Fission Signatures for Nuclear Material Detection

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Abstract—**Detection and interdiction of nuclear materials in all forms of transport is one of the most critical security issues facing the United States and the rest of the civilized world. Naturally emitted gamma rays by these materials, while abundant and detectable when unshielded, are low in energy and readily shielded. X-ray radiography is useful in detecting the possible presence of shielding material.**

Positive detection of concealed nuclear materials requires methods which unequivocally detect specific attributes of the materials. These methods typically involve active interrogation by penetrating radiation of neutrons, photons or other particles. Fortunately, nuclear materials, probed by various types of radiation, yield very unique and often strong signatures. Paramount among them are the detectable fission signatures, namely prompt neutrons and gamma rays, and delayed neutrons gamma rays. Other useful signatures are the nuclear states excited by neutrons, via inelastic scattering, or photons, via nuclear resonance fluorescence and absorption.

The signatures are very different in magnitude, level of specificity, ease of excitation and detection, signal to background ratios, etc. For example, delayed neutrons are very unique to the fission process, but are scarce, have low energy, and hence are easily absorbed. Delayed gamma rays are more abundant but "featureless," and have a higher background from natural sources and more importantly, from activation due to the interrogation sources.

The prompt fission signatures need to be measured in the presence of the much higher levels of probing radiation. This requires taking special measures to look for the signatures, sometimes leading to a significant sensitivity loss or a complete inability to detect them.

Characteristic gamma rays induced in nuclear materials reflecting their nuclear structure, while rather unique, require very high intensity of interrogation radiation and very high resolution in energy and/or time.

The trade off of signatures, their means of stimulation, and methods of detection, will be reviewed.

Index Terms—**Delayed neutrons and gamma rays, neutron fission and photofission, nuclear fission detection, prompt neutron and gamma rays, SNM detection.**

I. Introduction

DETECTION and interdiction of nuclear materials in all forms of transport is one of the most critical security issues facing the United States and the rest of the civilized world. The detection techniques being investigated over the last several years cover most attributes of the nuclear materials: uranium and plutonium isotopes, in particular U235 and Pu239.

Naturally emitted gamma rays by nuclear materials form the basis for all passive detection techniques. While abundant and detectable, they are low in energy and readily shielded.

High energy x-ray radiography is most useful in detecting the presence of dense objects which include nuclear materials and means to shield them.

While the aforementioned techniques are fast and an essential part in an overall detection approach, positive detection of concealed nuclear materials requires methods that unequivocally detect specific and difficult to conceal attributes of the nuclear materials. These methods typically involve active interrogation by penetrating radiation of neutrons, photons or other particles. Fortunately, nuclear materials, probed by various types of radiation, yield very unique and often strong signatures. Paramount among them are the penetrating detectable fission signatures, namely prompt neutrons, prompt gamma rays, delayed neutrons and delayed gamma rays. Other useful signatures are nuclear states excited by neutrons via inelastic scattering or by photons via nuclear resonance fluorescence and absorption.

The signatures greatly vary in magnitude, level of specificity, ease of excitation and detection, type and intensity of backgrounds, etc. For example, delayed fission signatures (e.g., delayed neutrons) are probably the most unique of the fission process. The delayed signatures are measured after the fission has been created and hence when the interrogation source is off, which greatly facilitates the measurements and reduces the background. The delayed neutrons are unfortunately scarce, have low energy, and hence are easily absorbed. Delayed gamma rays are more abundant but they are "featureless," and have a higher background from natural sources and, more importantly, from activation due to the interrogation sources.

The prompt fission signatures, as the name implied, are created instantaneously by the interrogation radiation and measured in its presence. Thus the background radiation, in this case, is much more intense than the sought-after signature. This requires taking special steps that often lead to a significant sensitivity loss or to a complete inability to detect them.

Nuclear material signatures and their measurement methods will be reviewed in the rest of the paper. Some possible new approaches are also discussed.

