Applications of Nuclear Science for Stewardship Science

Jolie A. Cizewski
Department of Physics and Astronomy, Rutgers University
New Brunswick, NJ 08903 USA
E-mail: cizewski@rutgers.edu

Abstract. Stewardship science is research important to national security interests that include stockpile stewardship science, homeland security, nuclear forensics, and non-proliferation. To help address challenges in stewardship science and workforce development, the Stewardship Science Academic Alliances (SSAA) was inaugurated ten years ago by the National Nuclear Security Administration of the U.S. Department of Energy. The goal was to enhance connections between NNSA laboratories and the activities of university scientists and their students in research areas important to NNSA, including low-energy nuclear science. This paper presents an overview of recent research in low-energy nuclear science supported by the Stewardship Science Academic Alliances and the applications of this research to stewardship science.

1. Introduction
Nuclear science has a long tradition of impacts beyond basic research ranging from medicine to national security to energy. More recently the national security applications include stockpile stewardship science, homeland security, nuclear forensics, and non-proliferation, activities that can be broadly categorized as stewardship science. The Stewardship Science Academic Alliances (SSAA) Program [1] was inaugurated ten years ago by the National Nuclear Security Administration (NNSA) of the U.S. Department of Energy to enhance connections between NNSA laboratories and the activities of university scientists and their students in research areas important to NNSA, including low-energy nuclear science. The SSAA supports a large number of individual investigator projects, as well as one Centre of Excellence in low-energy nuclear science. SSAA research directions in low-energy nuclear science include: neutron, gamma, and ion-induced reactions with stable and unstable nuclei; advanced simulations and measurement techniques that improve radiation and particle detection in terms of energy, temporal and spatial resolution; and physics of the fission process and properties of prompt fission products. A new direction for 2012 is radiochemistry research.

Nuclear science research important to stewardship science spans the landscape of nuclei as summarized in Figure 1. It includes the few-body nuclei important in fusion and the actinides that have low barriers to fission. Properties of and reactions on unstable nuclei such as fission products are also important for stewardship science. For particular applications in science-based stockpile stewardship, we also need to understand reactions on the so-called radiochemical detectors, materials that were placed in specific locations in a nuclear weapon during the testing era to assess the performance of the device, in particular neutron fluences. Table 1 summarizes the importance of neutron-induced reactions and targets for stewardship science.
Figure 1. (color online) Chart of the nuclei highlighting regions of nuclei where reactions are important for stewardship science. Adopted from figure by J.T. Burke (LLNL) [2].

Table 1. Neutron-induced reactions on actinides, fission fragments, and radiochemical detectors

| Reaction | Neutron Energy | Importance | Accuracy |
|----------|---------------|------------|----------|
| (n,γ)    | 0.01-0.2 (MeV) | High       | 10%      |
| (n,2n)   | 10-16 MeV     | High       | 3-5%     |
| (n,f)    | 0.1-16 MeV    | High       | 1-2%     |

Adopted from L. Ahle, ref. [3]

The following sections present some of the recent highlights of stewardship science applications from neutron-induced reactions, including those from SSAA-supported groups.

2. Fission
Fission is a complicated nuclear many-body collective process as sketched in Figure 2. The ground state configuration of the actinide nucleus is excited in fission, with the decay of highly excited states competing between gamma decay and fission. On the path to fission are discrete transitions at the barrier, $E_A$. To further complicate fission are nuclei where there are second, $E_B$, (or more) minima in the potential energy surface at larger deformations, where again there is a competition between decay within the nucleus and fission, now with reduced barriers $E_B$.

2.1. Fission Theory.
Witek Nazarewicz and his collaborators, including graduate students and postdocs supported by the SSAA, are developing a new approach to predicting the potential energy surface of actinide nuclei, such as $^{230}$Pu. Displayed in Figure 2 is recent work [4] with a new energy density functional that is able to reproduce the potential energy surface of $^{230}$Pu, including the energies of the inner and outer fission barriers and the second minimum, as a function of the mass quadrupole moment $Q_{20}$. 
2.2. New tools for fission cross-section measurements.

