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Livermore Accelerator Source for Radionuclide Science (LASRS)

Revitalization Plan for B194

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Livermore Accelerator Source for Radionuclide Science (LASRS)

The Livermore Accelerator Source for Radionuclide Science (LASRS) will generate intense photon and neutron beams to address important gaps in the study of radionuclide science that directly impact Stockpile Stewardship, Nuclear Forensics, and Nuclear Material Detection. The co-location of MeV-scale neutron and photon sources with radiochemical analytics provides a unique facility to meet current and future challenges in nuclear security and nuclear science.

Executive Summary

More stringent requirements for understanding the experimental and theoretical basis for stockpile science combined with the need to develop more sensitive tools for nuclear forensics and nuclear security have created a demand for improved measurements of photon- and neutron-induced nuclear reactions. Specifically, there is a growing need for high-precision, fission-product and cross-section measurements for evaluation in nuclear data libraries, new measurements of nuclear reactions to benchmark theoretical calculations, increased precision for isotopic signatures, and new methods for the detection of special nuclear material.

These new demands are occurring at a time when options to perform measurements at offsite facilities are becoming more scarce due to budget constraints and the consolidation of the national nuclear science program into a few large facilities that are creating high demand and limited availability. New investments in neutron- and photon-accelerator capabilities in the B194 complex provide an opportunity to perform many of these measurements to satisfy internal program needs as well as attract funding from external sponsors. The co-location of high-intensity, accelerator-driven neutron and photon sources combined with radiochemical analysis capabilities provides an opportunity to perform essential measurements not achievable at any other laboratory.

The measurements to be addressed by LASRS are as follows:

- Fission cross-section and product yield measurements
- Nuclear data evaluations in support of predictive theory for nuclear reactions
- Activation measurements to support radiochemical forensics and diagnostics
- Neutron and photon induced signatures for nuclear material detection
- Neutron scattering, spectroscopy, and imaging for materials research, primarily for non-destructive diagnosis

In this document, we present a path to develop an on-site capability to measure neutron-and photon-induced nuclear reactions for LLNL programs and sponsors. We review the different sets of measurements needed and show how current investments in accelerator-driven neutron- and photon-source capabilities made by the Weapons Complex and Integration (WCI) Directorate and Nuclear and Chemical Sciences (NACS) Division within the Physical and Life Sciences (PLS) Directorate will help address the growing need for precision nuclear reaction measurements. As a demonstration exercise, we present sample rate estimates for neutron- and photon-induced plutonium fission reactions, and compare them to estimates for other facilities to show that LASRS is not only competitive, but would be a high-value institutional asset supporting laboratory missions. Finally, we discuss how the LASRS advances long-term nuclear science mission needs at LLNL.

I. Neutron and Photon Induced Nuclear Reactions

Over the years, WCI has continued to examine nuclear science needs for the stockpile-stewardship program, such as the study recently conducted by P. Bedrossian [1]. Nuclear reaction measurements, such as cross sections for neutron-induced reactions, play an important role across the entire stockpile-stewardship program. Two data areas still in need of improvement are fission cross-sections and fission product yield (FPY) distributions. Although previous investigations of ^{239}Pu(n,f) and ^{235}U(n,f) had showed very good agreement with data for thermal neutrons, very recent measurements of FPYs at higher neutron energies performed at the Triangle Universities Nuclear Laboratory (TUNL) by a LLNL/LANL collaboration have shown discrepancies that motivate an urgent need to revisit current theoretical approaches. A revitalized B194 building will enable comprehensive high-precision measurements of FPYs covering a broad range of neutron incident energies; thereby, providing unprecedented constraints for theoretical developments. Also among the top priorities is the need for additional reaction measurements to support efforts in nuclear theory as we move towards predictive models for nuclear structure and nuclear reactions for ever-larger nuclei. In addition to the neutron-induced reactions (n,n'), (n,2n), (n,f), (n,n'd), and (n,γ), new measurements are needed for electro-magnetic strength functions and transfer reactions for photo-nuclear reactions (γ,xn), (γ,p), (γ,f), and (γ,γ') [2]. These measurements are critical for determining the gamma-ray strength functions and level densities necessary to determine capture cross-sections and decay spectra. These quantities are important for understanding nucleosynthesis processes found in nuclear weapons, nuclear reactors, and stellar explosions. These measurements are needed to benchmark theoretical nuclear physics calculations and to provide input to nuclear level-densities for nuclear data evaluations. It should be noted that direct experiments are limited by the need to produce targets that are stable on the timescale of the measurements. Therefore, a complete understanding of fission chain yields in the stockpile, nuclear reactors, or stellar explosions will require a predictive theory of nuclear reactions as well access to indirect methods using surrogate reactions. Support for improved theoretical tools for nuclear reactions is one of the more important goals for developing the capabilities of LASRS.

