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Development of a "Fission-proxy" Method for the Measurement of 14-MeV Neutron Fission Yields at CAMS

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Abstract

Relative fission yield measurements were made for 50 fission products from 25.6±0.5 MeV alpha-induced fission of Th-232. Quantitative comparison of these experimentally measured fission yields with the evaluated fission yields from 14-MeV neutron-induced fission of U-235 demonstrates the feasibility of the proposed fission-proxy method. This new technique, based on the Bohr-independence hypothesis, permits the measurement of fission yields from an alternate reaction pathway (Th-232 + 25.6 MeV α → U-236* vs. U-235 + 14-MeV n → U-236*) given that the fission process associated with the same compound nucleus is independent of its formation. Other suitable systems that can potentially be investigated in this manner include (but are not limited to) Pu-239 and U-237.

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Background and Research Objectives

A total fission yield from a nuclear event can be directly measured through radiochemical methods that are independent of fission product cross sections but rely on the knowledge of a bomb fraction (the fraction of the total debris field for a given sample) and cumulative fission product yields (number of atoms of a nuclide produced directly and through its precursors per 100 fissions). Additionally, cumulative fission yields can provide information regarding the composition of the nuclear fuel of a device, which is crucial to the post-detonation nuclear forensic community. Since the fission products in a debris sample are convoluted from the fission yield distributions of the individual nuclear fuel components, analysis of the fission products with known cumulative fission yields can allow for the deconvolution of the fission product distributions, also referred to as the fission split. Thus, the fidelity of a radiochemically measured total fission yield and the deconvolution of the fission split from experimental data are limited by the accuracy of the cumulative fission yields.

The direct measurements of the cumulative fission yields with 14-MeV neutrons in traditional accelerator facilities are unavoidably accompanied by room return (down-scattered, low energy) neutrons. These low energy neutrons are more effective at inducing fission in fissile materials in comparison to 14-MeV neutrons due to the resonances observed in the fission cross sections. As a result, the cumulative fission yields for fissile materials have the highest associated uncertainties for 14-MeV neutrons compared to fission spectrum and thermal neutrons and compared to 14-MeV neutron fission yields from non-fissile materials (Nethaway 1985) (England 1993).

A new proposed method for measuring 14-MeV neutron fission yields is through a fission-proxy reaction method; an indirect method for creating the same compound nucleus as that produced from 14-MeV neutrons on fissile isotopes. This is based primarily on the Bohr-independence hypothesis, also known as the compound-nucleus model, which states that the "only memory that the system (the compound nucleus undergoing fission) maintains is that of the constants of the motion: momentum, angular momentum, number of nucleons, parity, energy and isotopic spin" (Fluss 1969). In other words, once the compound nucleus is formed, it proceeds to decay into its reaction products independent of its formation process.

This proposed fission-proxy method may be validated by measuring fission yields from 25.6 MeV α-induced fission of Th-232, which simulates the same excited compound nucleus (U-236*, E*=20.6 MeV) as that produced from 14-MeV neutrons on U-235. Initial calculations indicate a large overlap in the angular momentum distribution for this compound nucleus from both reactions (Figure 1). Therefore, it is expected to undergo fission in the same manner, independent of the production reaction pathway. The 14-MeV neutron fission yields for U-235 are well-characterized (England 1993), whereas the proposed proxy reaction fission yields, Th-232 + 25.6 MeV α, are not well known (no evaluated fission yields).

