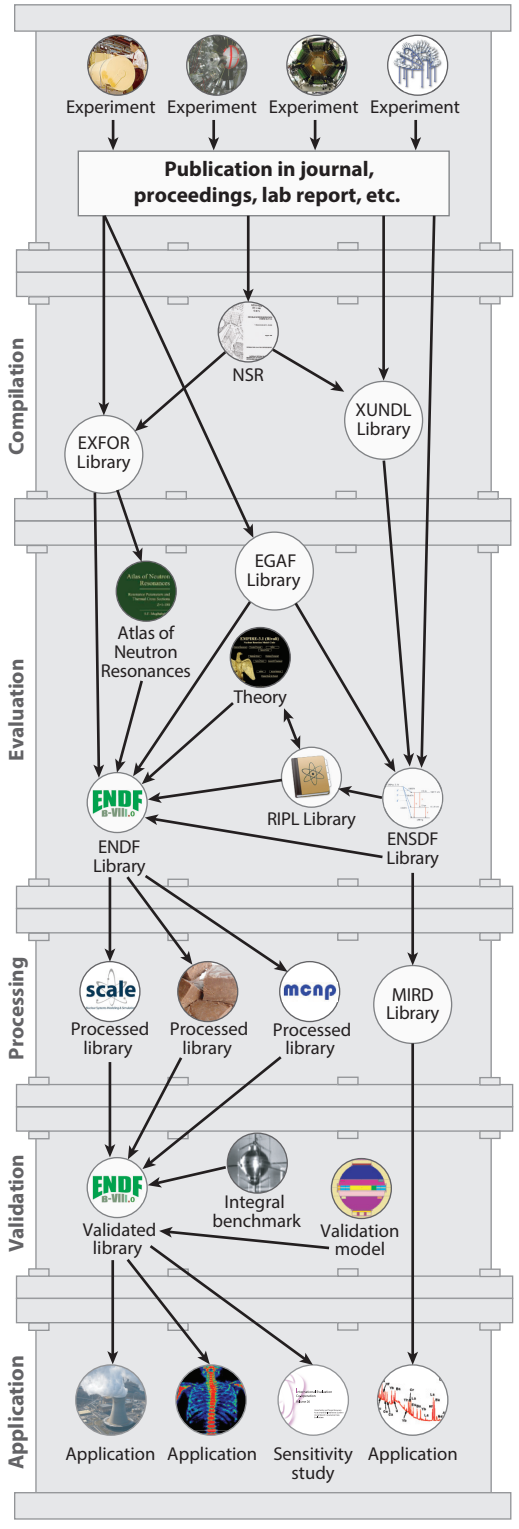
Thereafter, the data from the primary references contained in NSR, including both mean values and uncertainties, are extracted in numerical form and compiled into one of two databases, depending on whether the data are primarily regarding nuclear structure and decay or regarding nuclear reactions. This step may also include tracking down unpublished data from original sources before those data are lost.

The nuclear structure compilation database is called the Experimental Unevaluated Nuclear Data List (XUNDL) (2). The nuclear data in XUNDL are organized by nuclide, meaning that a single publication could lead to the production of more than one XUNDL data set. Each XUNDL data set is treated as a stand-alone work and is not required to agree with existing, evaluated nuclear data for the nuclide it concerns. However, during XUNDL compilation, basic consistency checks are performed and any internal inconsistencies are pointed out to the author to allow them to be reconciled in the database. XUNDL is used both by nuclear structure evaluators to aid in their work and by nuclear structure researchers as a quick way to get access to data from current publications in order to guide their own research.

The nuclear reaction compilation database is referred to as the Experimental Nuclear Reaction Data (EXFOR) Library (3). It includes not only reaction cross sections but also related data such as fission yields, resonance integrals, polarization data, and more. Given the wide range of data types present in EXFOR, there is significantly less error checking than what is performed during XUNDL compilation. However, an online visualization tool is provided to allow users to plot and

Figure 1

The nuclear data pipeline.
Abbreviations: EGAF, Evaluated Gamma-ray Activation File; ENDF, Evaluated Nuclear Data File; ENSDF, Evaluated Nuclear Structure Data File; EXFOR, Experimental Nuclear Reaction Data; MIRD, Medical Internal Radiation Dose; NSR, Nuclear Science References; RIPL, Reference Input Parameter Library; XUNDL, Experimental Unevaluated Nuclear Data List.



manipulate the data. EXFOR is used by both nuclear reaction evaluators and the nuclear reaction research and application community.

The second step in the process is evaluation itself. In the case of nuclear structure data, this step involves reconciling multiple data set types (e.g., prompt γ and particle spectra, decay data) and deriving a recommended set of adopted values. These data form the basis of the Evaluated Nuclear Structure Data File (ENSDF) (4). While most of the ENSDF evaluation is performed in the United States, there is a significant and growing contribution from evaluators around the world. The governing body that determines the rules for the evaluation process is the Nuclear Structure and Decay Data Network (NSDD) administered by the International Atomic Energy Agency (IAEA).

In the case of nuclear reaction data, the process is markedly different. The data from EXFOR are used to guide a physics-based model calculation through a variety of variance minimization procedures, resulting in best estimates of mean values and their uncertainties, including covariances, which are put into one or more regional libraries such as the recently released Evaluated Nuclear Data File (ENDF), ENDF/B-VIII.0 (5). Most of the effort put into the production of ENDF is provided by applications-oriented users because of its importance to national security, international counterproliferation, and nuclear energy. A consequence of this application focus is that the vast majority of the data in ENDF concern neutron-induced reactions at thermal (25 meV), fast fission, or 14.1 MeV. This has profound implications for isotope production since many of the nuclides are produced through charged-particle-induced reactions.

While ENSDF and ENDF contain a vast quantity of nuclear data, there are significant amounts of data that are not contained in them either because they "fall between" nuclear structure and nuclear reactions or because they are present in a format that is not well suited to their use in applications. Examples include the recently updated Atlas of Neutron Resonances (6), which contains neutron capture resonance widths, centroids, and cross sections; the Evaluated Gamma-ray Activation File (EGAF) of discrete γ-rays emitted following neutron capture (7); and the Atlas of γ-ray Spectra from the Inelastic Scattering of Reactor Fast Neutrons (8). Another very important nuclear data resource for applications is the Reference Input Parameter Library (RIPL) (9), which contains both discrete (distilled from ENSDF) and continuous (nuclear level densities and γ-strength function) nuclear structure data needed for accurate reaction modeling.

With evaluations in hand, the data have to be processed into a form suitable for use in a particular application. This is a surprisingly nontrivial part of the process, requiring a deep knowledge of the physics used in the evaluation process, the formatting limitations of intermediate data formats, and the capabilities of downstream codes that will use the nuclear data. Processing is also needed for the validation of nuclear reaction data using the integral benchmarks described below. Processing also plays a role in the creation of application-specific databases, such as the Medical Internal Radiation Dose (MIRD) (10) database used to guide medical treatment and diagnosis.

The final step in the pipeline is validation. In this step, user codes and user-defined problems are used to validate that the data perform as expected. The class of user problems most often relied on consists of so-called integral benchmark data that depend on many different types of data, including cross sections and emitted particle spectra, and are also extremely precisely known. The canonical example of such a benchmark is a critical assembly, where the ratio of neutron production to neutron loss (k_{eff}) is known to several significant figures. However, there are many other kinds of integral benchmarks used to test neutron interaction data alone, including reaction rate measurements and 14 MeV and fission spectrum transmission experiments, and many more for other forms of nuclear data.

There are multiple ways to interact with the nuclear data evaluation process along the way, including through web interfaces hosted by the National Nuclear Data Center (NNDC)

and the IAEA Nuclear Data Section, a smartphone-based application (11), and publication in peer-reviewed journals including *Nuclear Data Sheets* (**https://www.sciencedirect.com/journal/nuclear-data-sheets**) and *Atomic Data and Nuclear Data Tables* (**https://www.sciencedirect.com/journal/atomic-data-and-nuclear-data-tables**).

Many different government agencies support nuclear data evaluation activities, including several components of the US Department of Energy National Nuclear Security Agency (DOE-NNSA) and the National Criticality Safety Program (NCSP). However, the primary domestic organization responsible for organizing the evaluation process is the US Nuclear Data Program (USNDP) within the DOE Office of Nuclear Physics (DOE-NP). The USNDP is headquartered at the NNDC at Brookhaven National Laboratory (BNL) and includes centers at Argonne (ANL), Oak Ridge (ORNL), Los Alamos (LANL), and Lawrence Livermore National Laboratory (LLNL), as well as joint university/lab programs at North Carolina State University/Triangle University Nuclear Laboratory, Michigan State University/National Superconducting Cyclotron Laboratory, and the University of California/Lawrence Berkeley National Laboratory (LBNL).

