

RADIATION DETECTION FROM FISSION

J. T. Mihalcz

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Nuclear Science and Technology Division

RADIATION DETECTION FROM FISSION

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ABSTRACT

This report briefly describes the neutrons and gamma rays emitted in fission, briefly discusses measurement methods, briefly discusses sources and detectors relevant to detection of shielded HEU in sealand containers, and lists the measurement possibilities for the various sources. The brief descriptions are supplemented by reference.

1. INTRODUCTION

This report briefly describes the neutrons and gamma rays emitted in fission, briefly discusses measurement methods, briefly discusses sources and detectors relevant to detection of shielded HEU in sealand containers, and lists the measurement possibilities for the various sources. The brief descriptions are supplemented by reference.

1.1 NEUTRONS AND GAMMA RAYS FROM FISSION

The fission process produces radiation that can be detected to indicate the presence of fissile material. Active interrogation with either photons or neutrons induces fission in the shielded HEU and a variety of radiation is produced. For the purpose of detection of fissile material, only gamma rays and neutrons are discussed in this report along with their time dependence emission characteristics. Neutrons and gamma rays that are emitted promptly ($<10^{-14}$ sec) are referred to as prompt emissions, while neutrons and gamma rays that are emitted after a fission event are referred to as delayed. For example, there are ~ 2.5 prompt neutrons emitted in ^{235}U fission and 0.016 delayed neutrons. For gamma rays there are ~ 7 prompt, and ~ 7 delayed gamma rays emitted per ^{235}U fission. However, each fission of ^{235}U emits different numbers of prompt neutrons (up to 6) and gamma rays (up to 20). These fluctuation distributions for prompt neutrons and prompt gamma rays are known. The fluctuations in the numbers of prompt emissions result in deviations from the Poisson statistics when measuring the number of particles emitted within a time interval. These deviations can indicate the presence of fissile material since fission produces coincident counts with higher multipliers.¹ The neutrons and gamma rays differ in that further fissions are induced by both delayed and prompt neutrons through fission chain multiplication processes. For metallic HEU, the delayed neutrons have a higher probability for inducing fission than the prompt neutrons since they have lower energy and higher fission cross section. However, they are much less in number (factor of ~ 150). The time dependence of fission chain multiplication processes has been measured by a variety of methods.^{2,3} The number of delayed neutrons and gamma rays is proportional to the number of prompt neutrons. The ratio of delayed neutron emissions to prompt emissions is fixed and does not depend on the multiplication, although the total number does.

1.2 TIME DEPENDENCE OF DELAYED NEUTRONS

A wide variety of measurements of the time dependence of delayed neutron emission have been performed. Data from the original measurements of Keepin, Wimmatt, and Zeigler⁴ are given in the following Table 1, where the time dependence has been represented by 6 delayed neutron groups with their relative yields and exponential decay constants.

Table 1. Delayed neutron data of Keepin, Wimmett, and Zeigler

Group	Relative yield	Decay constant (sec ⁻¹)
1	0.038	0.0127
2	0.213	0.0317
3	0.188	0.115
4	0.407	0.311
5	0.128	1.40
6	0.026	3.87

1.3 TIME DEPENDENCE OF DELAYED GAMMA RAYS

The time dependence of delayed gamma rays with energies above 300 keV has been measured at ORNL by March-Leuba, et al.⁵

The measured delayed emission data have been fitted to a five-group model to represent the decay. This model includes 300-keV energy-discrimination threshold that is accounted for in the overall detector efficiency. The detectors for these measurements were 6 × 6 × 2-in.-thick BiGeO scintillators. The parameters of the five-group model are summarized in Table 4. Delayed gamma data, and a sample measurement is shown in Fig. 1. The parameters in Table 2 correspond to a best fit to the decay gamma data following a fission event, so that

$$n_{\gamma}(\tau) = \sum_{i=1}^5 \alpha_i e^{-\lambda_i \tau},$$

where $n_{\gamma}(\tau)$ represents the average number of photons per second following a fission event, α_i is the group yield constant, which is related to the group precursor fraction, β_i , as $\alpha_i = \lambda_i \times \beta_i$. Figure 1 shows the results of applying the delayed gamma emission model to measured data obtained by irradiating a ²³⁵U fission chamber for 60 and 600 seconds and measuring the decay gammas with the actual flow-monitor hardware. As seen in Fig. 1, the delayed gamma emission model predicts the measured data accurately up to 500 seconds following the fission event. This decay model also benchmarks well against the impulse-response data published in the literature.

