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Energy dependence of fission product yields from ^{235}U , ^{238}U , and ^{239}Pu with monoenergetic neutrons between thermal and 14.8 MeV

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Abstract. Under a joint collaboration between TUNL-LANL-LLNL, a set of absolute fission product yield measurements has been performed. The energy dependence of a number of cumulative fission product yields (FPY) have been measured using quasi-monoenergetic neutron beams for three actinide targets, ^{235}U , ^{238}U and ^{239}Pu , between 0.5 and 14.8 MeV. The FPYs were measured by a combination of fission counting using specially designed dual-fission chambers and γ -ray counting. Each dual-fission chamber is a back-to-back ionization chamber encasing an activation target in the center with thin deposits of the same target isotope in each chamber. This method allows for the direct measurement of the total number of fissions in the activation target with no reference to the fission cross-section, thus reducing uncertainties. γ -ray counting of the activation target was performed on well-shielded HPGe detectors over a period of two months post irradiation to properly identify fission products. Reported are absolute cumulative fission product yields for incident neutron energies of 0.5, 1.37, 2.4, 3.6, 4.6, 5.5, 7.5, 8.9 and 14.8 MeV. Preliminary results from thermal irradiations at the MIT research reactor will also be presented and compared to present data and evaluations. This work was performed under the auspices of the U.S. Department of Energy by Los Alamos National Security, LLC under contract DE-AC52-06NA25396, Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344 and by Duke University and Triangle Universities Nuclear Laboratory through NNSA Stewardship Science Academic Alliance grant No. DE-FG52-09NA29465, DE-FG52-09NA29448 and Office of Nuclear Physics Grant No. DE-FG02-97ER41033.

1. Introduction

Precision measurements of fission observables such as fission product yields (FPY), kinetic energies, neutron emission and intrinsic angular momentum provides insights and constraints for theoretical modeling that attempts to understand this most collective of all nuclear phenomena. A primary motivation for the present work is to address issues raised [1-4] regarding possible energy dependencies of certain fission product yields. The paper by Chadwick et al. [1] presented compelling evidence for a positive energy dependence for the fission product yield of ^{147}Nd from neutron induced fission of ^{239}Pu in the low-energy region between 0.2 and 2 MeV incident neutron energy as measured using critical assemblies and fast reactors. By using monoenergetic neutrons we are able to independently corroborate data extracted from these continuous, broad spectrum neutron sources.

There also existed a lack of reliable measurements connecting the low-energy region (available to critical

assemblies/reactors) and the high-energy region at 14 MeV. Returning again to ^{147}Nd , which has long been used in the national laboratories to determine the number of fissions in a sample, we felt it was important to connect these two regions using a self consistent set of experiments that minimize possible systematic errors. In fact, in the design of modern reactors and advanced fuel cycles, accurate knowledge of the fission product yields as a function of incident neutron energy is crucial. Lastly, it was found that even though a number of measurements are available for the fission product yield of ^{147}Nd at 14 MeV [3], there are large discrepancies [1].

A joint collaboration between Los Alamos National Laboratory (LANL), Lawrence Livermore National Laboratory (LLNL) and the Triangle Universities Nuclear Laboratory (TUNL) was formed. The goal of this collaboration was to measure the energy dependence of various high-yield fission products using monoenergetic neutrons in an accurate and self-consistent manner. This has been accomplished utilizing specially constructed dual-fission chambers and γ -spectroscopy. This is in contrast to the previous data measured mainly radiochemically. Detailed information on our procedure is contained in [5].

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2. Experiment

The energy dependence of the cumulative fission product yields of three actinide targets (^{235}U , ^{238}U and ^{239}Pu) were studied in these measurements and to date have been measured at ten energies: thermal(preliminary), 0.56, 1.37, 2.37, 3.60, 4.56, 5.5, 7.5, 8.90 and 14.8 MeV. Due to the fission threshold of ^{238}U , the lowest energies (thermal and 0.56 MeV) were not measured for that isotope. The first of such measurements was performed at 8.90 MeV [6] using the $\text{D}(\text{d},\text{n})$ reaction and served as an exploratory effort to establish the feasibility of the method. New data is currently being taken at 8.9 MeV to improve statistics and the preliminary data from $^{239}\text{Pu}(\text{n}, \text{f})$ is given, along with recent data that has been measured at the MIT Research Reactor Laboratory at thermal (0.025 eV) neutron energy.

