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Title: **PHOTON AND NEUTRON ACTIVE INTERROGATION OF HIGHLY ENRICHED URANIUM**

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Photon and Neutron Active Interrogation of Highly Enriched Uranium

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Abstract. The physics of photon and neutron active interrogation of highly enriched uranium (HEU) using the delayed neutron reinterrogation method is described in this paper. Two sets of active interrogation experiments were performed using a set of subcritical configurations of cocentric HEU metal hemishells. One set of measurements utilized a pulsed 14-MeV neutron generator as the active source. The second set of measurements utilized a linear accelerator-based bremsstrahlung photon source as an active interrogation source. The neutron responses were measured for both sets of experiments. The operational details and results for both measurement sets are described.

INTRODUCTION

As part of our research using active interrogation techniques on uranium systems, a series of measurements was performed that have applications to nuclear materials criticality safety and nuclear material safeguards science. The intent of this research is to demonstrate the utilization of the delayed neutron reinterrogation technique[1,2] as a means of providing technological solutions for detection and evaluation of subcritical configurations of highly enriched uranium (HEU). Currently, there are no computational tools that have been completely benchmarked with experimental data and/or fully simulate the physics of the delayed neutron reinterrogation technique during a contiguous computational run. The data from these measurements can be utilized to develop and benchmark computational tools that could then be used as predictive or design tools for the application areas mentioned above.

Why Use Active Techniques For Detection and Assessment of HEU Systems?

Active interrogation techniques are employed because passive detection of HEU containing approximately 90 wt% ^{235}U is often difficult.[3] The strongest gamma rays from ^{235}U with energies of 144, 186, and 205 keV, as well as the x-rays, are easily attenuated by a few millimeters of lead or its equivalent. The neutron emission rate of ^{235}U of 3×10^{-4} neutrons per second per gram is too low to be useful for assessment with most nuclear material criticality and safeguards applications. The next most abundant isotope in HEU, ^{238}U, has a very weak gamma ray with energy of 111 keV, which is also easily attenuated. However, ^{238}U decays to ^{234}Th (24.1 days half-life), which subsequently decays to ^{234m}Pa (1.17 m half-life). If processing of the HEU was not recent and sufficient and ^{234}Th has built up, the 766- and 1001-keV gamma rays from ^{234m}Pa may be detectable. However, these gamma rays are also present in background. HEU often contains ^{232}U, which produces a 2.6-MeV gamma ray following the decay to ^{208}Tl. However, this signature is not reliable because ^{232}U is produced in a reactor and not all HEU has been in a reactor.

Delayed Neutron Reinterrogation Physics

The kinetic properties of delayed neutrons are very useful for interpreting dynamic neutron measurements that involve active interrogation of HEU. Keepin has extensively discussed these properties.[4] There are approximately 270 different fission products that emit delayed neutrons. The yield per fission of each delayed neutron precursor is dependent upon the isotope undergoing fission and the dynamic characteristic of the particle inducing the fission.

For a subcritical configuration of HEU, the passive neutron response from the system is difficult to distinguish from the passive background neutron response. The objective of the delayed neutron reinterrogation technique is to artificially create an intrinsic steady-state source of neutrons that is distributed throughout the fissile material and allow the utilization of analogous neutron noise analysis techniques that are used for evaluation of passive plutonium measurements.

The creation of an intrinsic steady-state source of delayed neutrons is accomplished by repetitious interrogation using a pulsed active source to create fission products throughout the uranium. Delayed neutron emission follows the beta decay of a fraction of these fission products. The emission of a delayed neutron occurs randomly and later in time with respect to the parent fission that produced the delayed neutron precursor. The delayed neutrons either leak from the system or initiate further fission chains. After a finite number of active pulses, the delayed neutron precursor populations reach apparent steady-state levels and the continued active interrogation results in creating a near steady-state source of neutrons that is distributed throughout the uranium that reinterrogates the material. The intrinsic neutron source strength is dependent upon the intensity and ability of the active source to induce fissions in the uranium during a pulse.

General Experimental Protocol and Equipment Description

The following experimental protocol has evolved from the Los Alamos National Laboratory Advanced Nuclear Technology Group's research and development experience. We have chosen to investigate the neutron response that occurs when performing an active measurement because information pertinent to criticality safety and nuclear material safeguards can be inferred by measuring the neutron response.

A typical active system would consist of the following components:

- a pulsed active source (photon or neutron) with controller
- a neutron detection system (with associated electronics to interface with data acquisition)
- a data acquisition system (computer or embedded processor-based system)

For comparison purposes, we have chosen to operate our pulsed sources at a repetition rate of 50 Hz. A pulse from an active source (neutrons or photons) will saturate a ^{3}He-based neutron detection system for a finite time lasting up to several milliseconds (dependent upon source intensity and geometry). We experimentally determined this saturation time with each active source and neutron detector configuration. Then we gated off the data acquisition system for a finite time to save data acquisition memory space. The neutron detection system response is measured between pulses of the active source and the data can be evaluated using any neutron noise analysis technique that is applicable to the physics of fission chains initiated by a random source.

