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Experimental Neutron Signature Measurements of U-233 Plates at the Device Assembly Facility

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Abstract

The thorium fuel cycle is emerging as an attractive alternative to conventional nuclear fuel cycles, as it does not require the enrichment of uranium for long-term sustainability. The operating principle of this fuel cycle is the irradiation of ^{232}Th to produce ^{233}U, which is fissile and sustains the fission chain reaction. ^{233}U poses novel challenges for nuclear safeguards, as it is associated with a uniquely extreme gamma-ray environment from ^{232}U contamination which limits the feasibility of gamma ray-based assay, as well as more conservative accountability requirements than for ^{235}U set by the International Atomic Energy Agency (IAEA). Consequently, instrumentation used for safeguarding ^{235}U in traditional fuel cycles may be unsuitable. It is essential that the nondestructive signatures of ^{233}U be characterized so that nuclear safeguards can be applied to thorium fuel cycle facilities as they come on-line. In this work, a set of $^{233}U_3O_8$ plates, containing 844 g ^{233}U, was measured at the Device Assembly Facility. A high-pressure ^{4}He gaseous scintillation detector, which is insensitive to gamma rays, was used to perform the first passive fast neutron spectral signature measurement of $^{233}U_3O_8$, and was used in conjunction with a pulsed deuterium-tritium neutron generator to demonstrate the first differential die-away signature of this material. Furthermore, an array of ^{3}He detectors was used in conjunction with the same neutron generator to measure the delayed neutron time profile of ^{233}U, which is characteristic of the isotope. These measurements provide a benchmark for future nondestructive assay instrumentation development, and demonstrate a set of key neutron signatures which may inform future nuclear safeguards for the thorium fuel cycle.

1 Introduction and Background

1.1 Unique characteristics of ^{233}U in the thorium fuel cycle

Uranium-233 produced in nuclear reactors by the absorption of a neutron in ^{232}Th and the subsequent beta-decays of ^{233}Th and ^{233}Pa is always accompanied by trace quantities of ^{232}U, ranging from approximately 100 to 5000 ppm. This is due to several (n,2n) reactions in ^{232}Th,

^{233}Pa, and ^{233}U which occur in the presence of fast neutrons. The presence of ^{232}U is important, as its decay chain is associated with high specific activity and high branching ratio for high-energy gamma ray emission, principally at 0.78 and 2.6 MeV. Since ^{232}U is typically not separated from ^{233}U in thorium-based fuel cycles, this intense gamma ray environment is inextricably linked to any macroscopic quantity of ^{233}U.

1.2 Challenges in measurement of ^{233}U-bearing items for safeguards

Many non-destructive assay (NDA) measurements of fissionable materials conducted for the purpose of nuclear safeguards involve measurement of gamma ray spectra. These measurements may quantify the total mass of some isotope, or measure the relative concentration of isotopes in thick samples. Gamma ray spectroscopy, however, is not feasible for NDA of ^{233}U items due to the extremely intense contribution of gamma rays in the ^{232}U decay chain: the intensity of the 0.78 and 2.6 MeV gamma ray lines does not strictly correlate with the mass of ^{233}U, and all gamma rays directly associated with the decay of ^{233}U are low energy and low intensity, and consequently not measurable above the compton continua emanating from the ^{232}U lines. As an alternative, neutron-based NDA methods are under investigation for safeguarding ^{233}U-bearing materials.

1.3 Neutron signatures of ^{233}U-bearing items

1.3.1 Spontaneous neutron emission

Many ^{233}U-bearing objects of safeguards relevance are in an oxide matrix, either as U_3O_8 or UO_2. Alpha radiation emanating from ^{233}U as well as ^{234}U, which is typically present as a significant contaminant, can be absorbed by natural oxygen isotopes which then emit a neutron. These (α,n) neutrons can go on to induce fission in ^{233}U or other fissionable species present, and therefore produce additional neutrons, with a probability determined by the multiplication k_{eff} of the matrix. Induced fission neutrons have a distinct multiplicity and spectrum from (α,n) neutrons, so the ratio of spontaneous (α,n) to induced fission neutrons can theoretically be measured. Since this ratio is related to multiplication it may therefore help indicate the fissionable matrix composition.

