ARTICLE IN PRESS

Nuclear Inst. and Methods in Physics Research, A ▮ (▮▮▮▮) ▮▮▮–▮▮▮

Contents lists available at ScienceDirect

Nuclear Inst. and Methods in Physics Research, A

journal homepage: www.elsevier.com/locate/nima

Fast-neutron multiplicity counter for active measurements of uranium oxide certified material

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ABSTRACT

We developed a fast-neutron multiplicity counter (FNMC) based on stilbene and EJ-309 organic scintillators. The system can detect and discriminate correlated photon and neutron multiplets emitted by fission reactions. We used the system to estimate the fissile mass of uranium oxide samples in active interrogation mode at the Zero Power Physics Reactor of Idaho National Laboratory (INL). Two sets of certified reference material (CRM) samples were characterized. The U-235 enrichment of the first set is constant at 93.2 wt%, and the UO_2 mass ranges between 0.5 and 4 kg. The second set includes samples of increasing enrichment (from 20 wt% to 97 wt%) and constant UO_2 mass of 230 g. We used two AmLi sources to induce fission reactions in the samples. Despite the intense gamma-ray background of the UO_2 and interrogating sources, the system could measure induced fission neutrons emerging from the interrogated samples without additional shielding surrounding the sample and only relying on pulse shape discrimination to classify neutron and gamma-ray pulses. The overall neutron count rate and time-correlated counts are well correlated with the sample fissile mass. We also proved that CRM samples can be used to build a calibration curve to assay the U-235 mass of unknown samples of different mass, geometry and enrichment, with an average bias error of 8%, for U-235 mass higher than 390 g.

1. Introduction and motivation

Plutonium, uranium enriched in the isotopes 235 or 233, as well as source material (e.g. natural and depleted uranium (DU)), are subject to inspections under the current safeguards agreements and protocols [1,2]. Neutron coincidence and multiplicity counting systems are currently used by the IAEA for the non-destructive assay of plutonium and uranium bearing materials to support verification of inventory declarations and aid in the prompt detection of material diversion. The non-destructive assay of these materials relies on two modes of operation: passive and active neutron multiplicity counting (NMC). Passive NMC techniques are typically implemented for materials with a high yield of spontaneous fission neutrons (e.g., plutonium bearing). Conversely, the low yield of uranium spontaneous fission neutrons makes it very difficult to characterize uranium with passive NMC measurements; however, neutron emission yield can be increased in active interrogation mode, which requires an external source to induce fission reactions in these samples.

Commercial active NMCs use He-3 proportional counters embedded in polyethylene, which rely on capture reactions to detect thermal neutrons and exhibit a low sensitivity to gamma rays. These systems have several positive attributes, such as high neutron detection efficiency in the energy range of interest (thermal to <15 MeV) and long-term robustness and stability. However, the time gate window for detecting correlated particles is of the order of the thermal-neutron die-away time within the system (e.g., 51 μs for the Active Well Coincidence Counter (AWCC) by Canberra Industries, Meriden, CT). This system feature results in large accidental count rates, which may overwhelm the characteristic fission signature subsequently requiring long assay times to achieve a desired measurement precision. The variance due to the accidental counts can be the main contribution to the overall measurement uncertainty [3–6].

Several counters were designed to reduce the system sensitivity to accidental counts, including epithermal-neutron multiplicity counters (ENMCs) and fast-neutron multiplicity counters (FNMCs) [4]. The ENMC uses He-3 detectors like the AWCC. However, ENMCs feature a higher He-3 gas pressure (10 atm) and a thinner polyethylene layer surrounding the detectors, compared to standard thermal systems. These two modifications have the effect of increasing the detection efficiency and avoiding the full thermalization of the neutrons, respectively. The neutron die-away time in an ENMC is approximately 20 μs [4].

FNMCs are based on detectors sensitive to fast neutrons, e.g., organic scintillators and He-4 proportional counters that do not require moderation and are currently being investigated by several research groups [7–11] . The time interval between neutron production inside the sample and its detection is of the order of tens of nanoseconds and is

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https://doi.org/10.1016/j.nima.2018.05.049
Received 2 December 2017; Received in revised form 20 May 2018; Accepted 23 May 2018
Available online xxxx
0168-9002/

Table 1
Active interrogation methods exploiting fission signatures.

Interrogating source	Sample	Detectors	Measured quantities	Reference
AmLi	Pressurized water reactor fresh fuel assemblies	EJ-309	Neutron doubles rate	[11]
DT and AmLi	Low-enriched uranium (LEU) (1%–5% U-235 wt%)	EJ-309	Neutron gross count and time-of-flight	[18]
AmLi	LEU UO_2 samples (4.2% and 10% U-235 wt%)	BC501A	Neutron multiplets	[19]
DT	Highly-enriched uranium (HEU) (93.19% U-235 wt%) and DU (0.3% U-235 wt%)	Plastic scintillators	Neutron singles and doubles ratio	[20]
AmLi	HEU (93.17% U-235 wt%) and DU (0.31% U-235 wt%)	EJ-299–34	Neutron doubles rate	[21]

