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Recent Developments In Fast Neutron Detection And Multiplicity Counting With Verification With Liquid Scintillator

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Abstract. For many years at LLNL, we have been developing time-correlated neutron detection techniques and algorithms for applications such as Arms Control, Threat Detection and Nuclear Material Assay. Many of our techniques have been developed specifically for the relatively low efficiency (a few percent) attainable by detector systems limited to man-portability. Historically, we used thermal neutron detectors (mainly ^{3}He), taking advantage of the high thermal neutron interaction cross-sections. More recently, we have been investigating the use of fast neutron detection with liquid scintillators, inorganic crystals, and in the near future, pulse-shape discriminating plastics which respond over 1000 times faster (nanoseconds versus tens of microseconds) than thermal neutron detectors. Fast neutron detection offers considerable advantages, since the inherent nanosecond production time-scales of spontaneous fission and neutron-induced fission are preserved and measured instead of being lost by thermalization required for thermal neutron detectors. We are now applying fast neutron technology to the safeguards regime in the form of fast portable digital electronics as well as faster and less hazardous scintillator formulations. Faster detector response times and sensitivity to neutron momentum show promise for measuring, differentiating, and assaying samples that have modest to very high count rates, as well as mixed fission sources like Cm and Pu. We report on measured results with our existing liquid scintillator array, and progress on the design of a nuclear material assay system that incorporates fast neutron detection, including the surprising result that fast liquid scintillator detectors become competitive and even surpass the precision of ^{3}He-based counters measuring correlated pairs in modest (kg) samples of plutonium.

Keywords: Nuclear Instrumentation, Fission, Fission Chain, Fast Neutron Detection, Time-Correlated particle Detection, Special Nuclear Material Detection, Assay.
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INTRODUCTION

The low natural background rates and the penetrating nature of neutron radiation make neutron detection (particularly time-correlated neutrons) a good method for quantifying and accounting for large amounts of special nuclear material (SNM) capable of undergoing neutron induced fission and supporting subsequent fission chains. Fission is one of the few natural processes that produces time-correlated neutrons -- the others are spallation-type processes, like (n,xn) and cosmic induced background -- that have low but measurable rates in common terrestrial material. The high rates of most transuranic spontaneous fission sources (like Pu) of even a gram or less usually swamp typical cosmic induced background. In comparison, kg quantities of natural uranium produce neutrons only on the same order as that of typical cosmic background.

The primary characteristic of special nuclear material is its ability to fission and to support fission chains through *neutron* (and particularly *slow* neutron) induced fission. This means that neutrons produced from fission are not produced randomly but rather in time-correlated bursts. Even more importantly, these bursts occur on the time-scale of the neutron transit time through the fissionable material. This transit time-scale can be very long in comparison to fission time-scales due to the ability of slow neutrons (possibly even thermal neutrons) to create more fast neutrons by inducing fission. This means that fast neutron detectors can detect these bursts of neutrons as slowly-developing bursts of fast neutrons. The ability to discern these time structures is simply not possible with thermal neutron detectors, which blur out all such structure to thermal timescales. Examples of this can be seen in Figure 1 which shows data from a non-multiplying ^{252}Cf source (Figure 1a) and a highly multiplying Pu source (Figure 1b). The figures show the number of neutron counts (dark squares) observed in repeated measurements of these sources using a fixed 0.512 millisecond window (on a log scale), and compared to an expected distribution arising from a random source (light circles) of the same count rate. The wider variance of data (dark squares) compared to Poisson distribution (light circles) of same count rate is a clear indication of fission and is easily seen after only 52s in the highly multiplying Pu source and barely seen after 18.5 hours of measuring a non-multiplying Cf source.