The experiments have been performed at the Triangle Universities Nuclear Laboratory (TUNL) with quasi-monoenergetic neutrons. TUNL possesses a 10 MV FN Tandem Van de Graaff accelerator, capable of accelerating beams of both protons, deuterons and other light ions. Quasi-monoenergetic neutrons are produced at TUNL via the $^{7}\text{Li}(\text{p},\text{n})^{7}\text{Be}$, $^{3}\text{H}(\text{p},\text{n})^{3}\text{He}$, $^{2}\text{H}(\text{d},\text{n})^{3}\text{He}$ and $^{3}\text{H}(\text{d},\text{n})^{4}\text{He}$ reactions. The measurements were performed in what is termed the Neutron Time-of-Flight (NTOF) room at TUNL. This is a large, 10 m \times 10 m room with high ceilings, that helps to reduce the effect of room-return scattered neutrons. Neutron emission is into an angular range of ± 15 degrees around zero degrees.

The cumulative fission product yields were measured using an activation technique. A dual-fission ionization chamber (DFC) (see Sect. 2.1) holding one of the targets of interest was placed in close proximity to the neutron production source. The target was then irradiated for a period of $\sim 4\text{--}6$ days to build-up sufficient activity of fission products. After irradiation, the target was removed from the DFC and γ -ray counted on a highly-shielded High Purity Germanium (HPGe) detector for a period of 1–2 months. The use of the DFC allows for an accurate measure of the absolute fission rate in the activation target by mass scaling from the thin-reference foils to that of the target; all three foils being the same isotope.

2.1. Fission chambers

Each measurement involved a single dual-fission chamber placed in close proximity to the neutron production source using a spacer (15.8 mm), to set the distance from the end of the beam line. A fixed telescope downstream at 0° , was used to adjust the horizontal and vertical positioning of the chamber so that it was centered on the same position as the neutron production source. This enabled reproducible positioning of the fission chambers between different measurements. Since ^{235}U and ^{239}Pu have substantial thermal cross sections, a cadmium cover (“hat”) was made that fit over the top of the fission chambers, taking advantage of the large thermal-neutron capture cross section of ^{113}Cd (2×10^4 b) to effectively absorb neutrons at or below thermal energies. A BC-501A liquid scintillator neutron detector was also placed ~ 3 m downstream from the neutron production source which served as flux monitor and was used during neutron time-of-flight measurements.

The absolute number of fissions in the activation target (^{235}U , ^{238}U or ^{239}Pu) was determined using specially

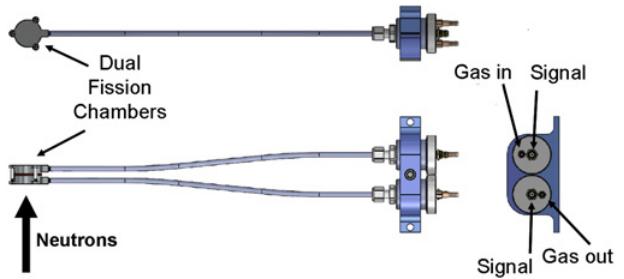


Figure 1. Side and top view of dual-fission chamber assembly showing the gas/signal lines and manifold.

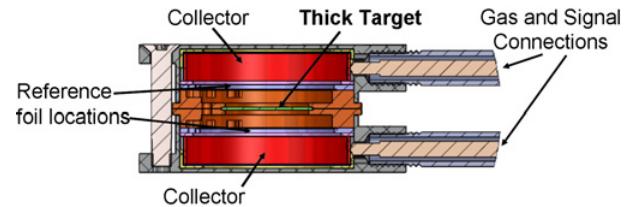


Figure 2. Cut-away view of dual-fission chamber. The green disk represents the activation target foil, while the purple disks show the thin neutron reference foils.

fabricated dual-fission ionization chambers [7], shown in Figs. 1 and 2. A single DFC contains a thick activation target and two thin reference foils of the same isotope. Since the activation target and reference foils in each DFC contain the same isotope, the number of fissions in the target can be accurately determined by a simple mass ratio, and thus a prior knowledge of either the absolute fission cross section or neutron flux, is not required. To avoid removing the reference foils in the chamber and introducing possible uncertainties, three identical chambers were created and each dedicated to one of the target isotopes.

The chambers were adopted from the design developed by Gilliam et al. at NIST [8]. The two individual FCs are mirror images of each other (as shown in Fig. 2). This separation required a gas connection to each FC, as well as separate signal connections. When the two FCs are clamped together, passages connecting the two FCs allow for gas to flow from one FC to the other. Under normal operation, the gas connection to one FC acts as the gas supply, and the gas connection to the second FC as the gas return (as shown in Fig. 1). The mating faces of the two FCs were carefully machined and then lapped so that the assembled device is leak free without requiring a gasket between the two FCs.

