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Testing of Liquid Scintillator Materials for Gamma and Neutron Detection

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BASIC PHYSICS AND CONCEPT

The key fact about fissile material is that a sufficient quantity of the material can produce chains of fissions, including some very long chains. A chain of fissions will give rise to a detected burst of neutrons with longer chains generally producing larger bursts. These bursts produce distinctive time correlations in a detector near the multiplying material.

These correlations are measurable and can be analyzed to infer attributes of the fissile material including fissile material mass, assembly neutron multiplication, characteristic fast fission chain evolution time scale, also known as the α time scale, thermalization time scale. The correlation signal is very robust with respect to background and to neutron absorbing material.

FISSIONS, CHAINS AND DETECTIONS

A fission chain is a chain reaction where neutrons induce fissions in nuclei such as uranium and plutonium. As these nuclei fission, they emit several neutrons which in turn induce themselves subsequent fissions and more neutrons. Two different kinds of fission chains exist: If a fast neutron fissions a nucleus, the time between consecutive fissions is of the order of tens of nanoseconds for small objects. We refer to these as fast fission. If the neutron slows down to thermal energy before fissioning a nucleus, the time between fissions is then of the order of the thermalizing time scale of the materials, which is typically greater than a microsecond. The fission chain then restarts after a relatively long time. We will refer to these as restart chains. In both cases, some of the neutrons produced in the fission chain will induce a detected signal pulse in the nearby detection system; some will be absorbed, while some others will escape the system.

In addition to the neutrons from fission chains there may also be single neutrons coming in from the outside and neutrons from non-fission processes in the assembly (e.g. (α ,n) reactions). These neutrons may be detected, contributing a Poisson component [see below], or may initiate fission chains. Environmental neutrons that are a bothersome background for a neutron multiplicity counter (NMC) are a source of signal for the more sophisticated techniques described here. Some other background sources such as rare cosmic-ray showers can generate a large number of correlated neutrons that are difficult to distinguish from fission chains.

For conventional helium-3 based detection systems, neutrons can only be detected at thermal energies. High-energy fission neutrons need to be thermalized over tens of microseconds by a moderating material for the helium-3 to capture them. This causes a long delay between the emission and detection of neutrons. Because the thermalization time is much larger than the fission chain evolution time scale α, detection systems based on thermal neutron capture cannot measure any phenomena of the order of the α time scale. If the source rate is greater than 10,000 neutrons/sec, it is also impossible to separate fission chains. With liquid scintillators on the contrary, neutrons are detected at energies of the order of 1 MeV and higher, need not be thermalized over a period of microseconds, and can be detected a few nanoseconds after their emission, i.e. the time it takes for fast neutrons to travel from the source to the detector, give or take a few possible collisions along their path.

This different in timing characteristics between the helium-3 tubes and the liquid scintillators is essential for SNM in the form of metal for instance, where all fissions are

fast fissions. The bursts of fissions from a chain are overlapping for a helium bases detection system, while they can be separated using liquid scintillators based detection systems.

CORRELATIONS

The simplest sort of radioactive decay produces a single decay quantum, which is completely independent of all past (or future) quanta. This kind of uncorrelated process is called a Poisson process and the distribution of the number of decays in a fixed time interval follows a Poisson distribution.

In contrast to a Poisson process a fissile material assembly will produce correlated neutrons. From a detection perspective, instead of each detection event being independent of all past or future events, detections will tend to occur in bunches (see Fig. 1), where several neutrons from a single fission chain will be detected over a short time frame, of the order of the fission chain time evolution time scale α.

