# A Survey of Nuclear Data Deficiencies Affecting Nuclear Non-Proliferation

Rian Bahran[1], Stephen Croft[2], Jesson Hutchinson[1], Morag Smith[1] and Avneet Sood[1]

[1]Los Alamos National Laboratory
X-Computational Physics Division, Monte Carlo Methods, Codes and Applications Group (XCP-3)
Nuclear Engineering and Non-Proliferation Division, Advanced Nuclear Technology Group (NEN-2)
Los Alamos, New Mexico 87545

[2]Oak Ridge National Laboratory
Safeguards & Security Technology Group
Oak Ridge, Tennessee 37831

## ABSTRACT

A general survey was administered across various academic and research institutions in order to identify the most significant nuclear data deficiencies affecting applications in nuclear non-proliferation. In recent years, such deficiencies have become increasingly apparent in the nuclear non-proliferation community where the accurate interpretation of physical applied nuclear measurements rests on the availability of nuclear data which are fit for this purpose. Well-known examples include those in key applications in non-proliferation such as special nuclear material characterization through neutron multiplicity measurements, spent fuel assay techniques, and $\gamma$-ray spectroscopy for isotope identification among others. The preliminary results of the survey reported in this paper will serve as initial documentation of some of the informal - but informed - backroom discussions on both recurring and non-common nuclear data discrepancies as they relate to non-proliferation. This would help future differential/integral measurements and evaluation campaigns in finding additional justification for targeting specific reactions and nuclides to help improve the quality of existing nuclear data libraries relative to these applications.

## INTRODUCTION

High fidelity nuclear data are important in their application to radiation transport calculations used in the design and/or analysis of reactor fuel cycles, nuclear non-proliferation, fusion systems, astrophysical systems, safeguards, criticality safety, stockpile stewardship, radiation protection, shielding calculations, particle physics, medical applications and fundamental nuclear science research. The amount of currently available application-ready nuclear datasets is vast and covers hundreds of nuclides and spans large energy ranges. Much of this data is stored in evaluated nuclear data libraries. Evaluating nuclear data is a standalone science in itself that is loosely based on the statistical accuracy and perceived quality of a differential measurement and how it compares to existing theoretical nuclear reaction physics models and integral benchmark results. Some of the main general purpose nuclear data libraries that are widely employed and are publicly available include the US Evaluated Nuclear Data File or ENDF [1], the European Joint Evaluated Fission and Fusion File or JEFF [2] and Japanese Evaluated Nuclear Data File or JENDL [3]. The ENDF library represents the evaluation efforts of a group of experts in industry, academia, and government laboratories in North America known as the Cross Section Evaluation Working Group (CSWEG). In general, all of the major nuclear data libraries exhibit differences among their evaluations. These inconsistencies can vary in magnitude and are widespread across many isotopes. This is because although undeniably an astonishing achievement, the multitude of the existing nuclear reaction theories are limited as they cannot reliably predict exact values for nuclear parameters needed for practical applications to sufficient accuracy. Instead, high-fidelity experimental data are needed in order to provide an accurate representation of the actual underlying physical process. To satisfy the needs of the nuclear community, experimental nuclear data activity started picking up in the 1930s at various national and international laboratories, many of which continue to contribute measurements and evaluations to the nuclear data community. Experimental nuclear data activity has experienced widespread revival in the past few decades [4], partially in response to increased reliance on computer simulations which combine nuclear data with radiation transport methods to mimic physical behavior in computer space. In the past, the design of nuclear systems relied heavily on data from actual prototypes using actual special nuclear materials. Such benchmarks are both financially and administrable burdensome and can take a considerable amount of time to construct. On the other hand, the time it takes to simulate a nuclear system on a computer depends on the complexity of the design, the simplification assumptions, the algorithms implemented in the code, and the performance of the hardware. When these factors are optimized and as high-performance computing continues to improve exponentially, discretization errors and stochastic uncertainties in computational methods can be minimized. Thus, with increased reliance on cheap computer simulations (e.g. utility of continuous energy radiation transport Monte Carlo codes), the quality of the largely empirically-based nuclear data becomes the limiting factor on the accuracy of these methods. Today, the non-proliferation technical community continues to be wholly reliant on physical standards and evaluations of measured nuclear data quantities that include neutron-induced reaction, charged particle-induced reactions, photon-induced fission and others.

