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RADIATION DETECTION FOR ACTIVE INTERROGATION OF HEU

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J. T. Mihalczo

ABSTRACT

This report briefly describes the neutrons and gamma rays emitted by active interrogation of HEU, 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. All but one of the measurement methods detect radiation emitted by induced fission in the HEU; the exception utilizes nuclear resonance fluorescence. The brief descriptions are supplemented by references. This report presents some active interrogation possibilities but the status of understanding is not advanced enough to select particular methods. Additional research is needed to evaluate these possibilities.

RADIATION DETECTION FOR ACTIVE INTERROGATION OF HEU

J. T. Mihalczo

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 highly enriched uranium (HEU) in Sealand containers, and lists the measurement possibilities for the various sources. All but one of the measurement methods detect radiation emitted by induced fission in the HEU; the exception utilizes nuclear resonance fluorescence. It is not meant to be a comprehensive description with very many details. The brief descriptions are supplemented by references. This report presents some active interrogation possibilities, but the status of understanding is such that particular method can not be selected without further research.

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 dependent 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 on average on average ~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 multiplets.[1] 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, Wimmett, and Zeigler[4] 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 a 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

$$n_\gamma(\tau) = \sum_1^5 \alpha_i e^{-\lambda_i \tau} \quad ,$$

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 ^{235}U fission chamber for 60 and 600 seconds and measuring the decay gammas. 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

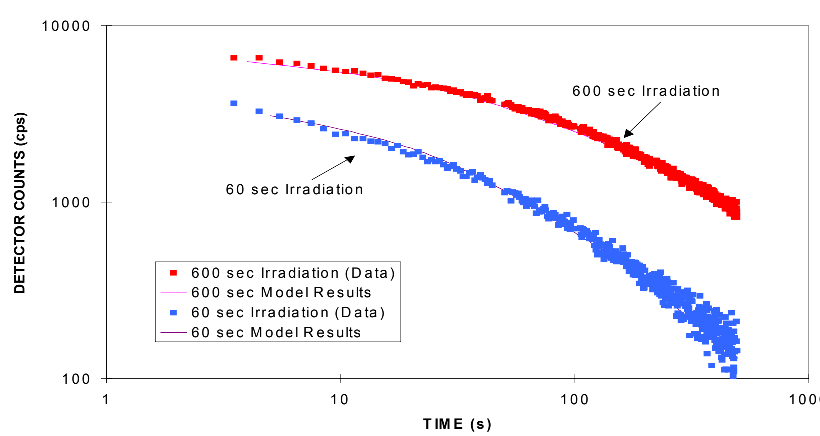

Fig. 1. Comparison between ORNL irradiation measurements and delayed gamma decay model predictions.

1.4 PROMPT NEUTRON EMISSIONS

The distribution of the 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 Brunson[7] 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.

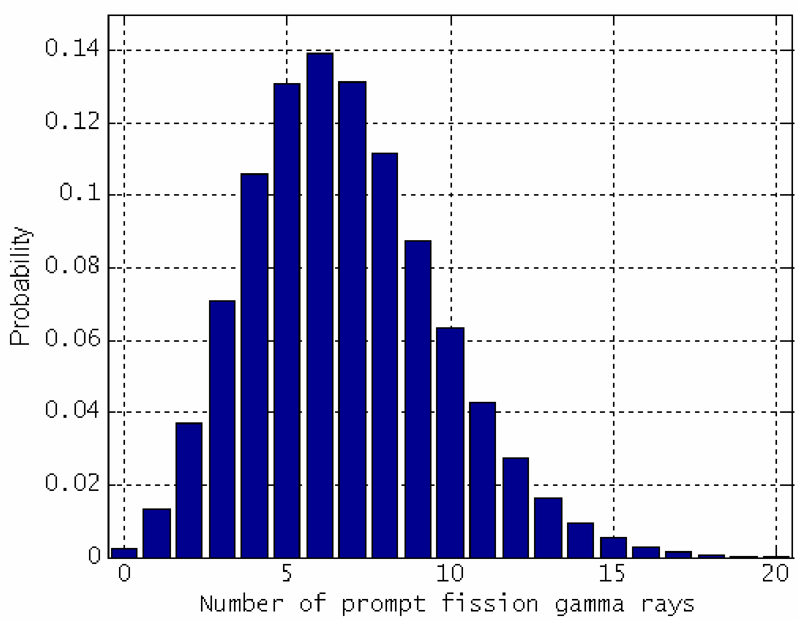

Fig. 2. Thermal neutron induced ^{235}U multiplicity of prompt gamma rays from the negative binomial distribution of Valentine

2. TYPES OF MEASUREMENTS

The measurements can be divided into detection of neutrons and gamma rays. Gamma ray detectors are well known. Gamma ray detection methods can utilize NaI for low energy resolution and HPGe for high resolution gamma ray spectrometry with commercial multichannel analyzers. The number of delayed gamma rays as a function of time could be measured after the sources are turned off. The measurements can be further 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 measurements[2] 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. The useful scalar count rate is 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 processor[3] with 512 one-nanosecond time channels.