II. Categories of Signatures and Cross Sections

SNM (Special Nuclear Materials) concealed in loaded cargo containers or trucks can, in principle, be detected by various techniques and multiple signatures; some are fission specific and some not:

Fission specific signatures are
- Prompt fission neutrons
- Prompt fission gamma rays
- Delayed fission neutrons
- Delayed fission gamma rays

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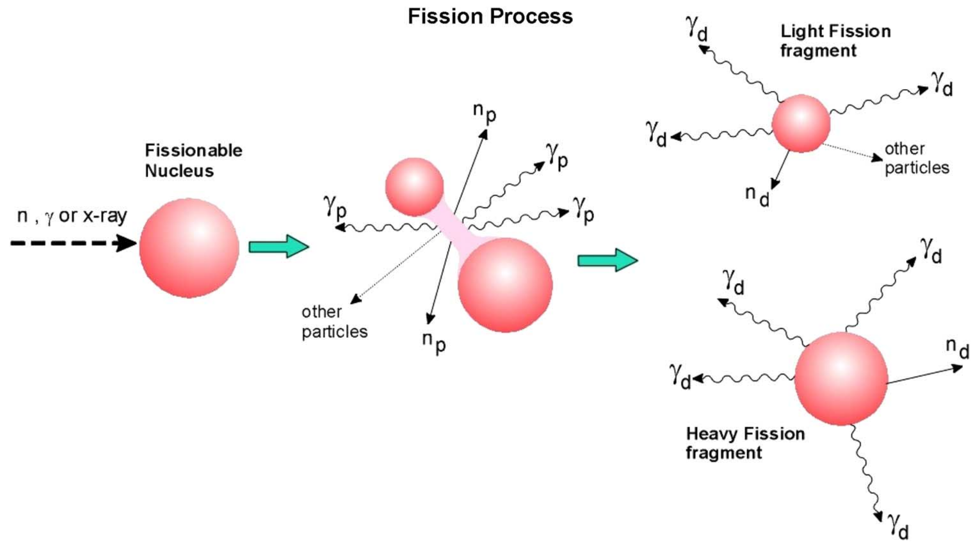

Fig. 1. Simple schematics of the fission process.

Non fission signatures are

- Isotopic nuclear levels
- High atomic number (Z)
- Others (muon induced fission, mesic atoms, muon radiography)

The fission specific signatures are the direct consequence of the nuclear fission reaction induced by the neutron or photon interaction with the material. A simple schematic of the process is shown in Fig. 1. The absorption of the probing radiation highly excites the nucleus which is then split into two smaller unequal fission fragments. The fragments are also highly unstable and decay towards stable nuclei by the emission over time of beta rays, with associated gamma rays (the "fission delayed gamma rays"), and in a few cases also neutrons (the "fission delayed neutrons"). Before the two fragments separate and fly apart (within 10^{-16} s of the initial interaction), they are connected by a neck of highly excited nuclear mass which releases part of the excitation energy first by "boiling off" neutrons and then gamma rays. Since all these processes are completed within less than 10^{-12} s, the emitted particles are called "prompt," hence "fission prompt neutrons" and "fission prompt gamma rays." Gamma-ray emission continues from the freshly created fission fragments by a rapid isomeric transition in the time scale of less than a nanosecond to several hundred microseconds. Depending on the time resolution of a prompt gamma detection device, part or all of these gamma rays may be included or excluded as part of the prompt γ rays signature.

A key parameter for a fission based SNM technique is the fission cross section. It expresses the ability of the interrogation radiation to induce fission in the nuclear material. It varies greatly with energy and type of the probing radiation. Fig. 2 shows this for U235. The cross section ranges from about 600 b $(\mathrm{barn} = 10^{-24}\ \mathrm{cm}^2)$ for thermal neutrons and a few hundred barns for the resonance neutrons (≈0.4 eV to10 KeV), down to about 1.2 b for 2.2 MeV (and fission spectrum) neutrons. It is much lower for the photofission process, about 0.001 b at 6 MeV rising to about 0.1 b at 10 MeV photons.