1.3. International Collaboration

Much of the work of nuclear data evaluation takes place as a part of international collaborations covered under the Organisation of Economic Cooperation and Development's Nuclear Energy Agency (OECD-NEA) and the IAEA. The NEA addresses the nuclear technology interests of its 33 member states, and in the area of nuclear reaction data, the Working Party on Evaluation Coordination (WPEC) is the main forum for collaborative effort between the nuclear data library projects from the NEA countries, namely ENDF/B (United States), JENDL (Japan), Joint European Fission Fusion (JEFF) (NEA), TENDL (Europe), and BROND (Russia), as well as the non-OECD file project CENDL (China).

A powerful instrument of WPEC to drive progress in nuclear data is the so-called Subgroup, or SG: Members of WPEC identify a common area in nuclear data that requires improvement, and if enough support from the various data library projects is present, a Subgroup is formed. Subgroups typically operate on a 3–5-year time frame. A recent successful example of a WPEC Subgroup, with specific relevance to the United States, includes the large-scale horizontal Collaborative International Evaluated Library Organisation (CIELO) effort (SG40) on worldwide nuclear data evaluation for the most important fission energy–related materials. The CIELO initiative has led to new ENDF/B-VIII.0 evaluations for ^{235,238}U, ^{56}Fe, and ^{16}O, among others. Other successful Subgroups include one centered on developing a new Generalized Nuclear Database Structure (GNDS) format (SG38) (**https://www.oecd-nea.org/science/wpec/sg38**) and one focused on covariance adjustment for improvement of nuclear data files (SG39) (**https://www.oecd-nea.org/science/wpec/sg39**).

WPEC also hosts long-term expert groups. Current groups are working on the development of the GNDS format mentioned above, which will provide a data library interface between nuclear physics and applications more modern than the ENDF-6 format, and the High-Priority Request List, which assembles the most important nuclear data requests from applications in a unified format to stimulate experimentalists and evaluators to provide these data. A full list of past and current Subgroups of WPEC is available elsewhere (12).

A key player in nuclear data evaluation is the IAEA, which covers the interests of its 170 member states. The main task of the IAEA Nuclear Data Section is to provide fundamental nuclear databases for basic and applied use, with data originating from experiments and theoretical simulations covering both nuclear structure and nuclear reaction data. An important collaboration coordinated by the IAEA is the Nuclear Reaction Data Center Network, which is responsible for

keeping the EXFOR database of experimental nuclear reaction data up to date. The NNDC is responsible for the US input to EXFOR.

In addition, the IAEA organizes Coordinated Research Projects (CRPs) and technical meetings as instruments to align international nuclear data efforts toward the production of validated databases ready for applied use. Examples of recent and current CRPs include a 2018 venture centered on nuclear data for primary radiation damage, which has been completed; an ongoing effort to improve nuclear model parameters for fission reaction calculations by modern nuclear model codes such as EMPIRE (13), CCONE (14), COH3 (15), and TALYS (16); and an effort to create the first-ever evaluated database of radiative strength. In 2019, a CRP on fission yields will start; it aims to produce updated fission yield libraries for the major actinides to respond to requests from the areas of reactor technology, safeguards, and nonproliferation.

Nuclear data evaluations of neutron-induced reactions for fission applications are covered by the International Nuclear Data Evaluation Network (INDEN), an IAEA initiative that continues the CIELO efforts of the NEA for differential nuclear data developments, and evaluations, for the most important materials relevant for fission technology. Other long-term projects are the NSDD, cross sections for beam current monitor reactions used in medical isotope production cross-section measurements, neutron standards, and the Fusion Evaluated Nuclear Data Library (FENDL). **Table 1** lists the major databases currently maintained by the nuclear data community.

2. IMPROVED (n, x) REACTION MODELING: A CROSS-CUTTING NEED

Accurate, physics-based modeling of neutron-induced reactions, particularly on the "Big Three" (^{235,238}U and ^{239}Pu), is key to virtually all nuclear science and technology applications. While a great deal of effort has been made to improve (n, f) data, a fundamental understanding of this most important of reaction mechanisms remains elusive. Furthermore, since fission leads to the production of neutrons with energies up to several MeV, elastic and inelastic scattering also play a critical role. Unfortunately, efforts to improve neutron scattering data have tended to take a back seat to fission due to a deficit of high-quality experimental data. Lastly, while all modeling of (n, x) reactions requires a good understanding of unbound nuclear states populated in (n, x) reactions, it remains one of the least-understood topics in LENS. In this section, we explore all three of these cross-cutting topics and present plans to improve them.

2.1. A Deeper Understanding of Fission

Not all fission data needed in nuclear physics applications can be readily measured in the laboratory. In many cases, the targets may be too short-lived, the analysis might yield unacceptably large uncertainties, or separate measurements of the same quantity may produce discrepant results. As with other reaction data, fission evaluation proceeds through the use of a model designed to reproduce the data as closely as possible. However, in the case of fission, we still lack a predictive theoretical framework to match all types of measured data within experimental errors. A truly predictive theory of fission will require advances in theoretical techniques and computational machinery, along with improved experimental data.

For fission theory, we can usually divide our description of the process into the events that come before scission (prescission physics) and those that follow scission (postscission physics). Eventually, one could envision a comprehensive model that accurately follows the entire fission process from the formation of the parent nucleus to the last decays of the final product nuclei within a single internally consistent theoretical framework. Such a model is not yet feasible, so

Table 1 Names, locations, and descriptions of the major databases maintained by the international nuclear data community

Database	Comments	Type	Website
Nuclear Science References (NSR)	List of published nuclear data articles	Compilation	**https://www.nndc.bnl.gov/nsr/**
Experimental Nuclear Reaction Data (EXFOR)	Compiled reaction data	Compilation	**https://www.nndc.bnl.gov/exfor/exfor.htm**
Experimental Unevaluated Nuclear Data List (XUNDL)	Compiled structure data	Compilation	**https://www.nndc.bnl.gov/ensdf/ensdf/xundl.jsp**
Evaluated Nuclear Data File (ENDF)	Evaluated reaction data	Evaluated	**https://www.nndc.bnl.gov/exfor/endf00.jsp**
Evaluated Nuclear Structure Data File (ENSDF)	Evaluated structure and decay data	Evaluated	**https://www.nndc.bnl.gov/ensdf/**
Reference Input Parameter Library (RIPL)	Data for nuclear model calculations	Derived	**https://www-nds.iaea.org/RIPL-3/**
Atlas of Neutron Resonances	Evaluated neutron data	Evaluated	None
Atlas of Gamma-Ray Spectra from the Inelastic Scattering of Reactor Fast Neutrons	Compiled reaction data	Compilation	**http://nucleardata.berkeley.edu**
Medical Internal Radiation Dose (MIRD)	Derived decay data	Derived	**https://www.nndc.bnl.gov/mird/**
Nuclear Structure and Decay Data (NUDAT)	Graphical interface for structure and decay data	Derived	**https://www.nndc.bnl.gov/nudat2/**
Evaluated Gamma-ray Activation File (EGAF)	Evaluated thermal capture γ-ray data	Evaluated	**https://www-nds.iaea.org/pgaa/egaf.html**
Java-Based Nuclear Data Information System (JANIS)	Graphical interface for reaction, structure, and decay data	Derived	**https://www.oecd-nea.org/janis/**
Joint Evaluated Fission and Fusion Nuclear Data Library (JEFF)	Evaluated reaction data	Evaluated	**https://www.oecd-nea.org/dbdata/jeff/jeff33/**
Japanese Evaluated Nuclear Data Library (JENDL)	Evaluated reaction data	Evaluated	**https://wwwndc.jaea.go.jp/jendl/j40/j40.html**
Computer Index of Nuclear Reaction Data (CINDA)	Compiled neutron reaction data	Compilation	**https://www.nndc.bnl.gov/exfor/cinda.htm**

(*Continued*)

Table 1 (*Continued*)