## NUCLEAR DATA FOR NON-PROLIFERATION

Generally speaking, many technical applications in the nuclear non-proliferation subspace that involve passive, active methods and emerging (mainly advanced active) methods are reliant on evaluations of measured nuclear data quantities at some esoteric level. Some examples include isotopic determination for nuclear safeguards which requires data on half-lives, relative emission probabilities (branching ratios), and accurate energy differences. Passive neutron multiplicity counting for special nuclear material characterization is limited by the accuracy of certain data on multiplicity distributions, energy spectra, isotopic correlations asymmetric neutron emission, spectrum-multiplicity correlation, and fission n-$\gamma$ correlations and ($\alpha$,n) yields. The need for good nuclear (and atomic) data is also apparent in applications for arms control and emergency response where some analysis codes get very good results by essentially hard-wiring x-ray lines that are clearly present in measurements of items even if the underlying reference data files do not contain them. For neutron detector characterization $^{252}$Cf has become a ubiquitous surrogate for Pu and opportunities exist to improve its database. There is also interest in improving nuclear data for in Cm, $^{244}$Cm and $^{248}$Cm. This stems both from their presence in the fuel cycle but and their potential application as a longer lived alternative to $^{252}$Cf. Other examples are given in the results section of this paper.

## SURVEY FEEDBACK MECHANISM

In order to help provide direction to support future high fidelity nuclear data measurements capabilities for non-proliferation applications and to help improve the quality of existing nuclear data libraries relative to these applications, a survey was developed. This survey aims to provide initial quantification of potential areas of improvement relative to non-proliferation based on community-wide reported issues that have arisen when comparing measurements to simulations. Such feedback will help future measurements and evaluation campaigns in finding additional justification for targeting specific reactions and nuclides to help improve the quality of existing nuclear data libraries relative to these applications. This type of feedback validation as illustrated in Figure 1 is commonplace in the nuclear data community.

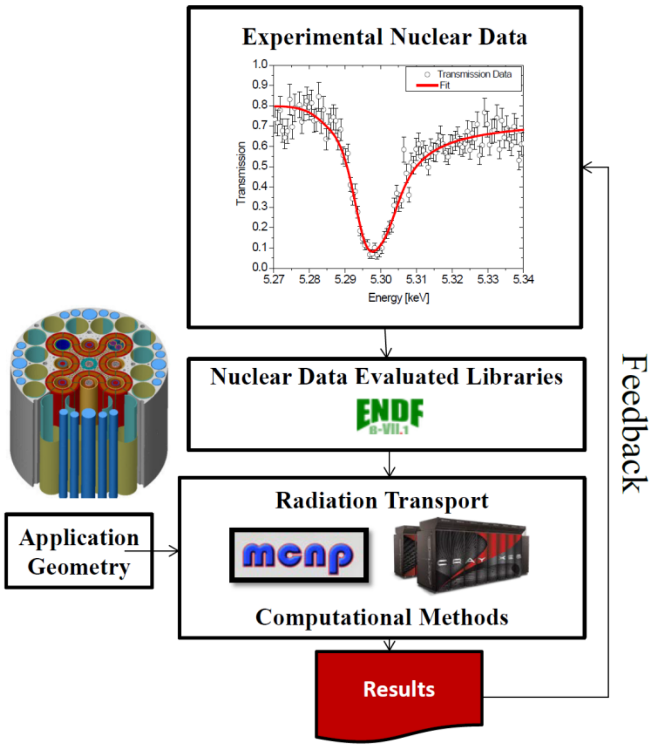


**Fig. 1.** When computational results are compared to experimental data, a feedback opportunity is presented that can help guide nuclear data measurements and evaluation campaigns.

Although the nuclear data evaluation process is expected to be application independent, most of the available feedback has historically been driven by other larger application-specific communities. For example the International Handbook of Evaluated Criticality Safety Benchmark Experiments [5] contains over 500 evaluations with benchmark specifications for almost 5000 critical, near-critical, or subcritical configurations intended for use by criticality safety engineers. A similar database or mechanism for non-proliferation feedback does not currently exist.

