LA-UR-23-24185 SRNL-STI-2023-00179

Neutron Coincidence Measurements of Uranium-233 Oxide

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Abstract

Renewed international interest in thorium-fueled advanced reactors has challenged the safeguards community to address future proliferation concerns. Thorium-based technology presents many benefits but does not eliminate the proliferation risks associated with producing and processing fissile material. A byproduct of thorium-fueled reactors is uranium-233, which is classified as a direct-use material. As a result, the development of new or improved methods to characterize and measure materials containing ^{233}U must mirror the pace of development of reactors and facilities that produce such material. Research is underway to assess, develop, and test approaches for safeguarding nuclear materials within the thorium fuel cycle. Neutron signatures from the nondestructive assay (NDA) of materials containing ^{233}U are being quantified to inform the potential characterization of these materials. Using a traditional neutron coincidence counter and a series of well-documented ^{233}U oxide samples, initial measurements have been made to assess the feasibility of ^{233}U characterization and discrimination from other uranium isotopes, primarily ^{235}U, using a combination of measurement techniques and analysis methods. Data acquisition is performed in list mode, allowing for a variety of analyses to be performed on the raw data that is not available using traditional shift register technology. Measurements were performed in passive and active configurations to quantify the strength of signal and to validate simulations in support of this work. This paper presents and discusses the results of the initial measurements of ^{233}U oxide performed at Oak Ridge National Laboratory.

1. Introduction

Neutron coincidence counting is a common tool used in the characterization of special nuclear material (SNM) for safeguards. There are well-established methods for measuring items containing ^{235}U, ^{238}U and Pu, but there is not a standard method for the characterization of ^{233}U. However. ^{233}U is often collocated with other fertile and fissile isotopes or contaminated with ^{232}U. Before tackling the larger challenge of determining isotopic ratios and quantifying the amount of SNM present in items containing more than one isotope, the basic signatures of ^{233}U must be studied before additional factors are considered. The first step of a multi-year effort to understand this problem further and develop new methods to characterize SNM containing ^{233}U is to take a systematic assay of well-known ^{233}U items with instrumentation and data acquisition that allows further exploration and deeper analysis of potentially unique signatures.

The mass calibration curve is a common technique used in the assay of uranium. If a series of similar items (similar geometry and composition) but varying masses are measured in the same configuration, a calibration curve can be established and used to calculate the mass of an unknown item. Uranium items composed of primarily ^{235}U and ^{238}U do not have a strong passive neutron signatures and are traditionally measured using an Active Well Coincidence Counter (AWCC) or comparable detection system. When performing an active measurement, AmLi interrogation sources induce fission in the item and a coincidence count rate (doubles) can be measured and plotted as a function of fissile mass.

^{233}U has a much shorter half-life ($t_{1/2} = 160{,}000$ yr) than ^{235}U and ^{238}U and ^{233}U primarily undergoes alpha decay. In oxide form, the alpha particles cause an (α, n) reaction to occur when they interact with the oxygen and emit a measurable neutron signature. A fraction of the (α, n) neutrons may also induce fission in the ^{233}U, called self-interrogation. However, self-interrogation is likely negligible in smaller mass items.

Passive and active neutron coincidence measurements of ^{233}U oxide items ranging from 2 to 80 g were taken in a Large Volume Active Coincidence Counter (LV-AWCC). The design and operating parameters for the counter, the ^{233}U item descriptions, and data acquisition are described in Section 2. The results of both passive and active measurements, including count rates and mass calibration curves, are provided in Section 3. A discussion of the results and ongoing work is provided in Section 4.

2. Method

a. Large Volume Active Well Coincidence Counter

The Active Well Coincidence Counter (AWCC) is a gas-filled well counter that is common among the safeguards community and was developed to assay uranium fuel through active interrogation with americium lithium (AmLi) neutron sources. The AmLi sources are removable, allowing the detector to operate in passive mode. The traditional AWCC design contains 42 ^{3}He tubes arranged in two concentric rings around a central cavity [1]. However, this experiment was conducted with a Large Volume Active Well Coincidence Counter (LV-AWCC) that is simply a scaled-up version of the AWCC. The LV-AWCC contains 48 ^{3}He tubes and its dimensions are scaled by a factor. Removable nickel reflectors and a cadmium liner allow the user to optimize the system configuration depending on the application. LV-AWCC configuration without the Cd liner and Ni reflector was used in these measurements. A schematic diagram of AWCC is shown in Figure 1 and an MCNP model of the LV-AWCC is shown in Figure 2. The nominal operating parameters for both detector designs are given in Table 1 [2].