Database	Comments	Type	Website
Chinese Evaluated Nuclear Data Library (CENDL)	Evaluated reaction data	Evaluated	None
Russian File of Evaluated Neutron Data (ROSFOND)	Evaluated reaction data	Evaluated	**https://www.ippe.ru/reactors/reactor-constants-datacenter/abbn-reactor-group-constant-database**
European Activation File (EAF)	Derived decay data	Evaluated	**https://www.oecd-nea.org/dbforms/data/eva/evatapes/eaf_2010/**
International Reactor Dosimetry File (IRDFF)	Evaluated neutron reaction data with uncertainties	Evaluated	**https://www.oecd-nea.org/dbforms/data/eva/evatapes/irdf_2002/**
International Criticality Safety Benchmark Evaluation Project (ICSBEP)	Compiled critical and subcritical assembly data	Compilation	**https://www.oecd-nea.org/science/wpncs/icsbep/handbook.html**
TALYS Evaluated Nuclear Data Library (TENDL)	Evaluated reaction data	Evaluated	**https://tendl.web.psi.ch/tendl_2017/tendl2017.html**
Russian Evaluated Neutron Data Library (BROND)	Evaluated reaction data	Evaluated	**https://www.oecd-nea.org/dbdata/data/nds_eval_libs.htm**
Fusion Evaluated Nuclear Data Library (FENDL)	Evaluated reaction data	Evaluated	**https://www-nds.iaea.org/fendl/**
International Reactor Physics Experiment Evaluation (IRPhE) Project	More complex experiments than the ICSBEP but still useful for validation	Compilation	**https://www.oecd-nea.org/science/wprs/irphe**
Shielding Integral Benchmark Archive and Database (SINBAD)	Database of LLNL pulsed spheres and other shielding/transmission experiments	Compilation	**https://www.oecd-nea.org/science/wprs/shielding**
Measured Isotopic Concentrations of Spent Nuclear Fuel (SFCOMPO)	Database of measured isotopic concentrations of spent nuclear fuel with operational histories and design data	Compilation	**https://www.oecd-nea.org/sfcompo**

different physics models are used to treat pre- and postscission processes. A wide range of theoretical approaches have been developed since the late 1930s to describe prescission physics, including scission-point models (17, 18), microscopic–macroscopic models (19) with random-walk dynamics (20), Langevin dynamics (21), time-dependent Hartree–Fock and Hartree–Fock–Bogoliubov theory (22–25), the time-dependent superfluid local density approximation (26–29), and the time-dependent generator–coordinate method (30–32). Several models have also been developed to describe postscission physics and built into computer codes, such as GEF (33), FIFRELIN (34), CGMF (35), and FREYA (36, 37).

The division between pre- and postscission physics in modeling efforts suggests a corresponding division in nuclear data needs to improve our understanding of the fission process. Postscission observables are generally easier to measure, and include the properties of fission fragments and the neutrons and γ-rays they emit. Recently, Bertsch et al. (38) suggested a subset of fission data that tend to have relatively small uncertainties and could therefore be used to test and validate fission models. These data consist of fission barriers, fragment mass distributions, fragment total kinetic energies, spontaneous-fission lifetimes, and fission isomer excitation energies. In addition to these quantities, the angular distribution of fission fragments can also provide useful information about the angular momentum of the fissioning nucleus and further constrain model calculations.

In addition to standard experiments looking at individual fission properties, multiparameter measurements that give a comprehensive view of a fission event should be pursued as facilities and detectors improve. Ideally, a multiparameter experiment would record all possible fission observables (e.g., fragment charge, mass, and energy distributions as well as neutron and γ spectra and multiplicities) on an event-by-event basis, thereby making it possible to reconstruct the state of the nucleus at scission. In practice, each additional observable reduces statistics, making such measurements exceedingly challenging. Finally, there are experimental measurements that directly probe the prescission dynamics of the fission process, although they carry large uncertainties and suffer from various ambiguities and limitations. These include measurements of scission neutrons (39), fission timescales (40), and muon-induced fission (41). Probes of prescission physics can provide unique insights into the details of the fission process, and are therefore extremely valuable to fission theory.

Both prescission and postscission theory and data will be needed to reach a satisfactory understanding of the fission process, and thereby make reliable predictions of nuclear quantities that cannot be measured in the laboratory. Prescission models can provide input to postscission models wherever the data are lacking, and postscission models can be used to improve prescission calculations using a wealth of available experimental data.

The ultimate goal for theorists—a comprehensive description of fission starting from protons, neutrons, and their interactions that provides accurate values for all fission data of interest—remains elusive. Many questions about the fission process remain open, such as how the initial partition of energy between kinetic and internal excitation occurs, what the relevant collective and single-particle degrees of freedom are near scission, and how they evolve and couple throughout the process. Lastly, a compelling question that remains is how to best quantify uncertainties (both epistemic and aleatory) in fission calculations. Improved experiments and theoretical models are needed to address all of these questions, and to provide the deeper understanding of the fission process needed for most applications.

2.2. Improved ^{235,238}U and ^{239}Pu(n, n') Data for Neutron Transport

Elastic and inelastic scattering profoundly affect neutron populations in any nuclear application by changing the directions and energies of the neutrons in the system. However, improvements in

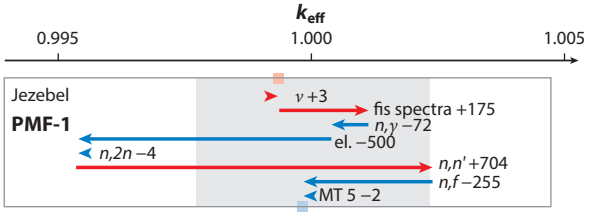


Figure 2

Simulations of criticality k_{eff} for ^{239}Pu for the fast assembly Jezebel PMF-1. The figure shows that both LANL CIELO-1 (ENDF/B-VIII.0) and CEA CIELO-2 (JEFF-3.3) predict similar k_{eff} values, but do so for very different reasons. The changes in criticality are evident when individual cross-section channels are substituted between the two evaluations. The shaded region shows the resulting range of values for k_{eff}. Abbreviations: CEA, Commissariat à l'Energie Atomique; CIELO, Collaborative International Evaluated Library Organisation; ENDF, Evaluated Nuclear Data File; JEFF, Joint European Fission Fusion; LANL, Los Alamos National Laboratory.

neutron scattering data have been hampered by the fact that they do not produce easily measured signals such as fission or a radiochemical observable such as neutron capture. This has resulted in significant disagreements between different reaction evaluations. These disagreements manifest as compensating errors that are resistant to the validation process. This issue has been recognized by the nuclear data community since the early days of the WPEC (42) and, more recently, by Maslov et al. (43).

A compelling example of compensating errors involves the Jezebel ^{239}Pu critical assembly (PMF-1). Models of Jezebel using LANL CIELO-1 (ENDF/B-VIII.0) and Commissariat à l'Energie Atomique (CEA) CIELO-2 (JEFF-3.3) reproduce a unity value of k_{eff} to better than 1 part in 104 (44). However, when the authors of this work substituted one nuclear quantity at a time between the two databases, the value of k_{eff} varied considerably. The greatest changes came from varying the elastic and inelastic scattering cross sections, which led to changes in the k_{eff} of −500 pcm and +704 pcm, respectively (**Figure 2**). Similar examples of such compensating errors were also observed in earlier research by Bauge et al. (45).

The CIELO CRP referred to above (44) was begun to have the international nuclear data evaluation community agree on a set of evaluated cross sections and output spectra for six critically important nuclei—^{1}H, ^{16}O, ^{56}Fe, ^{235,238}U, and ^{239}Pu—in order to address the issue of compensating errors. This list has recently been expanded by the IAEA, via CIELO's follow-up, INDEN, to include ^{14,15}N, ^{9}Be, ^{23}Na, ^{59}Co, ^{58}Ni, and $^{238-242}$Pu, among other isotopes.

CIELO has relatively few energy or angle differential data to draw upon to aid in its evaluation of (n, n') on the Big Three actinides, leading to "significant differences among the evaluations for ^{239}Pu in the fast energy range, with ENDF/B-VII.1 and JENDL-4.0 lying significantly above the JEFF-3.1 evaluation" (46, p. 16). The ^{239}Pu data provided in Reference 46 are all from indirect measurements made prior to 1970 (47, 48). The case is similar for ^{235}U, where differences for $E_n > 50$ keV "have a large impact on the fast criticality (Godiva), leading to a 540 pcm swing in calculated criticality, as shown by Go Chiba" (49, p. 12).

A significant source of $(n, xn\gamma)$ partial cross-section data from 1997 to 2003 was the GEANIE (Germanium Array for Neutron-Induced Excitations) Collaboration at the Los Alamos Neutron Science Center/Weapons Neutron Research (LANSCE/WNR) facility. While the primary goal of the GEANIE Collaboration was to determine the ^{239}Pu$(n, 2n)$ cross section (50), its research resulted in the publication of more than two dozen papers over the course of more than 15 years. Experiments were run on all three of the major actinides: ^{235}U (51), ^{239}Pu (50), and ^{238}U

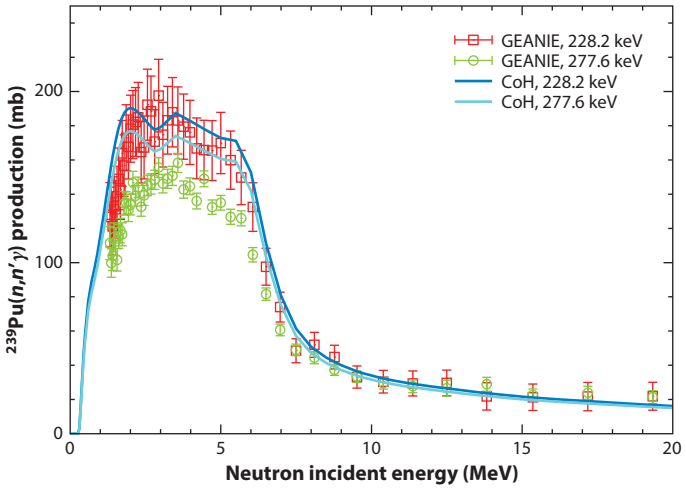


Figure 3

A selection of the ^{239}Pu$(n, n'\gamma)$ partial γ-ray cross sections (53) for the two transitions de-exciting the $5/2^+$ level at $E_x = 285.46$ keV.