There is also a need for improved measurements of activation cross-sections for test diagnostics and short-lived isotopes for nuclear forensics. For the latter, the B194 site at

LLNL offers proximity to on-site radiochemical laboratories and to the Nuclear Counting Facility (located in B151), which is key to collecting and analyzing data from short-lived isotopes to support a variety of laboratory missions related to nonproliferation and incident response. Radiochemistry at LLNL has taken on the role of Research Laboratory within the National Technical Nuclear Forensics (NTNF) community, which includes the fabrication of synthetic debris for the nuclear forensics program. In recent years this scope has involved performing high-energy neutron irradiations (20+ MeV) at the UC Davis Crocker Nuclear Laboratory cyclotron on structural materials of interest to the nuclear forensic community to create activation products of high debris-diagnostic value. LLNL has been able to irradiate similar materials at the National Criticality Experiments Research Center (NCERC) at the Nevada Nuclear Security Site (NNSS, formerly the Nevada Test Site) and irradiate uranium at the High Flux Isotope Reactor at Oak Ridge National Laboratory. These methods have allowed LLNL to make activation products, isolate fresh fission products, and then recombine them in specific proportions. LASRS offers a unique opportunity to perform these activations in-house and reduce transit time and the associated loss due to radioactive decay, thereby significantly extending the useful range of the technique. B194's neutron beam is expected to be an order of magnitude greater than the Crocker facility at UC Davis. This provides radiochemists with the means to produce more trace isotopes that can be used in the NTNF mission here at LLNL. The charged-particle accelerator that drives the neutron source also provides a high-brightness research opportunity to produce otherwise inaccessible isotopes of interest.

The detection of nuclear materials presents another demand for improved nuclear data from LASRS. One specific example is the detection of ^{235}U through the measurement of time-correlated fission neutrons. The photo-fission distribution of neutrons is essentially unmeasured, making it difficult to apply this technique for the detection of sub-kilogram quantities of HEU. The bremsstrahlung photon source in LASRS provides the capability to perform these measurements. A second example involves the use of nuclear resonance fluorescence (NRF) for isotopic identification of nuclear materials. These large, narrow resonances are excited by highly penetrating MeV photons and provide the primary motivation for the development of mono-energetic photon beams through the Compton scattering on electron beams. While research on the generation of these sources continues, access to an intense bremsstrahlung source will enable the continued study of NRF probes on nuclear materials.

In support of its missions, the laboratory has spent over 10 years advancing the technology for a novel and innovative non-destructive diagnostic called Fast Neutron Imaging, or simply Neutron Imaging (NI). This technique enables generating high-resolution radiographs of details hidden inside heavily shielded objects that X-ray technology is ineffective at imaging. Neutron Imaging is effective because neutrons are highly penetrating in dense materials like uranium; enabling the creation of images in regions that X-rays cannot access. A major challenge with advancing neutron imaging to date is the lack of fast-neutron sources with sufficient luminosity. The NI project has designed an affordable, compact commercially-rendered system for reliably generating and efficiently detecting neutrons in order to render radiographic images for tomographic reconstruction. With the physics demonstration of the concept solidly proven by over a decade of modeling and experiments, the accelerator design has matured through years of development and peer-review processes. Efforts are now underway to build an engineering prototype demonstrator system as the next step in making this new diagnostic technique a reality.