The fission cross section of Pu239 has a similar behavior but it is higher than that of U235 by about a factor of 1.2 to

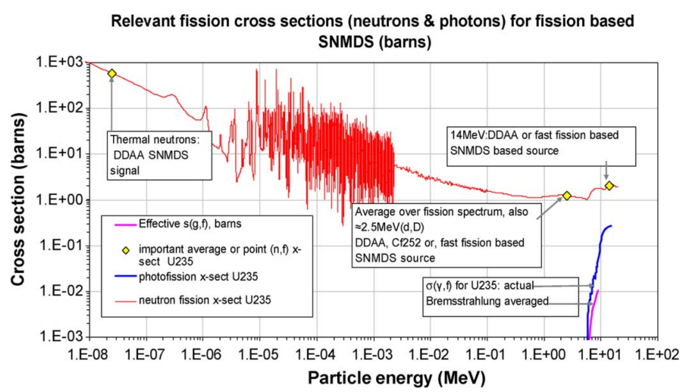

Fig. 2. Neutron and photon fission cross section for U235.

2. Most other actinide isotopes, such as Th232, U238, Np238, Pu240 etc are fissioned by higher energy neutrons (the thresholds range about 0.5–1.5 MeV) but not by thermal neutrons. Their fission cross sections are generally lower than those of U235 and Pu239.

III. Prompt Fission Signatures

The prompt neutrons and gamma rays are practically emitted instantaneously at the moment of inspection. They are amongst the strongest signatures available: about 2.5 and 5 neutrons per fission event, corresponding to fission by thermal and 14 MeV neutrons, respectively. About 3.5 neutrons per fission are generated in x-ray photon induced fission with an x-ray end point energy of 8 to 10 MeV (see Table I).

Since the background due to the interrogation particle is always very high and overwhelms the signal, one tries, in these cases, to use some kind of spectroscopic discrimination.

A. *Prompt Neutron Signature*

In the case of neutrons, one may choose to induce fissions with neutrons with energy lower than at least part of the fission spectrum. Since (d,D) neutrons produced by low voltage deuteron accelerators, with energy less than ≈2.8 MeV are

TABLE I
PROMPT FISSION NEUTRONS PER FISSION FROM NEUTRON AND PHOTON INDUCED FISSION [1]

Isotope	$\langle \nu_p \rangle$	$\langle \nu_p \rangle$ for (γ,f) at 9 MVX*	$\langle \nu_p \rangle$ E>3 MeV
^{235}U	2.4-4.4	≈2.6	0.6-1.1
^{238}U	2.6-4.4	≈2.6	0.7-1.1
^{239}Pu	2.9-4.9	≈3.2	0.7-1.2

*MVX=end point energy of x-ray in MeV. 2nd column is based on [2].

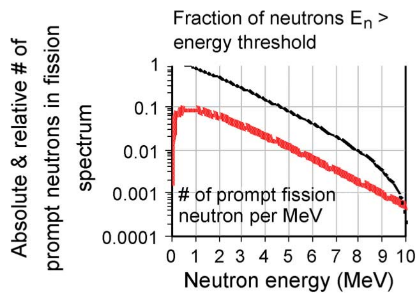

Fig. 3. Prompt neutron fission spectrum.

available, prompt fission neutrons with energy higher than 3 MeV could, in principle be detected. About 25% of the fission neutrons are above this energy, and the signature strength is still pretty good, above 0.6 neutrons per fission (Table I and Fig. 3).