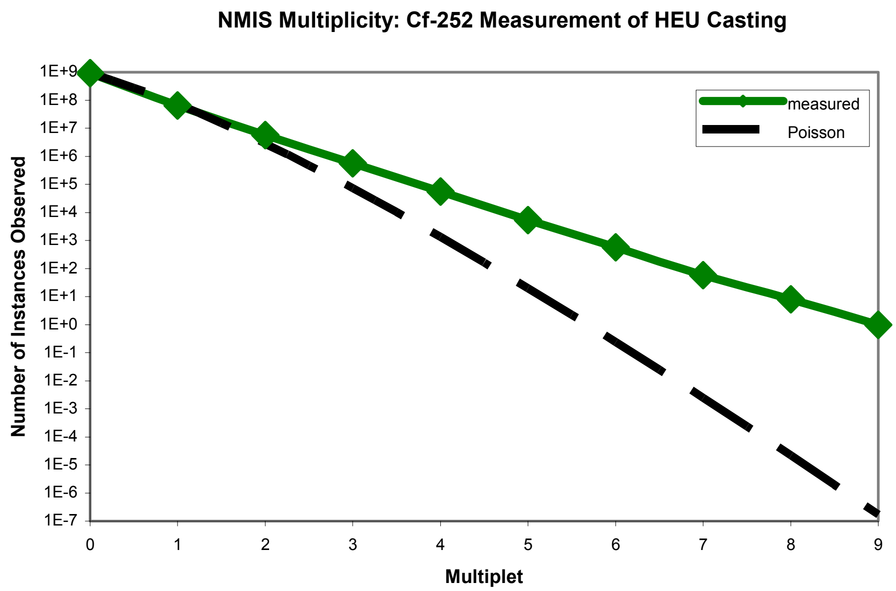

Fig. 3. Number of times n pulses occur in a 512 nsec window from plastic scintillators (from NMIS ~8.5 minute measurement in 1997) for a HEU (93.2 ^{235}U) 18.75 kg metal casting.

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 basis for a widely used nuclear safeguards method using ^{3}He 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 of the same type as described in Sect. 3.2 (but smaller) with a NMIS processor is given in Fig. 4.[11] In this measurement a Cf source was adjacent to the casting and the detector were on the opposite side. 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.

