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First Fission Yield Measurements at the National Ignition Facility: 14-MeV Neutron Fission of ^{238}U

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Abstract: The use of the Solid Radiochemistry (SRC) diagnostic platform at the National Ignition Facility (NIF) has allowed the development and implementation of the Target Option Activation Device (TOAD) assembly for fielding materials of interest inside the NIF chamber during high yield neutron shots. Preliminary studies with ultra-depleted uranium have led to the measurement of neutron-induced fission yields for ^{238}U after irradiation with the unique DT neutron source at NIF. The irradiated ^{238}U samples were examined for fission products via gamma spectroscopy, with spectral analysis by the GAMANAL code. Radiochemical procedures were utilized for the separation of fission and reaction products from the target material to provide higher sensitivity measurements. Cumulative fission mass-yields for 32 different isobars, 72≤A≤153, were measured from experiments fielded on two separate NIF shots.

Keywords (4-6 words): 14-MeV neutrons, fission yields, ^{238}U, TOAD, NIF

Introduction

The study of fission product distributions is a fundamental aspect of radiochemistry being revitalized by the current interest in nuclear forensic applications. The identification, development, and certification of new methods for the production of fission products are essential for the fabrication of realistic nuclear debris samples to baseline nuclear forensic capabilities. A special interest is given to fission-yield distributions of actinides from high-energy neutrons (14.1 MeV). These are more difficult to characterize in traditional accelerator facilities due to interferences from down-scattered low energy-neutrons and the saturation of short-lived fission products from long irradiation times.

The National Ignition Facility (NIF) located at the Lawrence Livermore National Laboratory (LLNL) utilizes an indirect-drive, inertial-confinement fusion (ICF) process to induce thermonuclear burn in a deuterium-tritium (DT) filled capsule [1] [2]. The fusion of DT fuel produces a distinct neutron spectrum dominated by 14.1-MeV neutrons from the direct fusion of deuterium and tritium. The capsule is placed at the center of the NIF chamber (10-meter-diameter sphere), along with a number of diagnostic insertion modules (DIMs) that house various diagnostic devices. One of these diagnostics, Solid Radiochemistry (SRC), was developed for the assessment of various performance parameters associated with the capsule implosion, such as areal fuel density and ablator-fuel mixing, through the collection and characterization of debris from the capsule and surrounding material [3]. This debris is collected on various metal foils, 5 cm in diameter, mounted on DIMs at 50 cm from the target center.

A special sealed sample container, Target Option Activation Device (TOAD), has been developed to field materials of interest directly behind SRC collector foils without requiring modifications to the SRC diagnostic platform. The use of the TOAD assembly has been demonstrated by successfully measuring fission yields of ^{238}U in two separate irradiations of uranium-filled TOAD assemblies at NIF. Results presented in this work provide the first fission-yield measurements at NIF and establish the capability for performing other fundamental nuclear measurements.

Experimental

Uranium-loaded TOAD assemblies on NIF shots

The TOAD assembly is a cylindrically shaped aluminum sample holder, 5 cm in diameter and 2 mm in thickness (see Figure 1). The aluminum disc has two centered circular cavities of different dimensions: 26.5 mm in diameter at a depth of 1.35 mm and 36.1 mm in diameter at a depth of 0.7 mm. As a result, the smaller cavity (26.5 mm in diameter, 0.65 mm in thickness) can be covered by another circular aluminum disc of 35 mm diameter and 0.5 mm thickness. This cover plate will isolate any material(s) placed within the smaller cavity (up to 0.36 mL) with the application of a low-vapor-pressure, NIF-approved, epoxy (Torr Seal®). The reliability of this epoxy as a sealant was established through investigations of He-filled TOAD assemblies with a Pfeiffer Smart Test He leak-check unit, before and after testing them on NIF shots. Due to the rigidity of the epoxy when dried, the TOAD assembly is easily opened through the application of mechanical torque.

For ^{238}U fission-yield measurements, an ultra-depleted uranium disc (>99% purity, 99.96/0.039/0.0003 wt% ^{238}U/^{235}U/^{234}U, 239 mg/cm^2) was loaded inside the TOAD assembly along with front-back catcher foils (Al, 99.999%, 6.75 mg/cm^2) and a flux monitor foil (Au, 99.95%, 483 mg/cm^2). Two individual U-TOAD assemblies of identical configuration were prepared and fielded on two separate NIF shots: Al-U-Al-Au-Al. The ultra-depleted uranium was legacy material from LLNL's archival storage that was cleaned prior to use with the following reagents: dilute nitric acid, ethanol, and water. The high-purity Al and Au foils were purchased from Goodfellow and Alfa Aesar, respectively, and used as received.