(52). GEANIE produced the first high-resolution partial γ-ray cross-section data on the Big Three. These include one of the first results for ^{239}Pu$(n, n'\gamma)$, shown in **Figure 3** together with calculations using the CoH reaction modeling code developed by Kawano et al. (15). Note that GEANIE did not include any neutron detectors, producing only angle-integrated partial γ-ray cross sections.

The need for new scattering data has motivated many new experiments. These have included energy-differential/angle-integrated measurements of the ^{238}U(n, n') cross section using the GELINA spallation neutron source in Geel, Belgium, by the CIELO Collaboration (54) and quasi-angle-differential measurements using the Rensselaer Polytechnic Institute Linac by Daskalakis et al. (55).

The ideal way to improve (n, n') data on ^{235,238}U and ^{239}Pu would be to perform measurements of the scattered neutron in coincidence with transitions emitted by the excited nucleus. In the case of the actinides, this could require the observation of conversion electrons, given that the lowest transitions in all actinides are almost entirely converted. Unfortunately, neutron beams usually have quite low intensity, with a fluence in the fast (1–10 MeV) region between 10^3 and 10^7 neutrons s^{-1} cm^{-2} at the target location. Furthermore, many spallation neutron sources have a significant high-energy spectral component that can cause interelement scattering in a neutron scintillator array, rendering precise determination of the scattered neutron energy difficult to perform. However, intense limited-energy neutron sources based on thick-target deuteron breakup (56) and forward-fitting techniques developed for use at spallation neutron sources (57) offer an opportunity to perform neutron-gamma coincident measurements in the fast fission energy spectrum. These techniques could provide valuable data to improve nuclear data evaluations of inelastic scattering, even on targets where fission is an open channel.

2.3. Improved Treatment of Continuum Nuclear Data Properties

Information regarding states in the unresolved continuum region above the particle separation energy is of central importance to accurately modeling the competition between particle and γ-ray emission. The most important of these are the nuclear level density (NLD), as a function of energy,

spin, and parity, and the radiative strength function (RSF), which describes the ability of excited nuclear matter to absorb or emit photons. Several recent studies have shown the importance of the RSF in modeling (n, γ) reactions for neutrons with energies between 1 and 100 keV, which correspond to the spectrum at the site of neutron capture nucleosynthesis in stars (58, 59) and energy generation in fast reactors and nuclear security applications.

Over the past several years, research led by the group at the University of Oslo has shown that there is a surprisingly large amount of M1 radiative strength believed to be a type of "scissors" collective mode throughout the actinide mass region near the valley of stability including ^{238}U, ^{239,240,242}Pu, and ^{232}Th (60–64). Other results from the Oslo group show that actinide NLDs appears to follow a constant-temperature form with a roughly Gaussian dependence on angular momentum that is often described via an energy-dependent width spin cutoff parameter (σ) of the form

$$\sigma(E_x) = 0.01389 \frac{A^{5/3}}{\tilde{a}} \sqrt{aU}, \qquad 1.$$

where U is roughly E_x minus the pairing gap Δ, a is the level density parameter, A is the nuclear mass, and $\tilde{a}$ is the asymptotic level density value one would obtain in the absence of any shell effects.

There is a great deal of uncertainty in the magnitude of the spin cutoff parameter. This is a significant issue since good knowledge of the J distribution is needed to accurately model the neutron versus γ-ray emission probability in nuclear reactions. This sensitivity was recently shown by Wiedeking et al. (65), who demonstrated that neutron emission from excited states in ^{95}Mo well above the neutron separation energy populated via (d, p) was hindered for states with high angular momentum.

In an interesting example of the interconnectedness of nuclear data, $(n, n'\gamma)$ provides a useful tool for studying the spin cutoff parameter since it proceeds largely through the population of a compound nucleus with a relatively wide range of angular momentum. **Figure 4** shows the ratio of the partial cross section for the population of the first two 2^+ levels in ^{238}U(n, n') as a

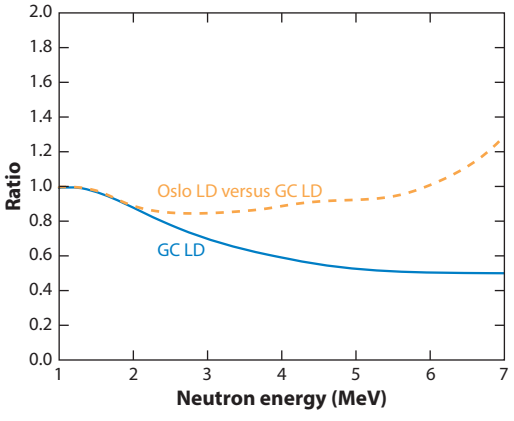


Figure 4

Ratio of the ^{238}U(n, n') cross sections for the first two 2^+ levels calculated using TALYS for two different sets of input parameters as a function of incident neutron energy. The solid line shows the ratio where the spin cutoff has been scaled to ±25% of its default value. The dashed line shows the case where the default value calculated using LD Model 1 (GC) is divided by the value obtained using the data from Guttormsen et al. (62). Abbreviations: GC, Gilbert and Cameron; LD, level density.

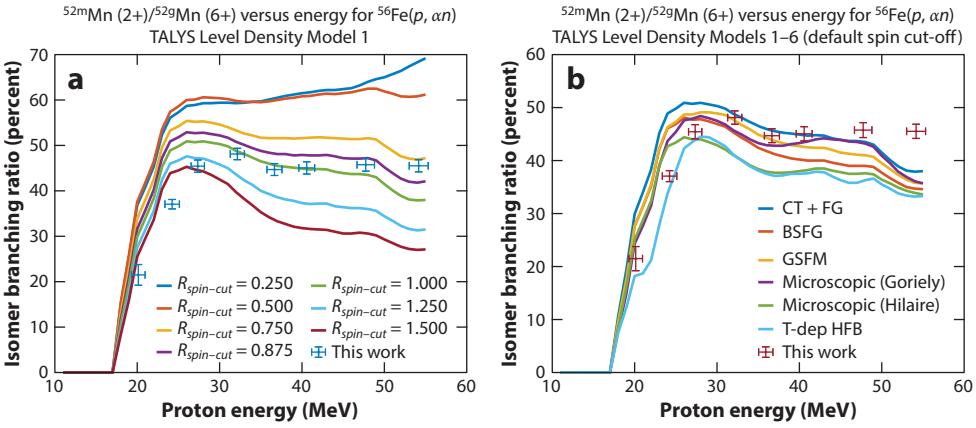


Figure 5

Predictions of the production of ^{52m}Mn($J_\pi = 2^+$)/^{52g}Mn($J_\pi = 6^+$) isomer-to-ground-state ratio as a function of energy (*a*) for the GC LD and (*b*) using the six-LD models available in TALYS. Abbreviations: BSFG, back-shifted Fermi gas; CT, constant temperature; FG, Fermi gas; GC, Gilbert and Cameron; GSFM, generalized superfluid model; HFB, Hartree–Fock–Bogliubov; LD, level density.

function of incident neutron energy calculated using the TALYS reaction code for two different choices of spin cutoff. The figure shows the case where a constant-temperature level density was assumed and the spin cutoff parameter was varied from 75% to 125% of its normal value given by Equation 1, above. The ratio decreases by nearly a factor of two with increasing energy. The figure also shows the ratio of the population of the state when the level density and RSF from the recent Oslo measurements (62) are divided by the results obtained using the default Gilbert and Cameron model in TALYS. In this case, the ratio decreases by nearly a factor or two. Similar behavior is observed in the partial cross sections for other off-yrast levels as well.

Information regarding the J dependence of the NLD can also be obtained through observation of the relative population of two low-lying states with very different spins via activation. This method has been used in mercury and gold by Chakravarty et al. (66) and more recently in mercury and platinum by Sudár & Qaim (67). **Figure 5** shows these data for the case of the ^{52m}Mn($J_\pi = 2^+$)/^{52g}Mn($J_\pi = 6^+$) ratio populated via ^{56}Fe($p, \alpha n$) measured via stacked-target activation at the LBNL 88-Inch Cyclotron in comparison to TALYS calculations for different values of the spin cutoff parameter (**Figure 5*a***) and the six level density models available for use in TALYS (**Figure 5*b***).

Note that both the ($n, n'\gamma$) and isomer-to-ground-state ratio measurements could be performed as part of a program that supports improved neutron transport, as described in Section 2.2, above, and a program of cross-section measurements in support of medical isotope production, described in Section 4, below. This synergy between efforts from many applications argues for continued coordination of nuclear data activities regardless of the program they are supporting.