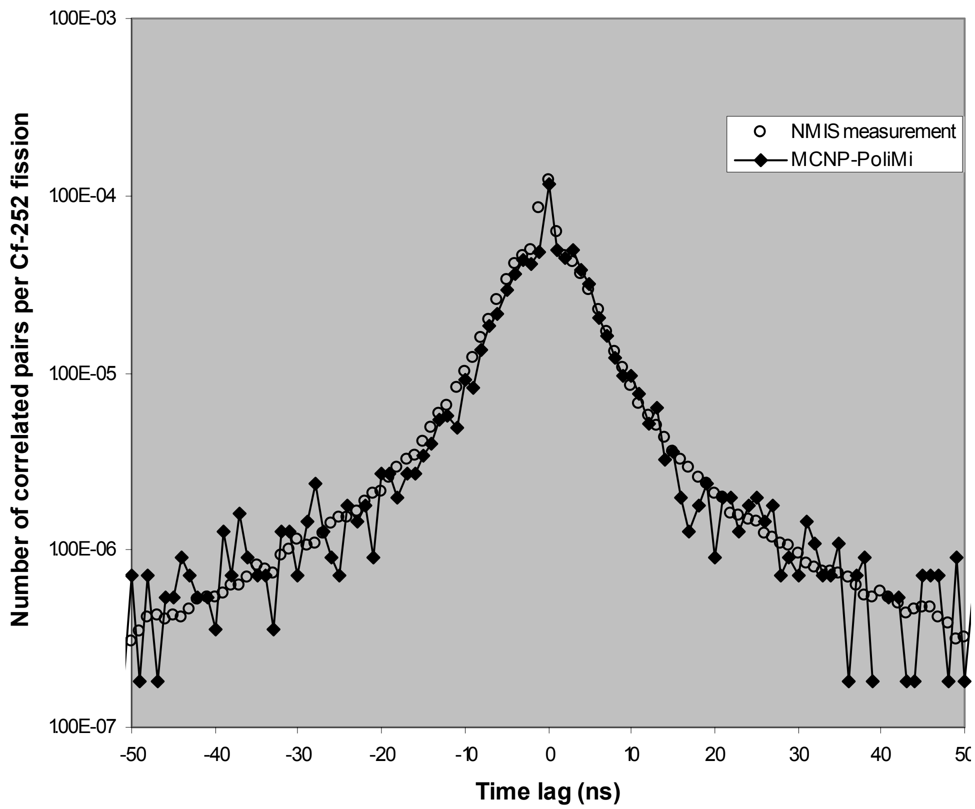

Fig. 4. Time distribution of coincidences between two plastic scintillation detectors for an annular 18.75 kg HEU (93.2 ^{235}U) metal storage casting.

This type of passive correlation measurement can also be performed with fissile material with inherent sources such as plutonium. A photograph of a typical arrangement of detectors around a 4 kg sphere of Pu metal with 1.77 wt. % 240Pu is given in Fig. 5 where one of the detectors has been removed for the photograph (courtesy of Dr. V. Dubinin of the All Russian Institute of Experimental Physics, VNIIEF). The time distribution of coincidences between two 6x6x4-in-thick plastic scintillation detectors on opposite sides is given in Fig. 6 where Monte Carlo calculations are compared to the measurements [6]. This data is included as typical for cases where delayed neutrons from previous fissions in HEU can act as an inherent source.

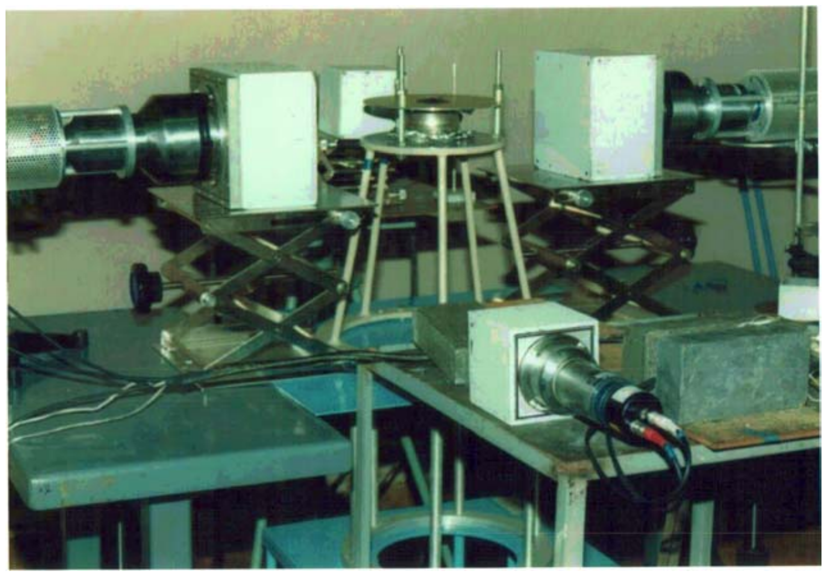

Fig. 5 Photograph of the plastic scintillator arrangement and a 4 kg Pu Metal (1.77% 240Pu) spherical shell with one detector removed.

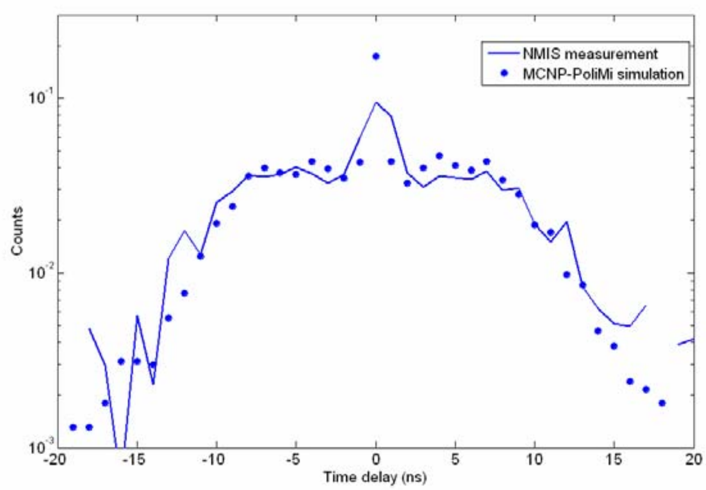

Fig. 6 Time distribution of coincidences between two plastic scintillation detectors for a 3.3 Kg Pu Metal (1.77 wt of ^{240}Pu) spherical shell

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) 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.