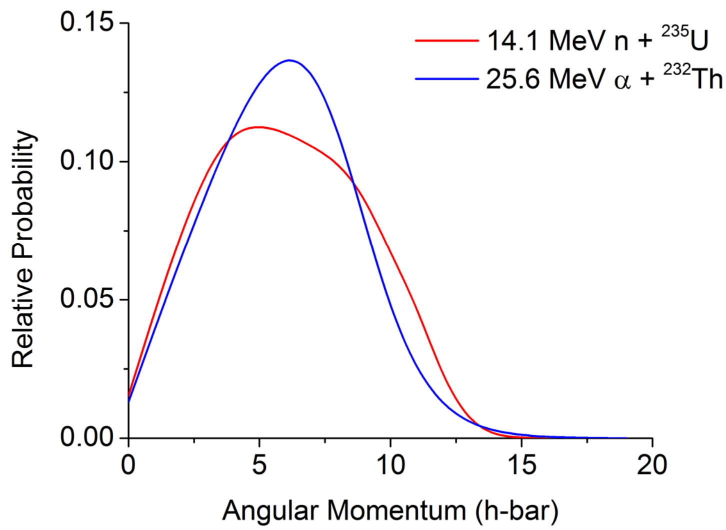

Figure 1. Calculation of the angular momentum for the compound nucleus, U-236*, from two different reaction pathways.

The α-induced fission yields for Th-232 in the literature are very limited (Foreman 1959) (Hicks 1962) (Ford 1965) (Nethaway 1965) (McHugh 1968), having been measured prior to the major developments in high-resolution gamma-ray detectors (Eberth 2008). Hence, these measurements were made strictly through beta counting following extensive radiochemistry separation procedures. Given the inadequate literature data for fission yields from Th-232 + 25.6 MeV α, it was difficult to assess the feasibility of the fission-proxy method without significant improvement of the available fission yields.

The objective of this project was to investigate the feasibility of the fission-proxy method by performing a detailed comparison of the fission yields from the two different reactions as outlined above (Th-232 + 25.6 MeV α vs. U-235 + 14-MeV n). Since the literature data for the Th-232 system is limited, a series of α-induced fission yield measurements on Th-232 were performed using the Tandem Van de Graaff accelerator located on-site at the Center for Accelerator Mass Spectrometry (CAMS), along with the use of the radiochemistry laboratories and the Nuclear Counting Facility (NCF) located in B151. These experimentally measured fission yields were then compared to the evaluated England and Rider (1993) fission yields for 14-MeV neutrons on U-235 to assess the fidelity of the fission-proxy method. The original proposal had outlined fission yield measurements for 45 fission products but the actual measurements exceeded that number – a total of 50 fission product yields were measured. The results, summarized in the next section, indicate that the proposed fission-proxy method is a suitable technique for measuring 14-MeV neutron fission yields through an appropriate alternate reaction pathway. As such, it appears that the application of the Bohr-independence hypothesis for the measurement of fission yields is acceptable.

Scientific Approach and Accomplishments

Fission yield measurements were performed by irradiating natural thorium foils (Th-232) with alpha particles in a foil stack configuration that included aluminum catcher foils to ensure the complete collection of the fission products due to recoil. Three irradiation experiments were conducted at the CAMS facility during FY16 (03/31/2016, 05/23/2016, 08/19/2016-08/22/2016). The first experiment, CAMS-N160331, utilized a thick thorium foil, 28 mg/cm^2, where the beam energy loss through the foil was significant, 2.6 MeV. Additionally, the thickness of catcher foils, 2.2 mg/cm^2, were not sufficient to stop the recoil of the fission products. Thus, the results from the first experiment were not included in the final fission yield calculations, but formed the basis of the final experimental design utilized for the next two irradiations – including radiation counting methods.

The second experiment, CAMS-N160523, used a thin thorium foil purchased from Goodfellow (11 mg/cm^2, 99.5% purity) with 4.3 mg/cm^2 and 5.8 mg/cm^2 aluminum catcher foils, front and back, respectively. The relative beam intensity on target was monitored throughout the 5.5 hours of beam-on-target at ~90 pnA current. Based on stopping-power tables generated through the SRIM module (Ziegler 2010), the alpha beam energy through the thorium foil was calculated to be 25.6±0.5 MeV with the uncertainty corresponding to the energy loss through the foil. This calculation assumes incident beam energy is accurate within 10 keV (conservative estimate for Van-de-Graaff accelerators) and does not take into consideration energy straggling through the foil stack. Following the irradiation, the thorium foil along with its catcher foils were dissolved, an aliquot was taken for direct counting (for analysis of the high yield refractory fission products), and separate radiochemical procedures were performed on the remaining solution to quantify low-yield fission products of interest (cadmium, silver, lanthanides). The last set of irradiation experiments, CAMS-N160819 (one irradiation on

08/19/16 and another on 08/22/16), which mirrored the experimental setup of CAMS-N160523, resulted in a whole thorium foil sample for direct counting and chemical fractions of zinc, niobium and palladium.

