Neutrons: It Is All in the Timing—The Physics of Nuclear Fission Chains and Their Detection

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Should a nuclear response team locate a potential threat object, it will be imperative for the team to quickly determine whether or not the object is an actual nuclear device. Only a nuclear device or a large amount of special nuclear material will sustain a significant number of fission chains. Therefore, the detection of fission chains constitutes a "smoking gun." This article is a technical introduction to the physics of fission chains and their detection using neutron time-correlation methods; it also touches on some of the Johns Hopkins University Applied Physics Laboratory's efforts to bring this important detection capability to the field.

INTRODUCTION

Since the 1970s the U.S. government has been concerned about the "loose nukes" scenario, in which a state with nuclear weapons loses control of one and it is used for a terrorist attack. In recent decades, several trends have heightened this fear. There has been widespread proliferation of nuclear weapons technology, most famously by the A. Q. Khan network.[1] Special nuclear material (SNM), the key component of nuclear weapons, has also proliferated. In the aftermath of the Soviet Union's dissolution, materials protection, control, and accountability for SNM in the former Soviet Union was poor. In the years since, materials protection, control, and accountability has been strengthened but not entirely fixed,[2] and there have been several documented cases of SNM disappearing from Russian stockpiles.

A third troubling trend has been the steadily growing number of nuclear weapon-owning states that are not signatories of the Treaty on the Non-Proliferation of Nuclear Weapons and that might act to further weaken the non-proliferation regime. Specifically, Pakistan has a growing stockpile of weapons but questionable political stability, and the government is having trouble contending with local terrorist organizations. Here, the loose nuke scenario is particularly worrisome. In addition, the Pakistani government has been complicit in the proliferation of nuclear weapon technology via the now-defunct smuggling network set up by A. Q. Khan, the founder of its nuclear weapons program. North Korea has also demonstrated a nuclear capability while behaving as a seemingly irrational international actor, heedless of international norms and unmoored by inter-

national commitments. Finally, Iran, a known sponsor of terrorism, is arguably working toward a nuclear capability, although recent diplomatic developments might ultimately limit the Iranian program.

Although it is impossible to estimate the likelihood that a terrorist organization could actually acquire a functioning nuclear device, the possibility exists via at least two different pathways.[3,4] Terrorists could steal a state weapon or they could acquire SNM and proliferated nuclear technology and then build their own improvised nuclear device.

Thus, the U.S. government maintains an assortment of assets and mechanisms for interdicting the movement of illicit SNM or an actual nuclear device. The U.S. Customs and Border Patrol operates radiation portal monitors at ports of entry throughout the United States. Radiation portal monitors are also operated in ports around the world, in collaboration with host nations as part of Customs and Border Patrol's Container Security Initiative and the Department of Energy's Megaports project.[5] In addition, a string of sensors was emplaced along the borders around Russia as part of the Department of Energy's Second Line of Defense program.[6] This patchwork of detection capabilities is coordinated under the Global Nuclear Detection Architecture managed by the Domestic Nuclear Detection Office in the Department of Homeland Security.[7]

Should any of these portals or sensors detect the passage of suspicious radiological material, a special rapid response team could be deployed to assess the threat. Alternatively, there might be an intelligence cue for the movement of SNM or a nuclear device, in which case the U.S. government would deploy national assets to search for, locate, and asses the threat.

Once the threat object is located, the response team would use diagnostic instrumentation to determine the level of threat it poses. The most pressing question for the response team would be whether or not the object was an actual nuclear device. Because the presence of nuclear chain reactions is the *sine qua non* of a nuclear device, their detection would provide the smoking gun that escalates the national response.

By their nature, these small rapid response teams necessarily have a limited load-out of equipment. The Johns Hopkins University Applied Physics Laboratory (APL) is developing technology that would give these teams the ability to detect fission chains without increasing their load-out.

THE PHYSICS OF FISSION CHAINS

To understand how fission chains can be detected, it is first necessary to delve into some aspects of the physics of fission chains. As is widely known, certain isotopes of the heaviest elements can fission into smaller nuclei. This process can happen spontaneously, or it can be induced by a free neutron colliding with the nucleus. When a nucleus fissions, it emits zero, one, or more fission neutrons with probabilities that depend on the particular isotope involved; on whether the fission is spontaneous or induced; and in the case of induced fission, on the kinetic energy of the incident neutron. The number of neutrons emitted in a given fission event is called its "multiplicity," and the probability distribution of different multiplicities is called a "multiplicity distribution." The multiplicity distributions for the spontaneous and induced fission of various important isotopes are plotted in Fig. 1.

It is possible for the neutrons emitted by the fission of one nucleus to collide with and induce fissions in other nuclei, which in turn emit more neutrons. But in order for this phenomenon to create a self-sustaining nuclear chain reaction, the probability that fission neutrons successfully induce additional fissions needs to be suf-

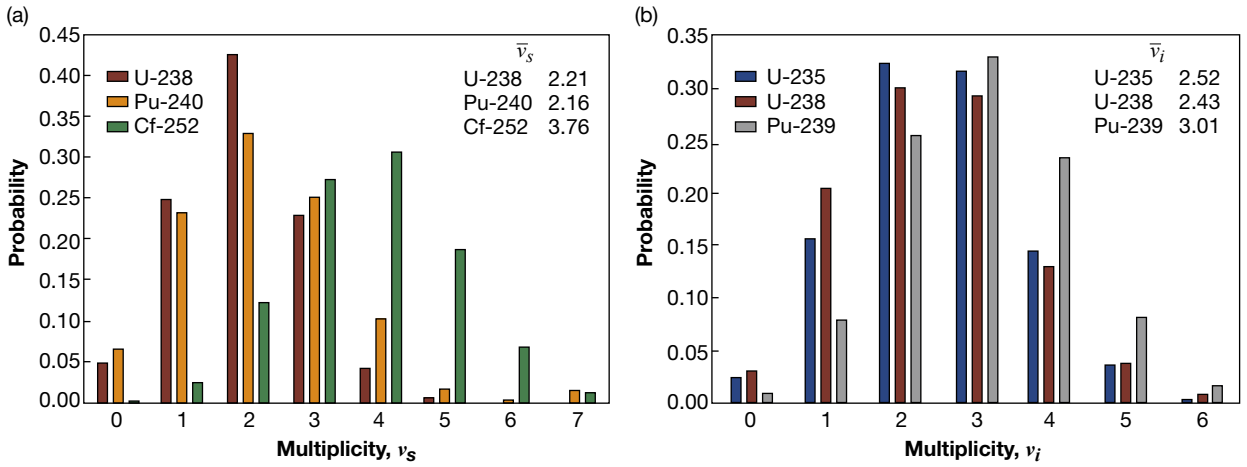

Figure 1. Multiplicity distributions and average multiplicity for various important isotopes for (a) spontaneous fission and (b) fission induced by 1-MeV neutrons.[8,9]

ficiently high. Although there are many isotopes that are fissionable, there are relatively few isotopes for which this probability is high enough to sustain chain reactions, and these select few are called "fissile."