3. NATIONAL SECURITY AND NONPROLIFERATION

3.1. Interpreting the Reactor Antineutrino Spectrum

A canonical example of a nonproliferation activity where improvements in multiple areas of nuclear data are needed is the interpretation of the antineutrino spectrum from the prompt decay of fission fragments in the core of a nuclear reactor. Nuclear reactors are prolific sources of electron

antineutrinos, producing approximately 10^{21} antineutrinos per second for a typical power reactor. These electron antineutrinos are produced by the β^- decay of the more than 800 neutron-rich fission fragments, which are the debris from the main source of energy generation in a reactor, the neutron-induced fission of actinide nuclides. These antineutrinos are also the only radiation escaping from the vessel of a safely operating nuclear reactor, making them ideally suited for reactor monitoring, even in the event that the actor operating the reactor refuses to cooperate with the international community. Recent technological advances in electron antineutrino detection have made another dream a reality, monitoring the performance of nuclear reactors and tracking ^{239}Pu for nonproliferation treaty verification. The WATCHMAN experiment in the United Kingdom has just begun to explore this field (68). Furthermore, the possible use of antineutrino detectors for the detection of nuclear explosions has been discussed (69).

Nuclear reactors have also been an essential tool for the study of the weak interaction. Cowan et al. (70) took advantage of their large antineutrino flux to discover antineutrinos in 1956, more than 25 years after they were first hypothesized by Pauli in 1930, in an open letter to his colleagues (see **https://www.symmetrymagazine.org/article/march-2007/neutrino-invention**), to explain the continuum electron spectra observed following β^- decay. In the last few years, the transformation of electron antineutrinos into the other two flavors was beautifully measured by three large-scale experimental efforts, Daya Bay (71), Double Chooz (72), and RENO (73). These experiments also confirmed a deficit of antineutrinos of approximately 5–9% at short distances that had been revealed in a 2011 reanalysis (74) of the conversion procedure to obtain antineutrino spectra from the measured electron spectra.

This intriguing deficit, as well as a spectrum distortion, has triggered a new generation of very short distance reactor experiments, such as NEOS (South Korea) (75), DANSS (Russia) (76), STEREO (France) (77), PROSPECT (United States) (78), and SoLid (Belgium) (79), whose first results are beginning to be made public. There are no signs of slowing down in the field of nuclear reactor antineutrinos, as the largest experiment to date, JUNO, designed to measure the neutrino mass hierarchy, should be ready for data taking by 2021 (80). JUNO will feature a 20-million-ton detector located 80 km from a 27 GW nuclear power plant in China.

The antineutrino spectrum produced in a nuclear reactor is calculated as the sum of the spectra produced by each of the nuclear fuels ^{235,238}U and ^{239,241}Pu, weighted by the respective fission fractions (81). The spectra for each fuel can be calculated using nuclear databases, in what is commonly known as the summation method. Alternatively, for ^{235}U and ^{239,241}Pu, the spectra can be calculated by converting the aggregate electron spectra measured at the Institut Laue-Langevin (ILL) in the 1980s (82–84). The current best predictions for ^{235}U and ^{239,241}Pu are those by Huber (85), who used the conversion method, whereas the best prediction for ^{238}U is that by Mueller et al. (86), who used a hybrid conversion–summation method.

The use of nuclear databases to calculate antineutrino spectra is considered less precise than the conversion method because of deficiencies in the underlying fission and decay data, such as β intensities (87), β shape factors due to first-forbidden transitions (88), evaluated library issues (89), and fission yields (90). There have been considerable efforts in the last few years to improve this situation, including (*a*) experimental campaigns at Jyväskylä (91), ORNL (92), and ANL (E.A. McCutchan, manuscript in preparation) to obtain highly precise values of β intensities, which have been incorporated in the ENDF/B-VIII.0 decay data sublibrary; (*b*) measurements of isomeric ratios at ILL, Jyväskylä, and ANL; (*c*) measurements of fission yields at GELINA (93), the GSI Helmholtz Centre for Heavy Ion Research (94), and LANL (95); and (*d*) a recent IAEA CRP on β-delayed neutron emitters (96) that would improve the derivation of cumulative fission yields from independent ones. Also planned are measurements of β shape factors as well as a new IAEA CRP to evaluate fission yields. Another issue affecting the summation method is the lack of fission

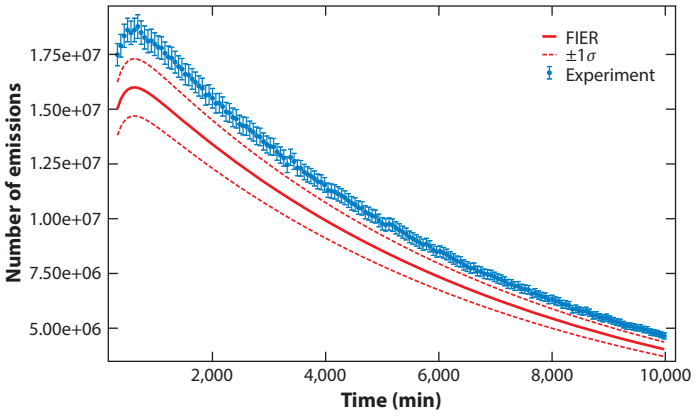


Figure 6

Difference in the FIER-predicted and observed yield of ^{132}I as a function of time for a ^{235}U sample irradiated at the Godiva critical assembly (98). Abbreviation: FIER, Fission Induced Electromagnetic Response code.

yield correlation matrices in the evaluated fission data libraries. As a result, uncertainties cannot be fully computed. There has been some progress in this area; for instance, the ongoing OECD-NEA SG44 will incorporate this topic (97).

The summation method has recently been applied in two different situations. First, the Daya Bay fuel evolution data (99) can be explained by summation calculations including an anomaly of approximately 4% (100). Second, a novel method to analyze the Daya Bay spectrum revealed the signature of four individual fission products, and the same method applied to the electron spectra revealed two additional fission products (101).

The conversion method is not fully free of issues, either. First, it is based on a single set of measurements; also, its most basic premise, the description of a level-to-level spectrum using a particular formalism, has not been tested, as highly precise measurements of electron spectra for fission products above the inverse β decay (IBD) cross-section threshold have not been published. As a result, although it is more precise than the summation method, it is considerably less reliable. A recent article (102) reviewed the different Fermi functions available for the calculation of spectra and showed that a linear correction factor of 6%/MeV could explain the anomaly.

Given the dependence of the antineutrino signal on both decay and fission product yield data, a series of sensitivity study–guided integral experiments, along the lines of critical assembly integral benchmarks, are needed to validate that the underlying nuclear data would provide a strong constraint on the underlying data. One approach would involve irradiating fissile and fissionable samples in facilities with well-understood neutron spectra and comparing the observed delayed γ-rays with a modeled spectrum produced using the Fission Induced Electromagnetic Response (FIER) code developed at Berkeley (98). FIER convolves fission product yield distributions and decay data to predict the γ-ray spectrum after neutron irradiation. The first example of its use involving a comparison to the delayed γ-ray spectrum from a ^{235}U foil irradiated in the Godiva critical assembly indicated several cases where the underlying fission yields or decay data required improvement. **Figure 6** shows an example in which the yield of the ^{132}I needed to be adjusted upward by 0.24σ.

These experiments would involve a sample of depleted ^{238}U since recent results from IBD indicate that the contribution to the observed anomaly in the antineutrino experiment can be attributed to issues with the ^{238}U(n, f) yields. These measurements could be performed at Godiva in the same fashion as the ^{235}U irradiation used to validate FIER, or at other well-documented fast

neutron facilities, including the thick-target deuteron break neutron source at the LBNL 88-Inch Cyclotron (56) and the High Flux Neutron Generator at Berkeley (103), and the results could be compared with the FIER predictions to inform both the fission yield and decay data for ^{238}U(n, f).

3.2. Improved (n, $x\gamma$) Data for Active Interrogation

An invaluable tool for determining the presence of fissile materials for nonproliferation involves prompt neutron and γ-ray spectroscopy coupled to a deuterium–tritium neutron source with associated particle imaging (DT-API). DT-API sources enable tracking of the trajectory of individual neutrons, thereby greatly decreasing background from scattered neutrons. The signal from DT-API generators includes long-chain γ-ray emission from multiple fission events (104, 105), as well as prompt γ-ray and neutron scattering on a wide range of low- and high-Z materials.

The interpretation of these data requires detailed knowledge of both the double-differential neutron scattering and γ-ray production cross sections from fast down to thermal neutron energies. EGAF (7) provides partial γ-ray cross-section data for transitions between discrete states following thermal neutron capture. At higher energies, the Atlas of Gamma-Ray Spectra from the Scattering of Reactor Fast Neutrons (106), which has recently been compiled into a database and reconciled to reflect state-of-the-art γ-ray energies from ENSDF (8), offers discrete γ-ray cross sections for irradiation with fast fission neutrons.