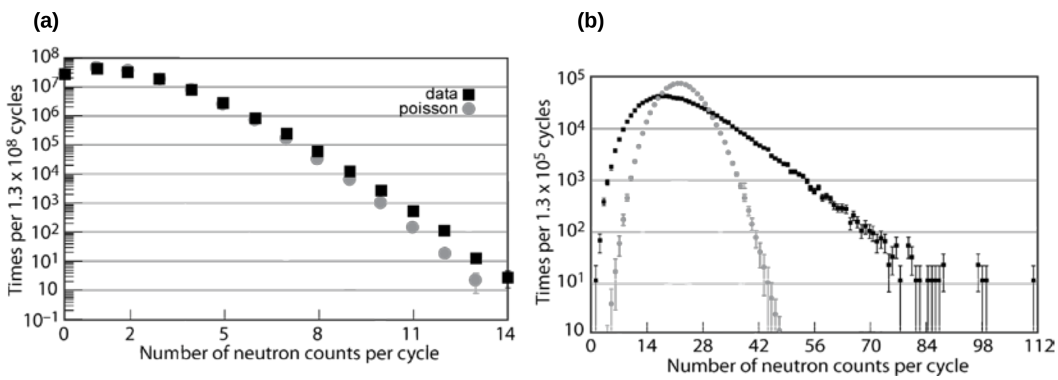

FIGURE 1. Data from (a) ^{252}Cf source, (18.5h) and (b) a highly multiplying plutonium ball (52s).

FAST LIQUID SCINTILLATION DETECTOR ARRAY

Recently, we have begun to apply our neutron analysis and assaying techniques to fast neutron detection with liquid scintillators[1,2]. Figure 2b. shows an array of liquid scintillators configured to cover about 2π of the solid angle surrounding a cylindrical chamber placed at the center of the array. Efficiency is an important factor when attempting to measure correlated events because the probability of detecting n neutrons goes as the n^{th} power of efficiency. Fast neutron detectors cannot be as efficient as the most efficient thermal detectors simply because of the minimum energy threshold of detection for fast neutron detector. However there are several important advantages to fast neutron detection that can be paramount, especially when thermal neutron detection is inadequate, as in high flux situations or cases in which neutron time history is important to the assay. In high fluxes, the probability of random correlations increases geometrically with the rate, which makes the ability to detect fission correlations increasingly difficult. Consider the measurement of ^{252}Cf in Figure 1a. ^{252}Cf only rarely fissions with a neutron multiplicity greater than 8, and yet a detector with 3% efficiency counted a non-negligible number of time-windows in which more than 10 neutrons were detected. This clearly means that even with a modest source flux of 10^5 n/s (detecting 2100 n/s) there is a significant amount of overlap of fission events within the gate window as shown in Figure 1a.

By comparison, the single most important characteristic of fast neutron detection is that it happens fast. Fast neutron detection allows the relevant detection time to shrink from tens of microseconds (detector thermalization time) to nanoseconds -- equivalent to reducing the effective flux by a factor of 10^4. Secondly, the fast detection preserves the timescale of the original neutron production. The prompt production of fission neutrons from a single fission and spallation-type processes occurs on a nanosecond timescale, while the neutron production of a multiplying body occurs in the neutron transit time, which can be tens of nanoseconds for pure metallic systems and up to many microseconds for moderated systems. Thermal neutron detection (using e.g. ^3He, BF_3, etc.) requires moderation of the neutrons for efficient detection, but moderation occurs on a timescale of tens of microseconds, which smears all the timescale details of the original neutron production.

Also more subtly, especially when combined with the introduction of, in particular, low energy neutrons below detection threshold, measuring the *change* in neutron flux can be most revealing with respect to the source of neutrons in a sample.