Table 2. Delayed gamma data

Group #	α_i (γ/s per fission)	λ_i (s ⁻¹)
1	0.35	0.4
2	0.06	0.04
3	0.015	0.008
4	0.0015	0.0008
5	0.0002	0.00005

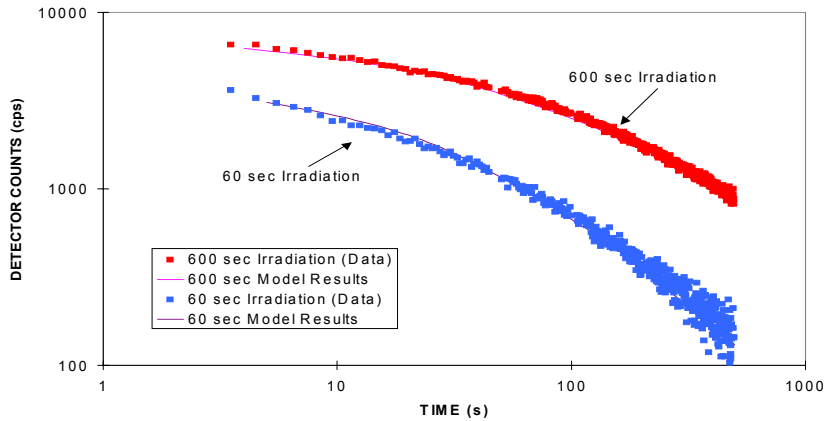


Fig. 1. Comparison between ORNL irradiation measurements and delayed gamma decay model predictions.

1.4 PROMPT NEUTRON EMISSIONS

The distribution of the neutron number of neutrons for ^{235}U fission induced by thermal neutrons is given in Table 3.⁶ In some fissions, there are no neutrons emitted and in some fissions up to 6. This distribution depends on the energy of the neutron inducing fission. More prompt neutrons are produced the higher the neutron energy inducing fission.

Table 3. Distribution of prompt neutron numbers for thermal fission of ^{235}U

Number of neutrons	Fraction of emissions
0	0.0313
1	0.1729
2	0.3336
3	0.3078
4	0.1232
5	0.0275
6	0.0038

1.5 PROMPT GAMMA RAY EMISSION

There is much less measured data for prompt gamma ray emission numbers. Measurements were performed by Brunson⁷ for ^{252}Cf , a spontaneously fissioning isotope, and theoretical estimates of the distribution are given by Valentine.⁸ Typical distributions of gamma ray multiplicities from Ref. 8 for ^{235}U are plotted in Fig. 2 where the highest gamma multiplet is 20 and the average is ~ 7 .

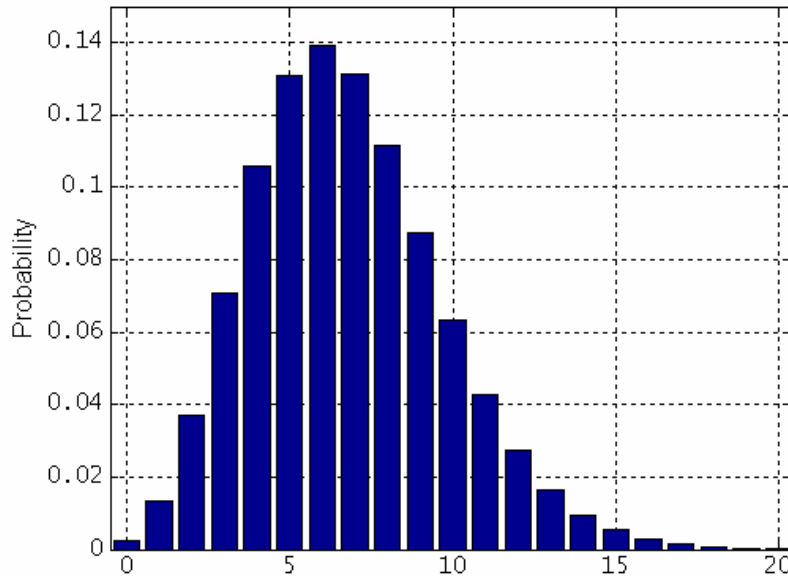


Fig. 2. Thermal neutron induced ^{235}U multiplicity of prompt gamma rays from the negative binomial distribution of Valentine

2. TYPES OF MEASUREMENTS

The measurements can be divided into two types: count rates and coincident count rates. The latter includes coincidences with the interrogating source, like pulsed neutron measurements and correlations within or between detectors, like Rossi alpha type measurements² and multiplet distributions.¹ Prompt measurements are more desirable but sometimes their detection is interfered with by high correlated backgrounds from cargo. Delayed neutrons can induce the fission chain multiplication processes between pulses and may not be subject to this correlated background. Before describing the types of measurements that may be useful for the various interrogation sources considered for this application, some examples of the various types of data are presented.

2.1 COUNT RATES

The count rate is the normal scalar detection rate seen by a detector. To be useful, this scalar count rate must be the count rate above background. The count rate in a detector could change with and without the fissile material present but in this application normal count rate measurements are not practical since there is so much background present that it would be difficult to distinguish the signal produced by the fissile material. However, count rates as a function of time between pulses from an accelerator are related to presence of fissile material and are discussed in Sect. 3.4.3.

2.2 PROMPT MULTIPLET DISTRIBUTIONS AND MULTIPLICITIES

In this type of measurement the number of times n pulses occur in a time interval (multiplets) is measured. Deviation of this distribution from Poisson statistics can be a strong indication of the presence of fissile material.¹ Typical data of this type from a Nuclear Materials Identification System (NMIS) measurement in 1997 with an 18.75 kg HEU casting is shown in Fig. 3. This data was acquired by a NMIS processor³ with 512 one-nanosecond time channels.