A program of measurements of γ-rays emitted from both discrete and quasi-continuum states would require a relatively modest effort to put in place. The data from these measurements could then be used to guide a physics-based reaction model, such as CoH, TALYS, or EMPIRE, to ensure that the total γ-ray production cross section is consistent with the total capture and scattering cross sections themselves. This effort would also dovetail nicely with measurements to improve data for the inelastic scattering and continuum level properties mentioned in Sections 2.2 and 2.3, above.

4. ISOTOPE PRODUCTION

The high energy density and wide range of decay lifetimes and chemical properties of radionuclides near the nuclear valley of stability make them an extremely versatile tool for applications ranging from the diagnosis and treatment of illness to nuclear nonproliferation and environmental studies to the powering of spacecraft to explore the outer reaches of the Solar System and beyond. Furthermore, given that there are hundreds of unstable nuclei with lifetimes of more than 1 h and less than 100 years between hydrogen and californium, it is clear that we have barely scratched the surface of potential applications.

However, there are significant uncertainties in the reactions used to produce these radionuclides and/or the intensities and energies of their decay radiation. Numerous papers produced over the past 5 years have detailed some of the nuclear data needs associated with producing radionuclides for applications. For a comprehensive list of nuclear data needs associated with radionuclides, we refer the reader to the summary white paper from the Nuclear Data Needs and Capabilities for Applications (NDNCA) Workshop held in Berkeley in 2015 (107), particularly the reactions and isotopes listed in appendix B. Additional guidance regarding nuclear data needs for medical isotopes can be found in a recent series of IAEA studies (108–110), as well as in an exhaustive paper by Qaim (111).

More than 20 million nuclear medicine procedures are performed each year in the United States (112), with the vast majority involving imaging. ^{99m}Tc [half-life ($t_{1/2}$) = 6.0067(5) h], which decays predominantly via the emission of a single 140 keV γ-ray, is used in 80% of of these procedures. ^{99m}Tc is produced via ^{235}U(n, f). However, this approach is less than ideal due to the

increasing age of many reactors (113) and a nuclear weapons proliferation risk associated with reactors utilizing highly enriched uranium. As a result, alternative ^{99m}Tc production pathways, as well as studies of the associated production of long-lived impurities (114), are under consideration (115).

Other imaging radionuclides are used in positron emission tomography (PET), which is now so common that there are numerous companies providing production capabilities built around low-energy medical cyclotrons. The utility of PET imaging, along with the recent rise in multimodal PET/MRI (magnetic resonance imaging) and PET/CT (computed tomography) imaging, has led to significant interest in the development of other β^+-emitting radionuclides. These include ^{18}F [$t_{1/2}$ = 109.771(20) min], which is produced locally via the ^{18}O(p, n) reaction; ^{82}Rb [$t_{1/2}$ = 1.2575(2) min], which is used in cardiac imaging; and ^{68}Ga [$t_{1/2}$ = 67.71(8) min], which is used in the detection of a wide range of tumors. The short half-lives of ^{68}Ga and ^{82}Rb necessitate that they be extracted from longer-lived radioactive parent nuclide generators, whose longer half-lives of 270.93(13) and 25.35(3) days for ^{68}Ge and ^{82}Sr, respectively, allow them to be produced at regional facilities and distributed to medical facilities throughout the world.

Numerous therapeutic radiopharmaceuticals are also in use or under development for cancer treatment. The principle of therapeutic radionuclide cancer therapy involves depositing a targeted dose, such as energy per unit mass, to a tumor cell, capable of producing damage to multiple strands of DNA, from which the cell cannot recover. The ideal dose delivery involves radiation with a high linear energy transfer. While some β^- nuclides, such as ^{177}Lu [$t_{1/2}$ = 6.6475(20) days], are already in use, the relatively long range of β^- particles makes them less favorable than other modes of radioactive decay. These include Auger and Coster–Kronig electron emitters, which typically have a range of less than 1 μm, or α-particle emitters, which have ranges of 2 to 10 μm.

The α-particle-emitting radionuclide ^{225}Ac has attracted particular interest. ^{225}Ac is especially well suited for medical applications in that it has a sufficiently long lifetime [$t_{1/2}$ = 10.0(1) days] to facilitate its incorporation into targeting molecules, and its daughters then decay relatively rapidly ($t_{1/2}$ < 3.2 h) to stable products, minimizing the concurrent dose to healthy tissue. Other promising α-particle therapeutic radionuclides include ^{211}At, ^{212}Pb/^{212}Bi, ^{213}Bi, ^{226}Th, and ^{227}Th, all of which have been identified as high-priority topics by the most recent Nuclear Science Advisory Committee's Long Range Plan for the DOE-NP Isotope Program (116).

A highly promising nuclear medical treatment modality involves coupling a pair of chemically similar radioisotopes, including a therapeutic nuclide capable of delivering a highly localized radiation dose and a chemically identical isotope that emits a positron for PET or a γ suitable for single-photon emission computed tomography (SPECT) imaging. Examples of these so-called theranostic pairs include ^{188}Pt or ^{191}Pt for use in imaging, together with ^{193m}Pt, ^{195m}Pt, and ^{197}Pt, which all have therapeutic potential. Another such promising pair involves the PET isotope ^{134}Ce [$t_{1/2}$ = 3.16(4) days] combined with ^{225}Ac. A recent Nuclear Science Advisory Committee report (116) highlights additional pairs of isotopes that have potential uses as theranostic agents.

As mentioned above, most of these promising radionuclides are made at a few regional facilities. These include the Isotope Production Facility at LANL (IPF-LANL) and the Brookhaven Linac Isotope Production facility at BNL (BLIP-BNL) in the United States, the TRIUMF laboratory in Canada, and iThemba LABS in South Africa (108, 116). In a compelling example of the synergy between fundamental nuclear research and societal benefit, IPF-LANL and BLIP-BNL operate symbiotically with LANSCE at LANL and the Relativistic Heavy Ion Collider at BNL. This dual-use approach provides an inexpensive, reliable source of radionuclides needed for the development of new drugs through a public–private partnership.

All of these regional sources utilize high-energy (≥100 MeV) (p, x) reactions on stable targets. In addition to producing the radionuclides of interest, these reactions create a large flux of

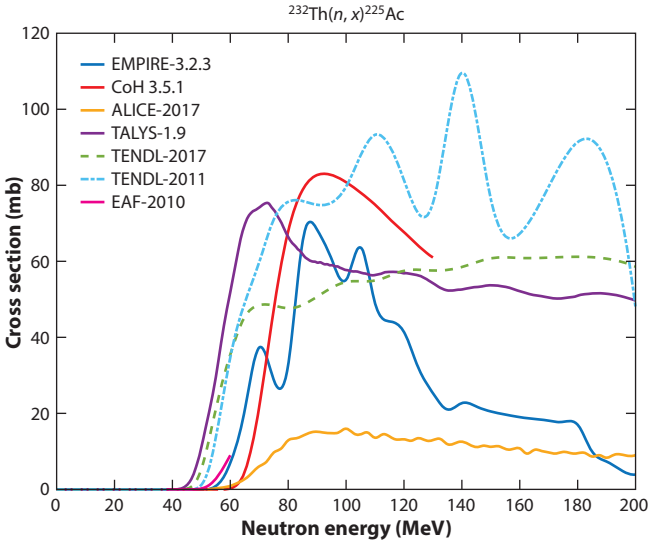


Figure 7

Predictions of several major reaction model codes for the (n, x) reactions leading to the production of ^{225}Ac. Differences of up to an order of magnitude are commonly observed.

secondary spallation protons and neutrons that can in turn initiate nuclear reactions that provide either a useful alternative source of valuable radionuclides or a troublesome chemically identical contaminant. The secondary neutron flux is particularly important since the lack of electronic stopping for neutrons makes them advantageous for radionuclide production (117). The energy spectrum of secondary neutrons from IPF-LANL was recently measured using spectral unfolding (118) and for thick-target deuteron breakup on beryllium at the LBNL 88-Inch Cyclotron (56) via a secondary time-of-flight technique. A number of therapeutic radionuclides can potentially be produced via secondary neutron-induced (n, p) reactions. Examples of possible targets include ^{32}S, ^{47}Ti, ^{nat}Zn (for the production of ^{64,67}Cu), ^{105}Pd, ^{149}Sm, ^{175}Lu, and ^{177}Hf. In addition, the production of α-particle-emitting radionuclides such as ^{225}Ac, ^{223}Ra, and ^{227}Th using neutrons has yet to be explored.

Unfortunately, accurate modeling of these high-energy reactions is notoriously difficult. **Figure 7** shows a comparison between cross sections for the production of ^{225}Ac from (n, x) reactions on ^{232}Th calculated with the reaction codes typically used to predict isotope production. These include EMPIRE-3.2.3 (119), CoH 3.5.1 (15), ALICE-2017 (120), and the TENDL-2011 and TENDL-2017 libraries produced using TALYS (121), as well as the most recent version of the code (TALYS 1.9) (16) and the European Activation File (EAF-2010) (122). Differences on the order of 50–100% are often observed.

