

# Overview of Experimental Nuclear Data Research at RPI

Y. DANON

*Rensselaer Polytechnic Institute, Troy, NY 12180*

## Abstract

Accurate nuclear data is an important ingredient required for accurate neutron transport calculations in a variety of applications. Current nuclear physics models require experimental data in order to provide nuclear data with sufficient accuracy for applications. The Gaerttner Linear Accelerator (LINAC) Center at Rensselaer Polytechnic Institute (RPI) has been actively measuring nuclear data for applications since the accelerator commenced operations in 1961. The electron LINAC is used to produce short neutron pulses required for neutron time-of-flight measurements. A major upgrade project that will increase the neutron production by a factor of about 10 is ongoing with expected completion in 2020. Current measurement capabilities cover the energy range from thermal to 20 MeV and include total, capture, fission, (n, alpha), and scattering reactions. The RPI gamma-multiplicity detector which is a nearly  $4\pi$  segmented gamma detector was recently used for simultaneous fission and capture measurement of  $^{235}\text{U}$  in the 0.01 eV to 3 keV energy region. This measurement helped resolve a discrepancy between ENDF/B-7.1 and JENDL-4.0 evaluations. A detector array for fast neutron scattering and angular distributions enabled measurement of  $^{238}\text{U}$ , Fe, Mo, Be, Zr, and Pb. Some of these measurements were used in the new ENDF/B-8.0 evaluation. Recently we performed a similar scattering experiment for  $^{235}\text{U}$  and  $^{239}\text{Pu}$  at Los Alamos National Laboratory. A unique capability at RPI is the Lead Slowing-Down Spectrometer (LSDS). This spectrometer creates an intense neutron flux in the eV to keV range that enables measurements on small samples or samples with small cross sections. Recent measurements with the LSDS include: fission cross section, fission fragment yields, (n, alpha) cross sections, and neutron capture yields. All described experimental systems were designed and constructed by graduate and undergraduate students which thus gain valuable experience in nuclear data, data acquisition and analysis, and neutron transport methods used to optimize and compare with the experiments. A review of some of the capabilities and recent results will be presented.

## I. INTRODUCTION

Nuclear Data (ND) in the context of this paper refers to fundamental physical quantities that describe neutron or photon interactions with matter. ND is required for applications-design and for interpretation of radiation measurements for example in safeguards and nonproliferation applications. The accuracy of the ND and the physics models embedded in computational tools determine the accuracy of the results, as such there is a need to improve the accuracy and reduce uncertainties in ND. In some cases the accuracy of calculations determines the safety margins of derived quantities, thus smaller uncertainties result in smaller safety margins that can lower cost without compromising safety.

Because of the complexity of the nuclear physics involved, theoretical models alone cannot provide the required accuracy and experimental data must be used. One such example is resonance region cross sections where experimental data must be fitted to a theoretical model to obtain resonance parameters with the accuracy required for applications. In order to perform ND measurements a variety of methods and experimental setups must be used. Constant improvement is driven by innovative methods and the use of state of the art facilities, detectors, electronics, and data analysis methods.

The Gaerttner LINAC Center at RPI has been producing ND for applications since 1961 when operations started. The majority of work over the years was related to neutron induced reactions. The measurement capacities at the Center are relevant to a variety of applications including: nuclear reactors, criticality safety, safeguards, medical isotopes, and nonproliferation. A significant upgrade of the Rensselaer Polytechnic Institute (RPI) accelerator is ongoing and will result in about 10x improvement in short pulse neutron production and will enable additional measurement capabilities with improved accuracy. This paper will highlight only some of the capabilities and measurements that are also relevant to nonproliferation.