Consider the case of uranium. The largest constituent of naturally abundant uranium is uranium-238 (U-238), which is fissionable but not fissile. The kinetic energies of most fission neutrons are not high enough to induce fission in U-238. Furthermore, U-238 has a strong predilection for absorbing intermediate energy neutrons, preventing them from inducing further fissions. (Incidentally, plutonium-239, or Pu-239, used in both power reactors and nuclear weapons, is produced using this absorption reaction: U-238 is placed inside a nuclear reactor, where it absorbs neutrons and coverts to U-239. U-239 is radioactively unstable and quickly decays into Pu-239.) On the other hand, a minor constituent, U-235, can be induced to fission by neutrons with any kinetic energy, and so it can sustain a chain reaction if there is enough of it present. Because naturally abundant uranium consists of 99.3% U-238, there is too little U-235 and too much neutron-absorbing U-238 to sustain chain reactions. Therefore, uranium needs to be enriched in its U-235 content before it can be used in a nuclear reactor or weapon. Weapons require highly enriched uranium (HEU), whereas reactors require lower enrichments and run on low enriched uranium.

We can construct a simple model for the average number of neutrons released by a single chain reaction (also called a "fission chain"). Any given free neutron can suffer one of several fates. It could, of course, induce a fission; it could be absorbed without inducing a fission; or it could do neither and escape from the nuclear material altogether. Let p be the probability that a given neutron will induce a fission. The value of p depends on the size and shape of the nuclear assembly, the type of nuclear material being used, and the fraction of fissile isotopes in the nuclear material. Furthermore, nuclear explosives are deliberately contrived such that the probability of absorption without inducing fission is small, so we will neglect absorption in our simple model. Thus the probability of a neutron escaping from the nuclear assembly is simply $1 - p$.

If a neutron induces a fission, on average $\bar{\nu}_i$ neutrons will be released. Taking into account that a neutron will successfully induce a fission only with probability p, the average number of fission neutrons in the first generation of a fission chain is then $p\bar{\nu}_i$ (see Fig. 2). Because each of these first-generation neutrons will induce their own fission with probability p, there are $p \cdot p\bar{\nu}_i$ second-generation fissions and $(p\bar{\nu}_i)^2$ second-generation fission neutrons. Thus, there are $(p\bar{\nu}_i)^n$ neutrons in the nth generation, and the total number of neutrons produced by the fission chain is

$$N = 1 + p\bar{\nu}_i + (p\bar{\nu}_i)^2 + (p\bar{\nu}_i)^3 + \ldots = \frac{1}{1 - p\bar{\nu}_i} = \frac{1}{1 - k}, \quad (1)$$

where $k = p\bar{\nu}_i$ is the multiplication factor. If $k \geq 1$, then the series in Eq. 1 diverges, and the assembly is said to be either critical or supercritical.

Each of these N fission neutrons has probability $1 - p$ of escaping the nuclear assembly, so the average number of neutrons from a fission chain that manage to escape is

$$(1 - p)N = \frac{1 - p}{1 - p\bar{\nu}_i} = M_L, \quad (2)$$

where M_L is called the leakage multiplication.

Note that this model does not take geometry completely into account. For instance, a neutron born near the surface of a nuclear assembly will have a greater probability of escaping than a neutron born at the center, and so p is not really a constant. This effect is often taken into account approximately by using a semiempirical effective value for k ($k_{eff} \cong k$) in Eq. 1. Regardless of whether or not the theoretical value ($k = p\bar{\nu}_i$) is used,

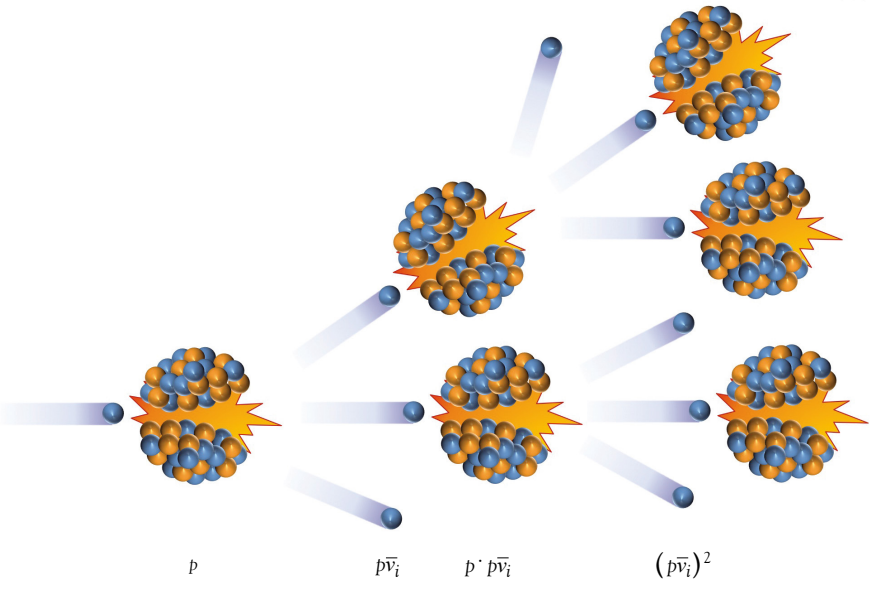

Figure 2. The evolution of a fission chain seeded by a single neutron. The average numbers of induced fissions occurring in the first and second generations are p and $p \cdot p\bar{\nu}_i$, respectively, and the numbers of fission neutrons are $p\bar{\nu}_i$ and $(p\bar{\nu}_i)^2$, where p is the probability that a given neutron will induce a fission and $\bar{\nu}_i$ is the average number of neutrons emitted by an induced fission.