These large uncertainties in reaction modeling mean that direct measurements of the reaction cross sections themselves are needed in order to design targets capable of efficiently producing low-contamination radionuclides. Fortunately, most of the reactions of interest take place on stable targets and are therefore experimentally accessible.

The preferred method for measuring these cross sections involves foil activation, in which one or more well-characterized thin foils are irradiated and the residual radioactivities are quantified through off-line α, β, γ, or electron spectroscopy (123, 124). The uncertainty in the deduced cross section depends strongly on accurate characterization of the target material (which is often

challenging to produce), the spectroscopic assaying techniques used, and precise determination of the incident particle flux. Access to well-characterized nuclear data can be useful in reducing the first two sources of uncertainty.

The flux is typically determined using a well-characterized monitor reaction on a coirradiated foil that leads to the production of an easily quantified, long-lived residual nucleus. However, for both charged-particle and neutron-induced reactions, there are relatively few monitor reactions that can be applied in the low- to intermediate-energy regimes of 30–70 MeV. The situation is worse above 70 MeV, where monitor reactions free from the potential influence of secondary neutron contributions are even harder to come by. Knowledge of these cross sections as a function of the incident particle energy is essential to isotope production, with recent efforts focused on developing ^{93}Nb$(p, 4n)^{90}$Mo [$t_{1/2} = 5.56(9)$ h] as a monitor reaction suitable for use at regional isotope production facilities (123, 125).

5. NUCLEAR ENERGY

Reliable modeling and simulations of nuclear energy systems are essential to the design, licensing, and operation of nuclear power stations as well as the fuel cycle facilities for enrichment and material processing and fuel fabrication, transportation, and storage. Additional predictive calculations are required for spent-fuel characterization for storage, transportation and disposal, as well as hardware activation, radiation shielding, and personnel protection. Nuclear data and their uncertainties are required for a wide range of calculations, including:

1. reactor core and fuel design,
2. safety parameter assessment,
3. criticality safety,
4. shielding,
5. material damage in structures,
6. decay heat at reactor shutdown,
7. decay heat in storage and transportation,
8. mass flow in the fuel cycle, and
9. material safeguards.

The nuclear physics of these systems are inferred from (n, x) cross sections, fission spectra, neutron multiplicity, fission product yields, decay constants, branching ratios, γ yields, delayed neutron precursors, and more. For light-water reactors (LWRs), nuclear data have been used, tested, and often tuned over many decades to provide acceptable results, and there is an abundance of documented benchmark quality experiments and plant operation data for the validation of common scenarios. However, for new applications such as fuels and materials in advanced reactors, accident-tolerant fuels, high-burnup fuel, and spent-fuel repository scenarios, it is necessary to improve nuclear data and assess the availability of benchmark experiments that could be applied for testing and validation.

An artifact of the nuclear data tuning process is the reduction in confidence due to the compensating errors referred to in Section 2.2, above. Where fission and capture reactions are modified to ensure that the integral value of k_{eff} is maintained, little confidence remains in the independent use of the fission cross section itself, which is essential to the prediction of reactor power distributions. These power distributions are crucial for the prediction of peak fuel temperatures during a reactor transient to ensure that the fuel does not exceed thermal limits that are set to ensure fuel integrity. Reduced confidence in the fission cross section can have real consequences for the allowed power level and operating regimes for these systems.

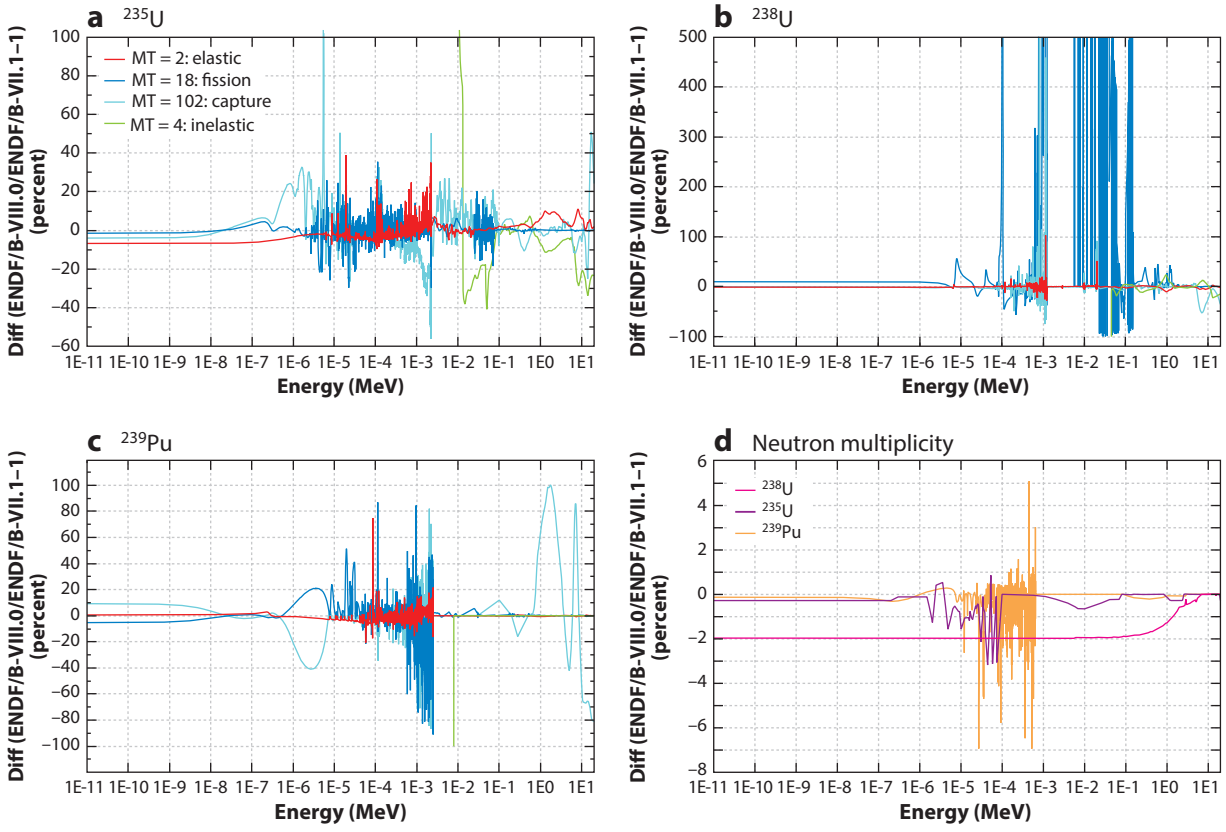


Figure 8

SG46 findings on changes in cross sections and prompt fission nubars from ENDF/B-VII.1 to ENDF/B-VIII.0 of key nuclides. Abbreviations: ENDF, Evaluated Nuclear Data File; SG, Subgroup.

A study conducted by SG46 (126) found that many individual reactions were modified by several percent from ENDF/B-VII.1 to ENDF/B-VIII.0. **Figure 8** shows some examples.

Many of these advanced systems also require so-called high-assay low enriched uranium (HA-LEU), which has a ^{235}U enrichment >5% and <20% by mass; the current fleet of LWRs utilize enrichments of 5% or less. With few benchmark experiments in the HA-LEU enrichment range, the interplay of the cross sections for multiple nuclei becomes important, especially for LEU experiments in which ^{1}H, ^{16}O, ^{235}U, ^{238}U, and others are applied simultaneously in the calculation of the integral k_{eff} response. Similar to the Chadwick et al. (44) (see **Figure 2**) and Bauge et al. (45) studies of Jezebel critical assembly, a team at ORNL performed an investigation of a hypothetical transportation package with 20% enriched ^{235}U in the form of UF_6 using the ENDF/B-VII.1 and ENDF/B-VIII.0 libraries by only exchanging the CIELO nuclides. A swing in k_{eff} of 450 pcm was observed. Future investigation and validation are required to gain confidence in either calculation.

Many specific nuclear data needs have been identified that are important for design and operating analysis for these advanced systems, including the following.

- Improved knowledge of the ^{238}U(n, n') cross section (discussed in Section 2.2, above) is needed to improve neutron transport calculations. ^{238}U(n, n') is a limiting factor in the design of fast spectrum reactors such as those with metallic cores or chloride salts. The 40%

uncertainty in this cross section in ENDF/B-VII.1 propagates to an uncertainty of 1.5% Δk in the criticality state of a sodium fast reactor (i.e., at the 2σ level, k_{eff} could range from 0.97 to 1.03 on the basis of this uncertainty prediction) (127).