Another way to detect the prompt neutrons is via the temporal effect that the neutron thermalization provides. If the interrogation neutrons are generated in microseconds (to hundreds of microseconds) wide pulses, they will rapidly slow down and thermalize in the source head (which is normally made, in part, of a neutron moderator) and in the cargo itself (if hydrogenous). The fast source neutrons, interrogating the cargo, create fissions in the fissile material, if present, however the thermal neutrons create many more because of their much higher fission cross section (hundreds of barns vs. a few barns). The thermal neutrons stay in the cargo much longer after the end of the source pulse. They decay with the characteristic time, called the thermal neutron "die-away time," which for large cargos, is of the order of a few hundred microseconds to milliseconds. The thermal neutron induced fission rate will decay with this die-away time. Thus a detector, which is insensitive to the numerous thermalized source neutrons (such as cadmium covered moderated He3) would detect fast neutrons only if a fissile material is present. This method is called Differential Die Away Analysis (D^2A^2) [3] and uses the full strength of the prompt neutron signature.

In the case of x-ray based photofission, the interrogation x-ray intensity is so high ("the gamma flash") that only neutron detectors which are entirely insensitive, one way or another, to the huge x-ray radiation can be employed to detect the prompt fission neutrons. Currently known detectors, such as the SiC [4], [5], which are insensitive enough to intense gamma radiations have very low neutron efficiency. Another problem with photofission based systems is the relatively strong photoneutron production by the (γ,n) reactions in the x-ray source itself and in the cargo by nuclides with (γ,n) thresholds lower than the

TABLE II
(a). PHOTONEUTRON PRODUCTION THRESHOLD IN CARGO MATERIAL, RELEVANT FOR 9 MeV ENDPOINT X-RAY INTERROGATION (USING NNDC Q-VALUE CALCULATOR); (b). PHOTONEUTRON PRODUCTION THRESHOLD IN X-RAY CONVERTER AND THREAT MATERIALS, RELEVANT FOR 9 MeV ENDPOINT X-RAY INTERROGATION (USING NNDC Q-VALUE CALCULATOR)

(a)

Isotope	Natural abundance (%)	(γ,n) threshold (MeV)	Where present
H2	0.015	2.225	cargo
C13	1.11	4.947	cargo
N14	99.634	10.559	cargo
N15	0.366	10.838	cargo
O16	99.762	15.672	cargo
O17	0.038	4.144	cargo
O18	0.2	8.046	cargo
Fe54	5.845	13.38	cargo & structural
Fe56	91.754	11.199	cargo & structural
Fe57	2.119	7.647	cargo & structural
Fe58	0.282	10.046	cargo & structural

(b)

Isotope	Natural abundance (%)	(γ,n) threshold (MeV)	Where present
Ta181	100	7.577	x-ray converter
W180	0.12	8.412	x-ray converter
W182	26.5	8.065	x-ray converter
W183	14.31	6.19	x-ray converter
W184	30.64	7.411	x-ray converter
W186	28.43	7.191	x-ray converter
Au197	100	8.072	x-ray converter
Pb204	1.4	8.394	x-ray shielding
Pb206	24.1	8.086	x-ray shielding
Pb207	22.1	6.738	x-ray shielding
Pb208	52.4	7.367	x-ray shielding
U235	0.72	5.297	SNM
U238	99.275	6.154	Fissionable
Th232	100	6.44	Fissionable
Pu239	100	5.646	SNM

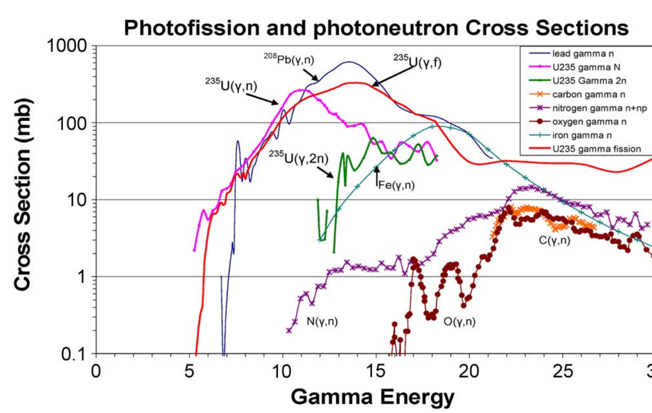

Fig. 4. Photofission and photoneutron cross sections for some key nuclides in the 5 to 30 MeV endpoint x-ray energy range.

end point energy of the x-ray. The most important (γ,n) thresholds relevant for a 9 MeV end point x-ray are listed in Table II.