Table 1: Operating parameters for Active Well Coincidence Counter

Detector	**LV-AWCC**	**AWCC**
Predelay	3 µs	4.5 µs
Gate length	64 µs	64 µs
High voltage	1700 V	1680 V
Die away time	50 µs	50 µs
Efficiency	0.32	0.28
Multiplicity DT (1e-9)	150	224
DT coefficient A (1e-6)	0.60	0.763
DT coefficient B (1e-12)	0.19	0.248

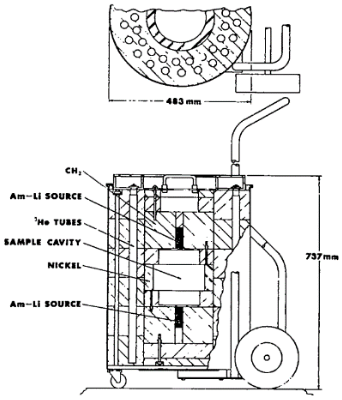

Figure 1: Diagram of the Active Well Coincidence Counter

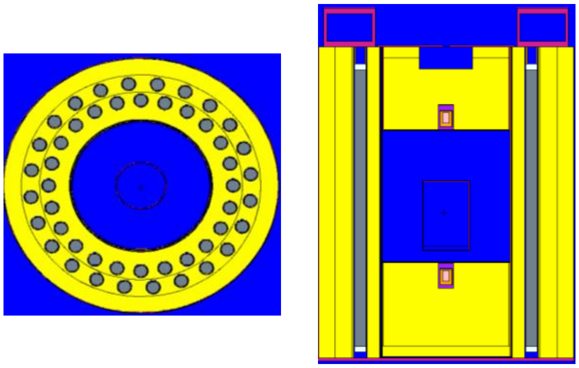

Figure 2: MCNP model of the Large Volume Active Well Coincidence Counter (LV-AWCC)

b. Uranium-233 Oxide Sources

A series of standardized ^{233}U oxide sources were used in these measurements to systematically measure up to 80 g of fissile mass. These sources were created as equivalent sphere radiation signature training devices (RSTDs), which are sealed sources (or assemblies of sources) that emulate the radiation signature of larger masses of special nuclear material (SNM) for passive detection [3]. Each source is an equilateral triangle shaped single encapsulation made of a titanium inner container and a stainless-steel outer encapsulation that is tungsten inert gas seal welded. Two sets of twenty triangles were measured. The uranium in the oxide is 99.75% ^{233}U. In the first set, each triangle contains 2.000 g of total uranium, and the second set is comprised of ten triangles with 1.805 g U, eight triangles with 2.200 g U, and two triangles with 2.196 g U. The total mass of each triangle is 59-63 g. They were designed to be arranged in a spherical configuration, but for these measurements, they were stacked vertically. Stacking the triangles allows the total mass to be increased and measured in increments from 2g to 80g. When stacking the triangles, 1 to 10 triangles (2-20 g U) were placed in one stack, 11-20 triangles (22-40 g U) were split between 2 stacks, and 21-40 triangles (42-80g U) were divided between 4 stacks. An image of the ^{233}U triangles is provided in Figure 3.

Figure 3: Eight ^{233}U oxide triangles in case

c. Data Acquisition

Data was acquired from the LV-AWCC using JSR-15, a traditional shift register, and the Advanced List Mode Module (ALMM). The LV-AWCC has three signal outputs: the total counts from all ^{3}He tubes, the total from the outer ring of tubes, and the total from the inner ring of tubes. The two signals from the inner and outer rings add up to the totals, which can be verified. The totals signal was received by the JSR-15 and the inner and outer ring signals were received by the ALMM on two independent channels. Both data acquisition devices (DAQs) were controlled through the International Neutron Coincidence Counting program (INCC). The latest version of the program (INCC6), which is under active development and undergoing testing towards eventual validation, will interface with list mode devices and enable additional analysis methods. An in-house software was also used for some of the list mode data acquisition.

The traditional shift register DAQs calculates the count rates in real time using a predetermined set of operating parameters. After the measurement is complete, the output contains the analysis parameters, calculated count rates, individual cycle data and multiplicity distributions. This information is useful in many applications but limited. For example, the user cannot study the effects of selected analysis parameters unless a measurement is repeated several times.

List mode DAQs record the raw pulse train, a list of times and channels for each neutron detection event. As a result, data from one measurement can be processed and analyzed with different parameters and additional techniques, including changing analysis parameters, time interval analysis, and Rossi-alpha distributions.

3. Results

a. Passive Measurements

Passive measurements of the ^{233}U triangles were taken in the AWCC with the AmLi sources removed. The measurement time was generally one hour divided into 20-second cycles. Measurements of one and two triangles were taken over a longer period to decrease the uncertainty. A summary of the passive measurements from the JSR-15 is in Table 2 and background has been subtracted.