Fission product separation and gamma spectroscopy

Following each NIF shot, the TOAD assembly was transported to a radiochemistry laboratory for sample preparation. TOAD assembly #1, from NIF shot N130331 (DT neutron yield: 2.97×10^{14} (±1.88%)), was opened, and two samples were prepared for initial counting: uranium foil sandwiched between two catcher foils and gold foil with the last aluminum catcher foil. In order to separate the fission and other reaction products from the source material, after initial counting (~16 hours of total count time), the uranium sample (along with its catcher foils) was dissolved in 6 M HCl/8 M HNO_3. A small amount of Cs carrier (~1 mg) was added to prevent sorption of fission products to the container walls. The sample solution was passed through an anion exchange column (Dowex 1x8, 140 mesh, 10 mL resin in 1 cm diameter column) and radiochemical fractions of interest were selectively eluted for analyses by HPGe detectors. The order of fraction collection was: fission products (8 M HNO_3), thorium (9 M HCl), neptunium (4 M HCl/0.1 M HF), and uranium (water). All reagents were AR grade or better and were used as received. For TOAD assembly #2, on NIF shot N130530 (DT neutron yield: 5.76×10^{14} (±2.05%)), uranium and gold samples were analyzed without any radiochemical separations.

Spectral analysis of the samples counted on HPGe multichannel analyzer systems was performed with the GAMANAL code [4]. The evaluation of the fission and reaction products was primarily based on nuclear data from the Table of Isotopes, 18th Edition [5]. Fission yields were calculated from the ratios of the decay-corrected atoms at shot time for individual fission products to the total number of fissions (extracted from three different reaction products and their corresponding reaction cross sections; Table 1). The uncertainties reported for the fission yields in this study incorporate those from counting statistics, detector efficiency, sample geometry, and the literature nuclear data.

Results and Discussion

A computed DT neutron spectrum from a NIF DT-filled capsule [6] is shown in Figure 2, along with the experimental cross-section data [7] for reactions of interest to calculate total fissions in each TOAD assembly (Table 1). Fusion of DT yields a neutron spectrum dominated by 14.1-MeV neutrons, with a continuum of lower-energy neutrons <1 MeV. Figure 2 indicates the expected neutron spectrum at the capsule, whereas the TOAD assemblies are fielded 50 cm from the capsule. The backscatter of neutrons from the NIF chamber wall and other diagnostic apparatuses fielded inside the chamber will undoubtedly increase the number of lower energy neutrons (<1 MeV). From Figure 2, the fluence of 14.1-MeV neutrons is 25-50x larger than that at lower MeV energies. In conjunction with the reaction thresholds listed in Table 1, it is clear that the calculated total fissions from the reaction products is independent of these low-energy neutrons and strongly based on the 14.1-MeV neutrons and corresponding reaction cross sections. The same holds true for total fissions in ^{238}U. Since the ^{238}U (n,f) cross section is constant over the 13-15 MeV region, the total neutron fluence determined from the monitor reaction products can be converted to total expected fissions, which in turn is used to determine final fission yields. These results are also summarized in Table 1.

Since the neutron yield at the location of the TOAD assembly was only 0.01-0.02% of the 4π shot output, fission yields <0.1% were difficult to measure, even with radiochemical separation of the fission products from the uranium source. A total of 62 individual fission products, from $72 \leq A \leq 161$, were evaluated for in the spectral analysis. For TOAD assembly #1, 32 specific fission products, over $72 \leq A \leq 153$, were reliably identified. However, for TOAD assembly #2, fission yields were determined for only 22 nuclides ($87 \leq A \leq 143$). As expected from the radiochemical separations of TOAD #1, more individual fission products were detected due to lower spectral backgrounds. The final fission-product yields from both TOAD assemblies were averaged to provide a more statistically worthy measurement of ^{238}U fission (Figure 3). Although these measurements were not true cumulative fission yields because each isobaric yield was extracted from only a single fission product, the results compare favorably with cumulative fission yields for 14.1-MeV neutron fission of ^{238}U in the literature [8].