II. B194 Facility Infrastructure

A. Current Utilization

The underground cave complex at B194, shown in Figure 1, was designed to house the 100 MeV electron linear accelerator (LINAC) and deliver a high-intensity electron beam to a number of separate experimental areas. The below ground layout, heavily shielded walls, ceilings, and doors were specifically designed to support the operation of accelerators producing very-high radiation doses. As such, the facility is perfectly suited to host the high-flux neutron and photon sources needed for nuclear and radiochemical science research. The caves currently house three different accelerator-based particle and photon sources:

1. The Neutron Imaging (NI) project is currently building high-current 4 and 7 MeV deuteron ion accelerators at 425 MHz to be used with a deuterium-gas target to generate a high-flux, quasi-monoenergetic neutron source. The 7 MeV accelerator is WCI-funded and, once demonstrated, will be delivered to an off-site facility. The 4 MeV accelerator will continue to be operable in B194 and can be configured as a source of neutrons, protons, deuterons, or alphas for precision cross section measurements and unique capabilities for development of novel radiochemical separations and chemistries. This program occupies the North and Magnet Caves.
2. The MEGa-Ray Test Station (MTS) is a compact, 11.424 GHz (X-band), photoinjector-driven linac capable of generating 30 MeV, ultra-low emittance electron beams and narrow-bandwidth, ultra-high peak brightness X-rays via Laser Compton-Scattering. The MEGa-Ray project is a continuing NIF led effort to develop an extremely narrow-bandwidth, tunable gamma-ray source capable of isotope-specific detection, assay and imaging as well as opening new fundamental nuclear and plasma science opportunities. Near future R&D activities will include an upgrade to the RF system to boost the beam energy up to 100 MeV and the installation of a NIF developed fiber laser system to generate electron-beam bunch trains and increase the light source average flux. The MTS occupies the South and Magnet Caves.
3. The original 2.8545 GHz (S-band) RF LINAC was reconfigured over the past 15 years into a high-precision electron accelerator by installing a photo-injector electron source and a new beam-focusing lattice. This high quality, low-average-current electron-beam source was used in the PLEIADES, TREX/FINDER, and IFEL experiments on intense laser/electron beam interactions. The LINAC RF power system is capable of driving much higher average current beams than are presently being generated. This capability can be exploited by developing a high

repetition rate, fiber laser-driven photo-gun. Coupled to the LINAC this source would provide a 3-100 MeV, 10s of μA average current electron beam suitable to drive intense bremsstrahlung and/or Compton-scattering based photon sources. This experimental apparatus uses the Accelerator, Detector, and Zero Degree Caves (see Figure 1). This LINAC is currently being partially dismantled to make room for the installation of the DTRA-developed PASS (Photon Active Search System) accelerator in the Accelerator Cave.

4. PASS is a state-of-the-art 2.998 GHz (S-band), electron accelerator with a thermionic source. PASS generates beams from 30-60 MeV with an average current up to 30 μA, three orders of magnitude more intense than the original 100 MeV LINAC. The machine has been simulated to run at energies as low as 12 MeV, and energies below this will be explored after the installation is complete. Coupled with a converter target, PASS will operate as a high-flux bremsstrahlung photon source.

In the near term, the realization of these machines as reliable radiation sources will enable and initiate the use of B194 as a user facility to pursue NACS basic science and application driven research as envisioned in the 2016 NACS Strategic Plan [3].

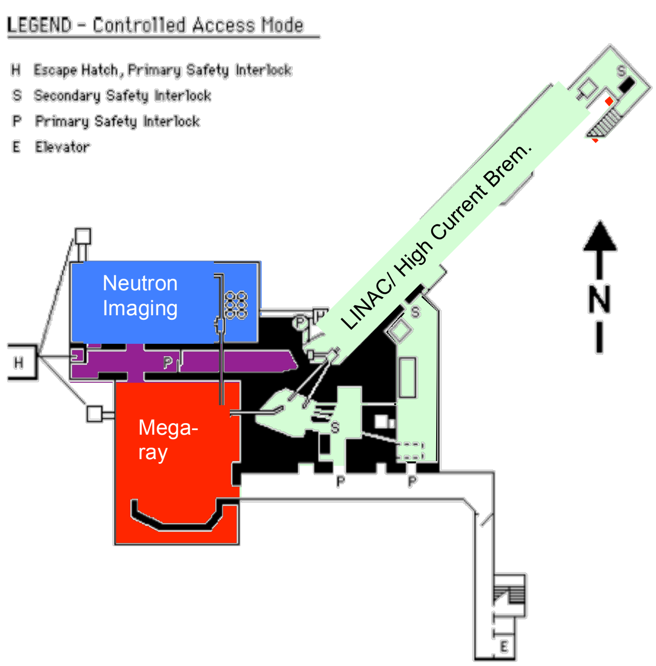

Figure 1. Sketch of B194 Underground Cave System.