Table 2: Summary of passive measurements of ^{233}U oxide triangles in the LV-AWCC

# of Triangles	**Total U (g)**	**^{233}U (g)**	**Measurement Time (min)**	**Singles (cps)**	**σ_S**	**Doubles (cps)**	**σ_D**
1	2.000	1.995	194	4.00	0.07	0.02	0.01
2	4.000	3.990	2880	9.37	0.06	0.03	0.01
4	8.000	7.980	60	18.81	0.11	0.06	0.01
8	16.000	15.960	60	38.99	0.13	0.12	0.02
12	24.000	23.940	60	57.90	0.13	0.25	0.02
16	32.000	31.920	60	76.24	0.17	0.33	0.02
20	40.000	39.900	60	97.23	0.19	0.52	0.03
24	48.010	47.890	60	117.08	0.21	0.68	0.03
28	56.020	55.880	60	137.25	0.21	0.90	0.04
36	72.040	71.860	60	175.56	0.26	1.32	0.05
40	80.033	79.833	41	195.58	0.32	1.43	0.06

b. Active Measurements

Active measurements of the ^{233}U triangles were taken in the AWCC with the AmLi sources in the top and bottom plugs. The measurement time was generally 30 minutes divided into 20-second cycles. Measurements of several configurations were taken over a longer period to decrease the uncertainty and some measurements were repeated to verify that the configuration was replicated when measurements were taken on different days. A summary of active measurements from the JSR-15 is shown in Table 3. Background was not subtracted.

Table 3: Summary of active measurements of ^{233}U oxide triangles in the LV-AWCC

# of triangles	**Total U (g)**	**233U (g)**	**Measurement time (min)**	**Singles (cps)**	**σ_S**	**Doubles (cps)**	**σ_D**
1	2.200	2.194	720	12758.36	0.55	14.92	0.70
2	4.399	4.388	30	12785.43	2.86	27.30	3.22
4	8.798	8.776	30	12871.11	2.83	42.70	3.60
6	13.197	13.164	30	12968.53	2.76	60.50	3.94
8	17.596	17.552	720	13023.62	0.56	74.43	0.71
10	21.205	21.152	30	13070.17	2.84	88.15	3.53
12	24.814	24.752	30	13114.46	2.54	97.90	3.74
14	28.815	28.743	30	13163.70	2.70	110.57	3.65
16	32.815	32.733	30	13206.18	2.72	124.13	3.57
18	36.424	36.333	30	13246.82	2.80	131.54	3.33
20	40.033	39.933	30	13285.09	2.97	147.09	3.23
24	48.033	47.913	43	13420.39	2.33	175.99	2.89
28	56.033	55.893	30	13481.04	2.96	200.83	3.66
32	64.033	63.873	30	13533.87	3.02	225.79	3.65
36	72.033	71.853	30	13590.56	2.45	240.12	3.59
40	80.033	79.833	30	13639.82	2.56	263.62	3.60

c. Mass Calibration Curves

Using the measurement data provided above, a mass calibration curve was calculated for passive singles and active doubles as a function of the fissile mass.

$$S = Am \tag{1}$$

$$\sigma_S = \sigma_A m \tag{2}$$

The passive singles calibration curve has a linear fit, described by Equations 1 and 2. The fit accounts for the uncertainty in the singles count rate, but not the uncertainty in the total fissile mass. The fit parameter was calculated to be $A = 2.436 \pm 0.004$. The measurements and corresponding fit are shown in Figure 4.

