Title: Energy Dependence of Fission Product Yields for 239Pu, 235U, and 238U

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Energy Dependence of Fission Product Yields for ^{239}Pu, ^{235}U, and ^{238}U

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Fission is undergoing a renaissance of interest as both new theoretical and experimental efforts are underway to obtain more precise fission cross section measurements; study fission yields and correlations of prompt fission gamma-rays and neutrons; and, as highlighted in this report, determine the energy dependence of fission product yields.

For the Stockpile Stewardship program the fission yield is an important performance metric. During the nearly 50 year nuclear testing program fission product yields provided the primary diagnostic for determining the fission yield of a device. In the underground testing environment, due to its relatively long half-life, low volatility, actinide similarity and minimal chemical fractionation, ^{147}Nd became the most utilized fission product for obtaining fission yield information. This necessitated an accurate knowledge of the number of times this isotope was produced per fission, i.e. its Fission Product Yield (FPY).

For high-yield fission products, such as ^{147}Nd, the fission product yield was generally assumed to be independent of incident neutron energy, however recently published data [1-3] indicate that the FPY for ^{147}Nd increases in the 0.2 to 1.9 MeV neutron energy region by 3.2%/MeV - 4.7%/MeV depending on what data set was used in the analysis. This is a significant finding that can impact the deduced fission yield. Given this importance, we are pursuing a complete, high-precision, self-consistent study of the energy dependence of fission product yields using mono-energetic neutron beams with the goal of providing accurate (<2% relative uncertainty) information on the energy dependent fission product yields covering an energy range from $E_{th} < E_n < 16$ MeV.

The irradiation setup is shown in Fig. 1. A dual fission chamber is used to count the number of fissions taking place in thin (10 - 100 μg) reference foils during the course of a neutron irradiation. A thick (200 - 400 mg) target placed between the reference foils is activated and then gamma counted for a period of 2 months to quantitatively extract the yields of characteristic gamma-rays from the specific isotopes of interest. Both the reference foils and the thick target are made of similar, isotope-enriched and chemically purified materials of ^{239}Pu, ^{235}U, or ^{238}U. The number of fissions occurring in the thick target is accurately determined by scaling the number of fissions measured in each fission chamber by their mass and solid angle acceptance relative to the thick target. Using standard activation analysis methods the yield of fission products are measured. The FPY for each fission product is then obtained by dividing the yield for each fission product by the total number of fissions in the target.

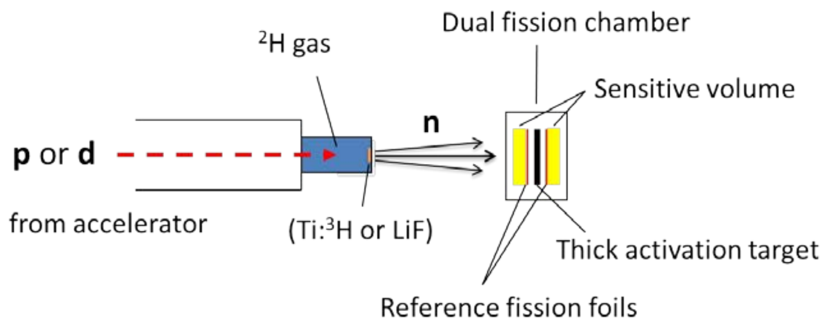

Fig. 1. Schematic of the irradiation setup.

Mono-energetic neutrons are produced at the 10 MV FN Tandem accelerator at the Triangle Universities Nuclear Laboratory (TUNL) on the campus of Duke University. Several different reactions, as listed in Table 1, have been used to produce neutrons of different energies. Typical beam currents were 1.5-2 μA for deuterons and 3-4 μA for protons. To obtain good counting statistics at these fluxes, target irradiations were done for 3 to 7 days. More details about the dual fission chamber and these neutron sources and their backgrounds can be found in Ref. [4].

Table 1. Reactions used to produce mono-energetic neutrons at the TUNL facility.

Reaction	Neutron Energy (MeV)	Typical Flux on Target (n/cm^2-s)
^{7}Li(p,n)	0.025 - 0.65	6 x 10^6
^{3}H(p,n)	1.4 – 3.6*	9 x 10^6
^{2}H(d,n)	4.6 – 14.5*	8 x 10^6 – 3 x 10^7
^{3}H(d,n)	14.8	5 x 10^6

*Off-energy neutrons are produced with increasing intensity above ~2 and 5.5 MeV, respectively, for these two reactions.