The photoneutron intensity rapidly increases, and so is the neutron energy, as the end point x-ray energy is increased beyond the 8–9 MeV range (see Fig. 4). These neutrons, especially

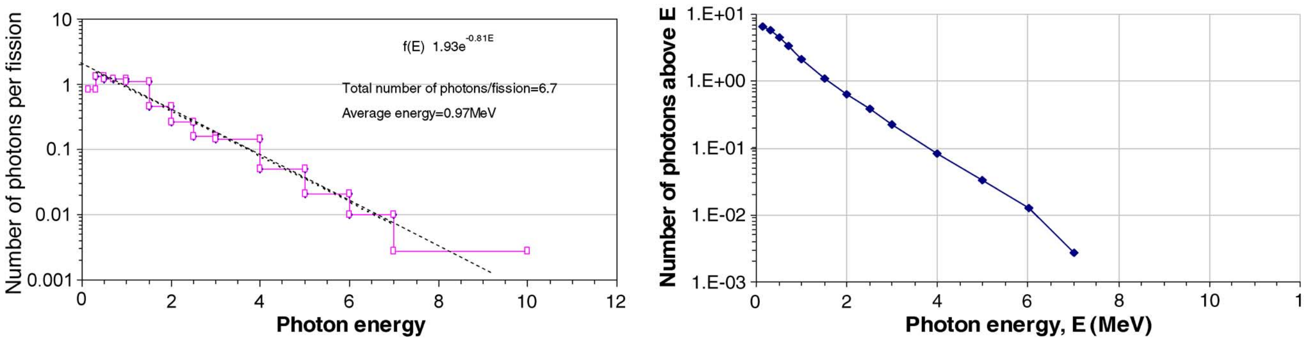

Fig. 5. (a) Prompt fission gamma from U235. (b). Integrated prompt fission gamma.

TABLE III
PROMPT FISSION GAMMA RAYS [7]

Isotope	$<\gamma_p>$	$<\gamma_p>$ E≥3MeV
^{235}U	6.7	0.21
^{238}U	6.7	0.21
^{239}Pu	7.2	0.23

TABLE IV
DELAYED NEUTRONS YIELD AND MEAN HALF LIFE FOR NEUTRON [1] AND PHOTOFISSION [2], [8]

Isotope	$<\nu_d>$, (n,f)	$<\nu_d>$, (γ,f)	$<\tau_{n1/2}>$ sec
^{235}U	0.017	0.01	8.8
^{238}U	0.045	0.029	6.4
^{239}Pu	0.0066	0.004	8.8

those with higher energy, constitute a significant background for the prompt fission neutrons. Significant reduction in this background is possible by proper design and choice of the x-ray converter and surrounding materials.

B. Prompt Gamma Ray Signature

This signature is the strongest amongst the fission signatures, on average about 7 photons are emitted per fission event (Table III). Their average energy is about 1 MeV with more than 3% above 3 MeV, namely 0.21 photon per fission [see Figs. 5(a) and 5(b)].

Detection of prompt gamma rays is a challenge. It has to be performed in the presence of the interrogation radiation. Even if the latter is a neutron beam, the prompt γ rays need to be distinguished from the gamma rays produced by the source neutrons in the cargo materials and in the gamma detector itself. These interactions, mostly made by inelastic neutron scattering and capture processes yield gamma rays with energies which overlap that of the fission gamma rays. Thus to detect them one needs to suppress the background or at least to account for it.