B. Near Term Upgrades

As described above, the construction of a 4-MeV neutron accelerator and the installation of the PASS accelerator constitute the current upgrades to the B194 Accelerator Complex. Figure 2 shows the current plan for work on both a 4 MeV source and a 7

MeV source that will be shipped offsite. The 4 MeV machine (DL4) will be online and ready for limited operation sometime in the middle of FY17. Per the current plan, final testing of DL4 is scheduled for May 2017. Both machines will be running (in alternate configuration) throughout FY18, and expectations are that it will be possible to perform measurements in coordination with the NI project during that time. Sometime thereafter the 7-MeV machine will be shipped offsite, leaving the 4-MeV available for continued, dedicated running at LLNL.

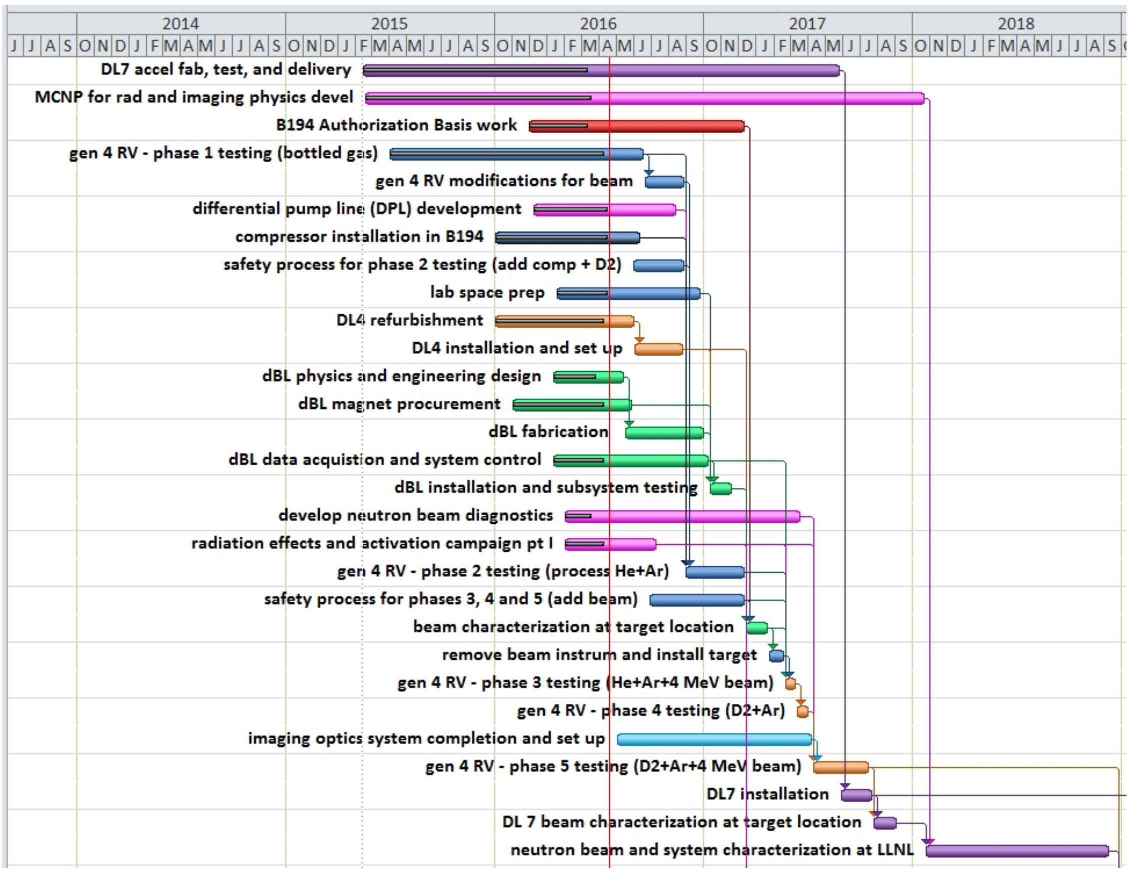

Figure 2. Design and installation schedule for 4-MeV and 7-MeV Neutron Imaging accelerators.