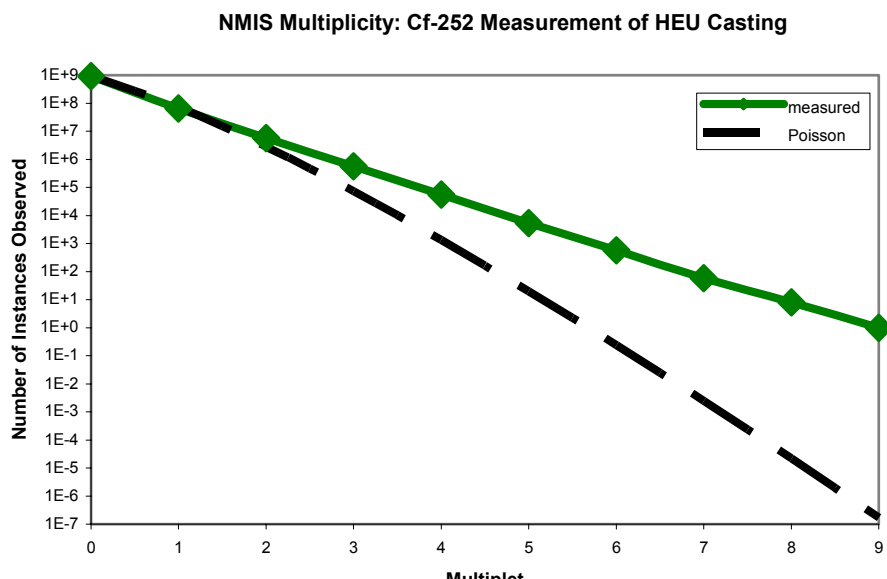


Fig. 3. Number of times n pulses occur in a 512 nsec window from plastic scintillators (from NMIS ~8.5 minute measurement in 1997) for a HEU (93.2 ²³⁵U) 18.75 kg metal casting.

The number of 512 nsec time windows for which the multiplets were acquired was 1.0×10^9 in 8.5 minutes. The 18.75 kg annular HEU storage casting in a stainless steel can had a ^{252}Cf source adjacent to one side and four 3×3 plastic scintillators on the other side. These detectors were sensitive to neutrons above 1.0 MeV and gamma rays above 150 keV. The measured distribution is compared to Poisson statistics and the deviation indicates the presence of fissile material.

In addition, the multiplicities can be obtained from the multiplet distributions. This has been done and is the bases for a widely used nuclear safeguards method using ^3He detectors embedded in polyethylene moderator, i.e., the neutron multiplicity method.⁹ NMIS also obtains multiplicities from the multiplet distributions and for plastic scintillators includes both neutron and gamma rays.¹⁰

2.3 TIME CORRELATION MEASUREMENTS BETWEEN DETECTORS

The initial measurements of this type were suggested by Rossi in the 1940s as a way of obtaining the prompt neutron decay constant without a pulsed neutron source.² Typical data of this type for the standard HEU storage casting obtained from two plastic scintillators described in Sect. 3.2 with a NMIS processor is given in Fig. 4.¹¹ This cross correlation function or time distribution of coincidence counts is composed of three components: (1) gamma-gamma coincidences at time lag 0 because the gamma rays travel at the same speed to the detectors, (2) gamma-neutron coincidences between 5 and 50 nanoseconds and neutron-gamma coincidences between -50 and -5 nanoseconds, and (3) neutron-neutron coincidences spread broadly about time lag zero from -50 to 50 nanoseconds and are much lower in amplitude.