Irradiated foils and chemical fractions were counted on coaxial and planar HPGe detectors where gamma-ray spectral evaluation was performed with the GAMANAL code (Gunnik 1972). When possible, decay curve analysis was utilized to further ensure the validity of the fission product measurement. The uncertainties associated with the measured fission yields (cumulative and independent/shielded) are one-sigma and include counting statistics, uncertainty in the half-life (NNDC 2016) and photon intensities (Firestone 1996). The final data set of measured relative fission yields (55 measurements covering 50 different fission products) for Th-232 + 25.6 MeV α is presented on Table 1.

Table 1. Measured relative fission yields (c=cumulative, c*=isomer/cumulative, s=shielded, i=independent (ingrowth correction from parent)) for Th-232 + 25.6±0.5 MeV α.

Fission Product	Reference	Fission Yield (rel. to Ref.)	% Error	Type
Zn-72	Sr-91	0.00249	18.1	c
Sr-91	Zr-97	0.901	7.0	c
Y-93	Zr-97	1.10	13.9	c
Y-93	Ce-141	1.13	15.2	c
Zr-95	Zr-97	1.05	0.5	c
Nb-96	Sr-91	0.000762	10.7	s
Mo-99	Zr-97	1.001	2.0	c
Ru-103	Zr-97	0.642	1.2	c
Ru-105	Ru-103	0.645	1.3	c
Rh-105	Zr-97	0.455	2.5	c
Ru-106	Ru-103	0.593	9.1	c
Ag-111	Zr-97	0.265	4.7	c
Pd-111m	Pd-109	0.0138	23.1	c*
Pd-112	Pd-109	1.14	15.3	c
Ag-113	Ag-111	1.25	6.9	c
Cd-115m	Cd-115	0.627	45.2	c*
Cd-117	Cd-115	0.591	4.0	c
Cd-117m	Cd-115	0.217	4.1	c*
Sb-124	Zr-97	0.00716	5.1	s
Sn-125	Zr-97	0.181	30.0	c
Sb-126	Zr-97	0.0596	2.2	s
Sb-127	Zr-97	0.388	2.5	c
Sb-128	Zr-97	0.174	5.5	c
Te-129m	Zr-97	0.164	36.2	c*

I-131	Zr-97	0.964	1.0	c
Te-131m	Zr-97	0.282	3.5	c*
Te-132	Zr-97	0.831	4.0	c
I-133	Zr-97	1.15	3.1	c
Xe-133	Zr-97	0.0751	35.4	i
Cs-134	Cs-136	0.280	29.0	s
Cs-136	Zr-97	0.0558	5.2	s
Cs-136	Cs-137	0.0523	6.3	s
Cs-137	Zr-97	1.05	3.3	c
Ba-140	Zr-97	0.877	1.1	c
La-140	Zr-97	0.0126	9.0	i
Ce-141	Zr-97	0.870	0.9	c
La-142	Ce-141	1.00	7.5	c
Ce-143	Zr-97	0.713	1.3	c
Ce-143	Ce-141	0.849	3.4	c
Ce-144	Ce-141	0.693	4.0	c
Pr-145	Ce-141	0.531	3.9	c
Nd-147	Zr-97	0.312	4.4	c
Nd-147	Ce-141	0.373	4.1	c
Nd-149	Ce-141	0.214	10.0	c
Pm-149	Ce-141	0.194	7.0	c
Pm-151	Zr-97	0.0850	5.6	c
Pm-151	Ce-141	0.103	4.8	c
Sm-153	Ce-141	0.0493	2.7	c
Eu-155	Ce-141	0.0175	19.1	c
Sm-156	Ce-141	0.0169	16.2	c
Eu-156	Zr-97	0.0123	10.3	c
Eu-156	Ce-141	0.0145	9.3	c
Eu-157	Ce-141	0.00950	19.1	c
Gd-159	Ce-141	0.00419	30.1	c
Tb-161	Ce-141	0.00255	6.8	c

