Fission Product Yield Measurements from Neutron-Induced Fission of ^{235,238}U and ^{239}Pu

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Abstract. Fission product yields (FPY) are one of the most fundamental quantities that can be measured for a fissioning nucleus and are important for basic and applied nuclear physics. Recent measurements using mono-energetic and pulsed neutron beams generated using Triangle Universities Nuclear Laboratory's tandem accelerator and employing a dual fission chamber setup have produced self-consistent, high-precision data critical for testing fission models for the neutron-induced fission of ^{235,238}U and ^{239}Pu between neutron energies of 0.5 to 15.0 MeV. These data have elucidated a low-energy dependence of FPY for several fission products using irradiations of varying lengths and neutron energies. This paper will discuss new measurements just beginning utilizing a RApid Belt-driven Irradiated Target Transfer System (RABITTS) to measure shorter-lived fission products and the time dependence of fission yields, expanding the measurements from cumulative towards independent fission yields. The uniqueness of these FPY data and the impact on the development of fission theory will be discussed.

1 Introduction

Nuclear fission is a collective phenomenon in which a heavy parent nucleus splits into two daughter nuclei either spontaneously or as a result of some inducement (neutrons, charged particles, photons, etc.). The distribution of fragments and their masses following fission is one of the most basic quantities that has been observed and quantified since the discovery of fission by Hahn and Strassmann [1], and these yields play an important role in many applications such as the estimation of decay heat and delayed neutron emission in nuclear reactors, reactor neutrino studies, radioisotope production for medical applications, development of advanced reactor and transmutation systems, fission in the galactic chemical evolution, and national security, among others. Unfortunately, due to the complexity of the fission phenomenon, the mechanism which determines the mass distribution is not simple and has been a challenge for fission theory using microscopic or phenomenological concepts [2]. While new applications will require accurate energy-dependent fission product yields (FPY) over a broad range of incident neutron energies, the current evaluated FPY data files contain only three energy points: thermal, fast (2 MeV), and 14-MeV incident energies. Currently there is a renewed effort from the fission community to extend and improve this FPY data. Prior work on FPY measurements has concentrated on long (days) irradiations followed by months of γ-ray counting, using an activation technique and fission counters that are described in more detail in [3–7]. In order to more completely understand the fission process, similar measurements have been extended to short-lived fission products in order to "bridge" from independent fission yields to cumulative yields [8, 9]. This is illustrated in Fig. 1 for the $A = 147$ decay chain. While the presentation provided several details of the longer and intermediate irradiation times, this paper describes nascent efforts to measure shorter-lived fission products using activation and subsequent γ-ray counting.

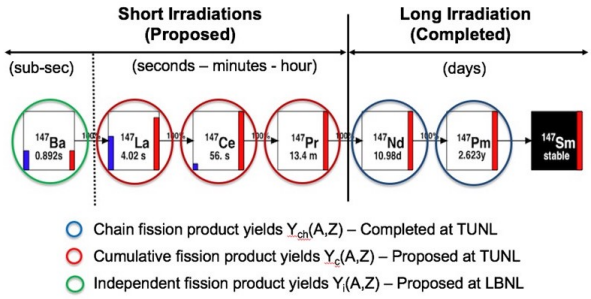

Figure 1. Example decay chain illustrating the complementary information one obtains by varying the lengths of irradiations in order to measure different isotopes with different half-lives.

2 Experimental Details

Since 2011, the LLNL-LANL-TUNL collaboration has undertaken experimental measurements of the energy evo-