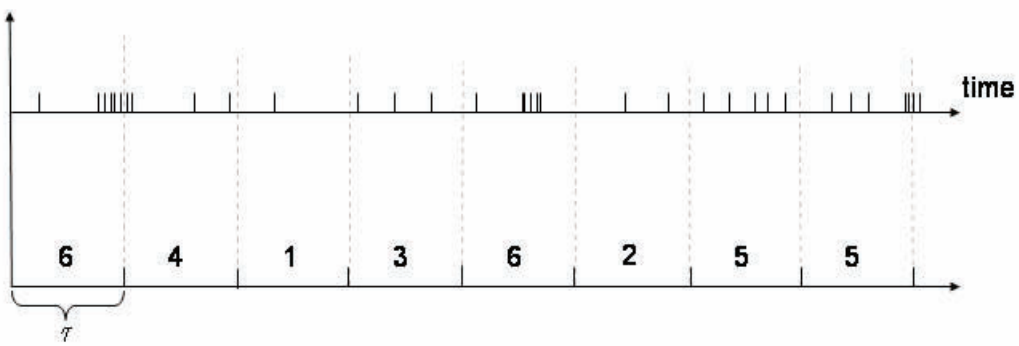


Figure 1: The upper line illustrates correlated pulses arriving in bunches as a function of time. The number of time tags within a time gates τ are reported underneath.

The key feature is that high multiplicity events (many pulses in a short time period) are much more likely for fissile material than for the corresponding Poisson process with the same total pulse rate.

DATA ANALYSIS

There are several ways to analyze and present correlation results, including correlation functions, moment methods and others. We will first focus on the discrepancy between measured and expected (Poisson) count distributions, and second on their time-depend moments.

The basic form of the data is the time of arrival of pulses (Fig. 1). Time gates are imposed on the data stream and the number of pulses within these time gates are recorded to build count distributions. One such count distribution is shown with blue points in Fig. 6. The red points represent the theoretical Poisson distribution of the same total neutron output. The discrepancy between the Poisson distribution and the multiplying source distribution shows how dramatically different a fissioning system is from what would be expected from a random Poisson process.

The most common approach until now for building count distributions (for thermal neutrons) has been to overlay a grid of time segments 512 μs in length over the data stream and to count the number of detector hits in the first 1 μs, 2 μs, 3 μs, etc. up to 512

μs. As opposed to building count distributions in which data are reused multiple times across all these time gates, the new proposed approach to building count distributions is such that different time gates do not share any of the data. In other words, time gates of different lengths never overlap. In practice, a first time gate length is chosen randomly among a fixed small set N of time gate lengths and the counts in that gate are accumulated until it closes. A second time gate is then chosen randomly and another set of events are counted within that second time gate, and so on until the data stream ends. The time gate lengths are chosen in logarithmic progression, so as to still have enough live-time/samples per time gate. The advantage of this method is that the N count distributions for these different time gates are all independent.

Moments of the count distributions are then computed. The first moment $\overline{C}$ determines the average number of counts in a time gate. The second moment $\left\langle \left(C-\overline{C}\right)^2 \right\rangle = \overline{C} + 2Y_2$ is the sum of two terms: $\overline{C}$, the variance of the count distribution of a random Poisson process, and (2 times) the Feynman variance Y_2, which is the probability to count two neutrons from the same fission event in a time gate. For a random neutron source, the Feynman variance to mean $Y_{2F} = Y_2 / \overline{C}$ is zero for all time gates. Deviation from zero indicates a correlated neutron source. Y_{2F} will be shown in the examples below.

The advantage of the new proposed approach to building count distribution is that the N moments of the count distributions are not correlated with each other. This enables us to use techniques such as chi-square (which requires independent/uncorrelated samples) to fit the theoretical/analytical expression for the moments to the data.

SIMULATION TOOLS/METHODS

The simulations presented here were done using the Monte Carlo (MC) radiation transport code MCNPX. For use in correlation applications MCNPX was modified. The correlation simulations depend on special purpose subroutines that generate particle tracking information in special files. These files record the time tags of interactions with nuclei within the liquid scintillator cells, along with information regarding the locally deposited energy, the incoming particle and the cell identification number.

MEASUREMENTS AND SIMULATIONS

To illustrate our capability to identify SNM in storage containers, we show examples of experimental data, and comparison to MC simulations. These examples are:

a) ^{252}Cf, a non-multiplying, spontaneous fission source in a close-pack detector configuration. The closeness of the detection system to the source results in a high detection efficiency. The quantity of californium is correctly determined using fast neutron counting. The agreement between the measured and predicted data sets demonstrate that the detector simulation and neutron transport are working properly. The measured data are non-Poisson as expected from a non-multiplying spontaneous fission source with $\overline{\nu}$=3.7.

b) a multiplying 2.35 kg ball of weapons grade plutonium in a AT400R metal container, with a less dense detector configuration. The efficiency is lower but the

multiplication and amount of Pu can still be quickly determined using fast neutron counting.