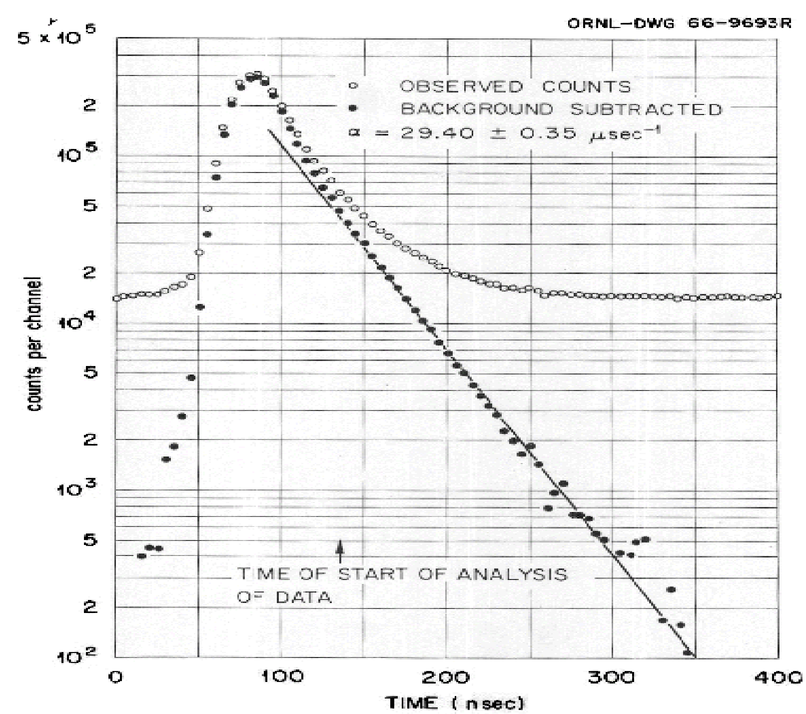

Fig. 7. Time distribution of count after 30 nsec accelerator pulses for an 11-in.-diam, 2.5-in.-high HEU cylinder.

Another way to perform the measurement of the time distribution of counts after fission 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

3×3×4-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 ~1 nanoseconds is from transmitted gamma rays from the source followed by scattered neutrons and then fission neutrons.