2a) **2b)**

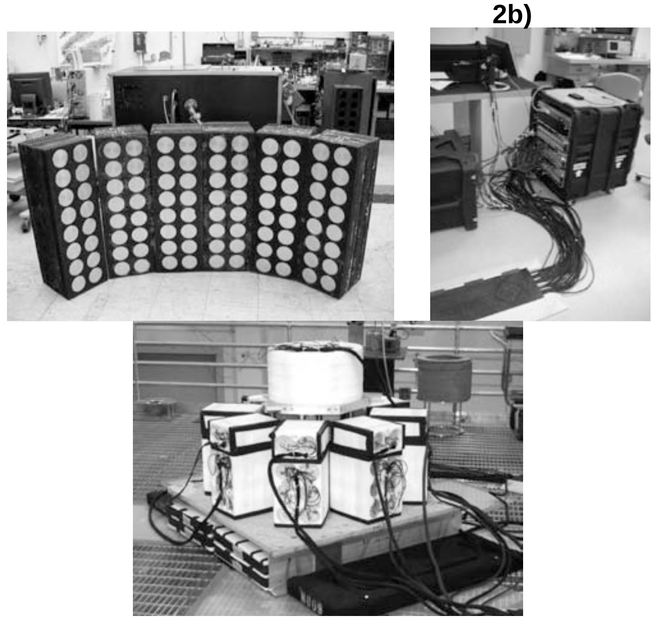

FIGURE 2. Liquid scintillator arrays (a) older configuration and (b) newer more efficient (approximately 2π solid angle) configuration.

Figures 3 and 4 are meant to illustrate the power of a fast neutron detection system with data taken with the liquid scintillator array. In Figure 3b, a pile of nearly one ton of lead bricks was measured with a thermal neutron detector of about 4% efficiency. Very large neutron correlations were seen at the tens of microseconds time scale. Comparing this to Figure 3a of a multiplying uranium system, one would be very hard pressed to tell the difference between the two. Assuming the pile of lead was in fact a multiplying uranium source, one could fit a neutron distribution which looked quite close.

We measured the pile of lead with our 1-2% efficient liquid scintillator array using the old configuration in Figure 2a, the result appears in Figure 4a. Here, the vertical axis is the log of the time interval between neutron counts in nanoseconds and the horizontal axis is linear running time of five minutes. The top wide band consists of intervals between consecutive events around the average count rate of 6 n/s, spread out between 10 microseconds and one second. The lower band consists of fast time correlations generated by cosmic interactions with the lead pile. They occur and are over in less than 10 or 20 ns. There are no time correlations occurring in the time scale between a few tens of ns and one µs. Figure 4b shows data from the same pile of lead with HEU hidden inside. For this configuration, you would see no gamma ray signatures and the thermal detector would look no more correlated than the data in Figure 3a, but the liquid scintillator measurement reveals time correlations of *fast neutrons* occurring in the intermediate time scale where nothing occurred in the pure lead pile. This is a clear indication of the presence of nuclear material because this intermediate time scale can only occur when *slower neutrons induce fission* in the uranium and producing more fast neutrons which cannot happen in lead alone.

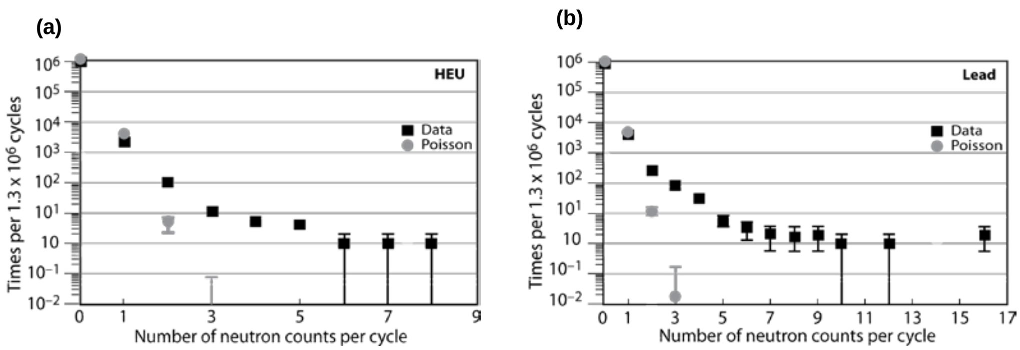

FIGURE 3. Thermal neutron data taken on 3a) an HEU object and 3b) on about a one tonne pile of lead interacting with cosmic rays.