As we move towards future generations of nuclear reactors, as well as for stockpile stewardship, it is important to measure fission cross sections to unprecedented levels of precision, as well as on minor actinides.

![Figure 3](image_url)

**Figure 3.** (color online) Fission event from the prototype Time Projection Chamber being developed by LLNL, LANL, and their collaborators. Private communication from M. Heffner (LLNL) [5].

A collaboration of scientists at Lawrence Livermore (LLNL), Los Alamos (LANL), and Idaho National Laboratories, joined by several universities, is developing time projection chambers (TPC) [5] to measurement fission cross sections using neutron beams at the Los Alamos Neutron Science Center (LANSCE). With the TPC, the tracks of the fission fragments are readily visualized, and can be distinguished from alpha decay, for example. An actual fission event in the prototype TPC is displayed in Figure 3. The precision fission measurements will focus on $^{235}\text{U}$ and $^{239}\text{Pu}$. 
It is also important to understand fission cross sections on rare actinides, such as $^{237}\text{U}$ ($t_{1/2}=6.75$ days) or $^{240}\text{Am}$ ($t_{1/2}=51$ hours). These measurements can be realized with a Lead Slowing Down Spectrometer [6] that can use samples as small as 20 ng. The measurements use a direct beam of protons, such as the 800-MeV protons from LANSCE, which interact with tungsten in the centre of lead cube. The reaction neutrons scatter within the lead, reducing the energies to 1 eV to 100 keV and then interact with the actinide samples and detectors in various locations in the spectrometer.

To create the samples and conduct the measurements requires a team of nuclear scientists. Key players are the nuclear chemists who develop the samples. One example [7] is the production of $^{240}\text{Am}$ samples for future measurements, an effort led by Professor Heino Nitsche at Berkeley and supported in part by an SSAA grant. The first challenge was to identify the reaction to produce $^{240}\text{Am}$, $^{242}\text{Pu}(p,3n)$ that was measured at the 88-Inch Cyclotron at Lawrence Berkeley National Laboratory (LBNL). The Nitsche group also had to develop the techniques to separate the Am. This work was also part of the Ph.D. project of Paul Ellison, who was supported in part by the NNSA as a Stewardship Science Graduate Fellow.

2.3. Surrogates for fission

While new techniques are being developed to measure fission cross sections of rare actinides, there are nuclei for which fission cross sections are important for stewardship science, but unlikely that a sample would ever be available for direct measurements. This requires a validated surrogate approach to deducing important cross sections. The theory of the surrogate mechanism and a summary of recent experimental results are summarized in ref. [8], which informs much of the following discussion.

The surrogate approach requires a compound nuclear process, such as fission, where the decay of the compound nucleus is independent of how it was formed. In general the cross section $\sigma$ as a function of neutron energy $E_n$ for a neutron-induced reaction on nucleus $A$ and decay channel $X$ in nucleus $A+1$ is

$$\sigma_{nX}(E_n) = \sum_{J,\pi} \sigma_{n}^{CN}(E_n, J, \pi) G_X^{CN}(E_n, J, \pi)$$

where the cross section is a sum over spins $J$ and parities $\pi$ for the product of the cross section that forms the compound nucleus and its decay, $G$, in channel $X$.

The probability $P$ for the surrogate reaction $B(a,b)$ to the compound nucleus $A+1$ is similar

$$P_{abX}(E_x) = \sum_{J,\pi} F_{ab}^{CN}(E_x, J, \pi) G_X^{CN}(E_x, J, \pi)$$

and depends upon the formation, $F$, of the compound nucleus and the same decay probability, $G$, both depending upon the excitation energy as well as spins and parities. To apply the surrogate approach has traditionally required two assumptions. The first is that the direct reaction forms the same compound nucleus as the one induced by neutrons and with probability unity, $F=1$. The second is that the population and decay of the compound nucleus is independent of spin and parity, the Weisskopf-Ewing limit [9]. With these assumptions the determination of the neutron-induced reaction cross section from the surrogate reaction is given by