^{252}CF SPONTANEOUS FISSION SOURCE

The ^{252}Cf sample is a spec, a spontaneous fission source that does not multiply the spontaneously created neutrons. A spontaneous fission, though, can emit no neutrons, or as many as about eight, with a probability distribution. The average number of emitted neutron is 3.757.

The detection system used for taking the ^{252}Cf data is the close-packed assembly shown in Fig. 2, it consists of 77 liquid scintillator cells. By ray-tracing, one can determine that this detector configuration has a 50% geometric efficiency for detecting particles of an isotropic source in the middle.

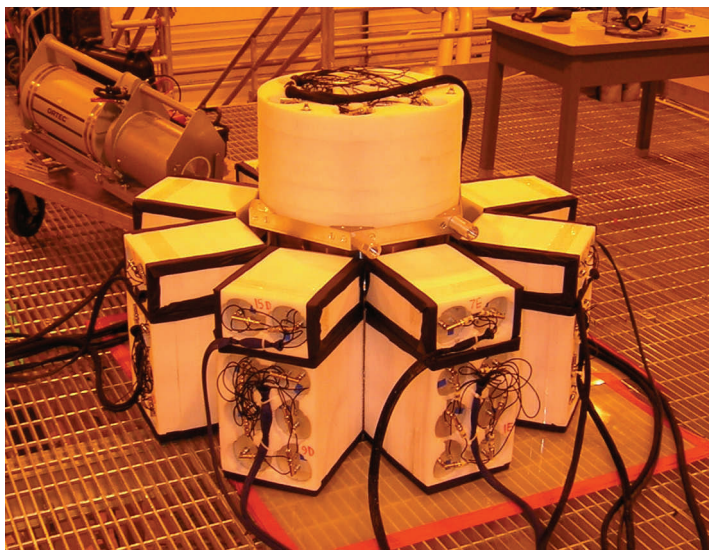


Figure 2 Liquid scintillator array used for ^{252}Cf experiment.

The count distribution for the ^{252}Cf experiment is shown in Fig. 3. The increased variance of the measured count distribution compared to a Poisson distribution is indicative of a fission source.

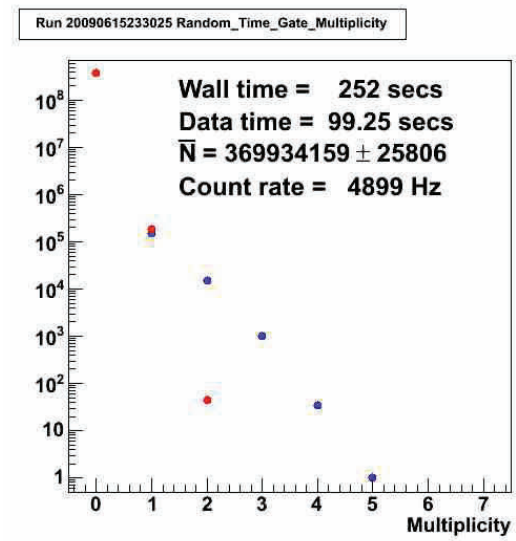


Figure 3 Measured ^{252}Cf count distribution for time gate 100 ns (blue), compared to a Poisson distribution (red) of the same count rate.

The mass of a spontaneous fission source was measured from the ratio of the count rate to the long time gate asymptote of Y_{2F}, shown in Fig. 5. The proportionality is (2.34 10^6 / D_{2S}) m_{Cf} [μg], where D_{2S}=1.6 is a parameter determined from the probability distribution that the spontaneous fission creates different numbers of neutrons (number of pairs to the average number). This mass was used for the MC simulation and the resulting count distribution is shown in Fig. 4. Apart from statistical fluctuations, the simulated count distribution matches the measured one in Fig. 3.