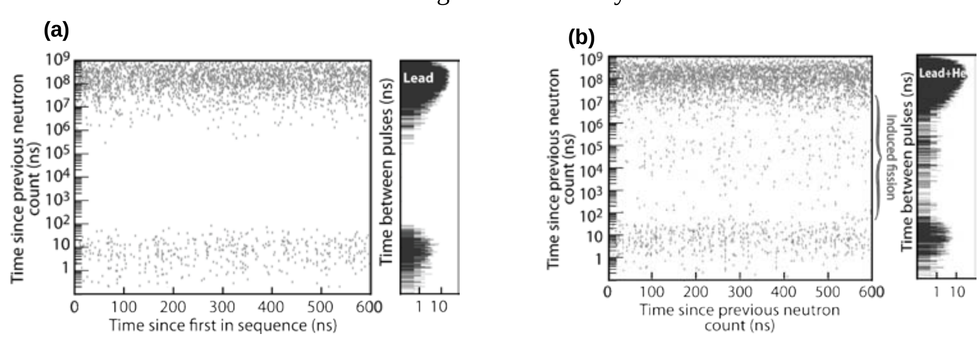

FIGURE 4. Fast liquid scintillator neutron data taken on (a) ~1 tonne pile of lead (old configuration) and (b) the Pb pile with a multiplying HEU object hidden inside. 600s of data plotted neutron arrival time (x axis) vs. log (running time) to next arrival (y axis). Note the absence of correlations in the 40 ns to 1 μs time scale in (a) but their presence in Fig. (b).

The next two figures represent assaying real objects with the liquid scintillators and illustrate the advantage of fast timing for suppressing the effect of random correlations. In Figure 5, a ^{252}Cf source was measured with the liquid scintillator array in the new configuration (shown in Figure 2b) that is about 6% overall true efficiency i.e. 6% of the neutrons emitted from the source were detected. Note the clean separation from a Poisson distribution for the same count rate because of the short time gate possible only with fast timing. Even with a higher count rate than was measured in the thermal neutron measurement of Figure 1a, there is clearly very little contamination from random correlations, and an assay was easily accomplished in 4 minutes of elapsed time (99.25 seconds of data), compared to the 18.5 hours of data shown in Figure 1b.

(a) (b)

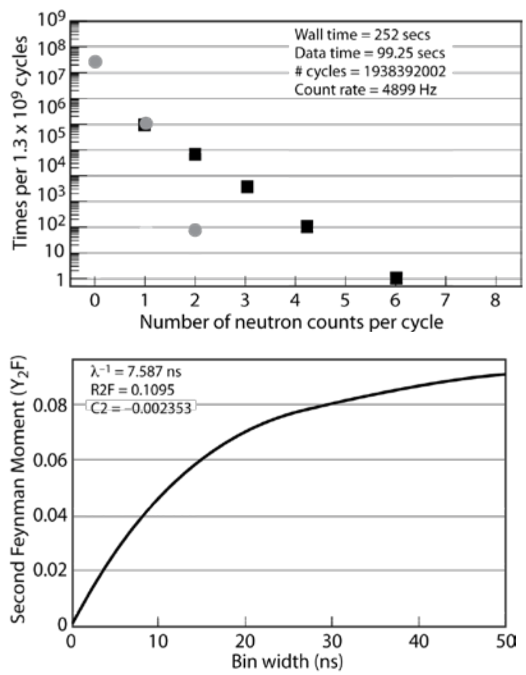

FIGURE 5. Fast liquid scintillator neutron data for ^{252}Cf source (99.25s) with a) counts in 50 ns width gate and b) Feynman second moment from 1-50ns width bins.