Qualitative comparison of these relative fission yields (normalized to Zr-97) to those from U-235 + 14-MeV n are presented in Figure 2. The similarities between the measured cumulative fission yields (black circles) and the U-235 literature data (gray line) is very obvious but in order to provide a more quantitative comparison, the same data set is converted to a ratio of ratios, S-value:

$$S_i = \frac{\left(\frac{FY_i}{FY_{ref}}\right)_{Th-232+25.6\ MeV\ \alpha}}{\left(\frac{FY_i}{FY_{ref}}\right)_{U-235+14\ MeV\ n}} \quad (1)$$

If the fission yields from both systems are in agreement, the population distribution of the S-value from the 55 measurements (Table 1) should be centered on unity, as is evident in Figure 3. Although this further confirms the similarities in the fission yields, it does not demonstrate the impact of the uncertainties associated with the experimental measurements and the evaluated fission yields. Thus, the distribution from the individual measurements is provided in Figure 4. The plot on the left is color-coded for the type of fission yield, where those furthest from unity are either shielded or isomer fission yields with large associated uncertainties. The plot on the right is color-coded to demonstrate the uncertainties associated with the evaluated fission yields from U-235 + 14-MeV n. The fission yield measurements with S-values furthest from unity correspond to those with the largest uncertainties in the literature data. Lastly, a reduced chi-squared of 2.4 was calculated for these 55 measurements as compared to the U-235 literature fission yields. If the extremely poor fission yield measurements from literature (those with >64% uncertainties) are excluded from consideration, a reduced chi-squared of 1.3 is obtained for 49 fission yield measurements.

Therefore, given the difficulties associated with measuring of fission yields from extremely complex gamma-ray spectra in addition to the large uncertainties in both experimental and literature data, the agreement between the fission yields from these two systems is significant. Although repeating some of these measurements (valley fission products, Pd/Ag/Cd, and the lightest/heaviest fission product, Zn/Tb) is necessary to confirm the few discrepancies, the measured fission yield set as a whole clearly supports the application of the compound-nucleus “fission-proxy” model for the measurement of fission yields.

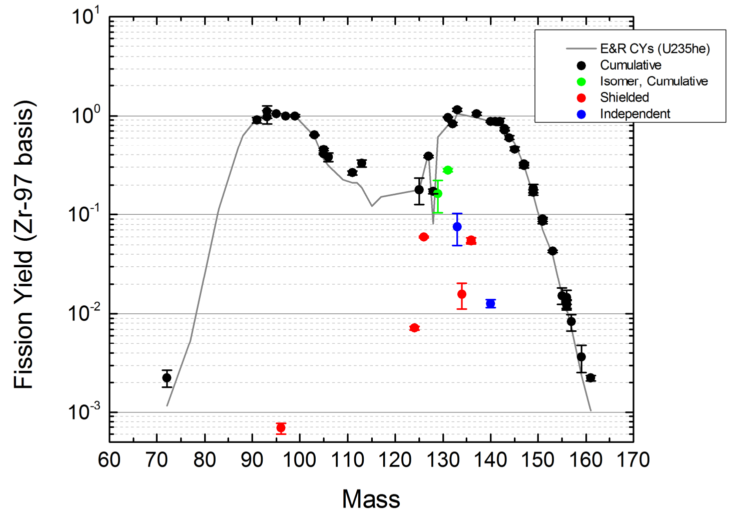

Figure 2. Experimentally measured fission yield distribution from Th-232 + 25.6 MeV α vs. U-235 + 14-MeV n (normalized to Zr-97).

