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LLNL-TR-648508

Final Report: Water-Based Neutron Detector Technology for Material Characterization Well Counters

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January 15, 2014

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Final Report

Water-Based Neutron Detector Technology for Material Characterization Well Counters

Project number LL13-Mat CharWellCount-PD2Lb

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Date: 1/9/2014

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Release number LLNL-xxx-xxxxxx

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1. INTRODUCTION

Coincidence counting of neutron pairs is an effective way to non-destructively determine the amount of fissile material within a sample of special nuclear material (SNM) [1]. Multiplicity counting is more versatile and precise, but also more demanding, requiring the detection of three or more neutrons per single fission event. Detecting a triple coincidence of neutrons depends on the 3rd power of the detection efficiency and so on. The detection efficiency quickly becomes the critical determining factor in evaluating the utility of a particular neutron multiplicity detection technique.

The purpose of this feasibility study was to characterize the performance of an LLNL-designed water Cherenkov based Multiplicity Well counter. Characterization tests included efficiency and background rejection, under both low and high background conditions, in order to justify inclusion of the technology in a 2-3 year in a DNN-supported venture, which started in 2014. We received funding early in FY2013, and have kept to our original 12-month timeline for completion of the study and reporting of results.

We report that for sources that produce a low rate of gamma ray emission, the absolute neutron detection efficiency of this detector is 28%. The ^{60}Co gamma ray rejection factor, a common metric for comparison of ^{3}He alternatives, is 10^{8} to 1. Both numbers are competitive with current ^{3}He-based systems and certainly compare favorably with non-^{3}He-based systems. This means that for fresh fuel and low activity waste samples the water-Cherenkov system is likely to perform very well when compared to current techniques. For high activity sources, such as spent fuel, the high gamma-rejection factor also implies that the system may be able to measure the content of spent fuel with a significant degree of burnup. However, further study is needed with real fuel samples quantify this latter expectation. We are proposing to perform this as a follow on task under the existing venture or through a separate project, as directed by the DNN Program Office.

2. MOTIVATION

In recent years the severe shortage of ^{3}He has been a great concern for governments and organizations involved in nuclear security. ([2],[3],[4]). ^{3}He detectors are uniquely suited for neutron detection, since they are insensitive to gamma rays, have a high neutron capture cross section, and are safe and non cryogenic. In particular, tightly packed arrays of ^{3}He tubes, surrounded by moderating material, are highly efficient. They have been in wide use since the 1970s to measure neutron multiplicities from fission chains, and hence the fissile content of both fresh and spent nuclear fuel, as well as other fissile material matrices. ^3He and polyethylene based well counting systems range in efficiency from 10% to 50%, depending on how tightly the tubes are packed and the gas density. Highly efficient and large systems, however, require the use of a large fraction of the yearly supply of ^3He and have become prohibitively expensive. In recent years the number of competing neutron detection techniques has proliferated in response to the ^3He shortage. Most are not ready for widespread use. Boron based systems such as BF_3 and ^{10}B tubes/planes are either toxic or relatively inefficient. Scintillator-based solutions generally 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. Given that the ^3He shortage is projected to continue for the foreseeable future, alternative techniques are clearly needed, and are being actively pursued by various end users.

3. THE DETECTOR

In order to perform this study, we reconfigured at low cost an existing LLNL detector, tailoring its form factor to multiplicity counting scenarios. The detector comprises 1.02 m^3 of pure DI water doped with 0.5% gadolinium-chloride ($GdCl_3$) contained within a stainless steel tank (121.9 cm x 91.4 cm x 119.4 cm). To protect the water from the corrosive effects of chlorine on stainless steel ([5]), the inside of the tank was coated with Teflon. Figure 1 shows a schematic and picture of the detector. There are eight waterproofed Hamamatsu R7081 10-inch 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 hemisphere, so that they are approximately neutrally buoyant. The PMT supports were constructed from inert clear acrylic or reflective white polypropylene, to maximize respectively the transmission and/or reflection of photons in the detector. A 19 cm diameter well or cavity was mounted from the top and center of the detector, extending 73 cm down into the tank (approximately 45 cm into the water). The well allows for the interrogation of samples as large as 15 cm across. In order to most efficiently capture and detect Cherenkov photons, the walls of the tank were also coated with a 0.5 mm reflective layer of GORE® DRP®, - a Teflon-based highly reflective material (> 99% at blue wavelengths).