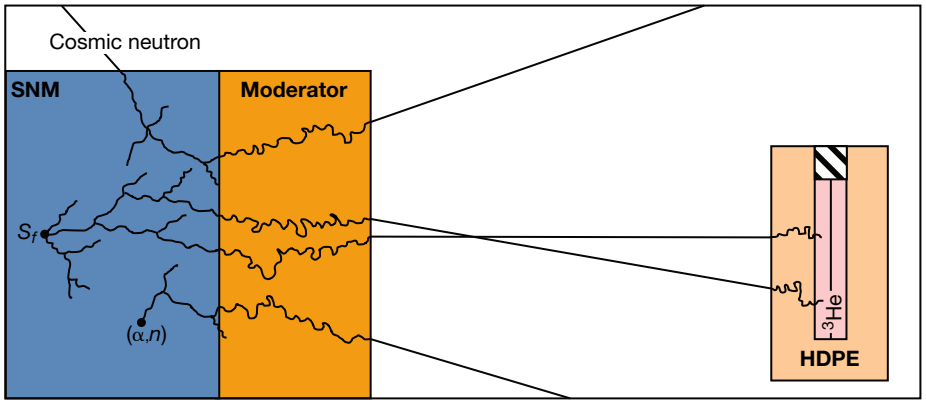

Figure 3. A schematic representation of processes occurring inside a multiplying nuclear assembly and the detection of the neutrons that leak out. Spontaneous fissions (S_f) produce multiple neutrons and (α,*n*) reactions produce only one. These neutrons or neutrons from cosmic showers seed multiple fission chains. Neutron trajectories are shown as crooked lines (indicating multiple scatterings), and the branching indicates induced fissions producing more neutrons. Neutrons that leak out of the nuclear assembly fly largely unimpeded to the He-3 neutron detector, where they scatter many times in the surrounding high-density polyethylene (HDPE) until they slow down to thermal energies, at which point they react with the He-3 in the proportional counter tube at the center and produce electrical detection pulses.

people often talk about the "*k*-effective" of a multiplying nuclear assembly.

Now suppose we start with a spontaneous fission. On average, a spontaneous fission event emits $\bar{\nu}_s$ neutrons. Each of these neutrons seeds its own fission chain, so then the total number of fission neutrons produced on average is

$$N_T = \frac{\bar{\nu}_s}{1 - k_{eff}}, \tag{3}$$

and $N_L = M_L \bar{\nu}_s$ of those neutrons leak from the assembly. Thus the original $\bar{\nu}_s$ neutrons are multiplied by a factor of $(1 - k_{eff})^{-1}$ overall and by a factor of M_L from the viewpoint of the leakage neutrons. Because only the leaked neutrons can actually be observed, the leakage multiplication is the property that is of central interest.

This simple model predicts only the average number of neutrons produced in a fission chain that leak from the nuclear assembly. In reality, this is a random number with a probability distribution that depends parametrically on the induced fission probability *p*. The mathematical theory of branching processes describes the evolution of populations whose members reproduce and die according to probabilistic laws, and the neutron population in a fission chain can be adequately described by this general theory. In particular, techniques from the theory of branching processes can be used to calculate the probability distribution of the number of neutrons leaked by a fission chain.[10–13]

Next let us turn to the sequence of events occurring inside a multiplying nuclear assembly and its time dependence (depicted schematically in Figs. 3 and 4). Free neutrons are needed to seed fission chains, and they can come from three sources. First there is spontaneous fission (indicated by S_f in Fig. 3), but the source of spontaneous fission neutrons does not have to be the same fissile isotope that supports the chain reaction. For example, a nuclear assembly made of HEU is composed of primarily U-235, but because the U-235 has a much longer spontaneous fission half-life than U-238, most of the spontaneous fission neutrons come from U-238, even though it is a minor constituent.

The second source of neutrons are so-called (α,*n*) reactions. Not only does SNM radioactively decay by spontaneous fission, it also decays by emitting α-particles. These α-particles can react with certain lighter nuclei, producing neutrons. For example, there may be trace amounts of oxygen in the metallic SNM used in a nuclear device, or the SNM may be in oxide form, as it often is in nuclear reactor fuel. Naturally abundant oxygen is composed of 0.2% oxygen-18, which can undergo the reaction

$$^{18}\mathrm{O} + \alpha \rightarrow {}^{21}\mathrm{Ne} + n, \tag{4}$$

producing neutrons that can seed fission chains.

The third source of neutrons comes from cosmic showers that impinge on the nuclear assembly. There is a flux of high-energy charged particles, consisting mostly of protons (~90%) coming from space that strike Earth's upper atmosphere. These particles interact with atmospheric atomic nuclei and generate a cascade of elementary particles. The particles in this "air shower" that reach the ground are mostly protons, neutrons, electrons, gamma-rays, and muons. At ground level, cosmic-ray showers typically have a short duration: less than 100 ns or so. So-called extensive air showers (tens to thousands of meters wide and N ~10^4–10^9) are infrequent but intense. Smaller air showers occur more frequently, with the frequency growing rapidly with shrinking size. The flux of particles at ground level depends on many factors including latitude, air pressure, and solar activity, but the average flux of neutrons is typically 80 m^{-2} s^{-1} and 1 m^{-2} s^{-1} for protons.[14]

Cosmic neutrons and protons that strike uranium nuclei in the nuclear assembly can knock neutrons free

from the nuclei through a process called nuclear spallation. These spallation neutrons have an energy spectrum very similar to fission neutrons, and they can go on to induce fission reactions. Alternatively, cosmic protons and neutrons can interact with nearby structural steel, also producing spallation neutrons that go on to strike the nuclear assembly and initiate fission chain reactions.

A single spontaneous fission, (α,n) reaction, or cosmic neutron incidence is generically called a "source event," and these events occur completely randomly in time (depicted as ×'s in Fig. 4a). That is, their occurrence times are a Poisson process (see for instance Ref. 15), and they are governed by the Poisson statistics of classical radioactive decay.

Each source event liberates one or more neutrons, and each neutron either escapes from the nuclear assembly with probability $1 - p$, or it seeds its own fission chain. In turn, each neutron in the fission chain either escapes with probability $1 - p$ or perpetuates the chain. In this way, a random number of neutrons from each chain leak from the nuclear assembly (depicted by bars of random heights in Figs. 4b–4d). The fission neutrons that leak from the nuclear assembly eventually reach an external neutron detector after suffering a random delay. (These randomly delayed arrival times are depicted by dots in Figs. 4c and 4d.)