The survey results reported in this paper are intended to provide initial documentation of the compilation of some of the informal - but informed - backroom discussions on both recurring and non-common nuclear data discrepancies as they relate to non-proliferation. Previous work that was similar in scope include the results of a 2005 report [6] from the United States Nuclear Data Program Task Force on Nuclear Data for Homeland Security which performed "an informal survey of homeland security technical programs at LLNL and LANL to provide a needs list for new nuclear data and new database capabilities" . In addition, Santi and his colleagues provided a similar assessment in a 2007 paper [7] on the role of nuclear data in advanced safeguards.

This work also overlaps with that of various international initiatives that are carrying on parallel concerted efforts aimed at identifying similar deficiencies and quantifying uncertainties, correlations and sensitivities for nuclear data. A recent overview of the current knowledge of nuclear fission data and its remaining uncertainties For example, the CIELO (Collaborative International Evaluated Library Organization) collaboration is working to identify, document and reconcile discrepancies among existing evaluated data libraries for (initially) a small number of their highest-priority isotopes which include several that are equally important to the non-proliferation community [8]. A recent overview of the current status of knowledge regarding expectedly high-priority nuclear fission data and its uncertainties can be found in Reference [9].

## SURVEY INSTRUMENT DEVELOPMENT

A self-administered written survey instrument was developed with the objective of effectively assessing deficiencies in nuclear data as they relate to nuclear non-proliferation applications. The survey was carefully crafted based on two metrics that were chosen in order to ensure survey content validity i.e. the extent to which the survey appropriately assesses the characteristics it intends to measure. The first metric was simplicity which was established by requesting open-ended responses for the following categories: type of radiation, nuclear data quantity, affected materials/isotopes, approximate energy, non-proliferation application and available references. Open-ended survey response options help maximize what can be learned while avoiding many of the typical errors or oversights. The second metric was the adoption of international nuclear data norms in the example answers provided. In addition, pilot testing of the survey questions for reliability was conducted in-house to ensure that survey instructions were clearly defined and easy to understand, and that the response categories were appropriate. The survey target demographic was chosen to encompass nuclear engineering faculty and staff scientists in major universities and laboratories who are working on nuclear non-proliferation projects that have a simulation component. Survey distribution was primarily delivered via electronic mail but in several cases was delivered in person. Geographically diverse responses were obtained from 30+ experts from 14 institutions.


**Fig. 2.** Responses were obtained from 30+ experts from 14 institutions including 6 national laboratories and 8 universities.

Although due diligence was taken in identifying this target demographic through peer-identification and literature reviews, it was still difficult to obtain a fully representative collection of survey responses with minimal non-response bias. Examples of non-response bias in this context that arose include:

- Obtaining many responses from those who are already interested in this specific topic.
- Obtaining redundant responses from individuals that are part of the same concerted research efforts.
- Overrepresentation of responses from the US national labs (specifically author affiliations) as compared to universities or international entities.

## SURVEY RESULTS OVERVIEW

The most widely reported general issues based on survey responses were related to correlated particle emissions from fissile nuclear material (neutron and gamma-ray multiplicity), fission product data, neutron total and partial cross sections of various isotopes and (α,n) yields from light elements. The most widely reported specific deficiencies were those related to nubar for $^{239}$Pu in the fast energy range, and data associated with fission products (yield, energy spectrum, half-lives, emission, branching ratios etc…). Other recurring issues included photonuclear data, and S(α,β) datasets. Standalone issues that were reported consisted of deficiencies in the electronic excitation cross section libraries, critical mass values and specific heat for nuclear calorimetry.  A quantitative summary of the results is shown graphically in Figure 3. A summarized list of bulleted responses is provided below. The full list of raw survey responses and additional references is provided in Appendix I.