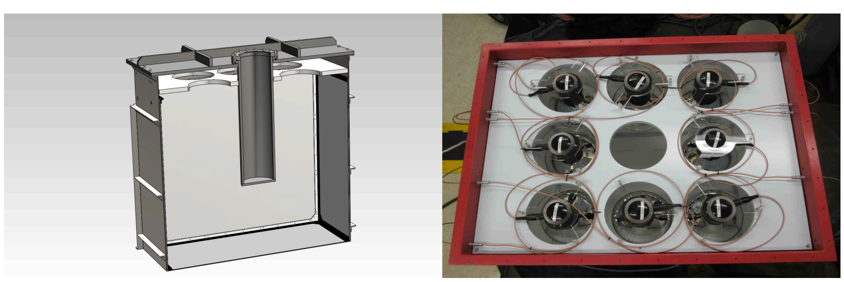

Figure 1: A schematic (left) of the detector showing a cut away of the 73 cm deep source deployment well/cavity and PMT placement (PMTs not shown). To the right is the finished detector immediately after PMT placement inside and prior to the installation of the lid and well.

While the detector is not fully optimized for this work, our study already reveals excellent performance, using the standard metrics that have been developed to evaluate ^{3}He alternatives. In this report, we describe the reconfigured detector and present our results. We believe these promising results offer a uniquely beneficial solution to the ^{3}He problem, and fully justify further development of the technology.

4. CHARACTERISTIC RESPONSE TO NEUTRONS AND GAMMA RAYS

The neutron source used in the following measurements was a spontaneous fission 1 μCi ^{252}Cf that emits 4400 neutrons per second. We compared the detector response to correlated events (mostly neutron pairs from single fissions), with uncorrelated events (accidental coincidences of independent gamma rays or neutrons), and generated a pure neutron capture detector response spectrum by statistically subtracting the normalized uncorrelated spectrum (see Figure 2 (left)). In Figure 2 (right) the spectral response of the detector to a 220 kBq (5.9 μCi) ^{60}Co source positioned inside the well for one hour is compared with a one-hour background run (no source), generating a pure ^{60}Co spectrum. The "pure" neutron capture spectrum is again included for comparison. The plot illustrates that nearly all of the ^{60}Co gamma-ray events can be removed by simply applying an energy cut at 50 photoelectrons. Remarkably, the gamma rejection/suppression factor is 10^{8} to 1 – competitive with ^{3}He-based detectors while the neutron efficiency remains high at 28% (see below). Unlike scintillator-based detectors, low energy gamma rays never generate a visible Cherenkov signal in the water, providing an intrinsic suppression mechanism against these backgrounds. For this feasibility study we employed the ^{60}Co source as a proxy for a source that emits a low intensity, ~1 MeV gamma ray background, which is a standard approach when evaluating ^{3}He alternatives (e.g. [6]). The pure ^{60}Co detector response spectrum shown in blue is background subtracted. Also shown for comparison is the spectrum of neutron capture event candidates. The neutron capture spectrum was then compared with the predicted detector response from our tuned GEANT4 detector simulation in Figure 3. The fit above 25 photoelectrons is very good, reinforcing the view that the detector is well described by our model. Below 25 photoelectrons the two curves diverge since the trigger is not modeled in our simulation.

We calculated the neutron efficiency of the detector when applying an energy cut at 50 photoelectrons, using both the simulated neutron capture curve and the nominal ^{252}Cf source intensity. In both cases the efficiency was 28% ± 0.5%. Both efficiency calculations include ALL neutrons emitted by the source, not simply the neutrons that hit the detector face (i.e. we are calculating the absolute, not intrinsic efficiency).

Figure 4 shows a highly important and useful feature of water Cherenkov detectors in the particular context of spent fuel assay. Unlike scintillator-based detectors, low energy gamma rays never generate Cherenkov photons, providing an intrinsic suppression mechanism against these backgrounds. In many cases, the 662 keV gamma ray emission from ^{137}Cs is the dominant background from a spent fuel sample. The detector response to ^{137}Cs is very small, indicating that Compton scattered electrons from these gamma rays rarely exceed the Cherenkov threshold in water. The ^{137}Cs source used in these measurements had an intensity of 335 kBq (9 μCi). The total detected event rate in the detector was less than 10 Hz, all of which was below 25 photoelectrons, and therefore not a neutron background.

It should be noted here that the 'no source background' events, which are present in Figures 2 (left and right), and Figure 4 are caused primarily by cosmic ray and local environment gamma rays, neutrons and muons incident on the detector. Water Cherenkov detectors are sensitive to these radiations if they deposit significant energy in the water (> ~2 MeV). Both these backgrounds, and the detector response to them have been remarkably stable over the six or so months we have been taking data. In fact we have so far not measured any significant change in the spectral shape or rate of this background in 6 months. What this means is that this background can be measured extremely accurately and subtracted from the source related data.