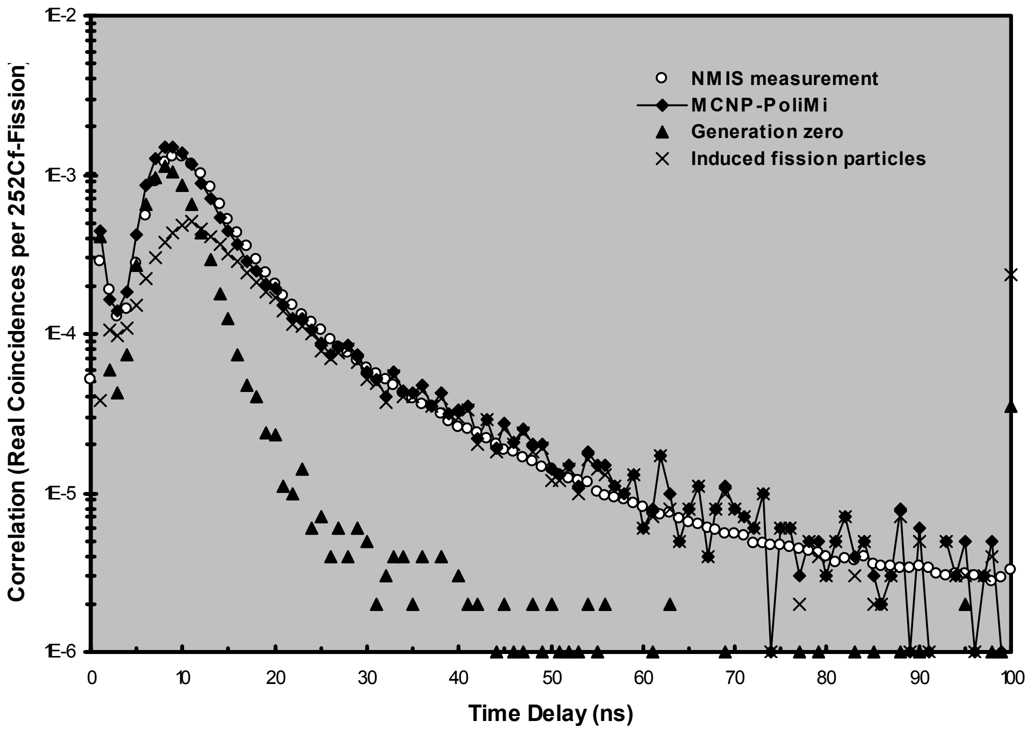

Fig. 8. Time distribution of counts in a plastic scintillator after Cf fission for an annular 18.75 kg HEU metal storage

2.5 DIFFERENTIAL DIEAWAY

In these measurements[14] the time dependence of the neutron count rate immediately after the accelerator pulse has been measured with ^{3}He counters embedded in moderators. Typical data from a package monitor at LANL is shown in Fig. 9 for various materials. 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. Another way to perform this measurement would be operate the source in a steady state mode, say for a few seconds and turn it off and measure the decay of the neutron population.

2.5.1 DELAYED NEUTRON COUNT RATE BETWEEN PULSES

In LINAC measurements, the count rate between pulses with ^{3}He counters embedded in moderators has been measured and a typical result for these measurements is given in Fig. 10.[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. The two components are equal for a 5 kg sample of HEU (~03 wt% ^{235}U). This type of measurement could be performed for any of the pulsed sources of interrogating radiation.

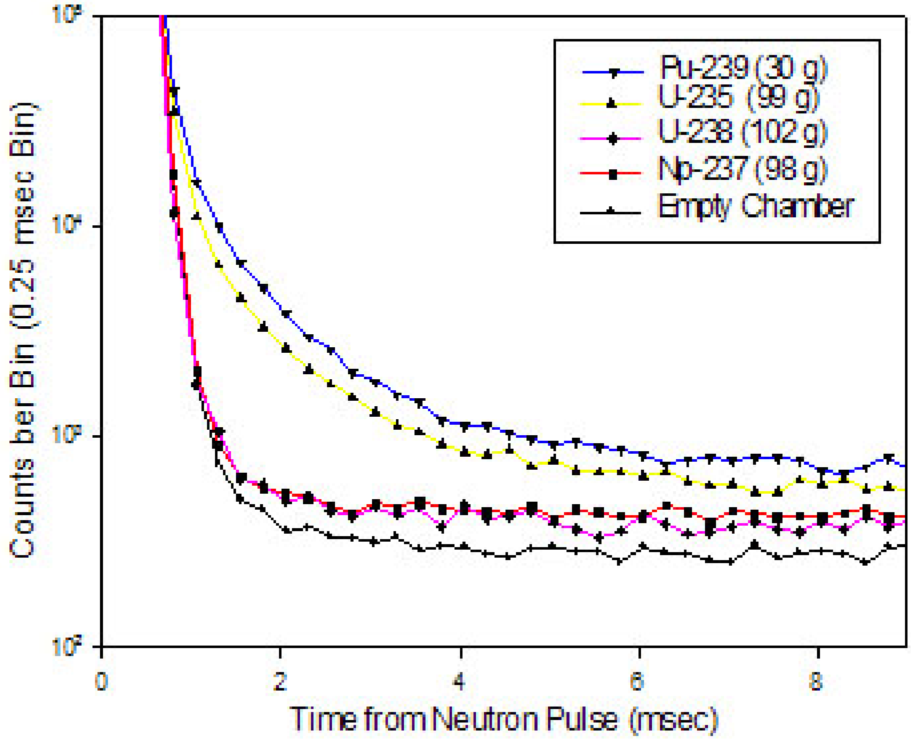

Fig. 9. Typical differential dieaway data from a package monitor at LANL (courtesy of C. Moss).casting