There are several causes for this delay. First, fission chains take a finite amount time to grow, wither, and die, and a leaked neutron could have been born at any time during this life span. Second, fission neutrons are emitted with a spectrum of kinetic energies. Hence, the leaked neutrons travel with a random distribution of velocities and require a random amount of time to cross the distance separating the nuclear assembly and an external neutron detector. Finally, a leaked neutron may scatter multiple times on its journey from the nuclear assembly to the external detector. When the neutron scatters, it gives up a random amount of energy, further broadening the distribution of neutron energies. Moreover, the length of the path traveled on the way to the detector is increased by a random amount. Both effects broaden the distribution of time-of-flight delays. This last phenomenon is particularly prominent if the nuclear assembly is surrounded by low-atomic-mass material (called a moderator), which is an efficient scatterer of neutrons. An example would be neutrons passing through high explosive before reaching the detector.

DETECTION SCHEMES FOR FISSION CHAINS

The upshot is that when fission chains are present, neutron detections occur in bursts. Although fission chains are spread out and occur at completely random times, each chain releases a burst of neutrons over a relatively short interval. This "burstiness" is the signature we use to discern the presence of fission chains, and we refer to it as neutron time-correlation.

Unfortunately, fission chains are not uniquely indicated by neutron time-correlations; there are non-multiplying, non-SNM sources that also exhibit correlations. As we have noted already, there are isotopes that can still spontaneously fission or undergo induced fission—even if they cannot support fission chains—and when they fission, they also release one or more neutrons in a short burst. Both californium-252

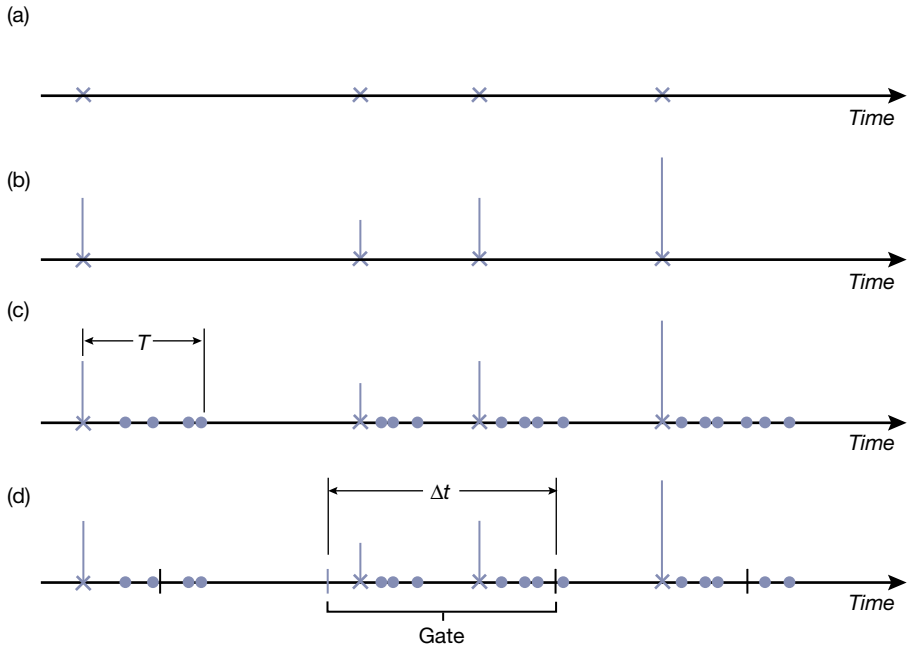

Figure 4. A schematic representation of the time dependence of events occurring inside a multiplying nuclear assembly. (a) The crosses (×) are source events, e.g., spontaneous fissions or (α,n) reactions, and they occur at completely random times (a Poisson process). (b) These source events seed fission chains that produce random numbers of neutrons, represented by vertical bars with random heights. (c) Not all of these neutrons leak from the assembly and are successfully detected, but those that do are detected after random delays from the source event times. The neutron detection times are indicated by filled circles. T is the total time spread of detection times for neutrons emitted by a single source event. (d) The time axis is partitioned into contiguous gates Δt seconds wide for generating the gated-count distributions used by one of the data analysis methods.

(Cf-252) and depleted uranium (U-238) are examples that are used industrially and that will be encountered in the field. Therefore, any detection technique will have to distinguish these materials from genuine SNM.

A Simple Detection Scheme

In principle, the most straightforward way to observe these time-correlations is to simply record the detection time of each neutron and then scan the resulting record, looking for the clumping of detection times. The multiplicity distributions for neutrons emitted by Cf-252 and U-238 spontaneous fission are plotted in Fig. 1. In the absence of fission chains, the number of neutron detections in each clump or burst would have that same distribution. Thus, the average numbers of neutrons emitted and detected would be 3.76 and 2.01, respectively. On the other hand, in fissile material, thanks to neutron multiplication occurring in fission chains, these averages would be about M_L times higher. Therefore, multiplying SNM assemblies with M_L somewhat larger than unity could be distinguished from nonfissile material by computing the average burst size in the data record, setting a threshold somewhat higher than ~3.76, and then determining whether the average burst size exceeds this threshold.

However, there are complications. Not every neutron leaking from the nuclear assembly will strike a detector, and not every neutron striking a detector will actually produce a detection pulse. The fraction, ε, of neutrons that is actually detected is called the absolute detection efficiency, and the average number of neutrons detected in each burst is reduced by this factor. If the detection efficiency is known, then the method just described can still be used if we scale the detection threshold by ε.

Unfortunately, there will never be an opportunity to independently calibrate the absolute detector efficiency in an actual field deployment. Thus, a more sophisticated approach to account for detector efficiency is required. Let N be the random number of neutrons leaked from a single source event. From our discussion in the previous section, we know that the probability distribution of this number depends parametrically on the induced fission probability p, i.e., $\Pr[N = n] = P_n(p)$. Let M be the random number of neutrons actually detected. The probability of detecting any one leaked neutron is simply ε. Therefore, the probability of detecting m out of n leaked neutrons follows the binomial distribution. This probability is, in fact, the conditional probability of $M = m$ given $N = n$, so

$$\Pr[M = m \,|\, N = n] = \binom{n}{m} \varepsilon^m (1 - \varepsilon)^{n-m}. \tag{5}$$

By applying the law of total probability, we can then calculate the probability distribution, Q_m, for the number of detected neutrons:

$$Q_m(p,\varepsilon) = \Pr[M = m] = \sum_n \Pr[M = m \,|\, N = n] \Pr[N = n] = \sum_n \binom{n}{m} \varepsilon^m (1-\varepsilon)^{n-m} P_n(p). \tag{6}$$

Because we can calculate P_n using the theory of branching processes, Eq. 6 yields a theoretical expression for Q_m that depends on two parameters, p and ε. This expression is the basis of a detection technique that handles unknown detector efficiencies. Rather than analyzing the time series of neutron detections to find merely the average size of a burst, instead histogram the number of neutrons in each burst (i.e., the "burst count") to make an empirical estimate of the probability distribution of M. Then fit the theoretical expression for $Q_m(p,\varepsilon)$ to estimate the parameters p and ε. Finally, use $k_{eff} = p_{est} \bar{\nu}_i$ to find k-effective, and then use Eq. 2 to calculate the leakage multiplication, M_L.