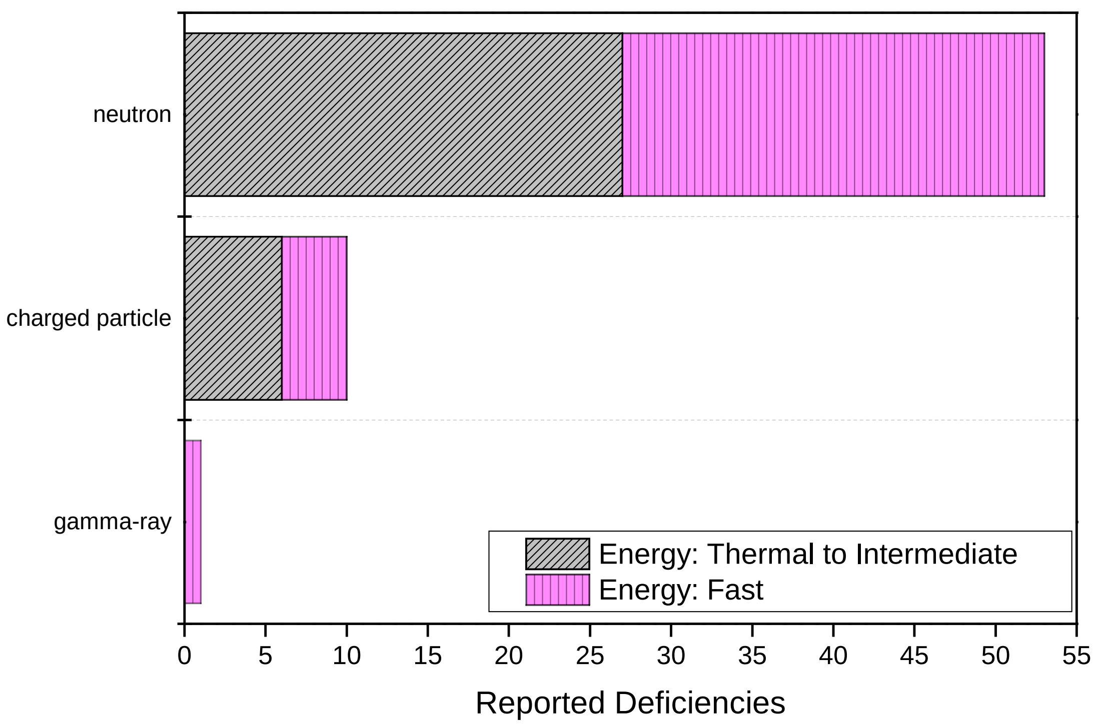


**Fig. 3.** A global overview of the survey responses that were obtained from the self-administered survey. Most of the responses were related to nuclear data from fast incident neutrons as expected.

**RECURRING DEFICIENCIES**

1. Correlated Particle Emissions: Neutron and Gamma-ray Multiplicity Data
    - Materials/Isotope: Pu isotopes
        - nubar (Fast), $P_{Nu}$ (Fast), Probability distribution of prompt gamma-rays
        - Inverse model e.g. affects counting rate in multiplicity measurements for special nuclear material characterization
        - Current ENDF evaluations unable to reproduce WGPu multiplicity measurements
    - Material/Isotopes: Cf
        - $P_{Nu}$ (Fast)
        - Improve “de facto standard”.
    - Material/Isotopes: Am, Cm (Spontaneous Fission)
        - Active and passive NDA for safeguarding fuel
2. Fission Product Data
    - Material/Isotope**:** All fission products
        - Fission yields, half-lives, branching ratios, capture emission spectra, peak energies, branching ratios, cross sections
        - Material characterization via neutron spectroscopy
        - Next generation safeguards techniques with active neutron interrogation
        - Post-detonation nuclear forensics-based fallout analysis (data required to decay-correct fission to t=0)
3. Neutron Interaction Probabilities: Total and Partial Neutron Cross Section Data
    - Material/Isotope: $^{239}$Pu
        - Total (n,total) and partial cross sections - Thermal to Fast
        - Inverse problem, neutron resonance densitometry, other applications
    - Material/Isotope: Short-lived fission fragments
        - Total (n,total) and partial cross sections - Thermal to Fast
        - Development of better physics models calculation-based nuclear forensic tools
    - Material/Isotope: Hundreds not available in ENDF (TENDL has ~2600 isotopes)
        - Total (n,total) and partial cross sections - Thermal to Fast
        - Correct Source term e.g. for burnup calculations
    - Material/Isotope: Non-common materials in explosives detection and containers/shielding.
        - Elastic scattering cross section (n,n’) - Fast
    - Material/Isotope: Np and other actinides
        - Capture cross section (n,g) - Thermal to Fast
        - Thermal to Fast
        - Safeguards e.g. accurate prediction of production or inventory
    - Material/Isotops: Cd
        - Capture cross section (n,g) - Thermal to Fast
        - Safeguards instruments that use Cd (e.g. to get flux ratios, PNAR, SINRD)
    - Material/Isotope: U, Np, Pu, Am and Cm
        - Fission cross section (n,f) - Thermal to Fast
        - Various e.g. SNM characterization through multiplicity measurements