(a) (b)

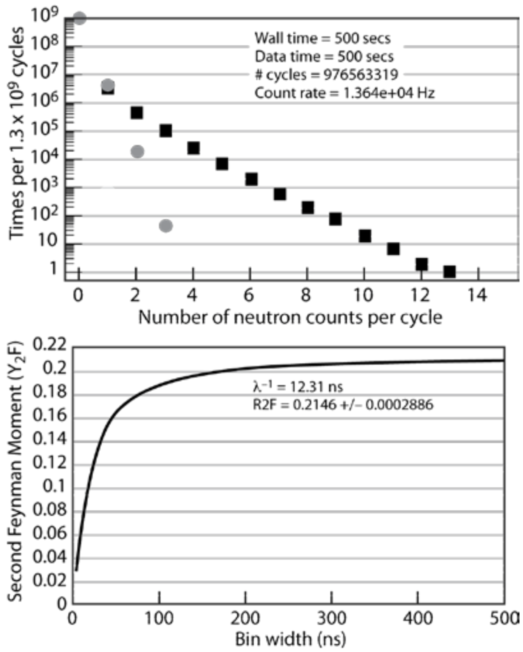

FIGURE 6. Fast liquid scintillator neutron data for plutonium source (500s) with a) counts in 500 ns width gate and b) Feynman second moment from 1-500ns with bins.

This advantage also extends to plutonium systems. Figure 6 shows the measurement of a small multiplying plutonium ball and an assay good to a few percent accuracy was completed with 5 minutes of data. Again, note the clean separation from a Poisson distribution for the same count rate because of the short time gate possible with fast timing. It is also important to note difference in time constants: for the ^{252}Cf source (a non-multiplying fission source) in Figure 5b, the time constant is 7 nanoseconds, and for the metal plutonium source in Figure 6b, the time constant is 12 nanoseconds, nearly twice as long, owing to slower timing as some neutrons are absorbed, causing additional fissions sustaining fission chains. This implies that the measured timescale of a system may be the most significant measurable difference between systems with significant material able to support induced fission from slower neutrons and those without.

USE OF LIQUID SCINTILLATORS IN SAFEGUARDS

The much lower random correlation rates in the faster liquid scintillator versus thermal ^{3}He is seen in both the ^{252}Cf data and the Pu data of Figure 1 compared with Figures 5 and 6. This lower rate of random correlations, strictly because of the shortened time bins attainable, has a profound implication for safeguards measurements in a world without ^{3}He detectors. In safeguards, most measurements use the “shift-register” method of obtaining the net neutron pair coincidences and then subtracting off the amount of random correlations seen a long time later in order to correct for the random neutron occurrences. This is a very robust way to measure, as long as the multiplication of the system and efficiency of the detector is known *and* the overall count rate is low enough that random correlations are small compared to the true correlations. When the random rates become comparable to the true pair signal, then the error in the difference of two large numbers increases the error of the measurement. So, even though ^{3}He based detectors can be 5-10 times more efficient than fast liquid scintillator detectors, as the count rate increases, the shorter time gates of the faster time-scale correlations observable in a liquid scintillator detector begin to win in the ratio of error over rate. This crossover point depends of course on the source and detector, specifically on how significant the source correlations actually are. For our current ~5% efficient liquid scintillator array versus a 50% efficient ^{3}He well counter, this occurs at about 10^{6} n/s for ^{252}Cf (Figure 7a) and 10^{5} n/s for pure ^{240}Pu samples (Figure 7b). Crossovers are lower for sources in oxide form or when including the amount of alpha-n random neutrons which degrades the significance of the correlated spontaneous fission sources.