A less detailed timeline for the installation of the PASS accelerator is shown in Figure 3. It begins with the removal of legacy magnets and then continues with work on the modulator and waveguides to be followed by a 4-6 month period of installation for the hardware.

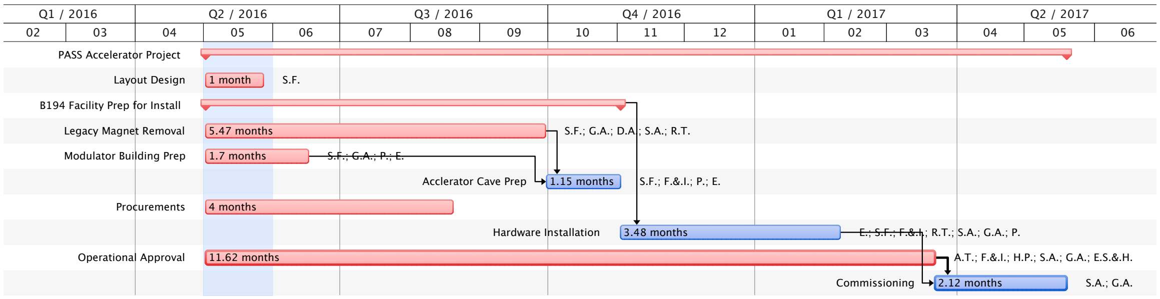

Figure 3. B194 installation schedule for the PASS accelerator.

The acquisition of the PASS accelerator will also open several opportunities in coherent γ-source development. These include coupling the high-current electron beam with a laser recirculation cavity to generate a high-flux coherent light source and developing a novel high efficiency Free-Electron Laser (FEL) capable of converting a significant fraction (on the order of ½) of the > 1 kW average electron beam power into laser power. The resultant FEL light would naturally be locked in timing and pulse format with the generating electrons for Compton scattering.

III. Rate Estimates

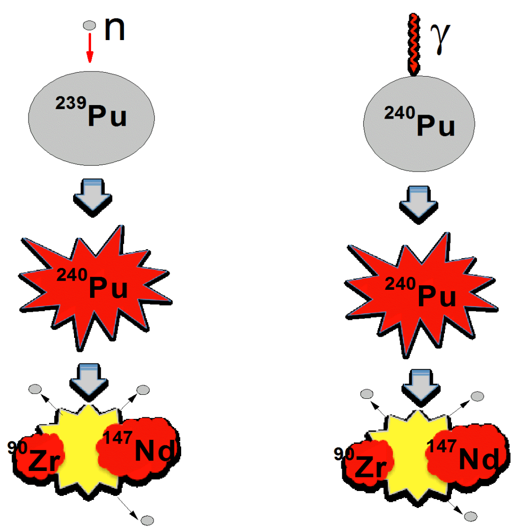

Figure 4. Induced fission reaction via a neutron (left) or photon (right) incident particle leading to the same compound ^{240}Pu nucleus.

In this section, we present rate estimates for measuring fission cross-sections for two reactions, (i) n + ^{239}Pu and (ii) γ + ^{240}Pu, shown in Figure 4. These reactions are related through the "Bohr hypothesis," which states that the decay of a compound nucleus for a given excitation energy, spin, and parity is independent of its formation [4,5]. Taken together, these two measurements would constitute a test of this hypothesis under the important caveat that the two excited states of Pu are likely to occupy different distributions of spin states that will still require some theoretical evaluation. If successfully demonstrated, this approach would allow reactions on *radioactive* targets, such as ^{237}U(n,f), ^{232}U(n,f), ^{238}Pu(n,f), and ^{231}Th(n,f), which are currently impossible because of safety and environmental concerns, to instead be replaced by photo-fission measurements on *stable* targets such as ^{238}U(γ,f), ^{233}U(γ,f), ^{239}Pu(γ,f), and ^{232}Th(γ,f), respectively. However, in this case the main reason for selecting the two Pu reactions is to provide a useful point of reference between the neutron and photon reactions in B194 and at other facilities.