To date we have measured 12-15 high-yield fission products for ^{239}Pu, ^{235}U, and ^{238}U at several different neutron energies (0.6, 1.4, 2.4, 4.6, 9, 14.8 MeV). Approximately 2000 hours of beam time have been used for these irradiations and the activated targets have been counted nearly continuously over the last three years. This is a large data set that cannot be presented in detail here (see [5]), but for illustrative purposes we highlight this work in Fig. 2 by presenting preliminary results for ^{147}Nd ($t_{1/2}$=10.98 d, E_γ=531.0 keV) from the neutron-induced fission of ^{239}Pu. These data show a significant increase in the FPY for ^{147}Nd at low energies. Fitting our three lowest data points (0.6, 1.4, and 2.4 MeV) to a line we obtain a slope of (7.1 +/- 2.1) %/MeV. This slope is larger than those determined from previous data by Chadwick and Thompson, but given the large uncertainties, they are not statistically inconsistent.

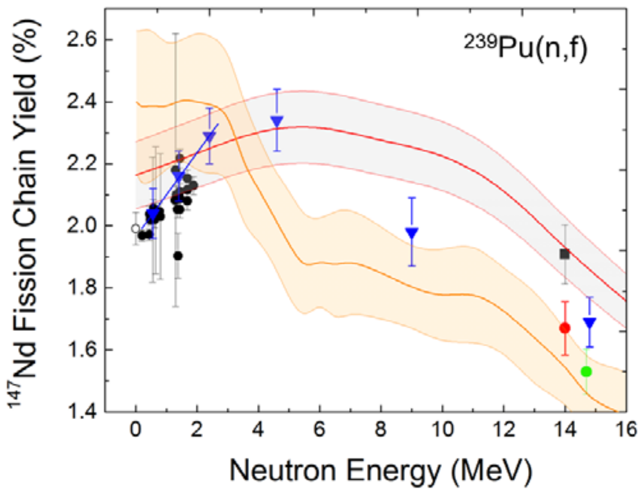

Fig. 2. The energy dependence of ^{147}Nd fission product yield from ^{239}Pu is shown from thermal to 14.8 MeV. TUNL data is shown in blue triangles with a fitted line through the three lowest energies; the open data point is the ENDF/B-VII.1 thermal value; the black, red, and green data points at 14-14.7 MeV are those of MacInnes [8], Nethaway [8], and Laurec [9], respectively. The red line and uncertainty band are the predictions of Lestone [6], and the orange line and shaded area are calculated using the GEF code [7].

At higher energies we see that the FPY for ^{147}Nd turns over and decreases. This trend is predicted by the calculations of Lestone [6], but the estimates of the GEF code [7] shows a flat FPY dependence below 3.5 MeV followed by a sharp decrease at higher energies. To better define the FPY energy dependence, we have recently performed a measurement at 3.5 MeV (in analysis) and are planning to make additional measurements between 4.6 and 14.8 MeV at TUNL as well as a thermal energy measurement at the MIT Nuclear Reactor Laboratory.

At the important 14.8 MeV neutron energy our ^{147}Nd FPY is in good agreement with Nethaway [8], but 2.3 σ below the value presented by MacInnes [8] and 2σ larger than the value of Laurec [9]. Our new measurement helps resolve this long-standing difference between these measurements. Compared to the FPY at 2.4 MeV, the FPY has decreased by 26% at 14.8 MeV. This general trend is expected since the FPYs for fission products in the valley region (A~110-128) and the high-/low-mass wings increase with increasing excitation energy in the fissioning system. In order to maintain the sum of all fission products to be 200%/fission, it is necessary for the higher yield "peak" products such as ^{147}Nd to decrease in relative yield.

Not only do these new results have important applied interest, the FPY increases observed at low-energies are providing valuable fundamental information about the various fission paths and microscopic shell structure and pairing effects that are in delicate balance at energies near or just above the fission barrier. We plan additional measurements to better define these dependences and encourage further theoretical development to improve our modeling and understanding of the nuclear fission process.

References

1. H.D. Selby, et al., Nucl. Data Sheets 111 (2010) 2891.
2. M.B. Chadwick, et al., Nucl. Data Sheets 111 (2010) 2923.

3. I. Thompson, et al., Nucl. Sci. Eng. 171 (2012) 85.
4. C. Bhatia, et al., Nucl. Instrum. Meth. Phys. Res. A 757 (2014) 7.
5. M.E. Gooden, Ph.D. thesis, North Carolina State University (2014); publications in preparation.
6. J. Lestone, Nucl. Data Sheets 112 (2011) 3120.
7. K-H. Schmidt, Europhy. Lett. 83 (2008) 32001, GEF v2.2 (2013).
8. M. MacInnes, et al., Nucl. Data Sheets 112 (2011) 3135.
9. J. Laurec, et al., Nucl. Data Sheets 111 (2010) 2965.