## II. DESCRIPTION OF THE WORK

The Center utilizes a 60 MeV electron linear accelerator (LINAC) to produce neutrons. In order to enable neutron time of flight (TOF) measurements, the LINAC was designed to work in short pulse mode (5 ns - 5  $\mu$ s) and variable repetition rate up to 500 pulses per second. Different neutron production targets were designed that tailor the neutron spectrum produced to the experiment requirements [1]. In order to perform different experiments several neutron detection systems were developed [1]; these included Li-Glass detectors for transmission, NaI and C<sub>6</sub>D<sub>6</sub> detector arrays for capture, and liquid scintillator and Li-Glass arrays for neutron scattering and transmission. A unique capability is a cube of lead with 1.8 m side length which forms the lead slowing down spectrometer (LSDS) [2]. The LSDS provides an intense neutron flux in the energy range from 0.1 eV to 100 keV and enables measurements of fission and capture cross sections on small samples, samples with small cross sections, and measurements of fission fragment mass and energy distributions [3]. The LINAC was also used for measurements of photoneutron production [4] cross sections, and

recently there is interest in additional measurements that are currently in progress.

### III. RESULTS

Three experimental setups and results will be reviewed, they provide an example of the measurement capabilities.

#### III.A. Capture and fission

Neutron capture and fission cross sections can be considered competing reactions in systems with fissile (or fissionable) material. Neutron transport calculations requires accurate cross sections in the resolved and unresolved resonance region in order to provide accurate results. In this energy region the evaluations include resonance parameters derived directly from experimental data. In order to improve the evaluation, new higher accuracy experiments are needed.

Two setups for time of flight neutron capture measurements are available, and additional capability using the LSDS was recently developed [5]. One of these detectors located at a 25 m flight path is the multiplicity detector, it is a cylindrical NaI detector divided to 16 segments and surrounds a sample that is being measured. The detector was originally built for neutron capture measurements and with some development was shown to be a very useful tool for simultaneous measurements of fission and capture [6]. Instead of the traditional method of using a fission chamber inside a gamma detector [7], the principle of this measurement was to use gammas to detect fission events and thus allow the use of a large sample which is not possible with a fission chamber. Another advantage was that the same sample was used for both transmission and capture and thus some of the systematic uncertainties are reduced. The method was implemented for  $^{235}\text{U}$  and utilized the total gamma energy deposition and the measured gamma multiplicity to separate fission from capture reaction rates. This measurement was detailed in reference [6] and resulted in fission and capture yields that together with the data of reference [7] helped resolved a discrepancy between ENDF-7.1 [8] and JENDL-4.0 [9] which resulted in the evaluation of  $^{235}\text{U}$  in ENDF-8.0 [10]. An example is shown in fig. 1 where the experimental data and evaluation were grouped in order to better show the differences. A reaction yield represents the probability that a reaction will occur as a result of a neutron interaction with the sample. The measured yield was normalized to the evaluation [6] which resulted in small differences between the experimental data normalized to ENDF-7.1 or 8.0. The measured fission yield is in very good agreement with the evaluations, however the measured capture yield is in much better agreement with ENDF-8.0.

One interesting observation from the experiment in reference [6] was related to neutron interactions with the NaI detector crystals. A measurement was performed with a Pb sample that scattered neutrons to the NaI segmented detector and the fraction of the scattered neutrons that were captured in the NaI was recorded. A simulation with MCNP-Polimi [11] of the same experiment using ENDF-7.1 [8] or JEFF-3.2 [12] evaluations is given in fig. 2. There is a large difference between the simulations, and between the simulations and experiment above 4 keV. This discrepancy was

attributed to a difference between the ENDF-7.1 (and 8.0) and JENDL-3.2 cross sections for  $^{127}\text{I}$ . The ENDF-7.1 evaluation transitions to the fast region at 4.05 keV, and JEFF-3.2 transitions to the unresolved resonance region at 5.2 keV and to the fast region at 57.6 keV. This result indicates a possible problem in the cross section of  $^{127}\text{I}$  above 4 keV which is important for applications that model the response of NaI detectors in a neutron environment.