To calculate the neutron rates for the NI source, double-differential (energy and angle) neutron flux spectra were calculated from Legendre polynomial fits to the d(d,n) cross section from Liskien et. al. [6], simulating stepwise energy loss of deuterons through 4 cm of deuterium gas at 3 atm. using stopping powers from ICRU49 [7].

For this calculation we used data for the 7-MeV machine to cover the physics region of interest and to make comparisons over a wider energy range. Figure 5 shows that monoenergetic neutrons from 2-10 MeV are produced as a function of angle from the beamline axis however the flux drops sharply at energies lower than 10 MeV (<20 degrees). For the fluxes considered in this study, it is assumed a three-volume gas cell will be installed, instituting a variable-pressure or variable-length argon degrader in front of the 4 cm of deuterium gas to tune the deuteron beam energy.

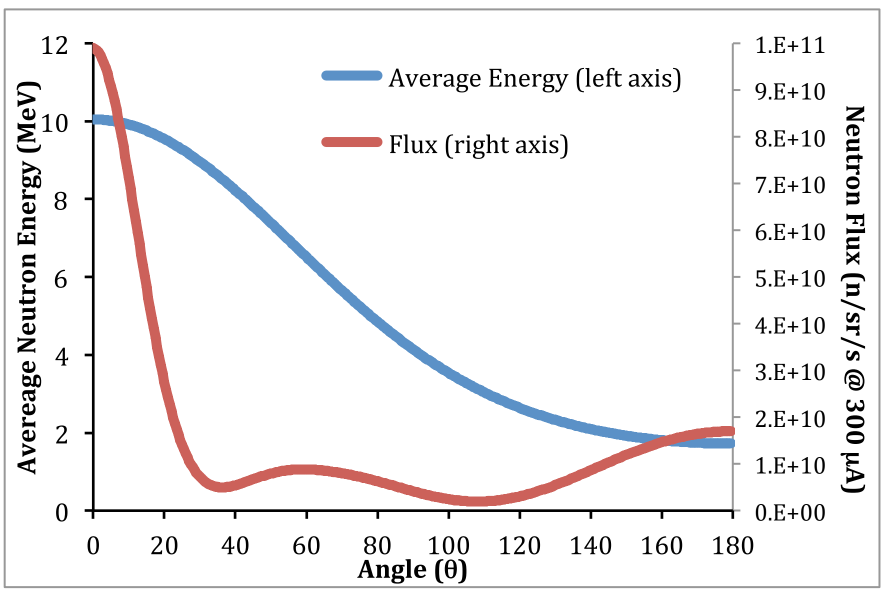

Figure 5. Neutron energy and flux vs. angle for 7-MeV Neutron Imaging machine.

Table 1 shows the energy bins for the accelerated deuteron, neutron, and excited nucleus as well as the incident nuclear flux at one meter, the fission cross-section, and the expected rate of fissions per gram of Pu. The data are divided into 6 energy bins, and cover a range of 2-5E+04 fissions per second. The actual measurement time would depend on additional factors including the detection efficiency. If we assume a 10% detection efficiency and require statistics of 10M events, then each energy would require approximately 10^4 seconds or about 3 hours of beam to complete for a 1-gram sample target. This measurement could easily be coordinated to fit within or around the Neutron Imaging program.

For comparison we show rates for similar measurements at LANSCE using the WNR beamline at 90 degrees and 10 m in Table 2. Because these data are collected simultaneously using gated neutron time-of-flight for the energy determination, the sum over an equivalent six bins is given as 4E+03, roughly an order of magnitude lower than the rates projected for the 7-MeV machine at LLNL. However, it should be noted that the WNR beam line provides simultaneous access to a wider range of neutron energies. Similar calculations for the 88-inch accelerator at LBNL also produce rates in the range of a few times 1E+04, slightly less than the projected rates at Livermore. Actual determination of where to perform this experiment would depend on several factors, including the precise geometry of the experimental setup and availability of beam-time. However, there are many cost and logistical advantages to performing measurements at a local facility.