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lution of the chain fission-product yields from neutron-induced fission of ^{235,238}U, and ^{239}Pu using quasimonoenergetic neutrons produced at the TUNL 10 MV Van de Graaf accelerator. The FPY measurements have concentrated on reducing experimental uncertainties and filling-in neutron energies for which no FPY data existed in a systematic way using a consistent method. Our method relies on direct (instrumental), post-irradiation γ-ray spectroscopy of γ rays emitted by fission products in actinide targets, with multiple γ-ray spectra taken for a period of at least a few days. High yields of fast monoenergetic neutrons between 0.5 and 15 MeV are produced utilizing the four nuclear reactions ^{7}Li(p,n)^{7}Be, ^{3}H(p,n)^{3}He, ^{2}H(d,n)^{3}He, and ^{3}H(d,n)^{4}He. This technique is described in detail in [3]. During exposure to the neutron beam, dual fission chambers (DFC) with the same isotope as the target of interest have been used to monitor the total fission rate [10]. The chambers contain two thin ($\sim$ 100 μg/cm^2) reference foils and a thicker ($\sim$ 200 mg/cm^2) actinide activation target. The activation target is contained in the center of the chamber while the thin reference foils are up- and down-stream from the activation target. The thick activation target is composed of the same actinide material as the thin reference foils in the adjacent chambers. The advantage of using the DFC method, compared to other methods such as radiochemistry, ratio methods, or mass separation, is that the fission chamber determines the total number of fissions in the target without having to explicitly know either the fission cross section or the neutron flux, greatly reducing the total uncertainty of the measurements. At the same time, performing high-resolution γ-ray counting of the actinide activation target provides the ultimate isotope resolution in (A) and (Z). Thus the ratio of the individual fission-fragment γ-ray yields and the total number of fissions, measured by the DFC, gives the FPYs.

In order to measure shorter-lived fission products, a RApid Belt-driven Irradiated Target Transfer System (RABITTS) was constructed to rapidly transport irradiated samples from the irradiation position to a counting position between two HPGe detectors. Because it isn't possible to transport the DFC due to gas-line and signal cable restrictions, the setup is slightly changed so that the DFC is positioned close to the sample but just downstream, typically within 1-2 cm of the sample position and 3-4 cm from the neutron source. RABITTS is comprised of a Macron Dynamic belt-driven aluminum-extruded linear actuator from iAutomation, controlled by a high power servomotor. Compared to traditional pneumatic systems, this approach has the advantages of an extremely high repeatability of $\pm$33 μm for the irradiation and counting positions. The system is fully automated, with user programmable irradiation time, counting time, and transit speeds up to 10 m/s and a controlled soft acceleration and deceleration. The system is designed to continuously cycle between irradiation and counting positions, performing cyclic activation, until the desired statistical accuracy has been reached. The irradiation setup is shown in Fig. 2(a).

The RABITTS system then transports the sample from the neutron irradiation position, through a shielding wall (see Fig. 2(b)), to a shielded detector counting position (see Fig. 2(c)). Note that the RABITTS is constructed at an angle due to space considerations, but that the sample is normal to the neutron source and to the detectors when in those locations. The distance between the irradiation position and counting position is approximately 7.5 m and the transit time is approximately 1 s. Various cycle times can be used to measure isotopes with shorter half-lives. For the Au measurements, a shorter (approximately 1.5 m) belt driven system was used, and a total cycle time of 81 s was used, consisting of 40 s irradiation time, 0.5 s transit time, 40 s measurement time, and 0.5 s transit time. These measurements are discussed in Section 3.1. Measurements were made with 4.6-MeV neutrons and a ^{238}U target. Several cycle times were used as detailed in Table 1. Note that many cycles are performed during several hours to increase statistics. The neutron-irradiation is discussed in Section 3.2.

Table 1. Cycle times for irradition of ^{238}U with 4.6-MeV neutrons. Note that the transition time refers to the time travelling between the irradiation and counting positions.

Irradiation (s)	Counting (s)	Transit (s)
4	8	1.09
10	20	1.09
60	90	1.09

3 RABITTS Results and Discussion

A test irradiation to produce the 7.7 s isomer in ^{197}Au via the (n,n') reaction provided a critical test of the RABITTS by measuring a previously well-measured short-lived γ-ray emitting isotope. These results are briefly described in Section 3.1. Some preliminary results of the 4.6-MeV neutron irradiation of ^{238}U are discussed in Section 3.2.