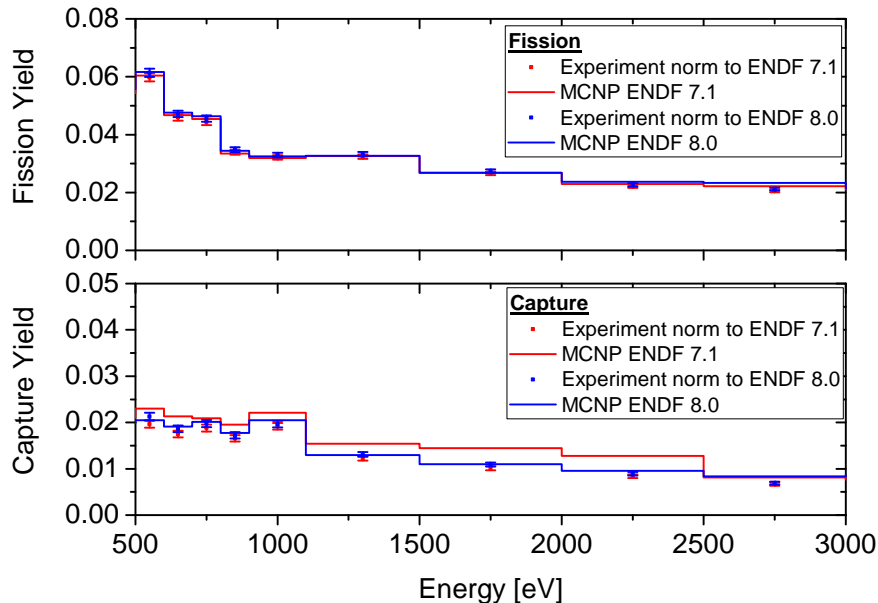


Fig. 1. Comparison of  $^{235}\text{U}$  measured fission (top) and capture (bottom) yield with MCNP [13] simulations using ENDF-7.1 and 8.0 evaluations

### III.B. Fast neutron scattering

Inelastic neutron scattering is one of the quantities that is the hardest to measure. A common method is to measure the first few states by measuring the gammas produced. An alternative method is to use a pulsed monoenergetic neutron source with a sample placed next to the source and measure the outgoing neutron energy as a function of TOF. At RPI a TOF method was developed that measures all scattered neutrons (elastically and inelastically). This method allows for an accurate measurement of the scattered neutron spectrum and angular distributions because analysis is not performed to separate the two scattering reactions. The method was successfully used to identify discrepancies with evaluations for several materials including Mo [14], [15], Be [15], Zr [16], Pb [17], and more recently  $^{238}\text{U}$  [18] and Fe [19]. This type of data was used for validation of the ENDF-8.0 evaluation [10] of  $^{238}\text{U}$  [20] and Be [10]. We are currently exploring the same methodology for  $^{235}\text{U}$  and  $^{239}\text{Pu}$  and performed measurements at Los Alamos National Laboratory [21]. In this case both scattered and fission neutrons are measured and thus this measurement is referred to as neutron-induced neutron emission. Initial results are shown in fig. 3 for carbon and  $^{239}\text{Pu}$  samples for a 30 deg angle. The carbon sample serves as a reference for the quality of the simulation and measurement, and shows excellent agreement between the experiment and