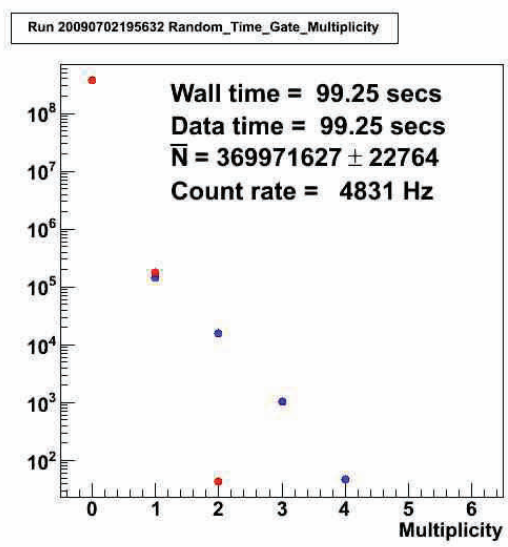
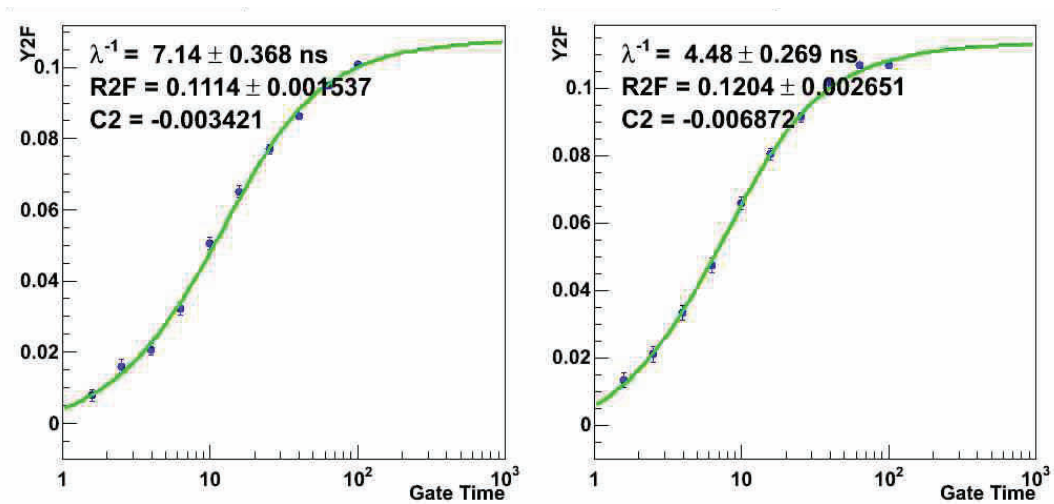


Figure 4 Simulated ^{252}Cf count distribution for time gate 100 ns (blue), compared to a Poisson distribution (red) of the same count rate.

The Feynman variance to mean Y_{2F} is shown in Fig. 5 Y_{2F} would be zero for all time gates τ for a random (non-fissioning) neutron source. Deviation from zero indicates a multiplying source.



(a) (b)

Figure 5 Time dependent Feynman variance to mean for ^{252}Cf spontaneous fission source (blue), determined from 10 (a) experimentally measured and (b) simulated count distributions varying from 1 ns to 100 ns. The red curve is a fit using a theoretical formula for the time dependence.

The red curve is a fit for the time dependence using a formula derived from a point-model theory. The fit determines a time constant, here ~7 ns. This time constant is associated with the spread in times of flight for the fission neutrons. Indeed, the closest detectors are about 30 cm away from the Cf source, while the furthest ones are about 45 cm from the source. Fast neutrons of energy 1 MeV travel 30 cm in approximately 20 ns. Given the wide energy spectrum of the spontaneous fission neutrons emitted by ^{252}Cf, the spread in arrival times is of the order of a few ns.

The efficiency of the detection system can be computed by taking the ratio of R_{2F} to D_{2S}, which is 7 % for this configuration. Given the geometrical efficiency of the detector assembly shown in Fig.2, approximately 14% of the fast neutrons crossing the liquid scintillators are detected. One should note that neutrons below ~1.3 MeV are invisible to the detectors at this moment, because they do not deposit enough energy in the liquid scintillator cells. More work on the pulse shape discrimination algorithm is currently being done to decrease that 1.3 MeV threshold. This will enable to see more of the fast neutrons and increase the efficiency.

Y_{2F} from the simulation is virtually indistinguishable from the measured data. Overall, the simulation for spontaneous fission of ^{252}Cf compares very well to the measured data.