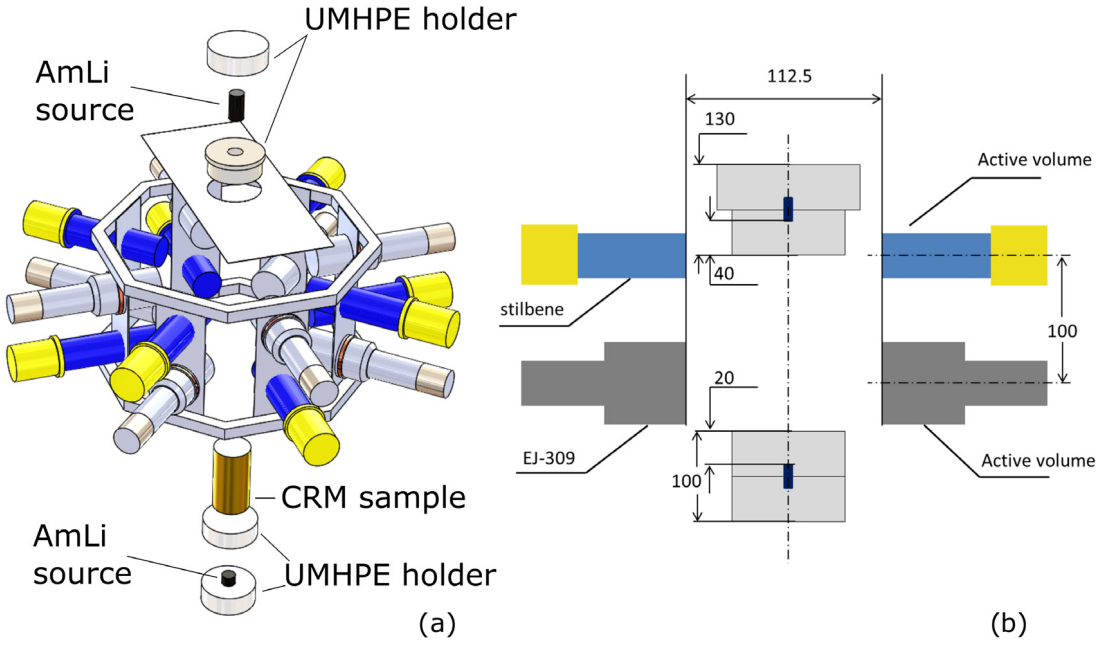

Fig. 1. FNMC exploded view diagram (a) and section-view (b, not to scale). All dimensions are in millimeters.

simply related to the neutron time-of-flight from its birth location inside the sample to the detector. Reduced sensitivity to accidental pulses and to low-energy interrogating neutrons make FNMCs particularly suitable for active interrogation applications as both time-correlated and single neutron counts are expected to correlate well with the sample fissile mass.

Several authors have proven the effectiveness of identifying uranium bearing material using fast-neutron detectors in active interrogation mode [11–17]. However, only a few experimental studies reported the characterization of uranium samples in terms of mass or multiplication, suitable for safeguards applications (Table 1).

In particular, the work by Beaumont et al. [11] describes the application of a fast-neutron multiplicity counter in active interrogation mode for the inspection of 3%–5% U-235 wt% enriched fresh fuel assemblies. In this work, we report the results of the active interrogation assay of uranium oxide certified reference material (CRM) with high enrichment and high mass with a newly developed FNMC [22], with no heavy metal shield between the source and the detectors. The FNMC includes sixteen organic scintillation detectors of two different types: EJ-309 liquid organic scintillators and stilbene crystals.

2. Materials and methods

This section describes the development of an FNMC for active interrogation of uranium bearing materials. After an overview of the full system, the interrogating AmLi sources and the measured uranium oxide samples are described in detail.

2.1. Description of the system

The FNMC includes eight EJ-309 liquid organic scintillators (7.62 cm diameter by 7.62 cm) and eight stilbene crystal scintillators (5.08 cm diameter by 5.08 cm) configured in a well-type geometry, arranged in an alternating "checkerboard" pattern. The distance between the front face of each detector and the virtual axis of the well, i.e. the radius of the well, can be adjusted in the range between 11.25 cm to 22.5 cm. In the present experiment, the well radius was 11.25 cm. A 16-channel waveform digitizer (V1730 by CAEN Technologies, Viareggio, Italy, 14-bit amplitude resolution, 500-MSps sampling rate, 2-V dynamic input range) directly acquires analog detector pulses, performs analog-to-digital conversion and transmits the data to a dedicated workstation. The operating thresholds of EJ-309 and stilbene detectors were 60 keVee and 40 keVee, respectively, corresponding to approximately 430 keV and 360 keV maximum energy deposited by a single neutron–hydrogen nucleus elastic collision, respectively [23].

Prior to assaying the UO_2 CRM samples, we calibrated the detectors using a 0.1-μCi Cs-137 source to ensure uniform detector response. The Compton edge was identified as the 80% of the maximum of the pulse height spectrum response to the Cs-137 source [23], and was calibrated to 0.4 V for all the detectors using a semi-automated script written in Matlab 6.1 (The MathWorks Inc., Natick, MA). For this calibration setting, and given the digitizer dynamic range, the maximum neutron energy that can be detected via a single neutron scatter with a hydrogen nucleus is 6.2 and 5.6 MeV for EJ-309 and stilbene, respectively.

Please cite this article in press as: A. Di Fulvio, et al., Fast-neutron multiplicity counter for active measurements of uranium oxide certified material, Nuclear Inst. and Methods in Physics Research, A (2018), https://doi.org/10.1016/j.nima.2018.05.049.