A computationally simpler alternative to calculating and fitting $Q_m(p,\varepsilon)$ directly is to compute its first two combinatorial moments instead. These two theoretically derived moments (which are necessarily dependent on p and ε also) are equated with the corresponding combinatorial moments of the measured distribution of M. The resulting two equations are then solved for p and ε.

In statistics, the shape of a distribution function is often characterized by its moments. There are several different classes of moments that can be used, depending on the context

of the problem. The most common is the set of central moments (which includes the mean and the variance), but other classes of moments are the factorial moments, cumulants, and combinatorial moments. The jth combinatorial moment of a discrete random variable N is defined as:

$$M_{(j)} = \left\langle \binom{N}{j} \right\rangle = \sum_{n \geq j} \frac{n!}{j!(n-j)!} \Pr[N = n]. \qquad (7)$$

In this alternative approach, the shape of the burst-count distribution, Q_m, is characterized by its moments—a common strategy in statistics. Because branching processes are essentially combinatorial in nature, the combinatorial moments have a relatively simple algebraic expression. This is the reason that combinatorial moments are used instead of the more familiar central moments, mean and variance.

Overlapping Fission Chains

A second complication occurs when the clumps of neutron detections from different fission chains overlap in time. When this happens, it is impossible to tell when one fission chain ends and the next chain begins. Thus, it becomes impossible to parse the time series of neutron detections into separate chains and then histogram the burst sizes to estimate Q_m.

Overlapping detections of fission chain neutrons occur when the typical time between source events (e.g., the spontaneous fissions that seed the fission chains) is smaller than the time spread, T, of the randomly delayed neutron detections (Fig. 4c). This circumstance arises for nuclear assemblies with high spontaneous fission rates. It can also occur in situations in which the time spread of detection delays becomes excessive—for example, when the neutrons pass through a lot of moderator.

The last case of large detection time spreads due to the presence of moderator occurs often, because the most commonly deployed detector type incorporates moderator in its design. The heart of this detector type is a proportional counter tube pressurized with helium-3 (He-3) gas. (Refer to Fig. 3 for a simplified drawing.) Incident neutrons react with the He-3 nuclei through an (n,p) reaction:

$$n + {}^3\mathrm{He} \rightarrow {}^3\mathrm{H} + p + 0.764\ \mathrm{MeV}. \qquad (8)$$

The ^{3}He and p reaction products fly apart at high velocities, carrying away the 0.764 MeV of energy released in the reaction and, by ionizing the helium gas, produce an electrical pulse at the tube's output. However, this reaction has a high probability of occurring only for slowly moving neutrons with energies $\lesssim$10 eV, whereas fission neutrons are fast moving, with energies on the order of 1 MeV. To combat this disparity, the proportional counter tube is encased in hydrogen-rich high-density polyethylene (HDPE) moderator. Incident neutrons repeatedly scatter in this moderator, losing energy with each collision, until they slow down to low energies, at which point they random-walk into the central tube and readily react with the He-3.

Figure 5 plots probability distributions for the time spreads, T, in the detection delays when a moderated He-3 detector (a) is used versus a fast detector that does not incorporate a moderator (b). This plot is generated from experimental data taken on a bare HEU object, and it shows that the moderator used in the He-3 detector significantly increases the spread in detection times. For the He-3 detector used in this experiment, detection delays extend out to about 150 μs. Therefore, if the spontaneous fission rate of a nuclear assembly were to be

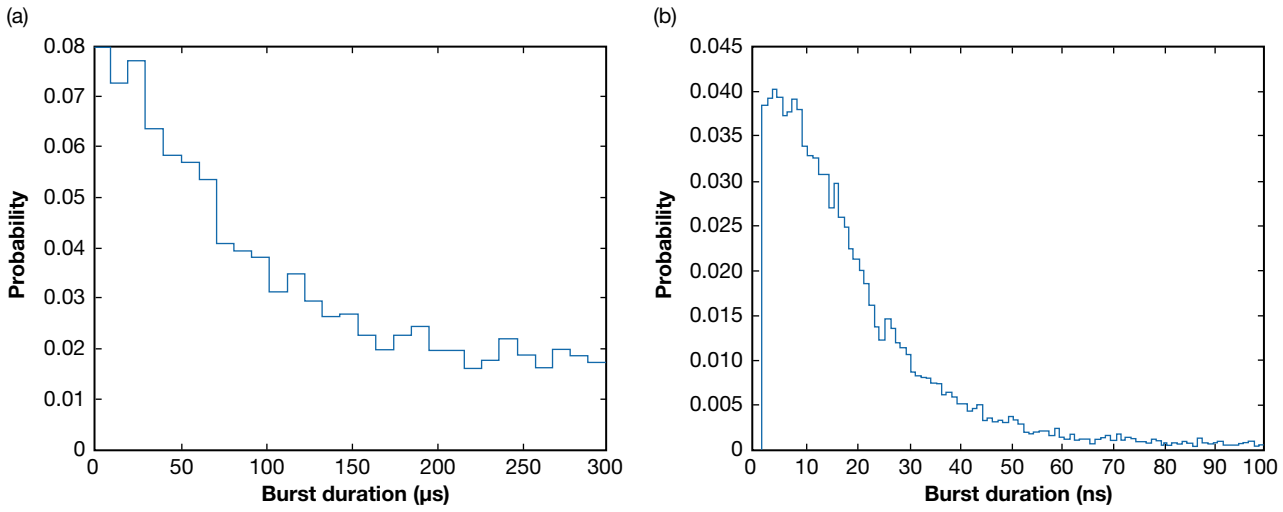

Figure 5. Probability distributions for the time spread, T, in the detection times for neutrons from the same fission chain, measured on the same HEU object using a moderated He-3 detector (a) and a fast neutron detector (b). Note the change in timescale—from microseconds to nanoseconds.

above ~1000 s^{-1} (which is not unrealistic in some cases), then more than 10% of the fission chains would occur at times separated by less than 100 μs, and overlapping fissions chains would be a problem.