PLUTONIUM BALL

The second example is a multiplying 2.35 kg plutonium ball in a AT400R metal container. The liquid scintillator assembly consists of 5 towers of 16 liquid scintillator cells each. Given the size of the AT400R, the liquid scintillators are further away from the Pu ball source.

The spontaneous fission rate for ^{240}Pu (489,000 fissions/sec/kg) is much larger than for other isotopes of U or Pu. This means that it is possible to acquire sufficient correlated data from a Pu-containing source very quickly compared to a uranium source of similar weight. The count distribution in Fig. 6 is for a 177.5 sec experiment.

It shows a large increase in variance compared to a Poisson distribution. This is because of enormous fluctuations in the number of neutrons created by fission chains, from one chain to the next. This system has a known multiplication of about M = 2.2.

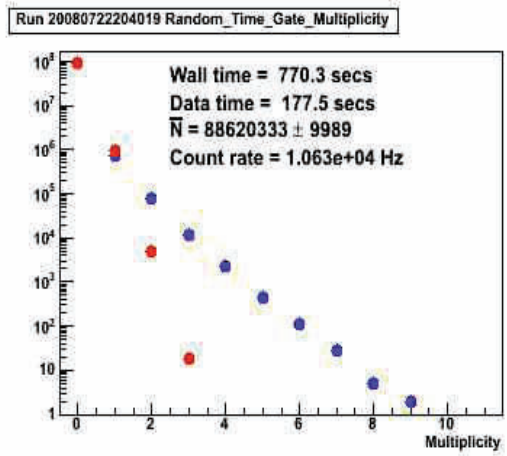


Figure 6: Measured count distribution from a moderated Pu ball for time gate 1000 ns (blue), compared to a Poisson distribution (red) of the same count rate.

The simulated count distribution in Fig. 7 is close to the measured data. The differences can be attributed to liquid scintillator detector thresholds.

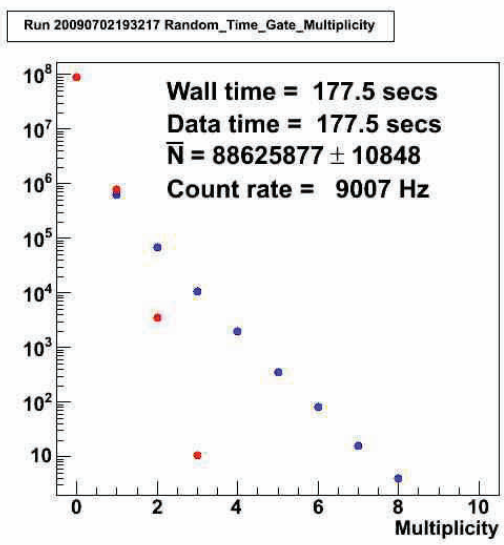


Figure 7 Simulated count distribution from a moderated Pu ball for time gate 1 μs (blue), compared to a Poisson distribution (red) of the same count rate.

The time-dependent moments of the measured count distributions are shown in Fig. 8. The multiplication of the source was determined to be 2.15 after 177.5 seconds. The standard deviation on the value of R_{3F} is however large enough so that the true multiplication 2.2 is within one standard deviation of the measured one. Longer measurement times or better pulse shape discrimination for lower neutron energies would give smaller standard deviations and better results. The efficiency for that experiment

was determined to be 3.4%. From these value, the plutonium ball mass was computed to be 2.5 kg, which is close to the true value of 2.35 kg.

The time-dependent moments of the simulated count distributions are shown in Fig. 9. The results are very close, and any difference is to be attributed to liquid scintillator thresholds, details in the source, AT400R, and detector configuration geometries.