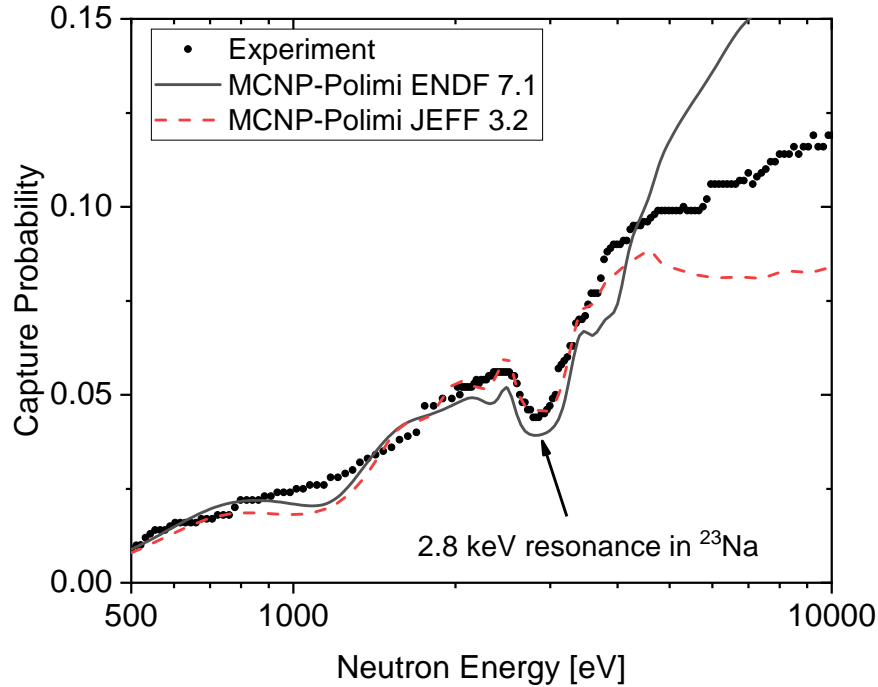


Fig. 2. Measurement of scattered neutron capture probability with a large NaI detector [6] compared with simulation.

MCNP simulation. Both the  $^{239}\text{Pu}$  experiment and simulation show some structure that is due to the sample encapsulation, and other differences possibly related to ND. More detailed analysis is required to understand the differences between the simulation and the experiment. An improved simulation tool that includes fission neutron angular distribution as a function of incident neutron energy is also required.

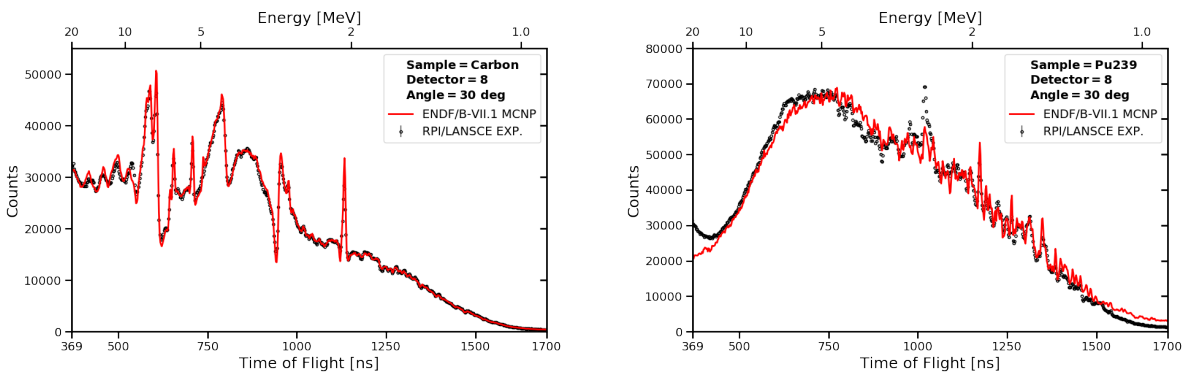


Fig. 3. Results from neutron-induced neutron emission performed by our group at Los Alamos National Laboratory. Left is a measurement and simulation for a carbon sample and right is the same for a  $^{239}\text{Pu}$  sample. The statistical uncertainties in the measurement were omitted from the plot because they are small.

### III.C. Fission fragment mass distribution

Measured data related to fission fragments are important for certain applications and also for better understanding of the fission process, and development of theoretical models to predict mass and charge yield. We were particularly interested in development of a double-gridded fission chamber and using the advantages of the LSDS to simultaneously measure fission cross sections and fission fragment mass and energy distributions. The advantage of this method is the very high neutron flux in the LSDS that enables the use of a very small sample of about 20-40  $\mu\text{g}$ , and the variable incident neutron energy between thermal to 50 keV. The disadvantages are the low mass resolution of 2-3 amu and the limited energy range. However, this method enables measurements of low mass actinide samples in a short experiment time. Results for  $^{252}\text{Cf}$ ,  $^{235}\text{U}$  and  $^{239}\text{Pu}$  were given in [3]. It was shown that useful data can be obtained when the sample was properly prepared. An example of the fragment Total Kinetic Energy (TKE) vs Mass matrix obtained for  $^{239}\text{Pu}$  fission is shown in fig. 4. The data was generated for neutrons with incident energy below 0.1 eV which makes the thermal group for the LSDS. The light and heavy fission fragment lobes are clearly visible and the TKE for each fragment mass can be obtained from such data.