For moderated He-3 detectors, a different approach for quantifying neutron time correlations is needed. This second approach dispenses with parsing the record of neutron detection times into fission chains. Instead, the time axis is partitioned into consecutive "gates" (Fig. 4d), and the number of neutron detections in each gate is tallied and histogrammed. The result is an empirical estimate of the probability distribution for getting K neutron detections within a gate Δt seconds wide. An experimentally measured gated-count distribution for a multiplying assembly is plotted in Fig. 6; also plotted is the best fit Poisson distribution. Clearly the probability distribution is non-Poisson.

On the other hand, there are industrial sources that produce neutrons solely from (α,n) reactions, such as americium-beryllium (AmBe) sources. In these sources, one and only one neutron is produced from each source event (Fig. 4a), and so the neutron count distribution will follow the Poisson distribution characteristic of α-decay. This suggests that deviations from Poisson statistics can serve as an alternative signature for fission chains.

Building on the theoretical expression for $Q_m(p,\varepsilon)$, it is possible to derive the probability distribution, $\Pr[K = k] = R_k(p,\varepsilon,\Delta t,S_f)$, for the random number of neutrons detected within a gate Δt seconds wide.[16] This theoretical distribution for K is compared with the empirically measured histogram to determine the source leakage multiplication—just as the empirical histogram for M is compared to the theoretical distribution $Q_m(p,\varepsilon)$ in the burst-count method.

There are a number of parallels between the two methods but also some differences. Like Q_m, R_k depends on p and ε, but it also depends on the gate width, Δt, and the spontaneous fission rate, S_f. As with the first method, the mathematical computations are considerably simplified if we work with the moments of R_k instead of the full distribution. However, because R_k depends on more parameters, more than two moments need to be computed. In the burst-count method, we compared the combinatorial moments of the theoretical and empirically measured distributions. In the gated-count method, it turns out that we instead equate the "correlation" moments of the theoretical and empirical count distributions. These correlation moments are defined recursively in terms of the combinatorial moments, $M_{(j)}$, and the first three are:

$$\begin{aligned} Y_1 &= M_{(1)} = \overline{K} \\ Y_2 &= M_{(2)} - \tfrac{1}{2!}Y_1^{\,2} \\ Y_3 &= M_{(3)} - Y_2 Y_1 - \tfrac{1}{3!}Y_1^{\,3} \end{aligned} \quad . \qquad (9)$$

The use of these arcane statistical moments may seem abstruse, but they actually have a simple physical interpretation.[11,17] Suppose we open a gate and count K neutrons arriving within the gate time. From Eq. 9, we see that $M_{(1)}$ and Y_1 are simply the expected number of neutrons. Now, any particular group of q neutrons selected from the K neutrons detected might have come from the same fission chain and are therefore "correlated." On the other hand, the neutrons in the group might have come from two or more different fission chains and, therefore, have been accidentally grouped. The qth correlation moment, Y_q, is simply the expected number of q-tuples of correlated neutrons in any given gate.

Given this physical interpretation, it is easy to see why combinatorial moments are used in the burst-count method, whereas correlation moments are used in the gated-count method. Because we can parse neutron detections into their respective fission chains in the burst-count method, we know that the resulting count

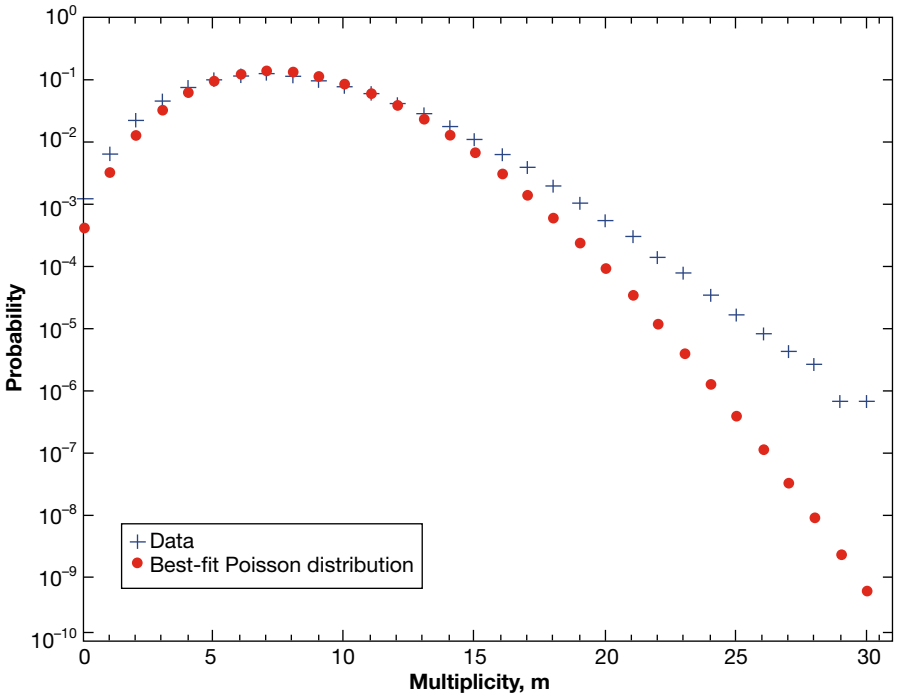

Figure 6. Measured neutron gated-count distribution from an HEU object for a 500-μs wide gates (blue) and the best-fit Poisson distribution (red).

distribution and its combinatorial moments pertain to only correlated neutrons. This is not the case for the gated-count method, and so the combinatorial moments have to be corrected for accidental correlations. The corrected moments are the correlation moments.

One last set of moments is frequently used when analyzing gated-count distributions: the Feynman moments, $Y_{qF} \doteq Y_q / Y_1$, which are essentially normalized correlation moments. These are so named because during the Manhattan Project in World War II, Feynman, Serber, and de Hoffman did the original work on neutron time correlations while studying neutron fluctuations in the so-called "water boiler" at Los Alamos.[18] In their analysis, which is attributed to Feynman, they defined the quantity that we now call Y_{2F}.