Photon Interrogation

The threshold energies of photons required to induce photoneutron and photofission reactions in the actinides begin near 5 MeV. Berman published an early compendium of references for applications that utilize photoneutron reactions[5] and compiled a comprehensive data set of the energy-dependent cross-section data for photoneutron reactions for different isotopes[6]. Hyde provides an early comprehensive literature review about the physics of photoneutron and photofission reactions of the heavy elements.[7] Research utilizing photoneutron reactions performed by Brunson and Coop[8] served as a basis for our current photon interrogation work.

The active source we have utilized as a source of interrogating photons for a majority of our work is a Linatron 2000™ linear electron accelerator that had been repackaged into transportable boxes for field deployment.[9] The accelerator can operate at energies of 6, 8, or 10 MeV and can operate at a pulse repetition rate in the range of 20–120 Hz. For the measurement reported below, we operated at 8 MeV with a pulse rate of 50 Hz. The pulse width was approximately 4.5 microseconds with the dose rate at 1 meter on axis being approximately 160 R/min. The electron current operates in the range of 80–100 milliamps on the target, dependent upon the energy setting of the accelerator.[10]

Neutron Interrogation

Neutron interrogation has been a common technique for assessing configurations of nuclear materials since the 1940s. Common sources of neutrons that have been employed during active interrogation measurements include isotopic sources, D-T (deuterium + tritium) neutron generators for

14 MeV neutron production, and D-D (deuterium + deuterium) neutron generators for 2.6 MeV neutron production.

The source of interrogating neutrons that we most commonly employ for our work are produced by an MF Physics model CC A-210™ neutron generator that is pulsed at 50 Hz. The generator uses the reaction (deuterium + tritium $\rightarrow$ neutron + ^{4}He) to produce 14 MeV neutrons. The neutron pulse width is 10–20 microseconds in length, and the output is approximately10^6 neutrons per pulse or 5×10^7 neutrons per second into 4π steradian.

Neutron Detection Systems

The size and configuration of the neutron detection systems used for our research were dependent upon their intended applications.[11] The general design characteristics of our neutron detection systems were that they are constructed using ^{3}He tubes that are surrounded by polyethylene with a thin layer of cadmium surrounding the outer surface of the polyethylene. The thin layer of cadmium helps minimize “room return” neutrons and helps to decouple the HEU object from the neutron detection system by absorbing neutrons that have been down-scattered in energy in the detector and prevent them from reinterrogating the HEU object. When possible, the electronics of the neutron detection system are segmented to minimize dead-time effects by operating each individual detector channel with its own preamplifier, amplifier, and discriminator.

Both sets of data presented below utilized a portable detector system that was designed for maximum efficiency in a small system that could be easily transported. The detector measures 10 cm × 43 cm × 51 cm and has a mass of 25 kg. It contains fifteen ^{3}He tubes, 2.54 cm (diameter) × 38.1 cm (length), pressurized to 10 atm, and inserted into holes in a polyethylene block. The polyethylene block, including the top except for the connectors, is completely covered by 0.81 mm of cadmium. The intrinsic efficiency is 20%. The total efficiency is approximately 1% and the detection system dead time was approximately 167 nanoseconds.

Data Acquisition

The data acquisition employed for the measurements was the custom Los Alamos-designed pulse arrival-time recording module (PATRM)[12]. The PATRM system can accept 15 channels of data. Signals from the ^{3}He neutron detectors are amplified and discriminated to form logic pulses that are used as input to the PATRM. The PATRM uses a logic pulse designated VETO to control data acquisition in active mode. At the onset of the VETO pulse, the PATRM disables all data inputs, writes a code marker to its internal memory at the next two memory locations, stops the internal clock counter, and resets this counter to zero. Upon lifting of the VETO, the clock counter and all data inputs are enabled. The result of the measurement is a history of the signal arrival times relative to the end of the VETO pulse. The clock frequency of the PATRM was 5 MHz, allowing the signal arrival time to be measured with an accuracy of 0.2 microseconds. Up to a million events can be recorded before transferring the data to a small computer for analysis and archiving. The VETO pulse length is chosen, dependent upon the ability of the active source to saturate the neutron detection system and the time needed by the neutron detection system to recover from this saturation. A typical VETO length chosen for our measurements was in the range of 700–2000 microseconds.

Data Analysis

The two data analysis techniques that are most frequently utilized by our research team are the Hanson-Dowdy formalism[13,14] and Hage-Cifarelli formalism[15] of Feynman’s Variance-To-Mean technique. These neutron noise analysis techniques use various moments of the neutron detection system counting distribution to infer or assess information from the measurements about subcritical neutron chain reacting systems.