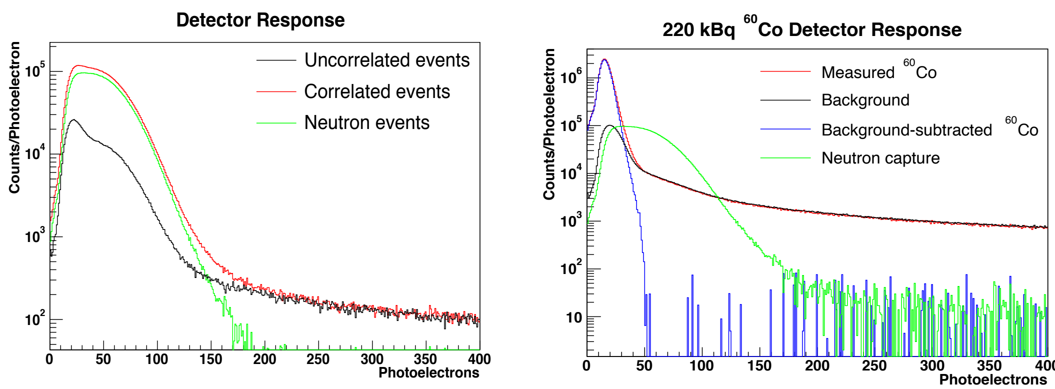

Figure 2: Left plot: Comparison of the detector response (in terms of the number of photoelectrons detected), of a group of correlated events (red, neutron rich) and uncorrelated background (black, neutron poor). The "pure" neutron capture spectrum, which is a statistical subtraction of the two, is shown in green. Right plot: The detector response of a 220 kBq ^{60}Co source for a one-hour well deployment (red). A one-hour no-source background run is also shown (black). The blue curve shows the pure background subtracted ^{60}Co component. For comparison with the ^{60}Co spectrum, the "pure" neutron capture spectral response is shown again in green.

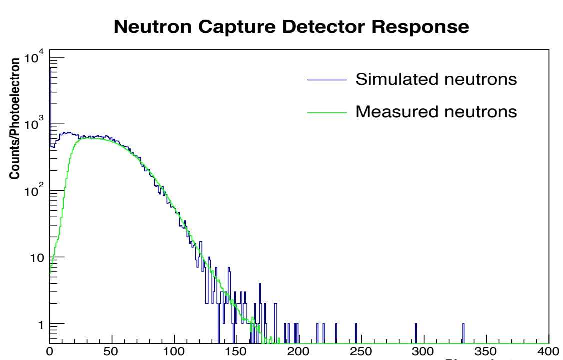

Figure 3: A comparison of the simulated (blue) and real data (green) neutron capture detector spectrum.

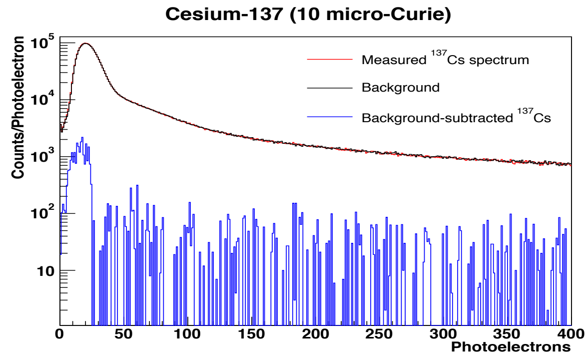

Figure 4: A one-hour 137Cs data acquisition (red) compared to a one-hour background (no source), run (black). The difference, which can be attributed to the 137Cs source alone is shown in blue.

Finally, we investigated the response of the detector to high intensity, low energy backgrounds – as one might expect from a spent fuel source. Even if low energy backgrounds are not energetic enough to trigger the detector on an event-by-event basis, it is possible for pileup to degrade the energy resolution of the detector at higher energies. This pileup may arise when the rate of low energy gamma-rays is so high as to create a small amplitude but nearly constant 'wash' of Cherenkov light, superimposed on the Cherenkov light created by real neutron captures. Recall that at low background intensities, an energy cut at 50 photoelectrons rejects nearly all of the ^{60}Co gamma rays. At high intensities however, it is necessary to reject these events with the trigger so that the DAQ is not swamped. We therefore increased the PMT trigger threshold somewhat from the nominal 60 mV setting, so that the detector triggers only on genuine neutron captures. We tested two high PMT threshold settings – 140 mV and 180mV, corresponding to neutron efficiencies of 22% and 12% respectively. A high background fissile source was simulated by a pair low energy event proxies, the ^{60}Co source and a fast pulsing LED, together with the 1 μCi ^{252}Cf source. The LED was biased to produce approximately the same charge per event in the DAQ as the ^{60}Co. We then measured the neutron sensitivity as a function of increasing LED pulse rate. Figure 5 shows the detector energy resolution performance as a function of increasing intensity of low energy backgrounds. We have subtracted the non-source related backgrounds such as those that result from muons traversing the detector or multi MeV gamma rays from the local environment. The ^{60}Co and LED related backgrounds are not subtracted. Resolution was reasonably consistent at ^{60}Co equivalent background levels up to ~4 MBq.