  - Material/Isotope: $^{238}U$
    - — Sub threshold fission
    - — Used in fast fission detectors e.g. LSDS for spent fuel assay
4. <u>Charged Particle Data: (α,n)</u>
  - Material/Isotope: Fluorine and other Low-Z elements: Li, N, B, C
    - — (α,n) yields - Fast
    - — Passive neutron measurements of fluorine compounds ($UF_6$, $UO_2F_2$)
    - — Pu oxide characterization at reprocessing facilities e.g. MOX
5. <u>Photonuclear Data</u>
  - Material/Isotope: U and Pu isotopes
    - — Photofission data and delayed gamma-rays after fission
    - — Photon-induced multiplicities , energy spectra, energy-angle correlations for active interrogation e.g. photon induced fission of SNM material/cargo
6. <u>S(α,β) datasets for Various Applications</u>
  - Material/Isotopes: Light elements

**NON-RECURRING DEFICIENCIES**

1. <u>Electronic excitation cross section, ($e^-$,gamma)</u>
  - Material/Isotope: $^{235}U$, $^{238}U$, $^{239}Pu$, $^{240}Pu$
    - — 0.5 to 4 MeV
    - — Potential method to produce monoenergetic photons from Nuclear Resonance Fluorescence
2. <u>Critical mass values</u>
  - Material/Isotopes: Np
    - — Determining non-proliferation activities since current values reported in the literature/simulation results vary widely
3. <u>Specific Heat for Nuclear Calorimetry</u>

## CONCLUSIONS

Given the increasing role of modeling and simulation in the non-proliferation technical subspace, it is crucial that the underlying data be accurate. Weaknesses in that data undercut the work and the conclusions based on that work including those that feed into high-stakes decision-making in the policy world. Based on the results of a community-wide self-administered survey, it can be concluded that the full list of nuclear data deficiencies for non-proliferation applications is quite large as expected. The most widely reported issues based on survey responses were related to correlated particle emissions, fission product data, neutron cross sections and alpha particle data. Other recurring issues included photonuclear data and S(α,β) datasets.

Prioritization of deficiencies is still needed regarding what would have a substantive effect. This can be based on sensitivity studies for impact determination and integral/differential measurement feasibility studies for predicting an achievable level of improvement. In addition, a better assessment and possible reduction of the uncertainty/covariance attributes of existing data is extremely important. Leveraging the existing capabilities from community-wide parallel efforts related to reconciling discrepancies, quantifying uncertainties, correlations and sensitivities for measured and evaluated nuclear data is key.

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Organizations: LANL, ORNL, SNL, PNNL, INL, Y-12, University of Michigan, NCSU, RPI, Penn State, Texas A&M, University of Tennessee, University of New Mexico, and Oregon State University.

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[7] P. Santi, D. Vo et al. "The Role of Nuclear Data in Advanced Safeguards" Proc. Of Global 2007: Advanced Nuclear Fuel Cycles and Systems, Boise, ID pg. 1670-1678 (2007)

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[9] P. Talou, T. Kawano et al. "Uncertainties in Nuclear Fission Data" To be published in the Special Issue of J. Phys. G: Nuclear and Particle Physics "Enhancing the interaction between nuclear experiment and theory through information and statistics".