The body of each DFC has an outer diameter of 2.50 cm and an overall thickness of 1.73 cm. The small size enables the dual-fission chamber to be placed into a reactor or critical assembly and, in our case, placed close to the neutron production source. This allows us to achieve higher fluxes with a minimum amount of scattering material. The dimensions of the active gas volume (2.21 cm diameter, 0.495 cm thick) were kept the same as in the original NIST design.

A cut-away view of one of the DFCs is shown in Fig. 2. The foils are electroplated on 0.013 cm thick Ti disks. The active area of the deposits are 1.27 cm in diameter. The reference foils for the ^{235}U and ^{238}U chambers are nominally $\sim 100 \mu\text{g}/\text{cm}^2$, while for the ^{239}Pu

Table 1. Fission products for which cumulative fission yields have been reported on.

Measured Fragments		
^{91}Sr	^{92}Sr	^{95}Zr
^{97}Zr	^{99}Mo	^{103}Ru
^{105}Ru	^{127}Sb	^{131}I
^{132}Te	^{133}I	^{135}Xe
^{136}Cs	^{140}Ba	^{143}Ce
^{147}Nd		

FC they are $\sim 10 \mu\text{g}/\text{cm}^2$. The activation targets also have a diameter of 1.27 cm. Their masses are 0.46 g and 0.26 g for the ^{238}U and ^{235}U targets, respectively, and 0.23 g for the ^{239}Pu target.

The efficiency of the chambers were determined using ^{252}Cf sources of known activity, in the same configuration as the reference foils. These measurements gave a fission detection efficiency for both halves of the chambers (up- and downstream) that was consistent with the value reported by NIST [8]. A fission detection efficiency of 98.5% is taken as the fission detection efficiency for the NIST chambers and we have likewise used the same value. A much more detailed discussion of the efficiency of the chambers, as well as a discussion of the effect of the more massive reference foils used for the measurements compared to the ultra-thin ^{252}Cf source used for calibration, can be found on page 326 of Ref. [5].

2.2. Neutron spectra

A series of measurements were made to measure the neutron spectrum at a number of the energies that have been measured, as well as, determine what the room-return contribution at the target position was. For each incident neutron energy reported on, neutron Time-Of-Flight (nTOF) measurements were performed by pulsing the incident proton/deuteron beams and the resulting neutron energy spectrum was measured with the BC-501A scintillator, mentioned above, positioned at 3 meters down stream of the target position. These spectra provided detailed information on the energy spectrum of the neutrons down to a few hundred keV. Activation foils were used at the target position and various nearby positions in an effort to determine the room-return contribution at the target position. A detailed discussion of the nTOF and room-return measurements can be found in [7].

3. Results and discussion

These measurements have provided for the first time the most complete and consistent set of cumulative fission product yields from 0.5 MeV through 14 MeV for ^{235}U , ^{238}U and ^{239}Pu . Table 1 lists all of the fission products for which cumulative yields have been determined in our measurements. Presented in Figs. 3, 4 and 5 are results for the cumulative fission product yield of ^{147}Nd for ^{239}Pu , ^{235}U and ^{238}U , respectively, that highlights the results of the present work. In the following figures, the data

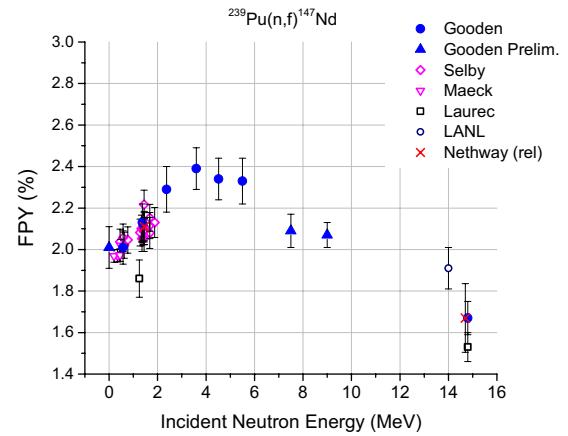


Figure 3. Cumulative fission product yields for ^{147}Nd from the fission of ^{239}Pu . Present data listed as Gooden has been published [5] and Gooden Prelim are unpublished preliminary data.