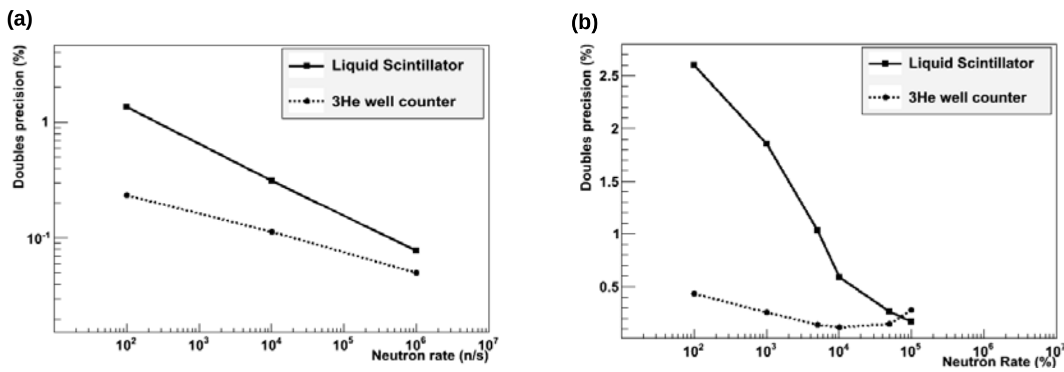

FIGURE 7. Doubles coincidence precision as a function of neutron rate for a) measured ^{252}Cf sources and b) simulated ^{240}Pu.

IMAGING WITH FAST NEUTRONS AND GAMMA RAYS

Another very interesting capability of fast detection is to exploit the joint production of gamma rays and neutrons from the same fission and use the difference in arrival times to “image” the fissioning source. This is possible because the nanosecond-scale fast timing allows separation of an individual fission from a following induced fission. Data shown in Figure 8 illustrate this concept. Two fission bursts are easily seen separated from each other and background. Zooming in on each fission burst, one can see the separation of individual fissions and following induced fissions. The gamma rays closely preceding the neutrons are from the same fission. Seeing how nanosecond timing enabled us to easily distinguish individual fissions from the following induced fission and distinguish fission-chain bursts from each other, we developed the following basic algorithm:

- search for pairs of fast time-correlated neutrons (within 10 ns) to tag a single fission event (because there are almost no random correlations occurring at that timescale);
- look for a preceding gamma ray above 1 MeV;

- for all such triples, assume that the gamma ray is infinitely fast (compared to the neutron) and take the time difference between the gamma ray and the neutron arrival at the speed of the measured neutron energy deposited in the detector;
- translate that into distance of the event from the detector;
- collect a large number of such events and populate a volume to create an image of the source, as shown in Figures 9 and 10.

Our biggest error arises from our assumption that the measured energy in the detector represents the real neutron energy -- it is only the minimum energy of the neutron given up in a proton recoil in the scintillating material. The Cf source in Figure 9 should look like a point source, its extent is mostly due to the uncertainty in the neutron energy. The asymmetry in the y-z and x-z planes comes from our detector geometry's insufficient resolution below the source. The difference in size between the Cf in Figure 9 and the Pu in Figure 10 is the actual difference between a point and extended source of a few centimeters diameter. The visual comparison shows we get reasonable images with resolution on the order of a few cm.

With Pu sources, these images can be made in seconds. It should also be pointed out that these images can be made with neutron interrogation sources as well as from intrinsic neutron sources. The neutron source can be pulsed, steady state, or our preferred way with a low-energy neutron source (in which case the source is invisible to our fast neutron detectors). For neutron-poor systems, the additional flux from a neutron source will speed up the measurement times. The ability to see fission and more importantly induced fission from events will allow evaluation of the quantity of fissionable nuclear material present by the change of the system from purely passive to interrogated, regardless of the strength of the intrinsic source. We also believe that spatial resolution can be greatly improved by considering events where the neutrons multiply scatter in the detector. This will likely lengthen the required measurement time considerably, but we can then use geometry as well as deposited energy to determine the true neutron energy and reduce our largest error.