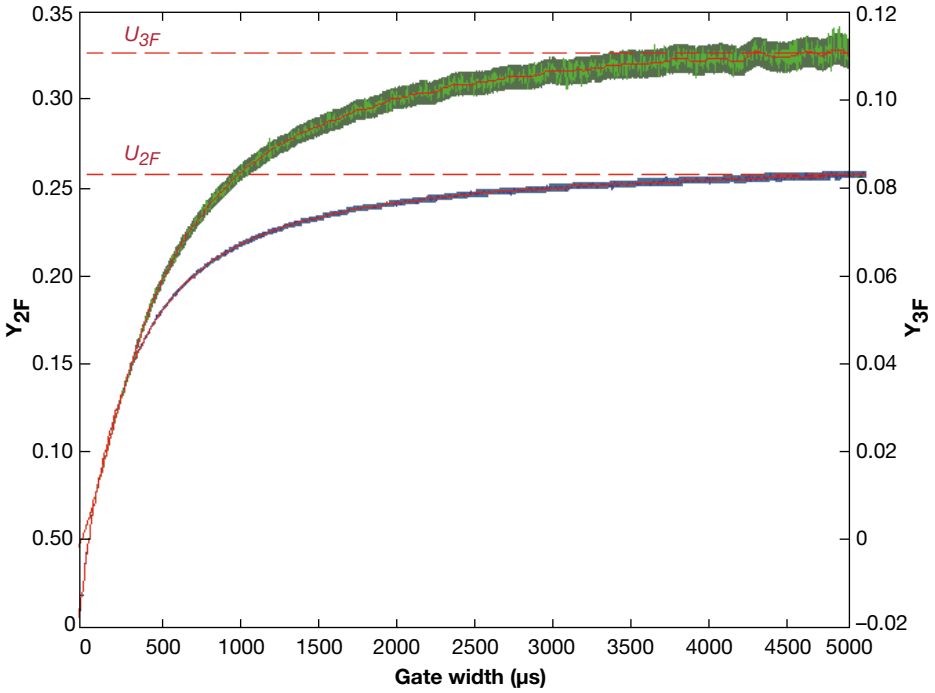

Figure 7. Feynman moments Y_{2F} (blue) and Y_{3F} (green) vs. gate width, measured on an HEU object.

Finally, the reason why the gated-count distribution, $R_k(p,\varepsilon,\Delta t,S_f)$, depends parametrically on the gate width, Δt, can be understood by referring to Fig. 4d. Because of the spreading of neutron detection times, it is possible that a fission chain occurring within one gate will have some of its neutrons detected in subsequent gates. Similarly, it is also possible to count neutrons from fission chains actually occurring in earlier gates. Clearly, the smaller the gate width, the more significant this "leakage" effect will be. This phenomenon can be observed in the data plotted in Fig. 7, which is a graph of the Y_{2F} and Y_{3F} moments versus gate width for measurements made on an HEU object. As the gates become wider, the relative significance of this effect becomes smaller, and the Y_{2F} and Y_{3F} values approach horizontal asymptotes.

Putting It All Together

Ultimately, we wish to find both the leakage multiplication, which will indicate the presence of fission chains, and the total mass of SNM present. We do this by estimating the values of parameters p and S_f from the measured gated-count distribution, $R_k(p,\varepsilon,\Delta t,S_f)$. The estimate for the spontaneous fission rate, S_f, along with the known spontaneous fission half-life and material density will give us an estimate for the SNM mass. Then, as was done in the burst-count method, we use the estimated value for the induced fission probability, p, in Eq. 2 to arrive at an estimate for the leakage multiplication M_L.

To estimate p and S_f, we use the correlation moments of $R_k(p,\varepsilon,\Delta t,S_f)$, which also necessarily depend on p, ε, Δt, and S_f. The dependence on gate width, Δt, can be eliminated by working with the asymptotic values of these moments:

$$U_q(p,\varepsilon,S_f) = \lim_{\Delta t \to \infty} Y_q(p,\varepsilon,\Delta t,S_f). \qquad (10)$$

These asymptotic values are estimated from the empirically measured count distributions and equated with their theoretical expressions. Doing this for the first three correlation moments gives us three equations in three unknowns, which can be solved for p, ε, and S_f.[19]

INSTRUMENTATION

Instruments called multiplicity counters have been used advantageously in materials accountability and international safeguards applications for a couple of decades now,[9] and they are available commercially (Canberra, http://www.canberra.com; and Ortec, http://ortec-online.com). These instruments use moderated He-3 neutron detectors and therefore use the gated-count method for characterizing neutron time correlations described above. Their design is usually carefully optimized in several respects:

- Neutron detection efficiency is maximized, because $Y_q \propto \varepsilon^q$, and so higher-order correlation moments

quickly become immeasurably small at low efficiencies. Most designs aim for 40–60% efficiency, which is achieved by surrounding the sample with neutron detectors. Therefore, these geometries are completely closed, with the sample lying in a cavity at the center of the instrument.

- Variation of neutron detection efficiency versus the precise location of the sample inside the sample cavity is minimized.
- Variation of neutron detection efficiency versus neutron energy is minimized. Fission neutrons are emitted with a continuous spectrum of energies, yet the models described above take into account neither this fact nor any energy dependence in the detection efficiency. However, if the instrument's response is designed to be independent of neutron energy, this oversimplification in the model becomes a nonissue.
- Detector dead time is minimized by using a highly segmented design. The He-3 proportional counter tubes produce electrical pulses that last several microseconds. If a second neutron enters the tube during this time, it will not be detected, and this presents a serious problem. The whole purpose of these instruments is to detect time-correlated neutrons that arrive in bursts; yet it is precisely these closely spaced neutrons that will be missed because of dead time. Multiplicity counters mitigate this problem by using many (15~50) independent channels of neutron detection, spread around the circumference of the instrument in a form of spatial multiplexing. Neutrons may be emitted in short bursts, but they leave the sample traveling in all directions. Thus, the odds of two neutrons striking the same channel within a few microseconds of each other are kept small.
- Immunity to background neutrons from the environment is maximized. The neutron background can be highly correlated and might fool the instrument into thinking that it is detecting higher levels of correlations from the sample. To guard against this problem, the detector can be shielded against external neutrons—a task made possible by placing the sample on the inside.

These instruments are used in industrial settings and can be quite large. For instance they may need to accommodate a drum of plutonium waste. Consequently, they are not man-portable and are not usable by a rapid-response deployment team. Furthermore, if such an instrument were to be used in a field deployment, the object under test would have to be lifted and placed inside the instrument—something a team would not do with a nuclear threat object.

In response, man-portable multiplicity counters have been specially developed. However, the constraints imposed by portability forces trade-offs in some of the design optimizations listed above. The most significant change is that the neutron detectors can no longer surround the sample or object under test. This means that the detection efficiency will be much lower and uncontrolled, except perhaps in terms of energy dependence. Moreover, because of the open sample-detector geometry, it is not possible to shield the instrument against the neutron background.