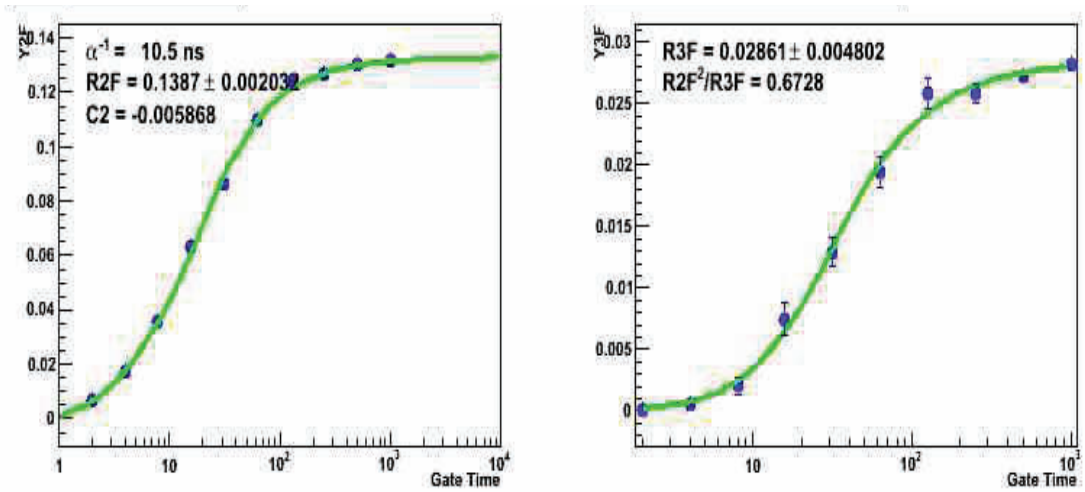


Figure 8: Time dependent Feynman moments for Pu ball (blue), determined from 10 experimentally measured count distributions varying from 1 ns to 1000 ns. The red curves are fits using a theoretical formula for the time dependence.

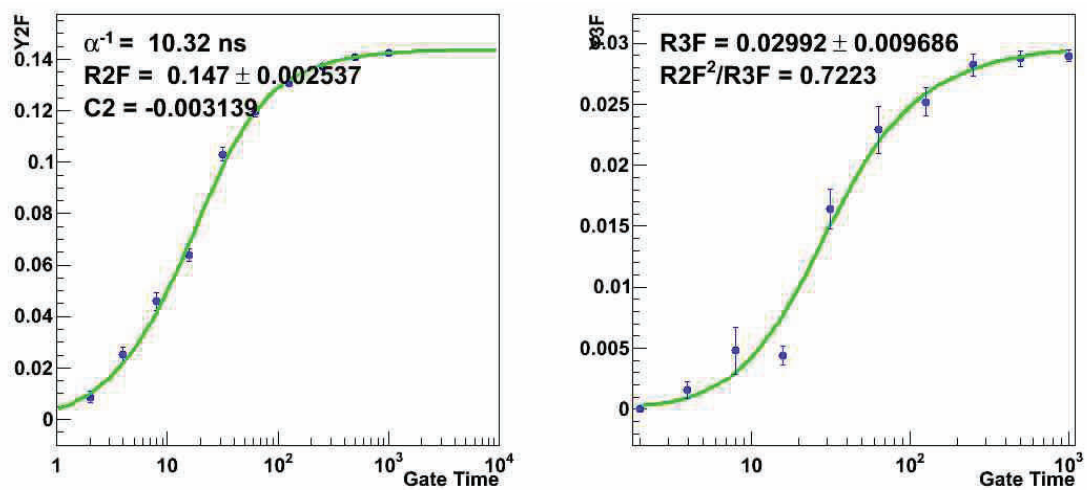


Figure 9: Time dependent Feynman moments for Pu ball (blue), determined from 10 simulated count distributions varying from 1 ns to 1000 ns. The red curves are fits using a theoretical formula for the time dependence.

CONCLUSIONS

Key lessons from these measurements and simulations are that efficiencies of liquid scintillators for detecting fast neutrons can be as high as 7% in a close-packed detector configuration, liquid scintillators can reliably measure fast neutrons and can be used to passively assess fissile materials characteristics of non-multiplying and low multiplying

systems using neutron correlation techniques, and that simulations can faithfully model the physical features of fissioning systems such as ^{252}Cf and Pu sources[2].

REFERENCES

1. J.M. Verbeke, K. Sale, A. Dougan, L. Nakae, G. Schmid, N. Snyderman, "Neutron Correlations in Special Nuclear Materials, Experiments and Simulation," INMM conference in Tucson, AZ in July 2007

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