**APPENDIX I: SURVEY DATA**

| **Nuclear Data Deficiency** | **Material or Isotope** | **Energy Range** | **Methodology/ Nonproliferation Application** | **Source**<br><br>**Relevant References** |
|---|---|---|---|---|
| Nubar<br><br>(n,gamma)<br><br>(n,fission)<br><br>Prompt fission neutron spectra<br><br>$P_{Nu}$ | $^{239}$Pu<br>$^{235}$U | Fast | Neutron multiplication measurements/ SNM characterization | North Carolina State University<br><br>Idaho National Laboratory<br><br>University of New Mexico<br><br>Oak Ridge National Laboratory<br><br>Los Alamos national Laboratory<br><br>University of Tennessee |
| P. Talou et al. “Uncertainties in Nuclear Fission Data” To be published in the Special Issue of J. Phys. G: Nuclear and Particle Physics (2014)<br>R. Evans et al. "Sensitivity Analysis and Data Assimilation in A Subcritical Plutonium Metal Benchmark" Journal of Nuclear Science and Engineering Volume 176 Number 3 pp. 325-338 (2014)<br>E. C. Miller et al. "Computational Evaluation of Neutron Multiplicity Measurements of Polyethylene-Reflected Plutonium Metal" Nuclear Science and Engineering Volume 176 Number 2 pp. 167-185 (2014)<br>S. Boldin et al. “Simulations of Multiplicity Distributions with Perturbations to Nuclear Data” Trans. Amer. Nucl. Soc., Washington DC (2013)<br>S. Noda et al. “Prompt fission neutron spectra from fission induced by 1 to 8 MeV neutrons on U-235 and Pu-239 using the double TOF technique” Phys Rev C. 2011 83 | | | | |
| Nubar, $P_{Nu}$ | $^{240}$Pu | Fast | Neutron Multiplicity measurements | Los Alamos National Laboratory<br><br>Idaho National Laboratory |
| n<br><br>half-life | $^{241}$Pu | N/A | Feeds Am-241 in-growth. | Oak Ridge National Laboratory |
| n<br><br>2$^{nd}$ 3$^{rd}$ moments | $^{241}$Pu | Fast | Neutron Multiplicity measurements for Spent fuel | Los Alamos National Laboratory<br><br>Idaho National Laboratory |
| n<br><br>(n,g) | Cd | Thermal to Fast | Safeguards instruments that use Cd to get flux ratios (e.g. PNAR, SINRD) | Los Alamos National Laboratory |
| n<br><br>(n,total), (n,f),(n,g), (n,inl) | Short-lived fission fragmenets (i.e. A=143 istopes such as Xe, CS, La, Ba, A=90 isotopes such as Br, Kr, Rb) | Thermal to Fast | Accurate prediction of inventory in used Nuclear Fuel assemblies, Development of better physics models calculation-based nuclear forensic tools, Neutron Resonance Transmission Analysis | University of Tennessee<br><br>Idaho National Laboratory<br><br>Texas A&M University |
| n<br><br>cross sections | Several thousands isotopes created during burnup that do not exist in ENDF (like TALLYS) | Thermal to Fast | Correct Source term for burnup calculation | Los Alamos National Laboratory |

| | | | | |
|---|---|---|---|---|
| n<br><br>(n,g) | Am, Cm isotopes $^{237,238,239}$Np | Thermal | Determining spent fuel isotopics (e.g. $^{238}$Pu and $^{244}$Cm production) | Los Alamos National Laboratory |
| n<br><br>decay<br><br>n, fission<br><br>inelastic scattering<br><br>Critical mass | Np | Thermal - Fast | Determining spent fuel isotopics<br><br>Weapons-usable material production assessment | Los Alamos National Laboratory<br><br>Oak Ridge National Laboratory<br><br>Idaho National Laboratory |
| I. Gauld et al. “Integral Testing of the $^{239}$Np Capture Cross Section” CSWEG 2013 | | | | |
| n<br><br>fission product yields and covariances<br><br>energy spectrum<br><br>half-lives<br><br>branching ratios | All actinides and Fission Fragments | Fast | Material characterization via neutron spectroscopy<br><br>Spent fuel measurements<br><br>Post detonation nuclear forensics-based fallout analysis (required to decay-correct fission to t=0)<br><br>Next generation safeguards techniques using active neutron interrogation | University of Michigan<br><br>Idaho National Laboratory<br><br>Pacific Northwest National Laboratory<br><br>Oak Ridge National Laboratory<br><br>Oregon State University<br><br>University of New Mexico |
| D. Rodriguez et al. “Measurement and analysis of gamma-rays emitted from spent nuclear fuel above 3 MeV” Applied Radiation and Isotopes 82 pp. 181-187 (2013)<br>R. Marrs et al. “Fission-product gamma-ray line pairs sensitive to fissile material and neutron energy” Nuclear Instruments and Methods in Physics Research A 592, 463 (2008) | | | | |
| n<br><br>correlated prompt emissions | All actinides | Thermal to Fast | Correlation measurements of prompt emissions from fission for various applications. | University of Michigan |
| n<br><br>Delayed neutron spectra | U, Pu | N/A | Explosives detection | Idaho National Laboratory |
| n<br><br>capture gamma | Gamma emission from Gd, Pb and structural materials | Thermal to 20 MeV | Neutron radiography, Prompt Neutron Activation Analysis | Texas A&M University |
| n<br><br>(n,fission) fission cross section | $^{238}$U | Sub-threshold fission | $^{238}$U is used in fast fission detectors. E.g. Non-Pro application LSDS for spent fuel assay. | Rensselaer Polytechnic Institute |
| B. Becker et al. “Nondestructive Assay Measurements Using the RPI Lead Slowing-Down Spectrometer” NSE 175 pg. 124-134 (2013) | | | | |
| n<br><br>S(α,β) | Light isotopes | Fast | Various applications | Oak Ridge National Laboratory<br><br>Los Alamos National Laboratory |