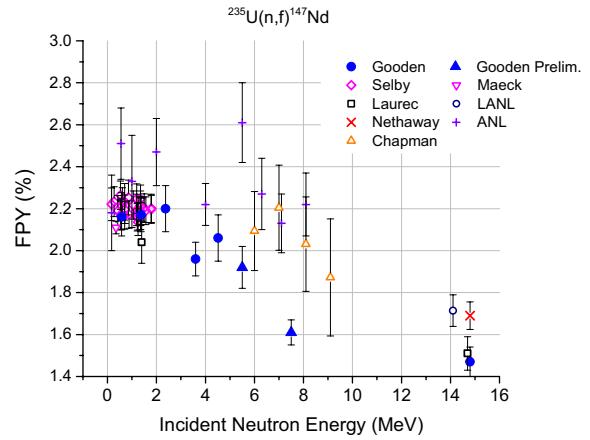


Figure 4. Cumulative fission product yields for ^{147}Nd from the fission of ^{235}U . Present data listed as Gooden has been published [5] and Gooden Prelim are unpublished preliminary data.

labeled Gooden has been previously published [5], while the newest data that has yet to be published is shown as 'Prelim.', and is given a different symbol.

The cumulative fission product yield of ^{147}Nd from ^{239}Pu is of particular interest due to a claimed energy dependence in data obtained from critical assemblies and fast reactors at low-energy ($0.5 \leq E_n \leq 2 \text{ MeV}$) [1]. The results obtained for this isotope are the strongest indication for energy dependencies in the fission product yields, as seen in Fig. 3. The present results with monoenergetic neutrons confirm the observed energy dependence at low-energy, as well as settling discrepant data previously obtained at 14 MeV for the fission product yield of ^{147}Nd from $^{239}\text{Pu}(n, f)$.

Unlike ^{239}Pu , the ^{147}Nd yield from ^{235}U shows essentially no energy dependence in the same low-energy region. In general it is true that for the fission products that show an energy dependence (positive slope) in ^{239}Pu , the same fission products in ^{235}U do not. The results from ^{238}U fall somewhere in-between the other two. It is more difficult in some case to determine an energy dependence in ^{238}U , since only two data points exist in the $0.5 \leq E_n \leq 2 \text{ MeV}$ region versus three for the other isotopes. This is due to the relatively high fission threshold of ^{238}U .

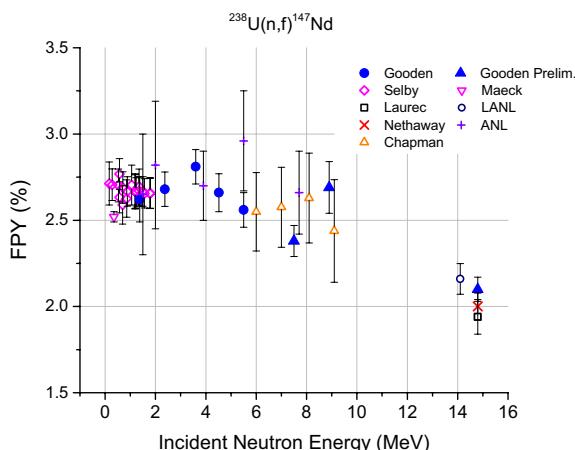


Figure 5. Cumulative fission product yields for ^{147}Nd from the fission of ^{238}U . Present data listed as Gooden has been published [5] and Gooden Prelim are unpublished preliminary data.

More data will be added in the future to this region to better define the energy dependence for all three targets.

Recently, new data was acquired at the thermal (0.025 eV) neutron energy and can be seen in Fig. 3 for ^{239}Pu . This data is the result of a preliminary test measurement. While not shown in Fig. 3, the agreement between the present data and ENDF/B-VII.1 for ^{147}Nd from ^{239}Pu at thermal is good (1–2%). At the time of publication, data from the ^{235}U thermal measurement was not yet available.

4. Conclusions

Data has been acquired on ^{235}U , ^{238}U and ^{239}Pu for nine incident neutron energies between 0.5 and 14.8 MeV. For ^{147}Nd from the neutron induced fission of ^{239}Pu , the present results have confirmed a positive slope to the fission product yield between $0.5 \leq E_n \leq 2$ MeV. Initial data at 8.9 MeV was published previously [6], but is being

updated to greatly improve statistics. This new data, only available for ^{239}Pu currently, has been shown in the figures of the previous section.

New data with thermal neutrons has been taken on ^{235}U and ^{239}Pu . This energy serves as a tie-in of the current series of measurements with what has historically been a well studied neutron energy. In the future, a joint measurement will be performed combining the present method of activation and gamma-ray spectroscopy with radiochemistry, which has traditionally been the main method of determining fission product yields at LANL.

The present data has, in a number of cases, demonstrated previously undetermined energy dependences in the fission product yields of ^{235}U , ^{238}U and ^{239}Pu , most dramatically seen in Fig. 3. More data will be acquired in the 6–12 MeV range to determine what, if any, are the effects of second chance fission on the fission product yields, as well as new data in the $0.5 \leq E_n \leq 4$ MeV range which will serve to strengthen the argument for the energy dependences observed. This will culminate in the most complete set of cumulative fission product yield data on the three major actinides measured.

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