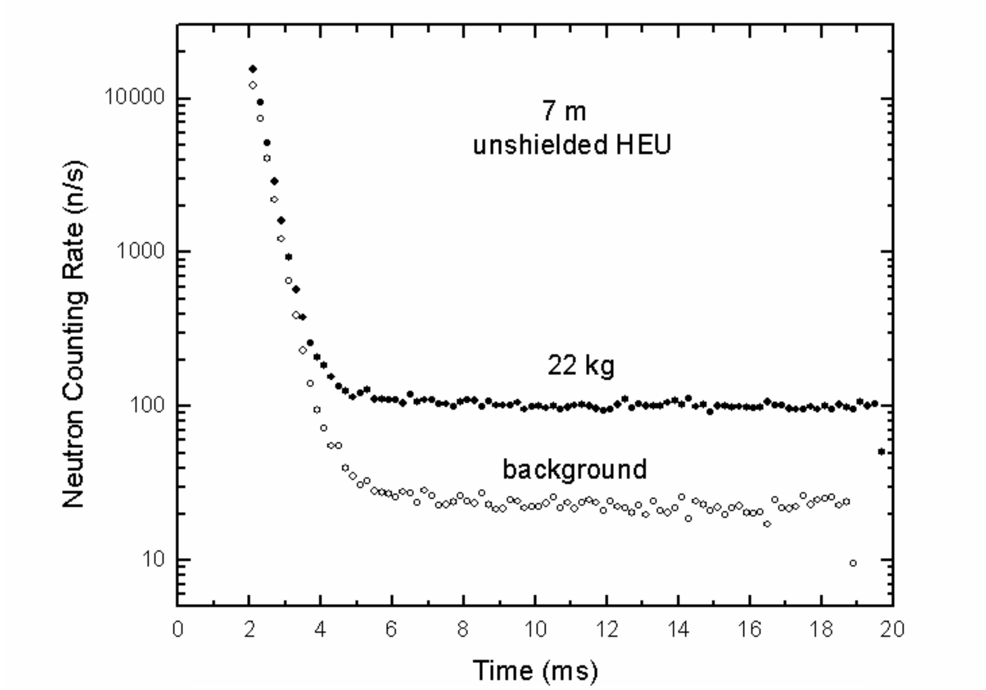

Fig. 10 Response with Linac and large-area detector.

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 ^{3}He proportional counters embedded in polyethylene with the moderator configured for the particular measurement. A photograph of a large 4×8 panel of ^{3}He detectors used at LANL is given in Fig. 11.[17] A smaller 51×43×10cm more portable system of several ^{3}He counters in moderation is shown in Fig. 12. A photograph of a single ^{3}He 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 ^{6}Li. A photograph of large panels of ^{6}Li containing glass fibers at PNNL sensitive to thermal neutrons is given in Fig. 14.[19]

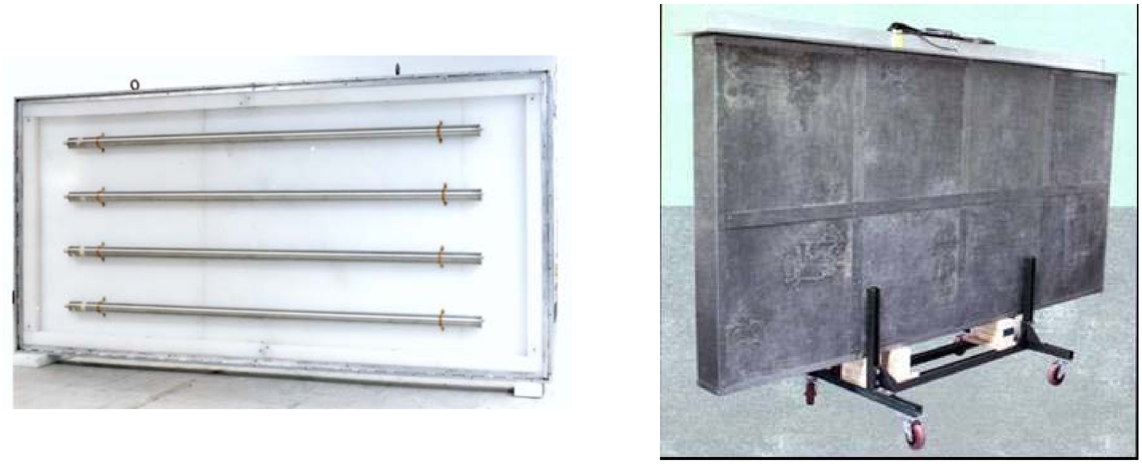

Fig. 11   Large-area (4 × 8 ft), ^{3}He neutron detector panel.

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

Fig. 13. INEL's ^{3}He proportional counter for pulsed LINAC photo fission measurements.