$$\sigma_{nX}^{WE}(E_n) = \sigma_{n}^{CN}(E_n) G_X^{CN}(E_n) = \sigma_{n}^{CN}(E_n) \frac{N(a,bX)}{\epsilon N(a,b)}$$

that depends upon the number $N(a,bX)$ of surrogate events in coincidence with the decay channel, $X$, of interest, relative to the total number of events $N(a,b)$, of course with appropriate efficiency $\epsilon$ corrections. The compound nucleus formation cross section $\sigma_{n}^{CN}$ is calculated with optical models.
The surrogate reaction approach to determining fission cross sections was pioneered by Britt, Cramer and Wilhelmy [10] using light-ion reactions, such as (t,p), to induce fission. More recently, scientists at LLNL and LBNL have developed a program to measure surrogate reaction induced fission with annular silicon-strip detector arrays to measure the reaction products and fission detectors [11]. University scientists, in particular the SSAA-supported group of Beausang at the University of Richmond, have played key roles in these efforts. A recent example [12] summarized in Figure 4 deduced the $^{238}\text{Pu}(n,f)$ cross section from the surrogate $^{239}\text{Pu}(\alpha,\alpha')f$, reaction, reducing uncertainties in the evaluations of this cross section that is important for stewardship science.

![Figure 4](image)

**Figure 4.** (color online) Neutron-induced fission cross sections for $^{238}\text{Pu}$ deduced from $^{239}\text{Pu}(\alpha,\alpha')f$ reaction studies by Ressler et al. [12] and compared to evaluations. Adopted from [12].

3. Neutron capture measurements

As highlighted in Table 1, neutron capture measurements are also important for stewardship science and on nuclei across the nuclear landscape. The DANCE array [13] of BaF$_2$ scintillator detectors is a superb instrument for measuring $(n,\gamma)$ cross sections with neutron energies from thermal up to about $\approx 200$ keV using the neutron beams at the Lujan Centre at LANSCE. Neutron capture cross section measurements have long attracted university collaborators, especially the North Carolina State University SSAA project which supports postdocs and graduate students who play key roles in all aspects of the experimental program, for example the results in ref. [14]. A recent example of the detailed results that can be obtained over a wide range of neutron energies is displayed in Figure 5.

3.1. Surrogates for neutron capture

Just as was the case for fission, there are many nuclear species for which direct measurements of neutron capture will not be feasible because of short half lives and associated high gamma radiation backgrounds. As in the case of the surrogate approach to fission, in the surrogate approach to neutron capture one needs to measure the decay of the compound nucleus. When combined with the formation probability of the compound nucleus, in principle the $(n,\gamma)$ cross section can be deduced.
Figure 5. (color online) Neutron capture cross sections measured with DANCE with a ≈200 µg $^{241}$Am ($t_{1/2} = 433$ years) sample. Data up to 320 keV in neutron energy were obtained and compared with evaluations. Adopted from [15].

The challenges with validating a surrogate for $(n,\gamma)$ are the assumptions that the decay is independent of spin [16] and that a direct reaction populates the same compound nucleus as in $(n,\gamma)$. To address the challenge there have been several studies [8 and references therein] to identify appropriate candidates for surrogates for $(n,\gamma)$ reactions, as well as a new concerted effort to validate $(d,p\gamma)$ as a surrogate for $(n,\gamma)$.

Figure 6. (color online) Schematic diagram of $(n,\gamma)$ and surrogate $(d,p\gamma)$ reactions. The decay probability is deduced by measuring the low-lying transition(s) that collect the great majority of the decay of the compound nucleus.

A schematic of the $(n,\gamma)$ and surrogate $(d,p\gamma)$ reactions is displayed in Figure 6. In $(n,\gamma)$, the compound nucleus $A+1$ is populated above the neutron separation energy and decays by statistical cascades down to the ground state, where one or more transitions collect most of the gamma-decay intensity. In a surrogate (such as $(d,p\gamma)$) the reaction particles are used to gate on the $A+1$ nucleus above the neutron separation energy and the intensity of the collecting transition(s) is used to deduce
the decay probability $G$ in equation (3). The surrogate ratio method is often used to reduce systematic uncertainties; in this case equation (3) becomes