| | | | | |
|---|---|---|---|---|
| n<br><br>(n,n’) | Light elements | >1.5 MeV (also 2.5 and 14.1 MeV) | Explosives detection and other applications | Sandia National Laboratories<br><br>Idaho National Laboratory |
| α<br><br>(α,n)<br>cross section | Fluorine | 0 – 3 MeV | Passive neutron measurements of fluorine compounds ($UF_6$, $UO_2F_2$) | `Sandia National Laboratories<br><br>Los Alamos National Laboratory |
| K. Miller et al. “Measured F(α,n) Yield from U234 in Uranium Hexafluoride” NSE 176 98-105 (2014) | | | | |
| α<br><br>Range | MOX fuel | 0 – 3 MeV | MOX holdup calculations | Los Alamos National Laboratory |
| α<br><br>(α,n)<br>target yields | Light elements | 0 – 3 MeV | Enrichment verification, Pu oxide characterization at reprocessing facilities, background in multiplicity measurements for SNM characterization | Idaho National Laboratory<br><br>Los Alamos National Laboratory |
| M. Fensin “Comparing Measured (a,n) Thick Target Yields to MCNP6 using TENDL 2012 Libraries” Proceedings of the 2014 RSPD Topical Meeting Knoxville, Tennessee, United States LA-UR-14-21392 | | | | |
| n<br><br>(n,n’) | Light elements | >1.5 MeV (also 2.5 and 14.1 MeV) | Explosives detection and other applications | Sandia National Laboratories<br><br>Idaho National Laboratory |
| γ<br><br>photofission | U<br>Pu | Fast | Active interrogation | Oak Ridge National Laboratory<br><br>Oregon State University<br><br>University of Tennessee |
| D. Morse et al. “Photofission in U by Nuclear Reaction Gamma-Rays” Nuclear Instruments and Methods in Physics Research B Vol 261 (2007)<br>J. Verbeke et al. “Simulation of Neutron and gamma-Ray Emission from Fission and Photofission” 2010 LLNLRidikas, D., et al., *Status of the photonuclear activation file: Reaction cross-sections, fission fragments and delayed neutrons.* Nuclear Instruments and Methods in Physics Research Section A, 2006. **562**(2): p. 710-713<br>Stevenson, J., et al. , *Linac based photofission inspection system employing novel detection concepts.* Nuclear Instruments and Methods in Physics Research Section A, 2011. **652**(1): p. 124-128.<br>Berman, B.L., et al.. *Photofission and photoneutron cross sections and photofission neutron multiplicities for $^{233}U$, $^{234}U$, $^{237}Np$, and $^{239}Pu$.* Physical Review C, 1986. **34**(6): p. 2201-2214. | | | | |
| e-<br><br>(e,gamma)<br>electronic excitation cross section | $^{235}U$<br>$^{238}U$<br>$^{239}Pu$<br>$^{240}Pu$ | 0.5 – 4 MeV | Potential method to produce monoenergetic photons from Nuclear Resonance Fluorescence | Penn State University |
| E.C . Morse et al. “Mechanical Doppler Compensation for Electron Excitation of NRF Photons” 2008 IEEE Nuclear Science Symposium Conference Record | | | | |