1.3.2 Delayed neutron emission

When fissionable nuclei undergo fission, the resultant neutron-rich fission fragments have some probability of emitting neutrons during their radioactive decay, a process referred to as β-delayed neutron emission, since these neutrons are emitted in coincidence with βdecay. There are many delayed neutron precursors, which are typically condensed into six groups determined by their half-lives, ranging from 1 to 60 seconds. Each fissionable isotope has a unique yield for each delayed neutron group, resulting in a unique delayed neutron time profile. When fission is induced during active neutron interrogation, this time profile can be measured to indicate the isotopic composition of fissionable material [1].

1.3.3 Differential die-away

Differential die-away (DDA) is a technique to measure the presence of fission neutrons emitted by a target after a pulsed active interrogation source is turned off [2]. Neutrons thermalizing in the environment typically have a long life-time compared to the duration of an active interrogation pulse, and the resultant thermal neutron population that persists beyond the fast neutron pulse has a high probability of inducing fission in fissile materials. Measurement of DDA is best done with a detector which is sensitive to fast neutrons only, as the signal-to-noise ratio (SNR) when

fissile material is present is orders of magnitude higher for fast neutrons after the pulse than for thermal neutrons.

2 Methods

The Device Assembly Facility (DAF) is an experimental venue at the Nevada National Security Site (NSSS) which holds a variety of test objects containing nuclear materials, including kilogram-scale quantities of ^{233}U. The DAF also has a pulsed deuterium-tritium (D-T) neutron generator available for active interrogation experiments.

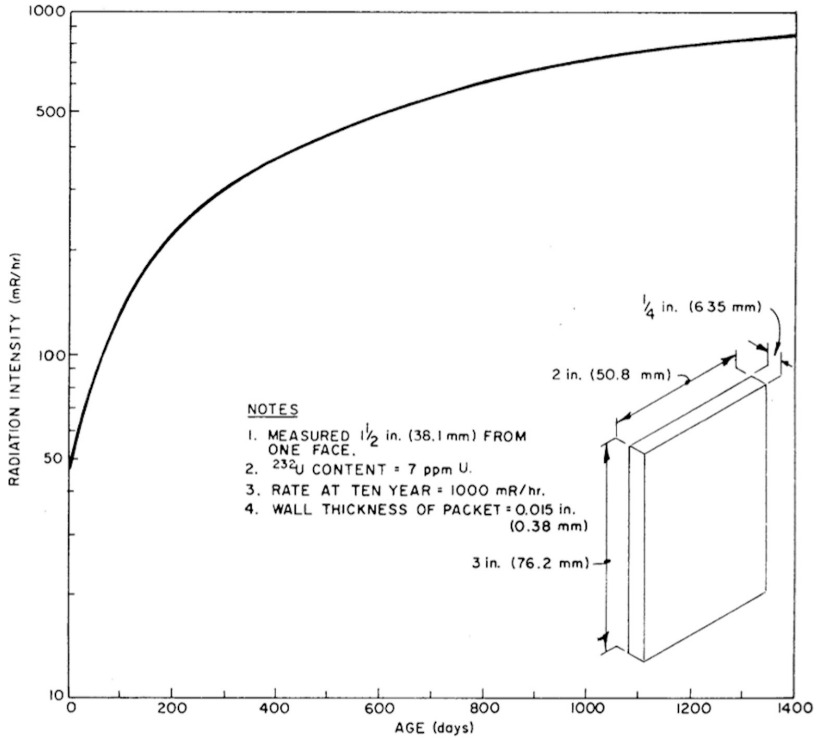

Figure 1: Gamma ray exposure due to a single ZPR plate at 1.5" from the plate surface [3].

