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A water-based neutron detector as a well multiplicity counter

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ABSTRACT

We report the performance characteristics of a water-based neutron detecting multiplicity counter for the non-destructive assay of fissile sources. This technique could replace or supplement existing ^{3}He-based multiplicity counters. The counter is a 1.02 m^3 tank containing pure deionized water doped with 0.5% $GdCl_3$. It has highly reflective walls and eight 10-in. PMTs mounted at the top. An unshielded source well of 19 cm diameter, mounted at the top and center, extends 73 cm down into the detector. The counter was evaluated using low intensity ^{252}Cf and ^{60}Co sources, and a fast pulsing LED to simulate higher intensity backgrounds. At low gamma ray intensities ($\sim$200 kBq or less) we report an absolute neutron detection efficiency of 28% and a ^{60}Co rejection/suppression factor of $\sim 10^8$ to 1. For sources with high gamma ray intensities, the neutron efficiency was 22% $\pm$ 1% up to a ^{60}Co equivalent activity of 4 MBq. The detector background event rate, primarily due to muons and other cosmogenic particles, was found to be stable over a period of almost three months. The minimum detectable neutron source intensity above background was 3.1 n/s, assuming a one-hour data acquisition.

1. Introduction

In recent years the severe shortage of ^{3}He has been a great concern for organizations involved in nuclear security ([1–3]). ^{3}He-based ionization tubes are uniquely suited for neutron detection: they are safe and non-cryogenic, exploit the high neutron capture cross-section of ^{3}He, and have excellent gamma ray rejection. Detector configurations comprising tightly packed arrays of ^{3}He tubes within a moderating material such as polyethylene are highly efficient and can be used to detect multiple neutrons arising from single fissions, and hence measure the fissile content of samples of special nuclear material (e.g. [4,5]) ^{3}He-based well counting systems range in efficiency from 10% to 50%, depending on how tightly tubes are packed and the ^{3}He gas density. Highly efficient and large systems, however, require the use of a large fraction of the yearly supply of ^{3}He and are prohibitively expensive. In recent years the number of competing neutron detection techniques has proliferated in response to the ^{3}He shortage, but many are not yet ready for widespread use. Boron-based systems such as BF_3 and ^{10}B tubes/planes present toxicity concerns and/or are relatively inefficient. Scintillator-based solutions often rely on differences in signal pulse shape to discriminate against gamma rays, placing severe limits on the event rate that can be tolerated before pileup issues dominate. Germanium or silicon-based detectors are small, reducing their overall efficiency. Due to deployment of large-volume neutron detectors at US borders and increased demand for medical imaging, US federal ^{3}He reserves have decreased from 220,000 l in 2001 to 50,000 l in 2010 [2]. The cost of ^{3}He has increased from $45 to $85 per liter prior to the shortage to $600–$1000 per liter in 2011. Since the ^{3}He shortage is projected to continue for the foreseeable future, alternative techniques are needed.

Coincidence counting of neutrons is an effective way to non-destructively determine the amount of fissile material within a sample of special nuclear material (SNM) [6]. The technique is to measure pairs of neutrons correlated in time from single fission events. For many fissile source configurations, multiplicity counting is a more powerful and general technique. However, it requires detection of three or more neutrons from a single fission event. Since the efficiency for detection of n coincident neutrons scales as the nth power of the efficiency for one neutron, the single neutron detection efficiency quickly becomes the most important criterion for evaluation of new technologies. Of the options available, despite their toxicity, BF_3 gas detectors have been considered the most viable alternative to ^{3}He for safeguards applications, because of the stringent efficiency requirements [7].

In evaluating the efficacy of a neutron coincidence counting technique, the safeguards figure-of-merit (FOM) is a standard metric ([7,8]):

$$\mathbf{FOM} = \frac{\varepsilon}{\sqrt{\tau}}.$$

ε is the single neutron detection efficiency and τ the mean thermal neutron capture time (often referred to as the die-away time). A second important performance criterion is the ability of the detector

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to maintain high neutron detection efficiency and low dead time in the presence of a high gamma ray dose rate. Dose rates at the detector face may be as high as 500 mR/h for spent nuclear fuel sources [9]. High gamma ray dose rates, however, are only significant in the context of a water Cherenkov detector if the gamma rays are of sufficiently high energy. Extreme levels of gamma ray emission from ^{137}Cs or ^{241}Am are likely to produce almost no water Cherenkov response whatsoever, as we demonstrate below.

In recent years we have studied a number of water-based detectors for the purpose of detecting neutrons ([10–12]), achieving neutron efficiencies in the 20% to 30% range depending on the materials used and the application. We present here an investigation into the utility of using a water-based neutron detector for the purpose of non-destructive assay (NDA) of special nuclear material.