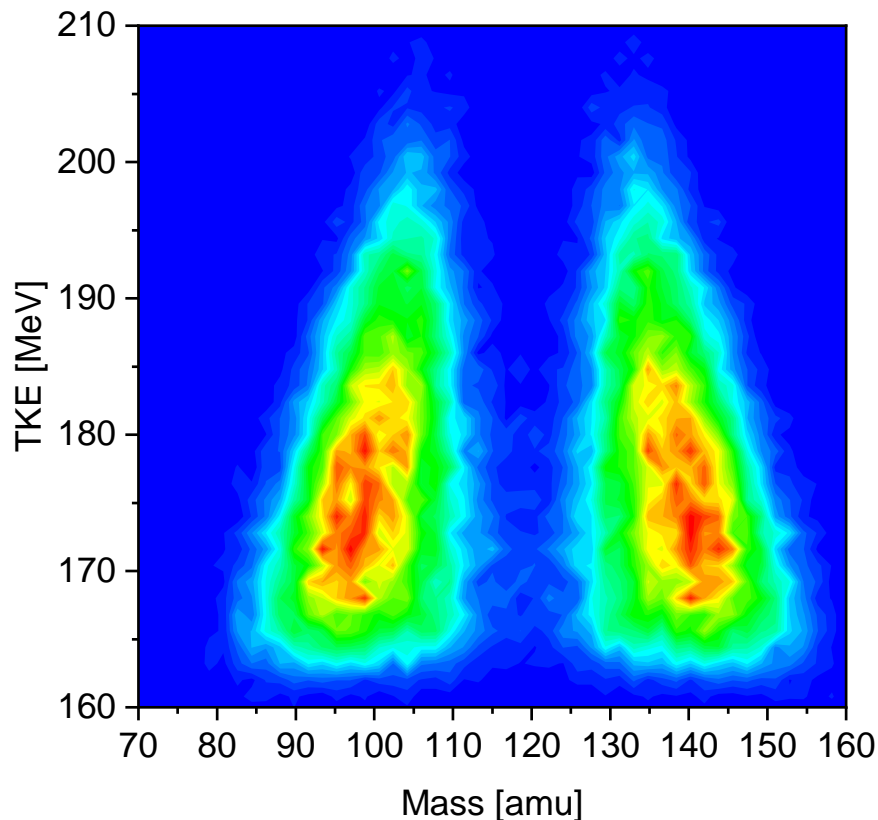


Fig. 4. Measured TKE vs fragment mass for  $^{239}\text{Pu}$  fission with neutron below 0.1 eV using the RPI LSDS

## IV. CONCLUSION

The Gaerttner LINAC Center at RPI combines an intense pulsed neutron source and multiple neutron and gamma detection systems. These capabilities enable measurements of neutron and gamma-induced reactions that are used to improve the accuracy of nuclear data. These include: neutron capture, scattering, and fission reactions. Measurements of (n,alpha) reactions were also performed with the LSDS [22]. Fission fragment mass and energy distributions and the fission cross section can be measured with the LSDS for small quantities (20-40  $\mu\text{g}$ ) of actinides.

In order to further improve the measurements a major upgrade and refurbishment project is in progress and will result in a modern accelerator with higher electron beam energy and power. This upgrade will provide about ten times improvement in short pulse (5 ns) neutron production. The new LINAC will enable new types of measurements with smaller isotopically enriched samples and will ensure the availability of a world class facility for resonance region measurements in the US.

It is important to note that having the LINAC at a technical university like RPI helps educate the next generation of nuclear engineers and scientists that understand the importance of ND and how it fits in different calculations. All of the experimental capabilities described here were designed and constructed by graduate students.