2.1 Description of target and interrogation source

The material upon which the experiment in this work was performed is a set of fuel plates originally intended for the Zero Power Reactor (ZPR) at Argonne National Laboratory (ANL). These plates are comprised of 33 g $^{233}U_3O_8$ charged into stainless steel packets measuring $2 \times 3 \times \frac{1}{4}$" [3]. The plates are stored in groups of ten in steel "soup cans", three of which are stored in a triangular lattice within an AT-400R radioisotope storage container. In the described measurement, the ZPR plates were not authorized to be removed from their AT-400R containers, which also contain some lead shielding of unknown geometry and thickness. Each AT-400R container holds a total of 999 g $^{233}U_3O_8$, 844 g of which is ^{233}U. The ^{233}U used to fabricate these ZPR plates was produced to minimize the concentration of ^{232}U, the average of which in all plates is 7 ppm. Even at this low concentration, the radiation exposure rate due to

^{232}U daughters at equilibrium in a single plate is 1000 mR/hr, measured 1.5" from the center of the plate surface, shown in figure 1. At this exposure level, most γ-sensitive detectors cannot be operated in pulse mode. In this experiment, a single AT-400R containing $^{233}U_3O_8$ ZPR was utilized.

The D-T generator used as an interrogation source was a Thermo Scientific model P211. This generator has a total output of 10^8n/s, in pulses of approximately 10 μs duration, operating at 100 Hz. This low duty factor and high pulse intensity is excellent for DDA analysis.

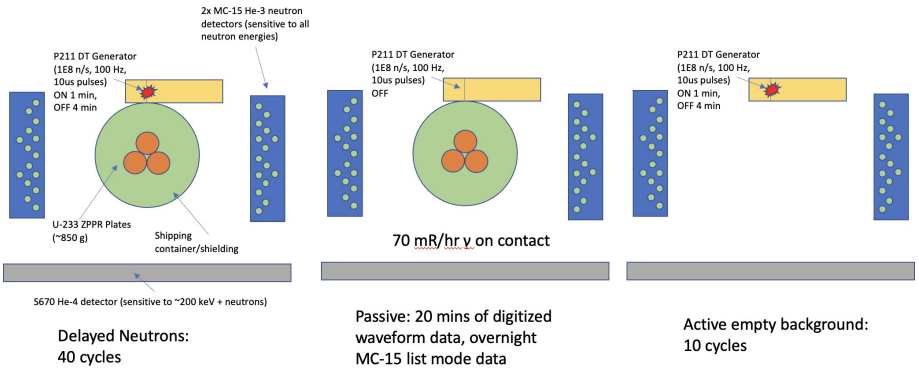

Figure 2: Conceptual diagram illustrating the measurement configurations used for active signature measurement, passive signature measurement, and active background.

2.2 Description of detectors used

In this work, two neutron detectors were used. The first is an Arktis Radiation Detectors S670 [4] high-pressure ^{4}He scintillation detector, which is intrinsically sensitive to only fast neutrons if an energy deposition threshold of 300 keV is used. This detector is sensitive to the energy of fast neutron-induced ^{4}He recoils, and can provide information on the neutron spectrum being measured. This detector has a faster response time, smaller dead-time, and smaller time resolution than most other gas-based detectors, since the ^{4}He scintillation pulses have a duration of approximately 1 μs. The detector and pulse analysis system applied is described further in Ref. [5].

Two MC-15 neutron detectors were also used. MC-15 detectors are comprised of an array of 15 ^{3}He proportional counters embedded in a slab of high-density polyethylene (HDPE) [6]. These detectors have high efficiency for fast neutrons, but since they are intrinsically sensitive to thermal neutrons, their time resolution for fast neutron detection is large compared to the S670 detector. Consequently, this detector was used to measure the time profile of delayed neutrons, but not DDA. The measurement configuration, showing the target material, interrogation source, and detectors are shown in Figure 2.

In active measurements, the D-T generator was operated in cycles of approximately 1 minute on (6,000 pulses) followed by 4 minutes off, to allow for the complete decay of delayed neutron precursors between cycles. Images of the actual measurement are shown in Figure 3.