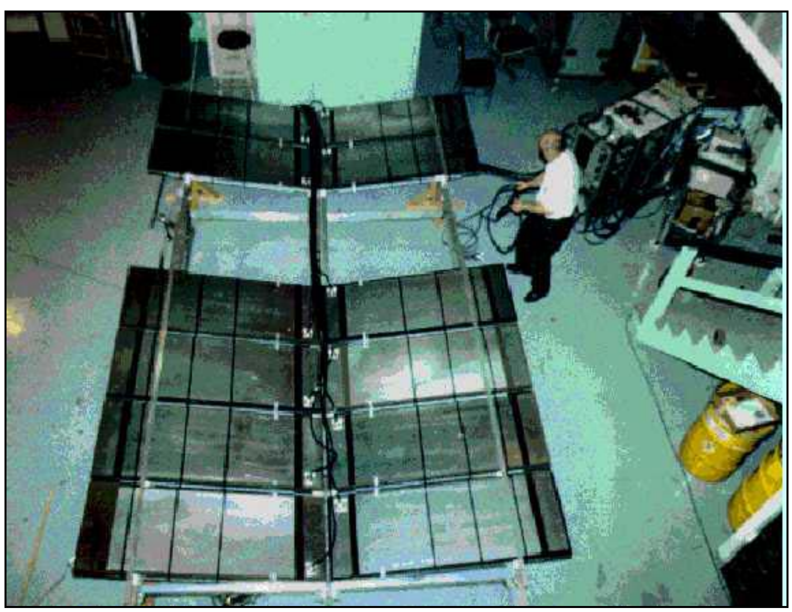

Fig. 14. Large panels of ^{6}Li glass fiber thermal neutron detectors.

3.2 PLASTIC SCINTILLATORS

Fast plastic scintillators, sensitive to fast neutrons and gamma rays without distinction, are available with nanosecond time resolution. They have 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 if a 1×1 meter plastic scintillator (8.0-cm-thick) covered with Lexan with two scintillation detectors centered on opposite thin surfaces is given in Fig. 15.[20]

In the photograph, a ^{22}Na 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 scintillation detectors tubes has ~2 nsec time resolution and satisfactory uniform response across the surface.

A combined detector which contains a 1×1 meter array of ^{6}Li 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.

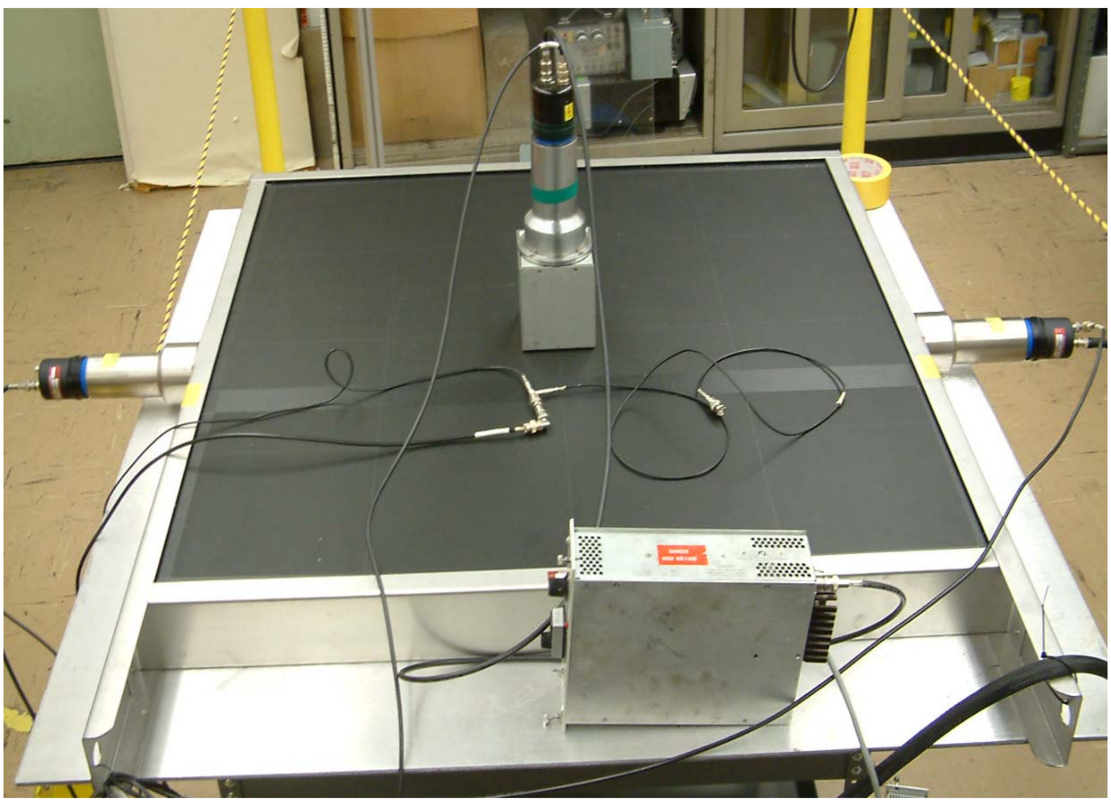

Fig. 15. Photograph of a 1 × 1 meter plastic scintillator in testing at ORNL.