## REFERENCES

- [1] Y. DANON, A. DASKALAKIS, B. MCDERMOTT et al., “Recent Developments in Nuclear Data Measurement capabilities at the Gaerttner LINAC Center at RPI,” *EPJ Web of Conferences*, **111**, 02001 (2016); <https://doi.org/10.1051/epjconf/201611102001>., URL <http://dx.doi.org/10.1051/epjconf/201611102001>.
- [2] Y. DANON, R. SLOVACEK, R. BLOCK, and M. MOORE, “Fission Cross-Section Measurements of  $^{247}\text{Cm}$ ,  $^{254}\text{Es}$ , and  $^{250}\text{Cf}$  from 0.1 eV to 80 keV,” *Nucl. Sci. Eng.*, **109**, 4, 341 (1991)URL <http://www.ans.org/store/article-23859/>.
- [3] C. ROMANO, Y. DANON, R. BLOCK, J. THOMPSON, E. BLAIN, and E. BOND, “Fission fragment mass and energy distributions as a function of incident neutron energy measured in a lead slowing-down spectrometer,” *Phys. Rev. C*, **81**, 014607 (2010); <https://doi.org/10.1103/PhysRevC.81.014607>., URL <http://link.aps.org/doi/10.1103/PhysRevC.81.014607>.
- [4] N. N. KAUSHAL, E. J. WINHOLD, P. F. YERGIN, H. A. MEDICUS, and R. H. AUGUSTSON, “Fast-Photoneutron Spectra due to 55-85-MeV Photons,” *Phys. Rev.*, **175**, 1330 (1968); [10.1103/PhysRev.175.1330](https://doi.org/10.1103/PhysRev.175.1330)., URL <https://link.aps.org/doi/10.1103/PhysRev.175.1330>.
- [5] N. THOMPSON, “Measuring and Validating Neutron Capture Cross Sections Using a Lead Slowing-Down Spectrometer,” *PhD Thesis, Rensselaer Polytechnic Institute* (2017).

- [6] Y. DANON, D. WILLIAMS, R. BAHRAN, E. BLAIN, B. McDERMOTT, D. BARRY, G. LEINWEBER, R. BLOCK, and M. RAPP, “Simultaneous Measurement of  $^{235}\text{U}$  Fission and Capture Cross Sections From 0.01 eV to 3 keV Using a Gamma Multiplicity Detector,” *Nucl. Sci. Eng.*, **187**, 3, 291 (2017); <https://doi.org/10.1080/00295639.2017.1312937>., URL <http://dx.doi.org/10.1080/00295639.2017.1312937>.
- [7] M. JANDEL, T. A. BREDEWEG, E. M. BOND et al., “New Precision Measurements of the  $^{235}\text{U}(n, \gamma)$  Cross Section,” *Phys. Rev. Lett.*, **109**, 202506 (2012); 10.1103/PhysRevLett.109.202506., URL <http://link.aps.org/doi/10.1103/PhysRevLett.109.202506>.
- [8] M. CHADWICK, M. HERMAN, P. OBLOZINSKY et al., “ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data,” *Nucl. Data Sheets*, **112**, 12, 2887 (2011); <http://dx.doi.org/10.1016/j.nds.2011.11.002>., URL <http://www.sciencedirect.com/science/article/pii/S009037521100113X>, special Issue on ENDF/B-VII.1 Library.
- [9] K. SHIBATA, O. IWAMOTO, T. NAKAGAWA et al., “JENDL-4.0: A New Library for Nuclear Science and Engineering,” *J. Nucl. Sci. Technol.*, **48**, 1, 1 (2011); 10.1080/18811248.2011.9711675., URL <http://www.tandfonline.com/doi/abs/10.1080/18811248.2011.9711675>.
- [10] D. BROWN, M. CHADWICK, R. CAPOTE et al., “ENDF/B-VIII.0: The 8th Major Release of the Nuclear Reaction Data Library with CIELO-project Cross Sections, New Standards and Thermal Scattering Data,” *Nucl. Data Sheets*, **148**, 1 (2018); <https://doi.org/10.1016/j.nds.2018.02.001>., URL <https://www.sciencedirect.com/science/article/pii/S0090375218300206>, special Issue on Nuclear Reaction Data.
- [11] S. A. POZZI, E. PADOVANI, and M. MARSEGUERRA, “MCNP-PoliMi: a Monte-Carlo code for correlation measurements,” *Nucl. Instrum. Methods Phys. Res., Sect. A*, **513**, 3, 550 (2003); <http://dx.doi.org/10.1016/j.nima.2003.06.012>., URL <http://www.sciencedirect.com/science/article/pii/S0168900203023027>.
- [12] A. SANTAMARINA, D. BERNARD, M. C. P. BLAISE et al., JEFF Report 22, Nuclear Energy Agency Organisation for Economic Co-operation and Development (2009).
- [13] T. GOORLEY, M. JAMES, T. BOOTH et al., “Initial MCNP6 Release Overview,” *Nucl. Technol.*, **180**, 3, 298 (2012); <http://dx.doi.org/10.13182/NT11-135>.
- [14] F. SAGLIME, Y. DANON, R. BLOCK, M. RAPP, R. BAHRAN, G. LEINWEBER, D. BARRY, and N. DRINDAK, “A system for differential neutron scattering experiments in the energy range from 0.5 to 20MeV,” *Nucl. Instrum. Methods Phys. Res., Sect. A*, **620**, 2-3, 401 (2010); <https://doi.org/10.1016/j.nima.2010.04.051>., URL <http://dx.doi.org/10.1016/j.nima.2010.04.051>.