Measurement Results

The uranium metal sphere used for the measurements was constructed using a set of uranium metal hemispherical shells, known as the Rocky Flats shells.[16] The Rocky Flats shells are reported to be enriched to 93.12 wt% with U^{235} and have an average density of 18.675 g/cm^3. The nesting hemispherical shells were manufactured to have approximately the same wall thickness (0.33 centimeters), but small gaps are present due to tolerances allowed on machining operations during manufacture. The configuration used for the measurements had an outer radius of 6.67 cm and was constructed using 30 hemispherical parts (15 shells total). The mass was 22.5 kg.

Figure 1 illustrates two separate measurements performed using pulsed 14-MeV neutrons. The geometry consisted of the 22.5 kg sphere of HEU metal placed midway between the neutron generator and neutron detector (separated by 1 meter). The upper curve represents the first moment of the neutron detector counting distribution as a function of time between bursts with the HEU sphere present. The lower curve is an active background measurement (same geometry with no HEU present). The data are averaged over 64000 bursts of the neutron generator.

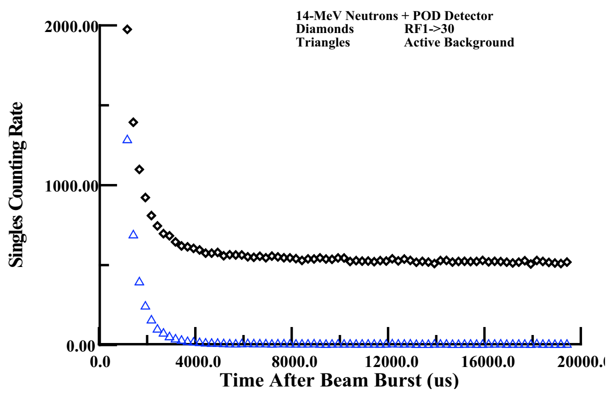

Figure 1. Neutron Interrogation of 22.5 kg Sphere of HEU Metal.

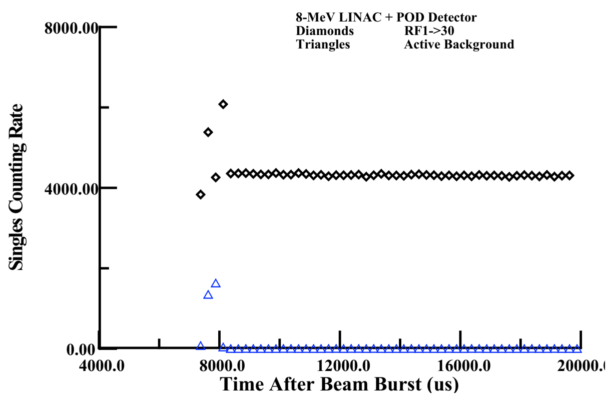

Figure 2. Bremsstrahlung Photon Interrogation of 22.5 kg Sphere of HEU Metal.

Figure 2 illustrates two separate measurements performed using the LINAC operating at 8 MeV. The geometry consisted of the 22.5-kg sphere of HEU metal placed 262 cm directly in front of the beam target and the detector placed 50 cm from the HEU sphere perpendicular to the photon beam central axis. No collimator was used during the measurement. The upper curve shows the first moment of the neutron detector counting distribution as a function of time between bursts with the HEU present. The lower curve is an active background measurement (same geometry with no HEU present). The data in Figure 2 illustrate an extreme case of detector saturation that can occur during an active measurement. The data are averaged over 64000 bursts of the LINAC.

How to Use Data to Benchmark Simulations

Estes and Goulding presented research aimed at better understanding the determination of the reactivity of subcritical systems from measurements of the apparent multiplication of the system.[17] They presented an experimental protocol and computational methodology for comparing simulated results with passive neutron measurements performed on subcritical configurations of plutonium. Mihalczo and Valentine presented a similar concept for utilization of ^{252}Cf-Source-Driven Noise Analysis technique as a way to validate calculational methods for criticality safety.[18] The main idea is to model the subcritical object, neutron detector configuration, and a portion of the environmental surroundings and then calculate the experimental observables. All this work utilized specially developed computational patches to be used with an analog version of the Los Alamos Monte Carlo Neutron and Photon (MCNP) transport code.[19]

To utilize our measurements for benchmarking and validation, the ability to simulate the physics of the delayed neutron reinterrogation method would need to be incorporated into the computational tool. This would include the ability to simulate the photoneutron reactions and the ability to include the dynamic and kinetic properties associated with delayed neutron production. The observables to simulate would consist of a list-mode data set representing the neutron detection system response, which includes markers to represent when each active burst occurred and a list of neutron detection times after each burst. Artificial corrections have to be made to the simulated data list to account for detector dead-time effects. One would then compare results by using the same neutron noise analysis technique to infer information from the simulated and experimentally measured list-mode data sets.

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