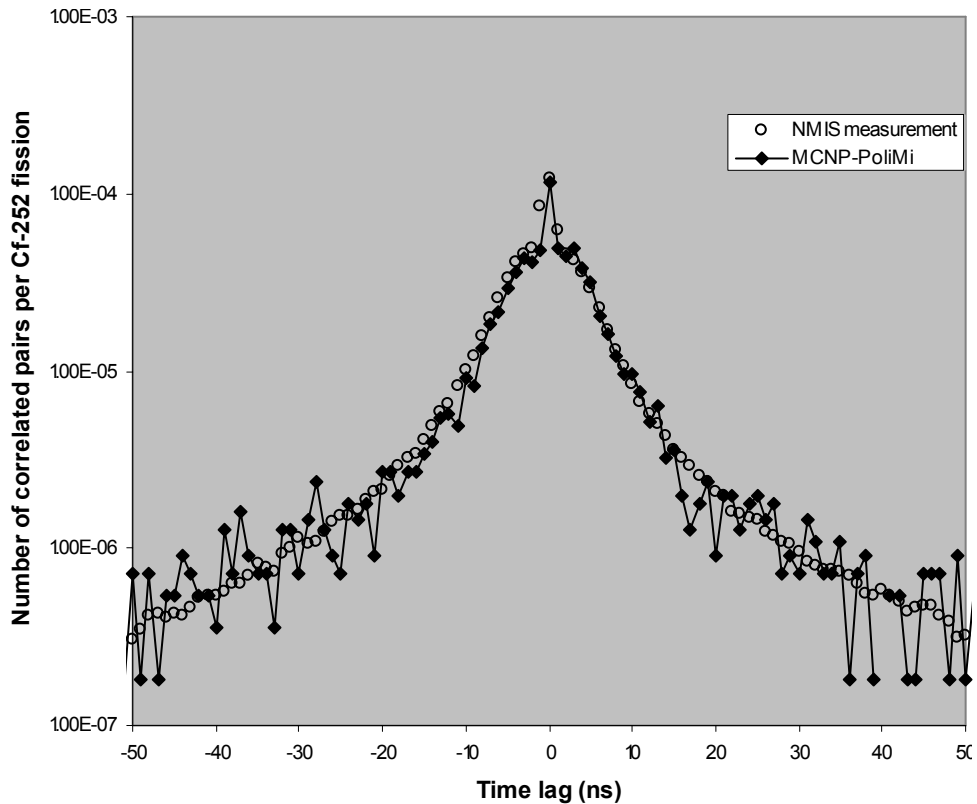


Fig. 4. Time distribution of coincidences between two plastic scintillation detectors for an annular 18.75 kg HEU (93.2 ^{235}U) metal storage casting.

A photograph of a typical arrangement of detectors around a 4 kg sphere of Pu metal with 1.77 wt. % ^{240}Pu is given in Fig. 4 where one of the detectors has been removed for the photograph. The time distribution of coincidences between two 6x6x4-in-thick detectors on opposite sides is given in Fig. 5 where Monte Carlo calculations are compared to the measurements^[6].



Fig. 5 Photograph of the plastic scintillator arrangement and a 4 kg Pu Metal (1.77% ^{240}Pu) spherical shell with one detector removed.

2.4 PULSED SOURCE MEASUREMENTS

In this type of measurement an interrogating source (neutron or high energy gamma rays) is pulsed and the time dependence of the detector response between pulses is measured in a variety of ways and for a variety of sources. The prompt or delayed time dependence of radiation from induced fission is measured between pulses after source initiation.

2.4.1 PROMPT TIME DEPENDENCE

The neutron or gamma count rate (or some combination) is measured after source initiation between pulses. Data from a pulsed neutron measurement with a Cockroft-Walton DT neutron generator for an HEU (93 wt % ^{235}U) metal assembly (11-in.-OD, 2.5-in.-high) measured with a plastic scintillation detector is given in Fig. 7.¹² The generator produced 30 nsec pulses at 0.9 MHz repetition rates and the prompt time response was measured between pulses. The detector was a thin plastic scintillator sensitive to neutron and gamma rays and the zero of time shifted in the electronics by delay of the signal with respect to the pulses to measure the background before the pulse and the rising portion of the pulse.

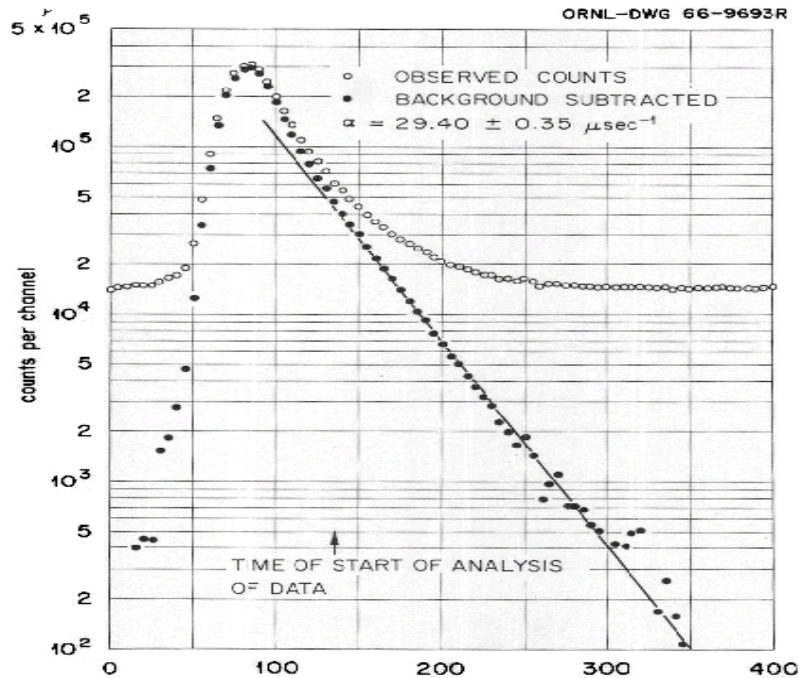


Fig. 7. Time distribution of count after 30 nsec accelerator pulses for an 11-in.-diam, 2.5-in.-high HEU cylinder.