\[
\frac{\sigma_n^{(1)}(E_n)}{\sigma_n^{(2)}(E_n)} = \frac{\sigma_n^{CN}(E_n)N^{\gamma(1)}_{dp}(E_n)e^{(2)}N^{\gamma(2)}_{dp}(E_n)}{\sigma_n^{CN}(E_n)N^{\gamma(1)}_{dp}(E_n)e^{(2)}N^{\gamma(2)}_{dp}(E_n)} = K \frac{N^{(1)}_{dp}(E_n)}{N^{(2)}_{dp}(E_n)} \tag{4}
\]

where it is assumed that the compound nucleus formation cross sections $\sigma_n^{CN}$ and the singles rates $N_{dp}$ are the same for both direct reactions. Therefore, by measuring surrogate reactions with two targets, the ratio of particle-gamma coincidences gives the ratio of $(n,\gamma)$ cross sections, allowing a straightforward determination of the unknown cross section relative to the known one. Surrogate reaction studies have attracted many SSAA-supported university groups, including the Centre of Excellence for Radioactive Ion Beam Studies, that is a consortium of universities and Oak Ridge National Laboratory.

**Figure 7.** (color online) Ratio of $^{171,173}$Yb$(n,\gamma)$ cross sections [17] compared to ratio of $^{171,173}$Yb$(d,p\gamma)$ cross sections [16] determined by the intensity of the $4^+ \rightarrow 2^+$ transitions, after subtracting the $6^+ \rightarrow 4^+$ feeding intensity. The measured cross section ratios are reproduced at higher neutron energies. The finite energy bins are a result of the finite proton energy resolution in the $(d,p)$ measurements. Taken from Hatarik et al. [16].

Hatarik et al., [16] was the first to assess the validity of the surrogate ratio method from the $(d,p\gamma)$ reaction. This work summarized in Figure 7 demonstrated that the Weisskopf-Ewing limit was not valid by comparing the ratio of measured [17] and $(d,p\gamma)$ deduced $(n,\gamma)$ cross sections. At the same time this work was able to develop a prescription to use the surrogate reaction to reproduce $(n,\gamma)$ cross section ratios by focussing on side feeding, rather than full feeding, of low-lying yrast transitions. More recently, Escher and Scielzo and their co-workers, [18,19] have demonstrated how $(n,\gamma)$ and surrogate $(p,p'\gamma)$ reactions have different entry spin distributions, as displayed in Figure 8. However, there are no data that can help quantify the entry spin distribution as a function of neutron energy in an $(n,\gamma)$ reaction.
3.2. Quantifying spin distributions in (n,γ) and (d,pγ) surrogate reactions with 95Mo targets and beams

There is currently a concerted effort to validate the (d,pγ) reaction as a surrogate for (n,γ) and in particular for future applications with radioactive beams of very short-lived isotopes. The (d,p) reaction is particularly well-suited for studies with 5-10 MeV/u beams. Not only does the (d,p) reaction bring in relatively low angular momentum, but the reaction protons are preferentially observed at angles greater than 90° in the laboratory, cleanly separated from dominant elastic scattering on CD₂ target materials. The 95Mo target was chosen because (n,γ) cross sections have been measured [20] up to 200 keV and the final nucleus is collective and even even, with the gamma cascade intensity expected to be concentrated in the 2⁺→0⁺ transition.

There are three components to the validation efforts. The first component is to measure the intensity of yrast transitions in the 95Mo(n,γ)96Mo reaction as a function of neutron energy. The first phase of these measurements, with neutron energies up to ≈200 keV is complete, using a small array of HPGe detectors on FP12 at the Lujan Centre at LANSCE. The next phase was measured in Fall 2012 with the GEANIE array of Compton-suppressed Ge detectors on the WNR flight path with 3.6-μsec beam timing to enable measurements with neutrons as low as 150 keV. The measured intensities of yrast transitions as a function of neutron energy would be modelled with spin distribution calculations as was done in refs. [18,19].
The second component is to measure the \((d,p\gamma)\) reaction in normal kinematics with \(^{95}\text{Mo}\) targets. These measurements [21] were completed at Texas A&M University in June 2012 with 13-MeV deuteron beams. Reaction products were measured with the STARS array of annular silicon-strip detectors and photons with 4-5 Compton-suppressed clover Ge detectors.