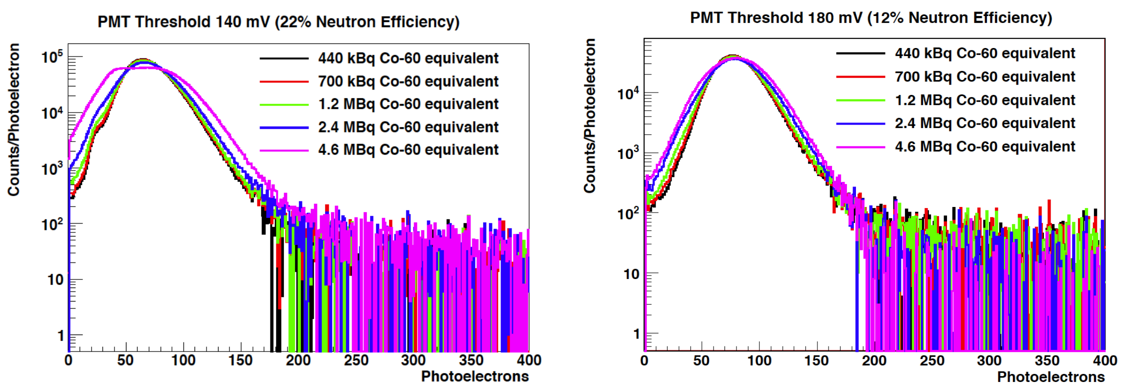

Figure 5: The detector spectral response for one-hour ^{252}Cf data acquisitions in the presence of progressively more intense low energy background. The ^{60}Co equivalent rates were modeled by the sum of a ^{60}Co source and a pulsing LED. We show the effect for two different PMT trigger thresholds, 140 mV and 180 mV, corresponding to 22% and 12% neutron efficiency respectively.

If, as before, we accept events between 50 and 200 photoelectrons as neutron candidates, the neutron count rate at each background level is given in Table 1. The data indicate that neutron efficiency is consistent to within 5% up to a ^{60}Co equivalent source intensity of ~4 MBq. Note also that the lower threshold (140 mV), capable of 22% neutron efficiency, is as effective at providing neutron detection consistency over a large range of background intensities as the higher threshold setting (180 mV).

Table 1: Measured neutron detection rate for steadily increasing rates of ^{60}Co equivalent background source intensity.

^{60}Co Equivalent Background Rate	Neutron Detection Count rates (Hz)	
	Threshold 140 mV (22% n Efficiency)	Threshold 180 mV (12% n Efficiency)
440 kBq	965 Hz	478 Hz
700 kBq	965 Hz	480 Hz
1.2 MBq	964 Hz	485 Hz
2.4 MBq	968 Hz	493 Hz
4.6 MBq	1010 Hz	558 Hz

5. CONCLUSIONS

We report that under low background rate conditions, such as for a fresh fuel or low level waste samples, the neutron detection efficiency of this detector is 28%. The ^{60}Co gamma ray rejection factor is 10^8 to 1. Both numbers are competitive with current ^{3}He-based systems and certainly compare favorably with any non-^{3}He-based systems. For high gamma ray intensity sources such as spent fuel we find that if we increase the trigger threshold such that the neutron detection efficiency is 22% (from 28%), the high-energy (neutron) detector resolution and efficiency isn't significantly affected until the background rate is equivalent to a ^{60}Co source greater than ~4 MBq. Since the use of real world spent fuel sources was outside the scope of this work, further study is needed to match spent fuel gamma ray intensity to this level of ^{60}Co background. We propose to perform this in Section 6 below.

6. PATH FORWARD

There are two obvious and urgent tasks that need to be performed with a water Cherenkov type detector in order to fully characterize it's performance under real world conditions. The first is to test a fresh fuel ^{235}U and/or Pu fission source and to directly measure the fissile content. The second is to perform the same test with a spent fuel source. The use of real world fresh or spent fuel fission sources was out of the scope of this feasibility study. We have proposed to perform this work as part of a LANL led 'venture' which was established recently to test promising non ^{3}He-based multiplicity counters. In the future, it may be possible to extend the background range over which water-Cherenkov based systems may be used by taking advantage of segmentation (reducing pileup within each segment), using lead shielding in the source well, increasing the detector energy resolution using water soluble wavelength shifter, high QE PMTs or more reflective materials.

7. PRESENTATIONS AND PUBLICATIONS

A paper will be submitted soon to a peer-reviewed journal to report the details of our measurement.

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