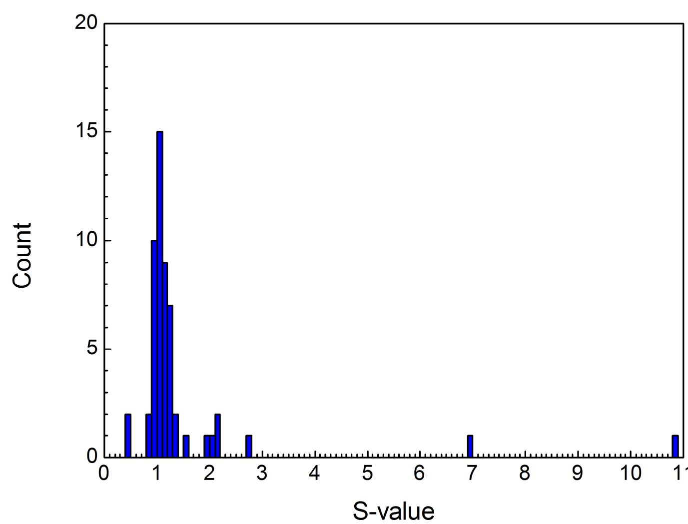

Figure 3. Population distribution of the S-value for the 55 fission yield measurements where 47% of those were within 10% of unity, 67% were within 20% and 80% were within 30%.

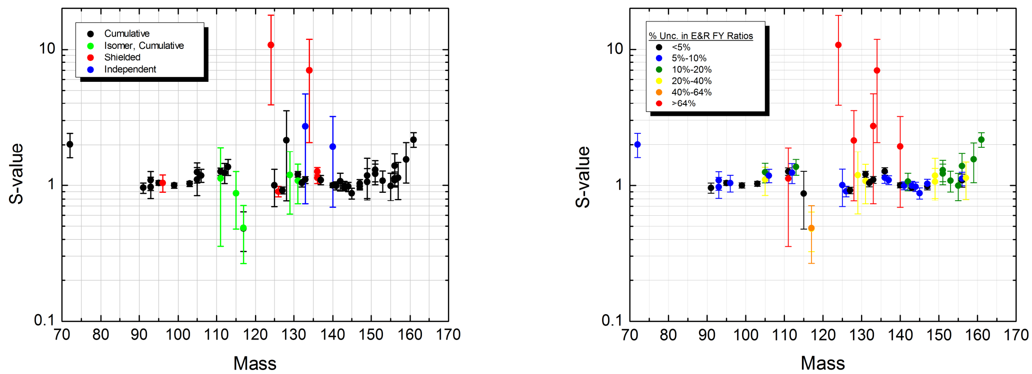

Figure 4. S-value for all 55 fission yield measurements – color-coded for type of fission yield (left) and uncertainty associated with the evaluated fission yields for U-235 (right).

Impact on Mission

By demonstrating the feasibility of the fission-proxy method, fission yield measurements for difficult-to-study systems relevant to Stockpile Stewardship are now possible (e.g., Pu-239 and U-237 for which proper fission-proxy reactions have been identified). Furthermore, applications of the fission-proxy method for the production of fission products that mimic unique fission product distributions are of interest for internal (Global Security) and external (DTRA) programs in support of the realistic post-detonation debris production and validation of such forensic debris data.

Conclusion

We have successfully demonstrated the feasibility of the proposed fission-proxy method. It will allow for the fission yield measurements of more difficult-to-study systems where the unavailability of target material (e.g., U-237) makes direct measurements with neutron-based experiments impossible. Publication of the results in a peer-reviewed journal (in preparation) will allow for further confirmation of this technique by the scientific community. We plan to reach out to external (DTRA) and internal (WCI and GS) programs for support of continued research and development of the fission-proxy method.

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