Another way to perform the measurement is with a randomly pulsed neutron source such as time tagged Cf,¹³ which spontaneously fissions, or with a DT generator with an alpha detector. Typical data of this type with a time tagged Cf source for the HEU casting described above in the multiplet section but obtained with one of the 3 x 3 x 4-in.-thick plastic scintillator is given in Fig. 8 where it is also compared to Monte Carlo calculations.¹¹ The time distribution consists of several components: (1) direct gamma rays which pass through the casting and occur at the flight time of the gamma rays, (2) transmitted high energy neutrons at about 5 nanoseconds, (3) scattered neutrons, and (4) neutrons and gamma rays from induced fission. The initial peak at

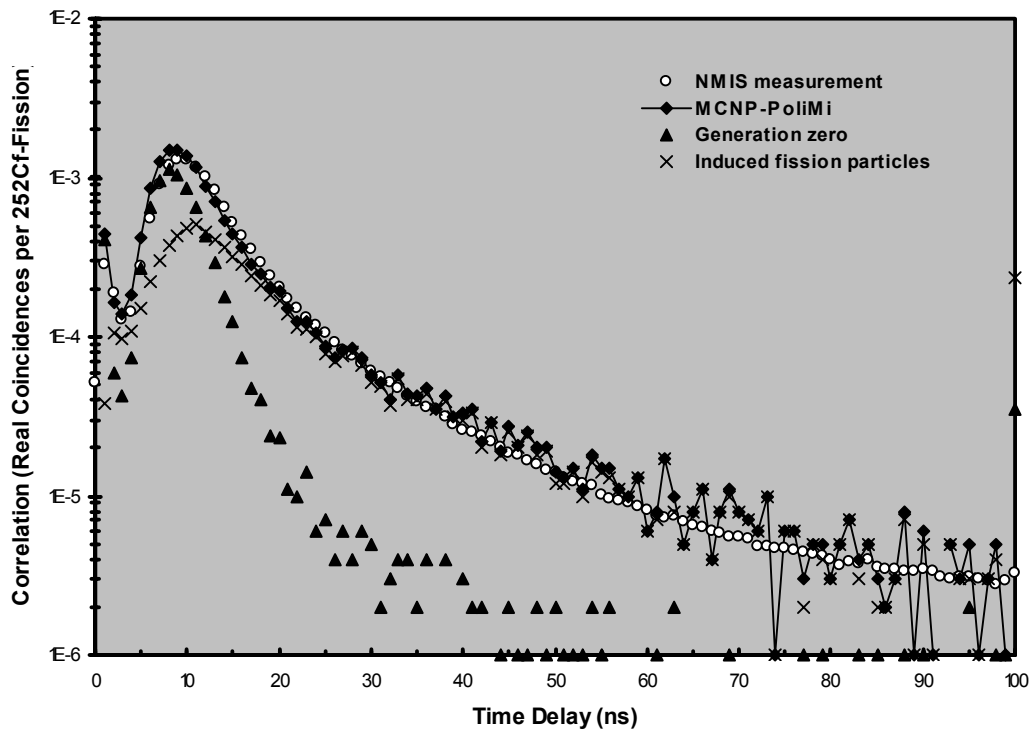


Fig. 8. Time distribution of counts in a plastic scintillator after Cf fission for an annular 18.75 kg HEU metal storage

~1 nanoseconds is from transmitted gamma rays from the source followed by scattered neutrons and then fission neutrons.

2.5 DIFFERENTIAL DIEAWAY

In these measurements¹⁴ the time dependence of the neutron count rate immediately after the accelerator pulse has been measured with ³He counters embedded in moderators. Typical data from a package monitor at LANL is shown in Fig. 9 for various material. In this measurement, the decay of the moderated neutrons is measured after the accelerator pulse of 14 MeV neutrons from a DT generator. In this case the accelerator source was repetitively pulsed. Say for a few seconds and turn it off and measure the decay of the neutron population. Another way to perform this measurement would be operate the source in a steady state mode.

2.5.1 DELAYED NEUTRON COUNT RATE BETWEEN PULSES

In LINAC measurements, the count rate between pulses with ³He counters embedded in moderators has been measured and a typical result for these measurements is given in Fig. 8.¹⁵ The count rate between pulses is from: (1) prompt neutrons generated from induced photo fissions in the nuclear material, (2) delayed neutrons from induced photo fission by the source that leak to the detectors, and (3) prompt neutrons from fissions induced by the delayed neutrons that do not leak from the system. The ratio of the two components to the signal depends on the fissile mass since the ratio of leakage to fission depends on mass. This type of measurement could be performed for any of the pulsed sources of interrogating radiation.