2. The water-based well counter

The active volume of the water-based well counter (WBWC) comprises 1.02 m^3 of pure 18 MΩ deionized water doped with 0.5% gadolinium-chloride ($GdCl_3$), contained within a stainless steel tank (121.9 cm × 91.4 cm × 119.4 cm). To protect the doped water from the corrosive effects of chlorine on stainless steel [13], the inside of the tank was coated with a baked on layer of Teflon. Fig. 1 shows a schematic and picture of the detector. There are eight waterproof Hamamatsu R7081 10-in. PMTs mounted at the top of the detector looking down into the water volume. The water level is filled to half way up the PMT bulbs so that they are approximately neutrally buoyant. All of the PMT supports were constructed from clear acrylic or reflective white polypropylene, relatively inert polymers that do not react with deionized water, to maximize the transmission and/or reflection of photons in the detector. Also mounted from the top in the center is a 19 cm diameter well, or source cavity, that extends 73 cm down into the tank (approximately 45 cm into the water). The well accommodates square samples as large as 15 × 15 cm^2. In order to efficiently transport Cherenkov photons to the PMTs the walls of the tank were also coated with a 1.0 mm reflective layer of GORE® DRP®,— a Teflon-based highly reflective material (> 99% in the blue and near UV).

Signals from each of the eight PMTs were sent into a CAEN V975 fast amplifier where they were amplified and split, with one signal sent to a CAEN V814 discriminator and the other to a Struck SIS3320 waveform digitizer (WFD). The trigger was generated by a CAEN V1495 FPGA from the simultaneous arrival of any three discriminator signals. Once a trigger is issued, the WFD can either record full microsecond (μs) long waveforms with a 5 ns (ns) sampling interval, or digitize a set of independently integrated waveform sections for each PMT.

The PMT gains were set relatively high ($\sim 10^7$) to resolve single photoelectron peaks, enabling easy gain calibration via a green LED permanently mounted inside the detector.

3. Characteristic response to neutrons and gamma rays

Spontaneous fission sources, such as ^{252}Cf, emit coincident gamma rays and neutrons with every fission, which can result in a set of correlated events in the detector. If the fission gamma rays are of sufficient energy ($\sim$1 MeV) and multiplicity, they may produce an instantaneous response in the WBWC. Neutrons are efficiently moderated in the active water volume and preferentially capture on gadolinium. The large gadolinium capture cross-section results in a short mean capture time of 16 μs. When multiple gamma rays and neutrons are produced simultaneously, the result is a sequence of correlated events, beginning with either Cherenkov light from above-threshold gamma ray(s), or with a neutron capture (if the prompt gamma rays failed to trigger the detector), and followed by delayed neutron captures. In any given correlated sequence, events occurring after the first event are more likely to be neutron captures. Uncorrelated event sequences may also arise, from the random arrivals of background gamma rays, or from two or more different source fissions.

Fig. 2 shows the distribution of time intervals between successive events from a one-hour calibration run with a 1.0 μCi ^{252}Cf fission source placed at the base of the source cavity. The inter-event time distribution has two exponential components—a fast decaying correlated component with mean inter-event time 12.3 μs, and an uncorrelated component with mean inter-event time of 395 μs. The short time constant exponential is associated with the correlated neutron bursts of interest. This is shown in Fig. 3, where events with small inter-event times have a spectral shape enhanced at higher energies by the excess of neutron captures. The underlying

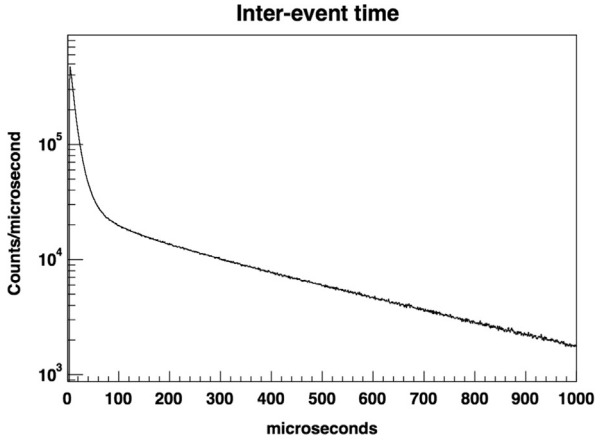

Fig. 2. A plot of the inter-event time distribution for a one-hour measurement of a ^{252}Cf source in the source well/cavity. The distribution is well fit by a double exponential function—indicating a correlated and non-correlated component. The correlated component, at small inter-event times, is due to the thermalization and capture of multiple simultaneously emitted neutrons in the detector. Its exponential has a time constant consistent with a mean inter-event time of 12.3 μs.

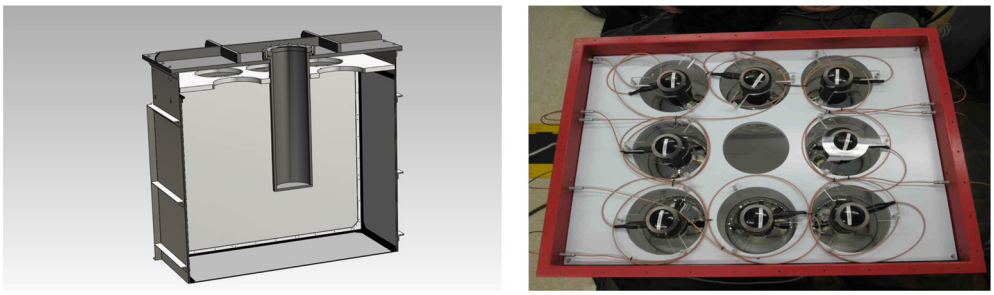

Fig. 1. A schematic (left) of the detector showing a cut away of the 73 cm deep source deployment well/cavity and PMT placement (PMTs not shown). To the right is the finished detector immediately after PMT placement inside and prior to the installation of the lid and well.