Conclusions

The development of a sealed-source sample holder, the TOAD assembly, has provided a new method for performing nuclear measurements with the distinct 14.1-MeV neutrons produced through the ICF process of DT fuel at NIF. Effective use of the TOAD was demonstrated through the measurement of neutron induced fission yields for ^{238}U. The yields of 32 different isobars were determined from the fission-product distribution produced in an irradiated uranium disc. Radiochemical separations of fission products were essential to measure some low-yield products that could not be observed with nondestructive analyses. The empirical fission mass yields reported here, although limited, are in good agreement with the literature data and comprise the first fission-yield measurements conducted at NIF.

Future work will entail further measurements of ^{238}U fission yields to provide a more comprehensive fission mass-yield distribution. This will be possible in the near future, as the evolution of NIF has recently produced higher neutron yields ($3x10^{15}$) [9], >5x the DT neutron yield in the experiments reported here. Additionally, the neutron spectrum at the TOAD location will be characterized in greater detail by examining (n,2n) and (n,γ) reaction products in different materials having various reaction thresholds. As NIF advances closer to ignition, higher-yield shots will provide more suitable experimental conditions for the study of basic nuclear science phenomena.

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Tables

Table 1. Total fissions in each uranium-filled TOAD assembly.

Reaction	E_{thr} (MeV)	$\sigma_{14.1\ MeV}$ (b) [7]	TOAD Assembly (fissions)	
			#1	#2
$^{238}U(n,2n)^{237}U$	6.18	0.84±0.01	3.04x10^{7}	7.19x10^{7}
$^{197}Au(n,2n)^{196g}Au$	8.11	1.93±0.07	3.20x10^{7}	7.23x10^{7}
$^{197}Au(n,2n)^{196m}Au$	8.71	0.118±0.007	3.58x10^{7}	7.93x10^{7}
Total fissions (average)			3.27x10^{7}	7.45x10^{7}

Figures

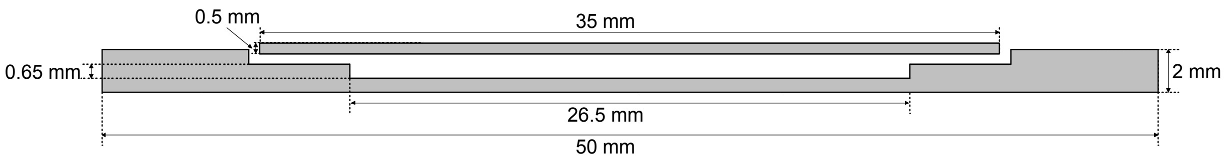

Figure 1. Cross sectional view of the aluminum-based TOAD assembly.