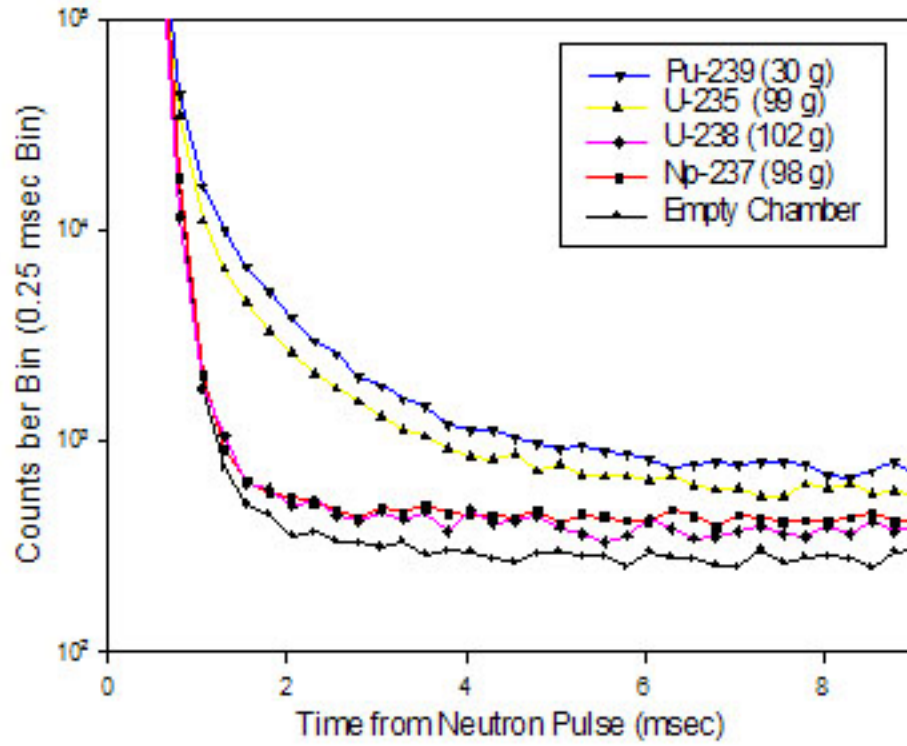


Fig. 9. Typical differential dieaway data from a package monitor at LANL (courtesy of C. Moss).casting

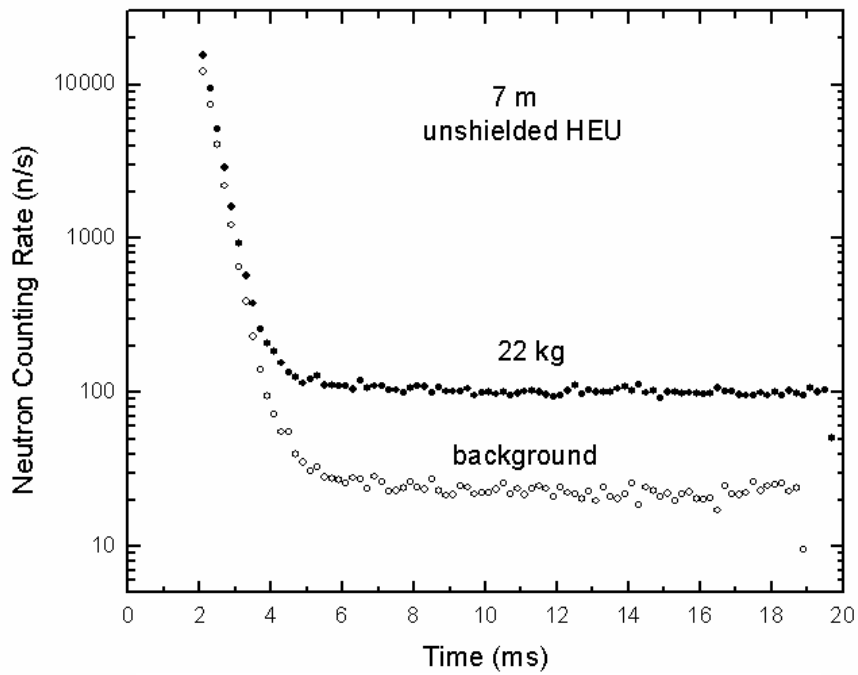


Fig. 10 Response with Linac and large-area detector.

2.6 DELAYED GAMMA ENERGY SPECTRA

The gamma ray energy spectra can be measured with commercial HPGe measurement systems with high resolution and can be measured with commercial NaI based systems for lower resolution measurements.¹⁶

3. DETECTORS

Detectors for this application are sensitive to fast neutron, gamma rays, and thermal neutrons. For this application the detectors have to be high sensitivity, large area systems, probably at least 8×8 feet on at least 2 sides of the sealand container.

3.1 THERMAL NEUTRON DETECTORS

Thermal neutron detectors have usually been ^3He proportional counters embedded in polyethylene with the moderator configured for the particular measurement. A photograph of a large 4×8 panel of ^3He detectors used at LANL is given in Fig. 11.¹⁷ A smaller $51 \times 43 \times 10$ cm more portable system of several ^3He counters in moderation is shown in Fig. 12. A photograph of a single ^3He counter used at INEL in photofission measurements with a LINAC is given in Fig. 13.¹⁸ Other types of thermal neutron detectors could contain boron or ^6Li . A photograph of large panels of ^6Li containing glass fibers at PNNL sensitive to thermal neutrons is given in Fig. 14.¹⁹



Fig. 11 Large-area (4×8 ft), ^3He neutron detector panel.