E_d (MeV)	E_n (MeV)	E_x (MeV)	n/0.5deg/d	n/cm^2/s	σ(b)	Rate (Hz/g)
7	10.05	16.6	1.24E-08	9.73E+06	2.240	5.49E+04
6	9.07	15.6	1.20E-08	9.38E+06	2.247	5.31E+04
5	8.08	14.6	1.07E-08	8.41E+06	2.281	4.83E+04
4	7.06	13.6	9.19E-09	7.19E+06	2.080	3.77E+04
3	6.00	12.5	7.49E-09	5.86E+06	1.772	2.62E+04
2	4.83	11.4	5.43E-09	4.25E+06	1.713	1.83E+04

Table 1. ^{239}Pu(n,f) fission rates at 0-deg. for 7-MeV neutron imaging machine at 1 m distance.

E_n (MeV)	E_x (MeV)	n/cm^2/s	<σ(b)>	Rate (Hz/g)
5—6	11.5—12.5	1.97E+05	1.66	8.24E+02
6—7	12.5—13.5	1.44E+05	1.93	7.01E+02
7—8	13.5—14.5	1.09E+05	2.19	6.00E+02
8—9	14.5—15.5	1.71E+05	2.25	9.67E+02
9—10	15.5—16.5	6.78E+04	2.24	3.83E+02
10—11	16.5—17.5	5.31E+04	2.24	2.99E+02
			TOTAL	**4.21E+03**

Table 2. ^{239}Pu(n,f) rates for WNR at 90-deg. and 10 m distance.

Fission rates for the $\gamma + {}^{240}$Pu reaction were calculated using estimated PASS beam parameters for 40 μA current in a bremsstrahlung MCNP calculation using a Cu target. The beam flux and photo-fission cross-section are shown in Figure 6. These were used to generate the fission rates shown in Table 3. For comparison, rates from the HIGS/TUNL facility are also calculated and shown in Table 4. For the PASS experiments the data from the bremsstrahlung source are collected simultaneously whereas the HIGS beam is mono-energetic with a beam width of approximately 5% of beam energy. The integrated rate for a measurement with PASS is 2E+04 per gram of target, and the average rate for measurements at HIGS approximately half this value. As with the neutron measurements, the choice of facility depends upon additional factors. The analysis of data from a mono-energetic source would be much simpler than for the integrated bremsstrahlung source. In principle, these measurements could be performed at either facility with comparable beam times, and given the rates, could be performed in a timeframe similar to that projected for the neutron induced fission measurements. One additional feature of performing these measurements at LASRS is the ability to use identical targets and detectors for both neutron and photon induced fission experiments thereby reducing systematic uncertainties when comparing results from the two experiments.