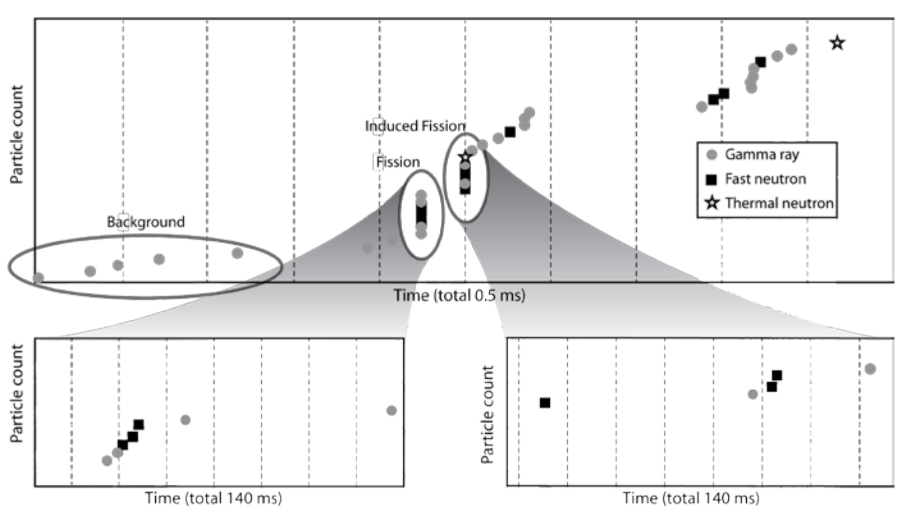

FIGURE 8. Fast liquid scintillator neutron data taken on plutonium source. The upper figure is 0.5 ms of data (horizontal axis is elapsed time, vertical axis is the order the particles were detected).

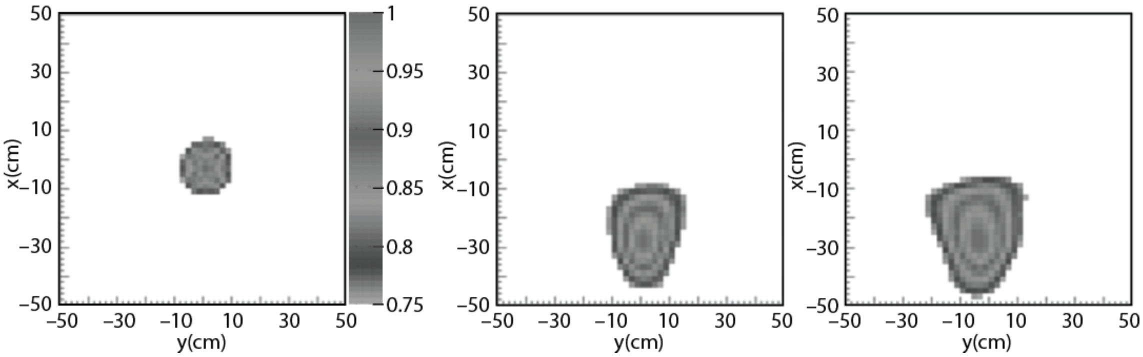

FIGURE 9. Image of a ^{252}Cf source made with gamma ray neutron time-correlations in our new detector system. The resolution (Cf should look like a point source) is mostly due to the uncertainty in the neutron energy (asymmetry in the yz and xz plains due to our detector geometry). ~5 min of data.