**Fig. 12. Portable ^3He detector in moderator
(51 × 43 × 10 cm dimensions).**



**Fig. 13. INL's ^3He proportional counter for pulsed LINAC
photo fission measurements.**

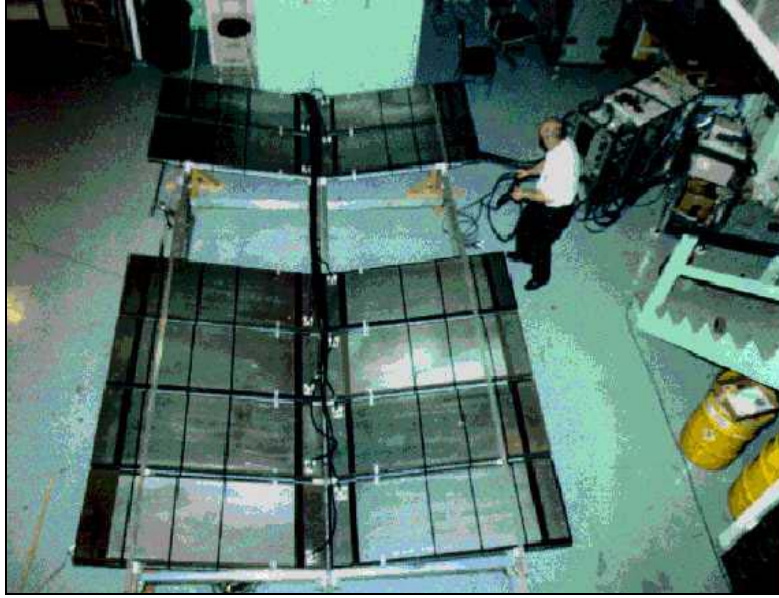


Fig. 14. Large panels of ⁶Li glass fiber thermal neutron detectors.

3.2 PLASTIC SCINTILLATORS

Fast plastic scintillators, sensitive to fast neutrons and gamma rays without distinction, are available with nanosecond time resolution with efficiency for 2 MeV neutron detection of 60% per hit on the front face of an 8-cm-thick detector and about 40% for unshielded fission gamma rays. Large plastic scintillator arrays could be made up of 1 × 1 meter plastic sheets with two photomultipliers per module. A photograph of a 1 × 1 meter plastic scintillator (8.0-cm-thick) covered with Lexan with two photomultiplier tubes centered on opposite thin surfaces is given in Fig. 13.²⁰

In the photograph, a ²²Na gamma ray source time tagged by a smaller plastic scintillator above it is shown near the center of the 1 × 1 meter detector during testing at ORNL. This 1 × 1 meter detector with coincident counting between the 2 photomultiplier tubes has ~2 nsec time resolution and satisfactory uniform response across the surface.

A combined detector which contains a 1 × 1 meter array of ⁶Li Glass fibers adjacent to a 1 × 1 meter plastic scintillator is under fabrication at the NUCSAFE Corporation and would be sensitive to fast neutrons, gamma rays and thermal neutrons.