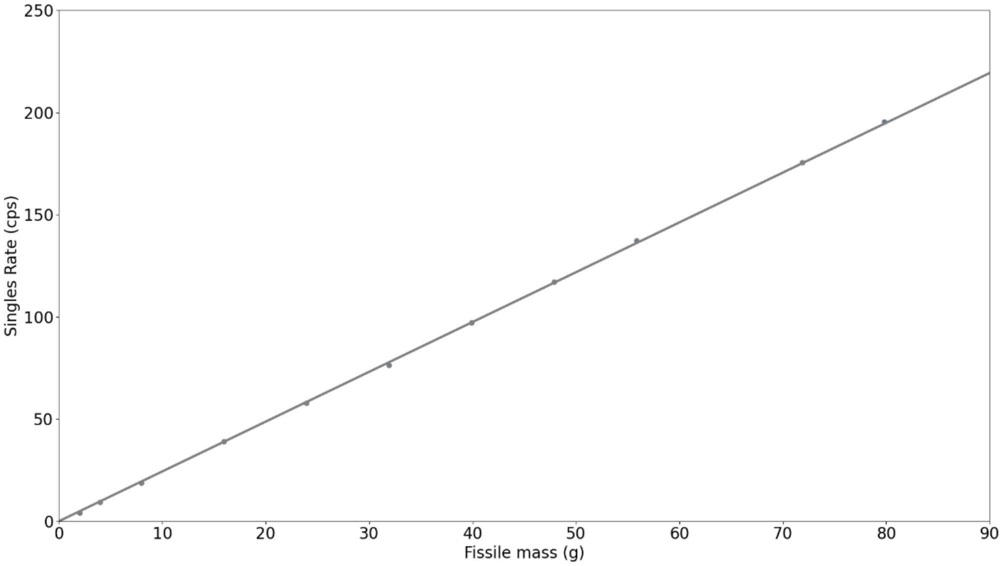

Figure 4: Passive Singles Mass Calibration Curve. Error bars are smaller than the marker size.

$$D = \frac{Am}{(1 + Bm)} \tag{3}$$

$$\sigma_D = D\sqrt{\left(\frac{\sigma_A}{A}\right)^2 + \left(\frac{\sigma_B}{1 + Bm}\right)^2} \tag{4}$$

The active doubles calibration curve has a fit described by Equations 3 and 4, which is the equation commonly used by practitioners when doing a mass calibration curve for uranium-235. The fit accounts for the uncertainty in the doubles count rate, but not the uncertainty in the total fissile mass. The fit parameters were calculated to be $A = 4.631 \pm 0.269$ and $B = 0.005327 \pm 0.000002$. The measurements and corresponding fit are shown in Figure 5. The shaded region indicates 1-σ error from the calculated fit parameters.

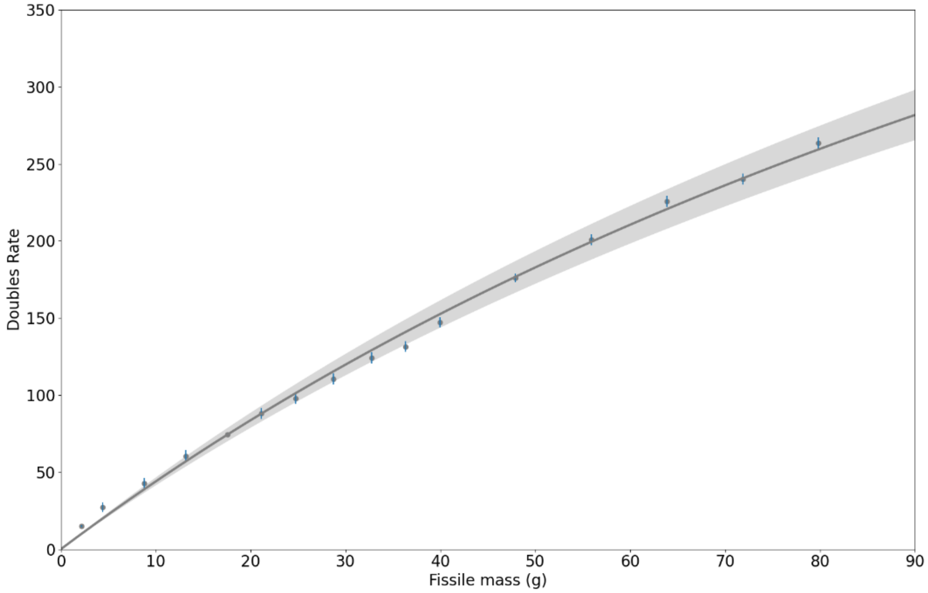

Figure 5: Active Doubles Mass Calibration Curve

4. Discussion

The purpose of this paper is to provide a summary of a series of well-documented systematic measurements of ^{233}U oxide samples that were performed at Oak Ridge National Laboratory using a Large Volume Active Well Coincidence Counter (LV-AWCC). The count rates and calibration curves for active and passive measurements are given, but further analysis into potentially unique signatures is still ongoing. All data was acquired in list mode, providing flexibility and room for creativity in post-measurement analysis.

The paper represents the first step in an multi-year collaboration that seeks to develop methods of characterizing and quantifying SNM that contains ^{233}U. The experimental results provided in this paper serve as a foundation for validation of models and simulations that will support this effort. Measurements of similar HEU triangles and mixtures of ^{233}U and HEU triangles have been performed and are currently being analyzed. Additional exploration of self-interrogation to determine the usefulness and limitations is also ongoing.

5. Acknowledgements

This research was performed under appointment to the Nuclear Nonproliferation International Safeguards Fellowship Program sponsored by the National Nuclear Security Administration’s Office of International Nuclear Safeguards (NA-241).

References

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