Neutron Time of Flight (TOF) is one of the techniques that allows for the detection of gamma rays (whether from fission or from another interaction e.g., inelastic scattering) of the source neutrons. It is founded on the fact that the speed of gamma rays (30 cm/ns) is much higher than that of fast neutrons (1.4 cm/ns for 1 MeV, to 5.2 cm/ns for 14 MeV). The large difference in the flight time between the two species (i.e., gamma rays and mono-energetic neutrons, generated in narrow nanosecond pulses) allows for detecting the gamma rays resulting from the neutron interaction in the cargo. If the fissile or fissionable materials are present the detected gamma ray spectrum will contain both inelastic scattering and fission gamma rays which have a distinctively different shape from spectra generated in benign materials [6].

IV. DELAYED FISSION SIGNATURES

The delayed fission signatures are characterized by a temporal behavior that allows measuring them well after the source, that induced the fission, is shut off. They originate from the β decay of the many fission products. Except for a small fraction of very fast decaying gamma rays (“isomeric” transitions), the delayed signatures slowly decay from a fraction of second to minutes. Only a small fraction of their maximum intensity (“equilibrium activation”) values can be induced in typical interrogation scanning systems. These signatures are however useful for alarm clearing application.

A. Delayed Neutrons

The delayed neutrons from neutron fissions and photofission in ^{235}U, ^{239}Pu and ^{238}U are given in Table IV. The first column lists the isotopes, the second and third columns give the number of delayed neutron per fission from neutron and photofission, respectively. The delayed neutrons are traditionally broken into 6 groups with half life ranging from 0.3 s to 80 s. The fourth column in Table IV gives the average half life of the delayed neutrons, which is the sum of the individual half lives each weighted with its relative abundance. The average delayed neutron energies are low in the range of 400 keV to 500 keV.

Delayed neutrons are unique and unequivocal signatures of fission. There is no other process which yields delayed neutrons, besides for a small background from the reaction $^{17}\mathrm{O(n,p)}^{17}\mathrm{N}$, which occur if the source neutrons are above 10.4 MeV, or $^{18}\mathrm{O}(\gamma,\mathrm{p})^{17}\mathrm{N}$ if the photon energy exceeds 15.9 MeV, yielding a delayed neutron with 4.1 s half life. It is relatively easy to measure them with high efficiency moderated neutron detectors such as He3.

The two main drawbacks of the delayed neutrons are their low abundance and low average energy, which cause excessive attenuation in hydrogenous cargo. Thus the use of this unique sig-

TABLE V
DELAYED GAMMA RAYS YIELD AND AVERAGE HALF LIFE FROM NEUTRON FISSION [1]

Isotope	$\langle\gamma_d\rangle$	$\langle\tau_{\gamma 1/2}\rangle$ sec
^{235}U	7	30
^{238}U	7	80
^{239}Pu	7	30

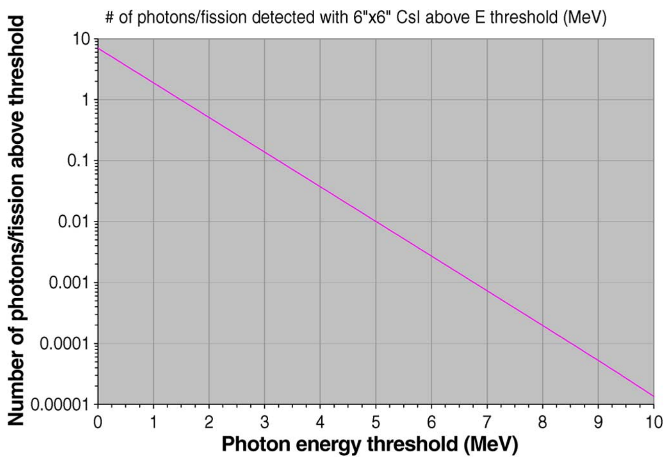

Fig. 6. Number of photons above the threshold energy (based on data in [12]).

nature requires very strong irradiation source and/or long measurement time. Because of the relative ease with which one can measure these neutrons, they can be readily added to any active interrogation system as a complimentary signature.