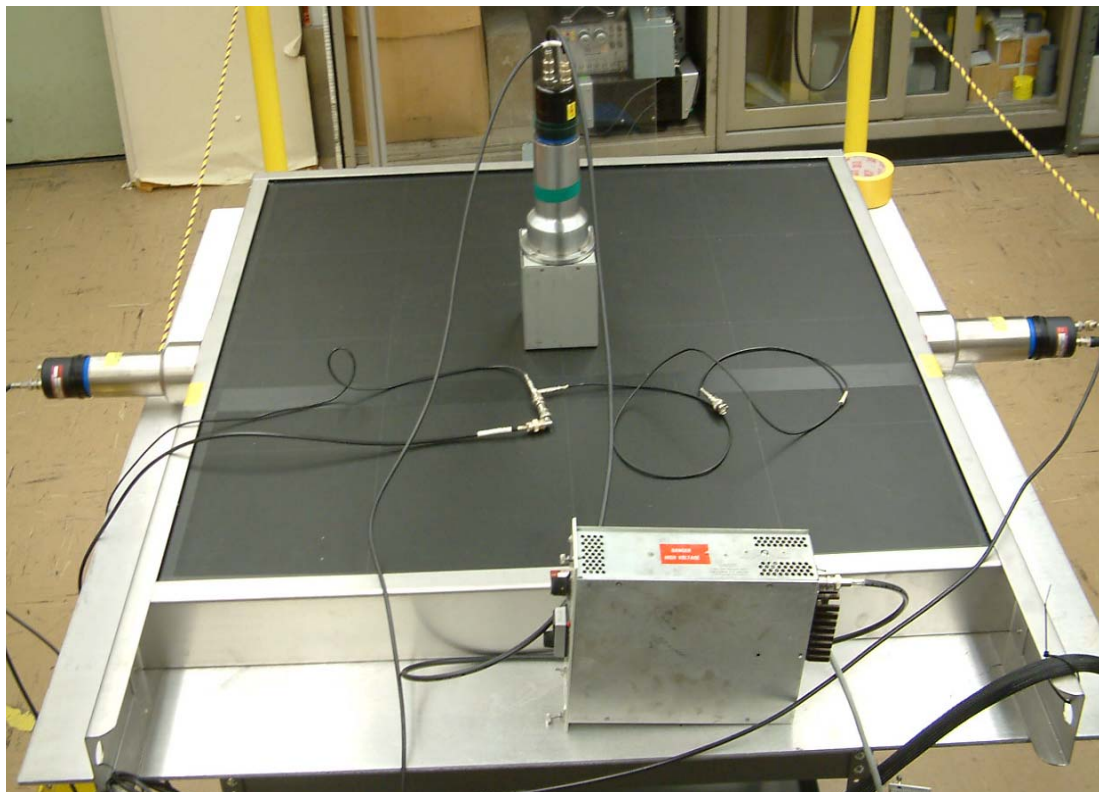


Fig. 15. Photograph of a 1 × 1 meter plastic scintillator in testing at ORNL.

3.3 LIQUID SCINTILLATORS

Liquid scintillators are another type of fast organic scintillator that are sensitive to fast neutrons and gamma rays with the ability to distinguish between fast neutrons and gamma rays and provide more information. A collection of $5.1 \times 5.1 \times 200$ cm liquid scintillators has been assembled into a 2×2 meter array for physics measurements by Michigan State University for use at Fermi Laboratory.²¹ However, they have the disadvantage in that the liquid can leak and may require a large number of photomultiplier tubes to maintain the pulse shape discrimination capability.

4. DATA PROCESSING

There are a variety of ways to acquire data: count rate, count rate as a function of time after interrogation source pulsing, and coincident count rate between multiple detectors during or between source pulses. Various commercial multichannel time analyzers can record the time distribution of counts after source initiation of fission chains. Standard microsecond resolution processing is available for multiplet measurements or multiplicity measurements with ^3He counters embedded in polyethylene. NMIS processing can obtain correlations between detectors, between a detector and the source, and the multiplet distributions for measurements on the time scale of the fission chain multiplication processes (1 nanosecond time channels), this processor for 10 channels of data requires 4 slots in a PC and the appropriate software. For 5 channels of operation the NMIS processor is capable of on line processing at rates of over 2×10^6 events in each channel without loss of any information and can calculate on-line all 15 cross correlations

and multiplets. The NMIS processor is deeply buffered to handle large fission chains. Short time scale measurements have the advantage of lower accidental coincidences and fission chain shape information is obtained. Measurements are routinely made with this processor for signal to noise ratios of $1 \times 10E^{-3}$.

5. SUMMARY MATRIX

The various interrogating sources and the methods of detection are summarized in the following table. Potential measurements are presented. For high time resolution measurements with organic scintillators, sensitive to neutrons above 0.5 MeV, the scintillators would not be sensitive to the 60 keV source neutrons and measurements could be performed when the source is on. Of course, for hydrogenous cargo there may be appreciable background. For other pulsed sources the scintillation detector would have to be gated off during the source interrogation pulse. Gamma-gamma, gamma-neutron, and neutron-neutron coincidences could be obtained for all sources, either between or in some cases during the active interrogation by the source. For some highly moderated cargo, only gamma rays from induced fission reach the detectors and produce all of the coincidences.

Table 4. Matrix of interrogating sources and potential measurements

Interrogation source	Prompt time correlations			Delayed time correlations			Count rate	Gamma ray spectrometry
	Multiplets	With source	Detector detector	Multiplets	With source	Detector detector		
1. Low energy stimulated multiplication (60 KeVn)	DP ^a	BP	DP	BP	BP	BP	BP	—
2. High energy stimulated multiplication (14.1 MeVn)	SS ^b	SS ^b	SS ^b	BP	BP	BP	BP	—
3. PFNA (14.1 MeVn)	—	—	—	BP	AP ^c	BP	BP	—
4. High energy monoenergetic photofission (4–14 MeVp)	—	—	—	BP	BP	BP	BP	HPGe
5. Neutron induced fission from thermalized 2.5 MeVn	DP	—	DP	BP	AP	BP	BP	—
6. Monoenergetic Photon induced fission (6–7 MeVp)	SS	—	SS	BP	AP	BP	BP	HPGe
7. Thermal neutron induced fission (14.1 MeVn)	—	—	—	BP	—	BP	BP	—
8. Thermal neutron induced fission (5–8 MeVn)	—	—	—	—	—	—	—	>3 MeV
9. Photon (bremstralung) induced fission, delayed neutron detection (7–9 MeVp)	—	AP	—	BP	AP	BP	BP	—
10. Pulsed photonuclear assessment (PPA) bremstrahlung (10–20 MeVp)	—	AP	—	BP	AP	BP	BP	HPGe

^aBP means between pulses, DP means during source interrogation, and AP means immediately after pulsing.

^bDT generator operated steady state with alpha detector to define a cone of neutrons.

^cDifferential decay.

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