The third component is to measure the \((d,p\gamma)\) reaction in inverse kinematics with a beam of \(^{95}\text{Mo}\). To realize this measurement, we are developing the Gammasphere-ORRUBA Dual Detectors for Experimental Structure Studies (GODDESS) system, which would couple the Oak Ridge Rutgers University Barrel Array (ORRUBA) [22] of position-sensitive silicon-strip detectors to the world-class array of Compton-suppressed Ge detectors at ALTAS at Argonne National Laboratory. A schematic showing the coupling of ORRUBA and Gammasphere is displayed in Figure 9. When completed GODDESS is well-suited to measure \((d,p\gamma)\) (and other) reactions with the \(^{252}\text{Cf}\) fission fragment beams that will become available in early 2013 with the CARIBU project at ATLAS [24]. The \(^{95}\text{Mo}(d,p\gamma)\) reaction study would be part of the commissioning of the GODDESS system.

Upon completion of the analysis of the three experiments, the entry spin distributions will be deduced from the intensities of yrast transitions from the \(^{95}\text{Mo}(n,\gamma)\) and the \(^{95}\text{Mo}(d,p\gamma)\) measurements in both normal and inverse kinematics, as was done in reference [19]. These results would be used to calculate \((n,\gamma)\) cross sections using the methods of Ref. [18] and compare them to the known cross sections [20]. In addition to using the intensities of the yrast transitions, an analogous analysis would be made using the side-feeding intensities of the yrast transitions, i.e., by subtracting the intensity of the feeding \(4^+\rightarrow 2^+\) transition from the observed \(2^+\rightarrow 0^+\) intensity. The work by Hatarik [16] showed that focusing on the side-feeding in the surrogate reaction may be a better analogue of the entry distribution in \((n,\gamma)\) and hence aid in deducing valid \((n,\gamma)\) cross sections from a surrogate reaction.

Figure 9. (color online) Schematic of how ORRUBA would be coupled to the 100-unit Gammasphere Compton-suppressed Ge detector array. The barrel array would be augmented by up to 4 annular strip detectors to be placed at forward and backward angles in the laboratory. All electronics signals and preamplifier boxes would be downstream of ORRUBA and before the quadrupole magnet of the Fragment Mass Analyzer. Provided by Ratkiewicz and Shand [23].
4. Summary and conclusions
Stewardship science is an important application of nuclear science and much of the stewardship needs are reactions induced by neutrons, in particular fission and neutron capture. This paper has highlighted some of the ways that the goals in low-energy nuclear science of the Stewardship Science Academic Alliances are being met:

• with neutron and light-ion induced reaction studies
• with advanced theoretical modelling
• with new radiation and particle detectors
• by helping to understand the physics of the fission process
• by probing the properties of fission fragments.

The fission and neutron capture activities highlighted in this report have attracted talented early career scientists and their university mentors in collaboration with staff scientists at the Los Alamos and Livermore NNSA laboratories and with support of the SSAA initiative of the NNSA. These efforts not only are educating early career scientists but they are developing the experimental and theoretical techniques, and new instruments, required to address challenges in fundamental nuclear science and its applications to stewardship science. These efforts are also the foundations for future research on nuclei away from stability that will become available in the next decade with the realization of the Facility for Rare Isotopes Beams [25] in the U.S.

Acknowledgements
The author would like to thank C.W. Beausang (Richmond), J.T. Burke, J. Escher, M. Heffner, J. Ressler and N. Scielzo (LLNL), A. Couture and M. Jandel (LANL), W. Nazarewicz (Univ. TN), P. Ellison and H. Nitsche (UC Berkeley) and her RIBSS Centre colleagues for contributing materials to this presentation. This research was sponsored in part by the National Nuclear Security Administration under the Stewardship Science Academic Alliances program through U.S. Department of Energy Cooperative Agreement No. DE-FG52-08NA28552.

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