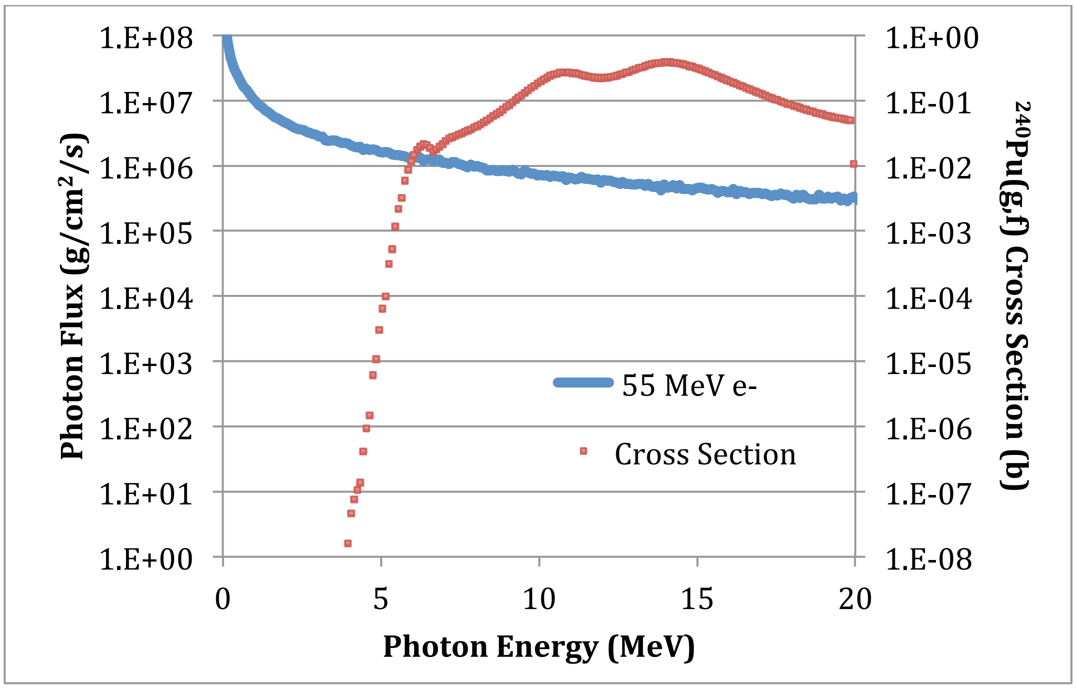

Figure 6. PASS bremsstrahlung photon energy and ^{239}Pu photo-fission cross-section.

Photon (1-MeV bins)	Cross section (b)	^{240}Pu(g,f) rate (Hz/cm)	PASS (MeV)	PASS (Hz/g)
11.4	0.253	1.13E+06	11.4	4.28E+03
12.5	0.239	1.07E+06	12.5	3.43E+03
13.6	0.353	1.58E+06	13.6	4.37E+03
14.6	0.362	1.62E+06	14.6	2.56E+03
15.6	0.252	1.13E+06	15.6	4.02E+03
16.6	0.160	7.15E+05	16.6	1.50E+03
			PASS Total	**2.02E+04**

Table 3. ^{239}Pu photo-fission rates for PASS beams per 1-MeV width of beam on target.

Photon (MeV)	dE/E (5%)	Intensity collimated (g/s)	Flux thru 2.54 cm collimator ($g/cm^2/s$)	Cross section (b)	$^{240}Pu(g,f)$ rate (Hz/cm)	HIGS (Hz/g)
11.4	0.57	9.00E+07	1.78E+07	0.253	1.13E+06	1.13E+04
12.5	0.63	9.00E+07	1.78E+07	0.239	1.07E+06	1.07E+04
13.6	0.68	9.00E+07	1.78E+07	0.353	1.58E+06	1.57E+04
14.6	0.73	9.00E+07	1.78E+07	0.362	1.62E+06	1.61E+04
15.6	0.78	9.00E+07	1.78E+07	0.252	1.13E+06	1.12E+04
16.6	0.83	9.00E+07	1.78E+07	0.160	7.15E+05	7.12E+03
					HIGs Ave	**1.20E+04**

Table 4. ^{239}Pu photo-fission rates for the HIGS beam.

IV. Summary

The Pu fission reactions have been provided as a guide to demonstrate the capability to perform meaningful measurements on-site at LLNL. However, the strength of LASRS lies in its ability to address multiple sponsor needs combined with co-located radiochemical analysis facilities. The measurements mentioned in Section 1 of this document are attractive to a number of LLNL sponsors, including NNSA campaigns, NA-22, DTRA, and DHS. Once current neutron and photon source capabilities are developed, we plan to embark on a series of demonstration projects that will serve as a basis to attract support from these sponsors. Measurements with higher risk and/or greater scientific impact may be proposed for LDRD support once the neutron- and photon-beams become available. The measurements of photo-fission neutron distributions and gamma-strength functions needed for nuclear-structure calculations are examples of projects that would be suitable for LDRD support.

The current upgrade/installation path executed by PLS/NACS and WCI will produce energetic (MeV) beams of mono-energetic neutrons and bremsstrahlung photons within the FY17-18 time frame. We anticipate performing a series of demonstration experiments shortly after beam commissioning. While these experiments are underway, laboratory staff will begin the process of submitting proposals to external sponsors to measure fission yields, evaluate nuclear diagnostics, develop new signatures for special nuclear materials, and provide essential support for the nuclear data and theory program.

V. References

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