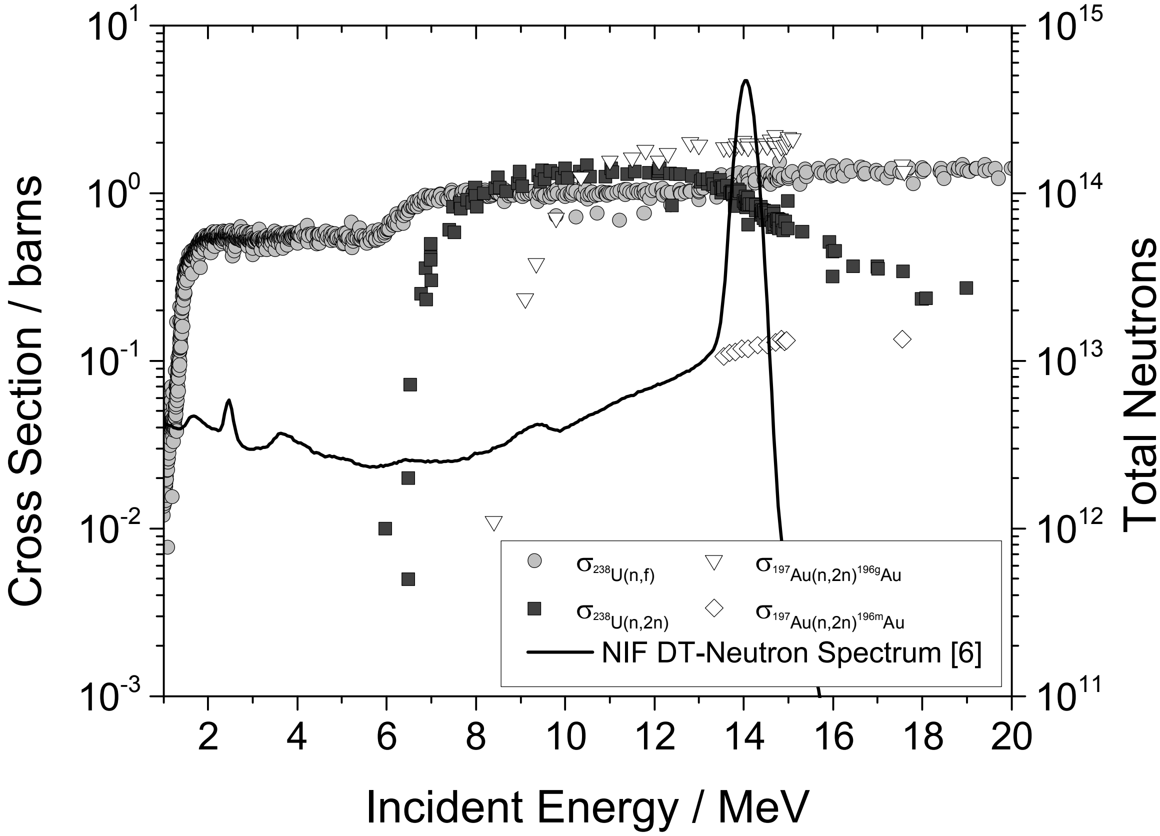

Figure 2. A typical DT neutron spectrum from a NIF shot (normalized to 4.50x10^{15} DT neutrons) [6] and the experimental cross-section data for reactions of interest [7].

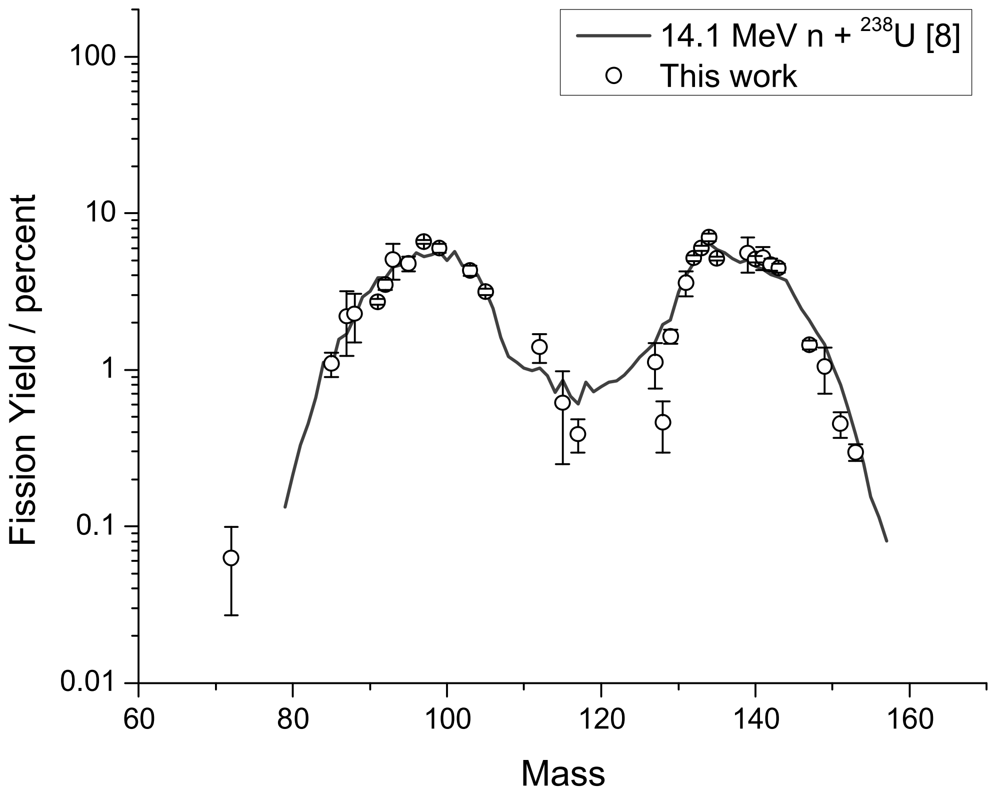

Figure 3. Average cumulative fission yields of ^{238}U(n,f) from two TOAD assemblies, compared to commonly accepted literature values from England and Rider [8].

Figure Captions

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