3.1 The Gold standard measurement

A 515.27 mg sample of ^{197}Au was irradiated with 2.0-MeV neutrons for 40 s and then transported in approximately 0.5 s to a well shielded γ-ray counting station consisting of two HPGe detectors placed approximately 10 cm apart and counted for 40 s. The neutron energy was chosen to match prior measurements for this well known (n,n') inelastic scattering measurement and the decay of the 7.7 s isomeric state back to the ground state was monitored with the 279-keV γ ray. The sample was cycled back and forth from the irradiation position, to the counting position for just over an hour (3969 s). The details of the experiments, together with the measured γ-ray spectrum and extracted half-life is shown in Fig. 3. The half-life measured in this experiment was 7.75 $\pm$ 0.09 s in agreement with the NNDC half-life of 7.73 $\pm$ 0.06 s. The measured (n,n') cross-section, using the DFC for dose determination, was 507 $\pm$ 32 mb in excellent agreement with the previously accepted value of 510 $\pm$ 42 mb. The results of this experiment provide validation that the technique works for measurement of shorter-lived isotopes and

Figure 2. RABITTS system. A picture of the irradiation position is shown in (a). Note the presence of the Cd-shrouded DFC, black sample holder (on plastic rod) used to minimize mass near the sample, and air-cooled neutron target (Cu tube). The sample is transported on a cart riding an Al extruded track through the shielding wall (b) to a separate cave for counting between two HPGe detectors (c). Neutron generation is automatically shut off using a rapid switching magnet when the sample is in the counting position to further reduce any backgrounds. The sample is normal to the neutron source and each HPGe detector.

encouraged experiments to quantify fission products produced in the neutron-induced fission of ^{235,238}U and ^{239}Pu, amongst other isotopes.

3.2 Results with 4.6 MeV neutrons on ^{238}U

Preliminary results for two different cycle times are shown in Figs. 4 and 5. Because the data are collected in list mode, the measurement time was divided into four equal time periods and the data binned in time to show the growth and decay of various fission product γ rays. In Fig. 4, the counting time was 8 s, so four 2-s duration spectra are compared. There are several γ-ray peaks which clearly decrease in intensity during the total 8 s counting time, indicating observation of fission products with half-lives on the order of a few seconds (see $\sim$ 810-keV and $\sim$ 836-keV for example). In addition, some γ-ray peaks do not change intensity, clearly indicating longer-lived isotopes (see $\sim$ 843-keV and $\sim$ 846-keV for example).

In comparison, spectra with longer irradiation and counting times are shown in Fig. 5. This highlights fission products with seconds to minutes half-lives. Comparing the spectra between Figs. 4 and 5 enables one to determine isotopes with slightly longer half-lives. Note the shorter-lived peaks in Fig. 4 have essentially decayed away because of the longer counting times. This data were collected in August 2019 and is under more detailed analysis to identify all of the short-lived fission products observed and quantify their yields.

4 Summary and Conclusions

We have begun performing self-consistent FPY measurements for isotopes with half-lives of seconds to minutes. The measurements will provide self-consistent, high-precision, and time-dependent FPY data for ^{235,238}U, and ^{239}Pu isotopes using monoenergetic and pulsed neutron beams for incident energies from 0.5 to 15.0 MeV. Using irradiations of varying duration, and a new RABITTS system for short-lived fission products, we will measure the energy dependence of the cumulative and independent yields for more than two dozen fission products. The new measurements will give a complete picture of the fission product yield landscape; from the initial distribution produced directly by fission, through the complex, time-dependent evolution of the yields from β-decay and neutron emission, and finally for the produced and often measured final distribution of stable/long-lived nuclei. The uniqueness of these FPY data will provide insights crucial for the development of fission theory as well as important benchmarks for new data evaluations based on high-precision fission products.

Document number LLNL-PROC-795698-DRAFT. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344, and in part by the US Department of Energy, Office of Nuclear Physics, under Grant No. DE-FG02-97ER41033, and the SSAA Program of the National Nuclear Security Administration under Grant No. DENA0003884.