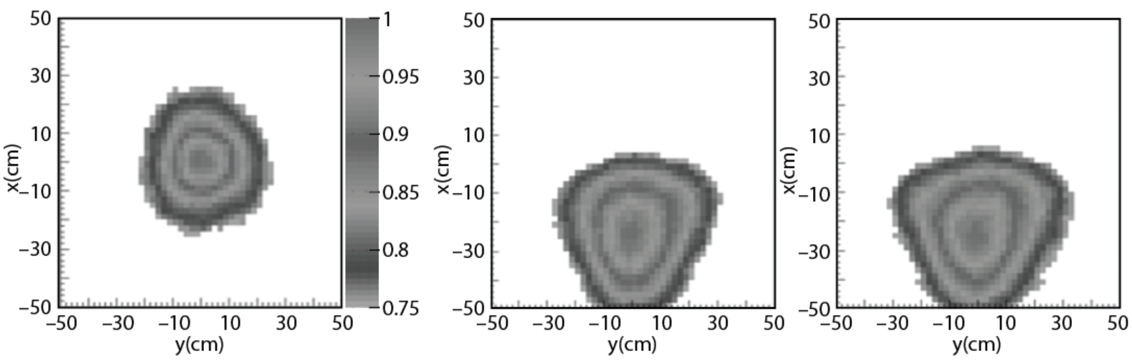

FIGURE 10. Image of a slightly multiplying Pu source made with gamma ray neutron time-correlations in our new detector system. The difference in size between Fig. 9 and Fig. 10 is the difference between a point and extended source of a few cm's diameter.

SUMMARY AND CONCLUSIONS

We have taken fast neutron data and performed assays with our liquid scintillation array that are good to a few % within a few minutes. We have been able to achieve 5 to 6% total efficiency with an approximately 2π solid angle detector by applying algorithms developed originally for low efficiency portable thermal detectors. The intrinsically fast detection time of the liquid scintillator arrays greatly reduce random time-correlations that typically plague time-correlation measurements. Fast timing also helps distinguish the time scale differences between individual fission or fission-like processes (such as cosmic induced background) and those of fission chains that are unique to nuclear material able to support induced fission from lower energy neutrons. We have shown that the intrinsically faster liquid scintillator can be competitive with the highest efficiency well-counters measuring pairs correlations using the shift-register method for modest-sized samples of Pu, and will be superior in precision to thermal neutron ^{3}He-based detectors for measuring multi-kg sized samples, such as MOX fuel rods. This makes liquid scintillators a viable replacement in the short term for ^{3}He coincidence counters proposed for in-line measurements at storage facilities, provided that questions of long term stability can be answered satisfactorily. In the longer term, plastic scintillators doped for pulse-shape discrimination (PSD) (currently under development at LLNL) can be expected to have even better stability and field performance.

We believe that it is also possible, with further development, to differentiate between complicated samples containing different fractions of spontaneous fission sources, for example Cm versus Pu or Cf, all of which produce a different average number of neutrons per fission, an unsolved problem in the safeguards community. We also believe there is a wealth of information in the detailed timing information (including imaging samples with fast neutron-gamma ray time-correlations) that can help with material control and accountability (MC&A), especially in samples with high fluxes and complicated neutron sources. Finally, the ability to see the detailed time structure of fission and induced fission makes neutron interrogation (particularly with low-energy neutrons) a very attractive technique that can certainly speed up measurement times when intrinsic neutron sources are lacking, and give useful information even when there is a strong intrinsic source. Fast neutron detection also has the potential to exploit neutron energy information not available to thermal detectors.

For the arms control and treaty verification arena, we have already shown the ability to determine just about any characteristic of the fissionable material of an object that would prove weapons origin. We can measure the amount, and through imaging we can count sources and show spatial extent. We are developing tools to show true density and thickness of material by examining the ratios of gamma rays from fission to neutrons (determined by their close time proximity). These capabilities all have profound implications for the quality of arms control verification. It remains to be seen which algorithms are to be used and how much information revealed may be too much. The choices will be determined by the verification agreements and the careful balancing of the exact way data are collected (for example adjusting the efficiency and time of the measurement) and the exact algorithms are employed.

There are many other as yet incompletely determined and developed applications for fast neutron technology. For example, related projects at LLNL have long expounded the insensitivity of fast liquid scintillators to sub-MeV neutrons as a positive feature for use of liquid scintillator with active or semi-active neutron interrogation in the search for HEU[3,4].The introduction of slow neutrons into a target system can be used to detect even small quantities of hidden HEU because the liquid scintillators are insensitive to slow neutrons and so any extra fast neutrons induced by the slow neutrons are easily detected and identified as hidden HEU. This technique has many implications for both search and material assays when dealing with unknown objects. We have only begun to tap the wealth of information available by employing fast neutron technology and the time scales of the measured-time correlations to detect and evaluate objects.

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