- Improved knowledge of the ^{35}Cl(n, p) cross section is needed to better predict criticality in molten salt systems. Changes from the 2006 release of ENDF/B-VII.0 to the 2011 release of ENDF/B-VII.1 are causing a 2–3% Δk change in reactivity in molten salt reactors. This data change will also affect repository analysis in salts, and there are no benchmark experiments available for validation. Recent results from activation using a deuteron–deuteron neutron source (117) indicate both resonant behavior in this cross section and a smaller average value for incident neutron energies between 2.2 and 2.7 MeV (128).
- Graphite data for high-temperature reactors, as well as the Transient Reactor Test Facility (TREAT), have realized a number of updates that cause dramatic changes in criticality. Updates from ENDF/B-VII.0 to ENDF/B-VII.1 have led to a 1% Δk change in reactivity for the High Temperature Engineering Test Reactor (HTTR), leading to calculations that better match experimental values. Proposed updates to the graphite thermal scattering data for the ENDF/B-VIII.0 release have been demonstrated to cause a 2% Δk change in reactivity for the TREAT M8CAL benchmark (129).
- A direct measurement of the thermal neutron scattering law in molten salts, such as FLiBe, is needed to improve transport calculations. There are no cross-section data that represent the effect of the molecular bonds, leading to a computational bias of unknown magnitude in all reactor design calculations. The molecular bonds in water versus polyethylene can lead to a computation bias of 1–2% Δk. In addition, this effect is temperature dependent and is not well studied or understood. A recently developed benchmark experiment evaluation of the Molten Salt Reactor Experiment, which consisted of uranium fuel FLiBe molten salt flowing through graphite channels, demonstrates a bias of more than 1% Δk from the experimental value (130), with other presented results showing biases greater than 3% Δk.
- (n, x) reactions on heavy actinides, which will build up in molten salt reactors and high-burnup fuel, are poorly quantified and benchmark data are rare, so these effects are almost completely unknown.
- Uncertainties provided with ENDF in the form of cross-section covariance data are often not mature and may or may not represent the true uncertainty in the nuclear data. For the recent ENDF/B-VIII.0 release, the README file provided by the NNDC provides a disclaimer regarding the use of covariance data without applying a sensitivity/uncertainty approach to adjust the uncertainties for application-specific analysis using relevant benchmark experiments. This specialized process is generally not available to users and presents many complications even for experts in the field, leaving a large gap between the data provided by nuclear data evaluators and those useful for application in safety and design calculations.
- Nuclear data are gathered from many different sources—not only ENDF but also the ENSDF as well as the JEFF library—which often have differing representations of the same data. The process of sorting through the data to correct inconsistencies and benchmark the consolidated library against benchmark experiments is complex.
- Uncertainty data are not available or are incomplete for fission product yields, decay constants, and angular distributions, making it difficult to have high confidence in predicting the performance of advanced systems. In the case of fission product yields, there is not even a format for covariance data in the ENDF format, harkening back to the admonition given in Section 1.2.

The generation and distribution of nuclear data, as well as the benchmarking of the performance of codes and data, are a global effort. In the United States, nuclear data are generated by teams supported by DOE-NP and DOE-NNSA. These teams gather at the Cross Section Evaluation Working Group meetings to create and test the ENDF libraries prior to distribution. They advocate for improved performance for their own applications, but because nuclear energy interests are not well represented at the review meetings, the impact of updates to nuclear data and associated uncertainty information on nuclear energy applications is often overlooked. A focus on data needed for modeling nuclear energy systems would lend confidence and support to the development of new reactors, providing a desperately needed low-carbon energy source for society.

6. FUTURE DIRECTIONS IN NUCLEAR DATA

In recent years, there has been growing recognition of the need to develop a plan that addresses nuclear data needs, from measurement through compilation and evaluation, in an organized, multiagency fashion. This process started in July 2014 with the first review of the USNDP, which is administered through the DOE-NP, in nearly two decades. The review stated that experimental efforts to address gaps in nuclear data are within the scope of the USNDP program, resulting in DOE-NP requesting that NDNCA be organized to compile a list of nuclear data needed for nuclear energy, national security, and isotope production. NDNCA was held in Berkeley in May 2015, and a white paper delineating these needs was released 5 months later (107). The white paper provides a long and comprehensive list of nuclear data needs, both cross-cutting and application specific, that has served as the basis for multiple subsequent roadmapping efforts.

Following NDNCA, Dr. Catherine Romano, working with the DOE-NP and the DOE-NNSA office on counterproliferation research and development (NA-22), organized a grassroots Nuclear Data Working Group (NDWG) composed of subject matter experts selected by program managers from DOE-NP, DOE–Nuclear Energy, the Isotopes Program, the Department of Homeland Security, and several offices within DOE-NNSA to identify a roadmap to address the most important and cross-cutting nuclear data needs, including all steps in the nuclear data pipeline from measurement through compilation, evaluation, and processing. The NDWG presented its results to participating program managers as well as other interested agencies during the Nuclear Data Exchange Meeting (NDEM) in April 2016. The recommendations included:

1. infrastructure modernization,
2. expanded covariance data,
3. inelastic scattering on ^{235}U, ^{238}U, and ^{239}Pu,
4. γ emission data,
5. fission yields, and
6. renewed nuclear data target production capabilities.

The NDEM provided a venue for valuable discussions between the nuclear data community and program managers and the tasks required to provide evaluated nuclear data to the database.

In response to the plan presented by the NDWG, the program managers formed the NDIAWG to pursue a collaborative effort to address several of these high-profile nuclear data needs in a cooperative fashion. The NDIAWG issued two Funding Opportunity Announcements in 2017 and 2018 that led to a total of 11 new projects covering topics from improving fission fragment yield and γ-ray decay data to measuring specific high-priority cross sections for isotope production and neutron transport for nuclear reactors.

A key element of the NDIAWG process is ensuring that the efforts to address these nuclear data needs result in user impact. This goal is accomplished by reviewing the progress made via

recurring interagency workshops. These workshops initially served as roadmapping discussions in which the nuclear data users explained their needs and the nuclear data community agreed upon a best path forward. Now that several projects are under way, the workshops also serve as a project review. The first of these was the Nuclear Data Roadmapping Enhancement Workshop (NDREW), held in Washington, DC, in January 2018, which focused on nuclear data needs relevant to nonproliferation (131) and resulted in a nuclear data roadmap for DOE-NNSA, Defense Nuclear Nonproliferation Research and Development. The second, the Workshop on Applied Nuclear Data Activities (WANDA), took place in January 2019 in Washington, DC, and covered topics relevant to nuclear energy, isotope production, safeguards, and the medical community.

The NDIAWG process reflects a growing recognition that improved nuclear data are essential to a wide range of human endeavors that transcend the needs of any one society or governmental mission. Its goal is the development of a new, multiagency national plan to address nuclear data needs in a coherent way. This plan, which is being developed in partnership with international collaborators at the IAEA and the OECD-NEA, represents a new paradigm for collaborative research.

7. CONCLUSION: RETAINING AND TRAINING A SKILLED NUCLEAR DATA WORKFORCE

A robust nuclear data process is needed to provide the information required for many applications ranging from nonproliferation to nuclear energy to isotope production. However, it takes many years of post-PhD experience to train nuclear data evaluators, and only a short break in support to lose this workforce to other pursuits. It is therefore critical for the many programs that rely on nuclear data to support individuals engaged in maintaining all functions that make up the nuclear data pipeline, from measuring and modeling through compilation, evaluation, validation, and processing. As more and more of the research in the United States moves to a project-driven model, it is imperative that sponsors coordinate their efforts to ensure that there is no loss of talent in the field. The NDIAWG, discussed above, is a significant step to help address these issues in a systematic fashion.

Furthermore, nuclear data evaluation is an aging field, with the mean age of an evaluator in the United States being well over 50. It is essential that the next generation of nuclear data evaluators be recruited and trained while the current set of experienced individuals are still active in the field. This is particularly challenging since there is, per se, no graduate education program in nuclear data evaluation, and the vast majority of the centers with evaluation expertise are not associated with degree-granting institutions.

Perhaps one of the most endangered species in nuclear data is the experimental nuclear data evaluator. This is neither an experimentalist doing a new, better experiment nor a data library creator making an ENDF-like library, but rather a person who knows which of the more than 22,000 measurements in the EXFOR library are to be taken seriously, discarded, or normalized to newer standards. This sort of hybrid individual would be capable of providing not only good mean values but also the well-quantified uncertainties (including covariances) that are needed for most applications. A concerted effort on the part of both academia and the international research community is needed to ensure that talented early-career individuals with the right combination of interest and inclination are afforded the opportunity to be trained by an "international village" of evaluators and application subject matter experts.

The dramatic increase in computational capabilities over the past several decades has allowed for the sort of detailed calculations needed to fulfill the promise of many applications, provided that the correct evaluated data are available. This would also open the door for application of

machine learning techniques for the evaluation of both nuclear structure and reaction data. Recent efforts in the United States offer the promise of a national nuclear data plan to provide the needed data. If these efforts are maintained, they could help usher in a world with a new sense of safety from enhanced nonproliferation capabilities; a new source of safe, carbon-neutral energy; and a new class of radiopharmaceuticals to diagnose and treat illness.

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