- [15] A. DASKALAKIS, E. BLAIN, G. LEINWEBER, M. RAPP, D. BARRY, R. BLOCK, and Y. DANON, “Assessment of beryllium and molybdenum nuclear data files with the RPI neutron scattering system in the energy region from 0.5 to 20 MeV,” *EPJ Web Conf.*, **146**, 11037 (2017); <https://doi.org/10.1051/epjconf/201714611037>., URL <https://doi.org/10.1051/epjconf/201714611037>.
- [16] D. P. BARRY, G. LEINWEBER, R. C. BLOCK, T. J. DONOVAN, Y. DANON, F. J. SAGLIME, A. M. DASKALAKIS, M. J. RAPP, and R. M. BAHRAN, “Quasi-Differential Neutron Scattering in Zirconium from 0.5 to 20 MeV,” *Nucl. Sci. Eng.*, **172**, 2, 188 (2013); [dx.doi.org/10.13182/NSE12-1](https://doi.org/10.13182/NSE12-1)., URL [http://www.ans.org/pubs/journals/nse/a\\_16857](http://www.ans.org/pubs/journals/nse/a_16857).
- [17] A. E. YOUMANS, J. BROWN, A. DASKALAKIS, N. THOMPSON, A. WELZ, Y. DANON, B. McDERMOTT, G. LEINWEBER, and M. RAPP, “Fast Neutron Scattering Measurements with Lead,” *AccApp 15, Washington, DC*, 355–360 (2015).
- [18] A. DASKALAKIS, R. BAHRAN, E. BLAIN et al., “Quasi-differential neutron scattering from <sup>238</sup>U from 0.5 to 20 MeV,” *Ann. Nucl. Energy*, **73**, 455 (2014); <https://doi.org/10.1016/j.anucene.2014.07.023>.
- [19] A. DASKALAKIS, E. BLAIN, B. McDERMOTT et al., “Quasi-differential elastic and inelastic neutron scattering from iron in the MeV energy range,” *Ann. Nucl. Energy*, **110**, 603 (2017); <https://doi.org/10.1016/j.anucene.2017.07.007>., URL <http://www.sciencedirect.com/science/article/pii/S0306454917301950>.
- [20] R. CAPOTE, A. TRKOV, M. SIN et al., “IAEA CIELO Evaluation of Neutron-induced Reactions on <sup>235</sup>U and <sup>238</sup>U Targets,” *Nucl. Data Sheets*, **148**, 254 (2018); <https://doi.org/10.1016/j.nds.2018.02.005>., URL <https://www.sciencedirect.com/science/article/pii/S0090375218300243>, special Issue on Nuclear Reaction Data.
- [21] K. MOHINDROO, E. BLAIN, Y. DANON, S. MOSBY, and M. DEVLIN, “Quasi-differential neutron induced neutron emission reaction measurements at WNR,” *Trans. Am. Nucl. Soc.*, **115**, 701 (2016).
- [22] J. THOMPSON, T. KELLEY, E. BLAIN, R. HAIGHT, J. O’DONNELL, and Y. DANON, “Measurement of (n,α) reactions on <sup>147</sup>Sm and <sup>149</sup>Sm using a lead slowing-down spectrometer,” *Nucl. Instrum. Methods Phys. Res., Sect. A*, **673**, 16 (2012)URL <http://www.sciencedirect.com/science/article/pii/S0168900212000198>.