Ad Hoc Multiplicity Counters

Although deployable, existing man-portable multiplicity counters are single-purpose instruments, and they necessarily add to the limited equipment load-out of a rapid-response team. APL is seeking to minimize this load-out, retaining its core capabilities while adding multiplicity counting for detecting fission chains.

The underlying approach is to concentrate on answering the most pressing question—is the threat object multiplying or not?—and to leave detailed assay to larger follow-on teams. Concentrating on this more limited question opens the possibility of using cruder instrumentation than is needed for traditional multiplicity counting assay.

Rapid-response deployment teams already carry neutron detectors with them, built into radiation backpacks or as discrete sensors, but these neutron detectors are configured as simple counters that measure only the average neutron count rate. Because these neutron detectors are already part of the load-out, the most convenient way to add neutron time-correlation capability is to retrofit the existing deployment kit with electronics that allow operators to gang individual neutron detectors together into a single ad hoc multiplicity counter.

The data from this ad hoc instrument will necessarily be lower fidelity than those obtained from a purpose-built assay instrument. Additionally, an operator in the field will seldom have the luxury of arranging the individual neutron counters optimally, further degrading the quality of the data. Therefore, novel algorithms are also needed to take the necessarily low-fidelity data from this ganged instrumentation and answer the limited yes/no question on neutron multiplication in the threat object.

In an effort to explore the viability of this approach, APL has developed the electronics for integrating arbitrary individual neutron counters into an ad hoc multiplicity counting instrument. In collaboration with the Idaho National Laboratory, APL has also just completed a measurement campaign in which two representative ad hoc multiplicity counters were fielded against a multiplying HEU test object. The purpose of this campaign was to gather data from a wide range of measurement scenarios to systematically map out the performance

envelope of these ad hoc multiplicity counters. The test object[20] was configurable so that the *k*-effective was variable from approximately 0.4 to 0.7. The test object was also sometimes configured with neutron reflectors made of steel, tungsten, and HDPE, which further boosted the *k*-effective up to 0.84. A radioisotope neutrons source, such as Cf-252, could also be inserted into the test object to boost the effective spontaneous fission rate. This allowed us to independently vary the multiplication and the rate at which fission chains were created. Altogether, varying the source configuration in all these ways allowed us to cover a wide range of source physics. In addition, we made measurements over a range of different detector arrangements and detector standoff distances (which affect the all-important neutron detection efficiency, ε).

The data that were collected in this series of measurements provide the foundation for developing binary classifier algorithms for determining whether or not the threat object under measurement is supporting significant fission chains. These algorithms will ascertain, with a stated level of statistical confidence, whether the object is exhibiting neutron multiplication above some exigent threshold, and they must be robust against the marginal data quality expected from an ad hoc instrument operated in less than ideal conditions in the field.

Fast Neutron Detectors

Another class of detectors, called proton recoil scintillators, can detect fast neutrons directly, needing no moderation. Free of the attendant moderation time, these detectors have a fast response. Therefore, these detectors have advantages for measuring neutron time correlations. Provided that the spontaneous fission rate is not too large, it is possible with these detectors to implement the burst-count method for determining neutron multiplication described first.

Even if the gated-count method is used, the fast detector response still has the advantage of needing a shorter measurement time. Because these fast detectors suffer no moderation time, the $Y_{2F}(\Delta t)$ and $Y_{3F}(\Delta t)$ curves reach their asymptotes at much shorter gate widths. The statistical error in estimating Y_{qF} is proportional to $\sqrt{N}$, where N is the number of gate periods for which data are collected. Thus, specifying the required error fixes N. Furthermore, reducing the maximum gate width by using a fast-response detector also reduces the total measurement time, $T = N\Delta t$.

Proton recoil scintillators are organic materials that are rich in hydrogen. When an incident neutron collides with one of these hydrogen nuclei, enough kinetic energy is imparted to it that it is ejected from its parent molecule. This recoil proton then traverses the scintillator, leaving a trail of ionization and molecular excitations in its wake. These excited molecules subsequently decay back to their ground states, emitting photons and producing a light pulse. The light pulses are then collected and converted into an electrical detection pulse by a high gain photodetector, such as a photomultiplier tube.[21]

These scintillators are also sensitive to gamma-rays via a parallel mechanism: incident gamma-rays Compton scatter off electrons in the scintillator and eject them from their parent molecule. Like the recoil protons, these Compton electrons also leave a path of ionization and molecular excitations.

The scintillation pulses produced by recoil protons decay more slowly than the pulses produced by Compton electrons. This difference in pulse length forms the basis of pulse shape discrimination for distinguishing incident neutrons from incident gamma-rays. Unfortunately, pulse shape discrimination is not error free, and one gamma-ray in 10^3 or 10^4 may be misclassified as a neutron. Since the gamma flux is often much higher than the neutron flux, then the number of misclassified gamma-rays can be a substantial fraction of the neutron counts, leading to errors. By comparison, only one gamma-ray in 10^6 or 10^7 causes a false neutron count in moderated He-3 detectors.

Not all scintillators exhibit a significant difference in the pulse lengths for neutrons versus gamma-rays. Unfortunately, scintillators that have good pulse shape discrimination properties typically do not lend themselves to building rugged deployment gear for many reasons. They are either liquids or fragile crystals. Most are flammable, many with low flash points, and they can be carcinogenic. However, recently there has been renewed interest in plastic scintillators capable of pulse shape discrimination.[22,23] Furthermore, a plastic developed by Lawrence Livermore National Laboratory[24] is now available commercially (Eljen Technology, http://www.eljentechnology.com). Plastic has many advantages for making deployable gear. It is not flammable; being plastic, it is intrinsically rugged; and it can be molded into any size or shape.

Therefore, APL is investigating these new plastics for potential use in field deployable systems. However, it remains to be seen whether the benefits of fast response outweigh the downside of gamma misclassifications that come with pulse shape discrimination.

SUMMARY

In this current period of increased nuclear proliferation risks, the United States is implementing a multilayered defense aimed at interdicting a smuggled nuclear device. If a potential threat object is found, it is imperative that the exact level of threat be assessed quickly. Because the presence of fission chains is hard evidence of either an actual nuclear device or a significant quantity of SNM, detecting fission chains is invaluable for this threat assessment.

The technology for detecting fission chains has been in use for several decades for various applications and has roots going back to the Manhattan Project in World War II. However, the standard instrumentation does not lend itself to field deployment by a small, rapid-response team that might be dispatched to assess a threat. APL is exploring several avenues to bring to the field better neutron time-correlation techniques for detecting fission chains.

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