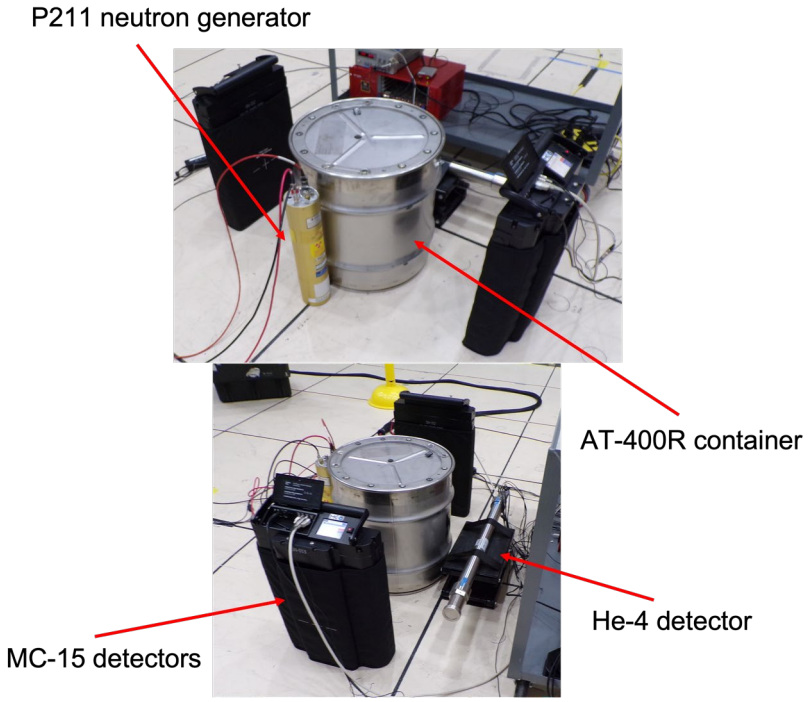

Figure 3: Images from DAF measurement.

3 Simulation of expected results & conclusions

The simulated β-delayed neutron emission decay time profiles of ^{233}U, ^{235}U, and ^{238}U are shown in Figure 4. These time profiles are well-separated, demonstrating that measurement of delayed neutron emission could be used to reconstruct the relative concentrations of each isotope in a mixed-isotope item.

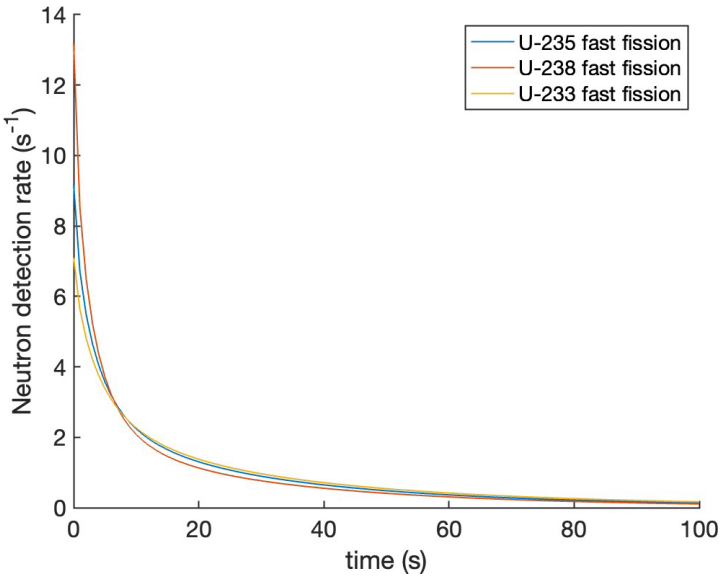

Figure 4: Simulated β-delayed neutron emission of uranium isotopes after 60 seconds of neutron irradiation. Each time profile normalized to 100 total neutrons detected.