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 5.1×5.1×200 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.

3.4 DETECTION FOR GAMMA RAY SPECTROMETRY

Gamma ray detectors are well known and gamma ray detection methods can utilize NaI for low energy resolution and HPGe for high resolution gamma ray spectrometry with commercial multichannel analyzers.

4. INTERROGATION SOURCES

Various interrogation sources are being considered for application to detection of shielded HEU in sealand containers. This section briefly describes he interrogation sources that may be useful for this application The sources are:

1) Proton accelerator using the H(p,n)7Li reaction to produce 60 keV neutrons in the forward direction[22].
2) DT generator producing 14.1 MeV neutrons isotropically[24], detecting prompt neutrons after the accelerator pulse is turned off or time correlation measurement with the source, and detection of multiplets and correlation between detectors between pulses.
3) DT generator producing 14.1 MeV neutrons isotropically[25,26] , differential dieaway between accelerator pulses (PFNA).
4) Photofission from 4-14 MeV discrete gamma rays from a 1-2 MeV proton accelerator[27]
5) Neutron inducd fission from thermalized 2.5 MeV neutrons from a DD generator
6) Photofission by 6-7 MeV gamma rays from a 3-4 MeV proton accelerator utilizing the $^{19}F(p\alpha\gamma)^{16}O$ reaction[28]
7) Thermal neutron induced fission from 14.1 MeV DT neutron generator[29,30] detection of delayed gamma rays above 3 MeV [31].
8) MeV Photon (bremstralung from a LINAC) induced fission with delayed neutron detection [15, 32]
9) Pulsed photonuclear assessment with a 10-20 Mev photon (bremstralung from a LINAC) induced fission [33, 34] with delayed neutron and delayed gamma detector.
10) Nuclear Resonance fluorescence induced by an 8 MeV LINAC [35] and detection of gamma rays with high purity Ge detectors.

The detection method for the last source listed does not detect fission emissions, but characteristic nuclear resonance fluoresense gamma rays emitted by the nucleus of a particular isotope. This source will also induce photofission in HEU. The sources can be divided into neutron, both 60 keV from proton accelerators and high energy neutrons from DD or DT neutron generators, and gamma ray produced by proton accelerators producing monoenergetic gamma emissions or LINAC emitting bremstrahlung spectra. In addition, a simple change of target in a LINAC would provide an interrogating neutron source.

5. DATA PROCESSING

Data processing methods include gamma ray spectrometry and time coincidence methods with the interrogating source or coincidences between detectors.

Gamma ray detectors are well known and gamma ray detection methods can utilize NaI for low energy resolution and HPGe for high resolution gamma ray spectrometry with commercial multichannel analyzers. Delayed gamma ray as a function of time could be measured after the sources are turned off. Similar measurements could be made for delayed neutrons.

There are a variety of ways to acquire other 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 ^{3}He 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 appropriate software. For 5 channels of operation the NMIS processor is capable of on-line processing at rates of over 2×10E6 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 fast processor for signal to noise ratios of $1\times10E^{-3}$.

6. SUMMARY MATRIX

The various interrogating sources and the methods of detection are summarized in the following Table 4. Potential measurements are presented. For high time resolution measurements with organic scintillators, sensitive to neutrons above 0.5MeV, the scintillators would not be sensitive to the 60 keV source neutrons and measurements could be performed when the 60keV source is on. Of course, for hydrogenous cargo there may be appreciable background from neutron capture. For other pulsed sources the scintillation detectors 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. The sources can be divided into two classes: neutron and gamma sources with a variety of detection methods. Many of the detection methods are common to many of the interrogating sources. LINAC sources can be used to produce neutrons or bremsstrahlung spectra by changing targets.

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
11. Nuclear Resonance Fluorescence (8 MeV brehmstralung)	—	—	—	—	—		—	HPGe

[a]BP means between pulses, DP means during source interrogation, and AP means immediately after pulsing.
[b]DT generator operated steady state with alpha detector to define a cone of neutrons.
[c]Differential dieaway.

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