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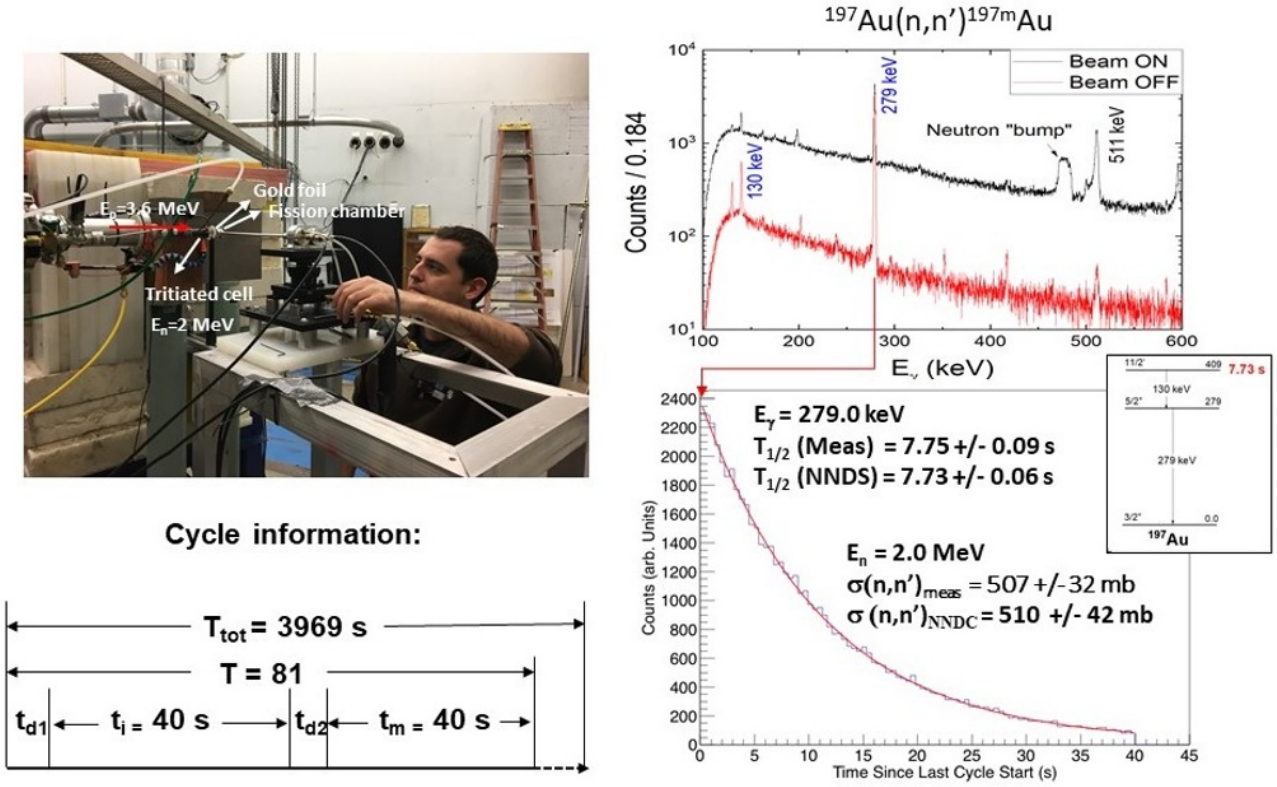

Figure 3. Au standard test of the system. Experimental details and results are discussed in the text.

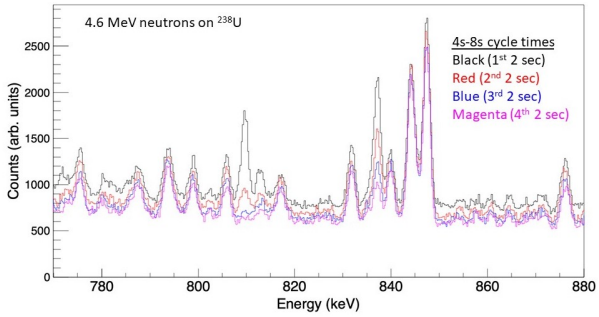

Figure 4. Preliminary data showing the presence of short-lived fission products for a 4 s irradiation/8 s measurement cycle.

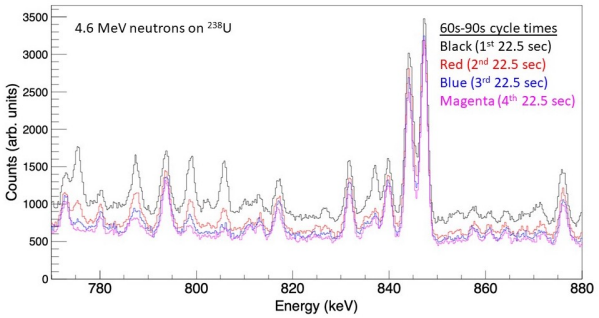

Figure 5. Preliminary data showing the presence of short-lived fission products for a longer 60 s irradiation/90 s measurement cycle.

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