The simulated DDA signature of the ZPR plates, measured with the ^{4}He detector, is shown in Figure 5. The simulation contains all relevant features of the laboratory structure, shielding, and containment. A clear exponential decay behavior is observable at the equivalent of 140 s of P211 generator operation, demonstrating that this detector can be used to measure this signature.

The (α,n) neutron spectrum of $^{233}U_3O_8$ has not been measured previously. However, the spectrum may be reasonably approximated with the $^{238}PuO_2$ (α,n) spectrum. The ^{4}He detector response to this neutron spectrum as well as a ^{252}Cf spontaneous fission spectrum is shown in Figure 6. These two spectra have clearly distinct shapes, demonstrating that this detector may be used to distinguish $^{233}U_3O_8$ neutrons from spontaneous or induced fission neutrons.

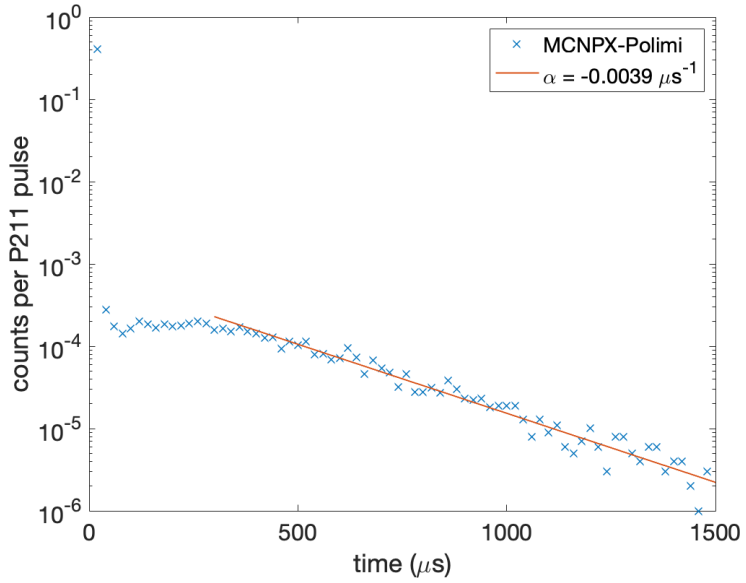

Figure 5: Simulated DDA signature of ^{233}U plates.

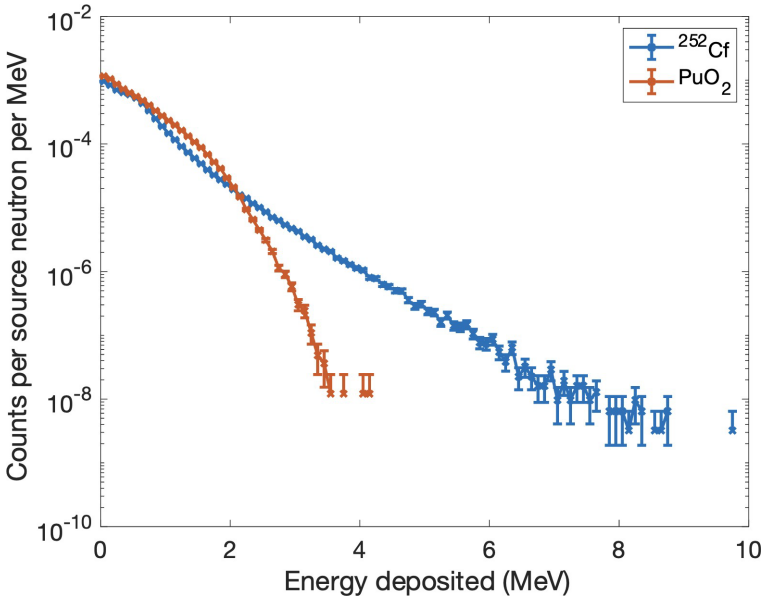

Figure 6: Simulated ^{4}He detector response to oxygen (α, n) neutrons and ^{252}Cf spontaneous fission neutrons. Note: spectra not broadened to account for energy resolution.

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