B. Delayed Gamma Rays

The key properties of the delayed gamma are given in Table V. The first column lists the fissile/fissionable isotopes, the second one gives the total number of delayed γ rays per fission, the third column gives the number of photons above 3 MeV (above most of the expected gamma ray background); see also Fig. 6. The last column gives the weighted average half life of the delayed γ rays, which is noticeably longer than that of delayed neutrons. The delayed γ-ray yields are much higher than those for the delayed neutrons but so are the competing backgrounds. During the "die-away" times of the thermal neutrons originated by the fast source neutrons, the background is dominated by capture gamma rays. But since the delayed γ rays decay slowly one can start their measurement many milliseconds after the interrogation beam shut off. In this time range the delayed activation of some isotopes that may be present in cargo and the natural background constitute the main source of background.

Typical activation backgrounds are from reactions such as: $^{16}\mathrm{O(n,p)}^{16}\mathrm{N}$, $^{19}\mathrm{F(n,\alpha)}^{16}\mathrm{N}$, $^{37}\mathrm{Cl(n,\alpha)}^{34}\mathrm{P}$, $^{48}\mathrm{Ca(n,p)}^{48}\mathrm{K}$, $^{34}\mathrm{S(n,p)}^{34}\mathrm{P}$.

To overcome these activation backgrounds, the source neutrons must be below 10 MeV, to eliminate the $\mathrm{strong}^{16}\mathrm{N}$ activation, and only delayed gamma rays with energy above 3 (or 4) MeV are measured. This still leaves a respectful signal of about .12 to 0.04 photons per fission. The delayed gamma signature cannot easily be used for a rapid scanning because of their relatively long half lives. However for slow scanning, or for alarm resolution with irradiation and measurement times of tens of seconds, close to 50% of the above photon yield can be measured. The high energy delayed γ rays are much more penetrating in hydrogenous cargo compared to delayed neutrons, and are somewhat more attenuated in inorganic metallic cargo. Thus the two signatures, e.g., delayed neutrons and gamma rays are complementary, though the neutron source requirement for the latter is much more demanding.

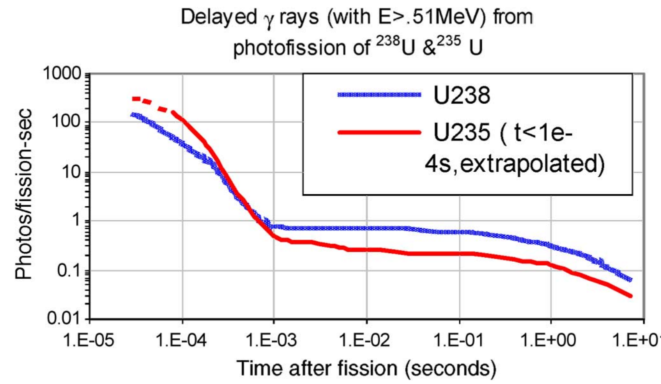

Fig. 7. Early decay of delayed gamma rays [13].

TABLE VI
EARLY DELAYED GAMMA RAY YEILD [13]

Early Delayed gamma rays (photons/fission)	238U	235U
0.1μs to 7s	2.86	1.25
≤1ms	0.04	0.09
≤10ms	0.06	0.10
≤0.1s	0.21	0.15
0.1s to 7s	2.71	1.11

C. Early Time Delayed Gamma Rays

Early fast decay of gamma rays following the occurrence of fission offers the possibility of detecting fission after short irradiation needed for faster scanning. This fast decay is the result of short live isomers formed directly in the fission process releasing gamma rays in the time range of less than nanosecond to milliseconds. The gamma decay from 50 ms over 6 orders of magnitude of time is shown in Fig. 7 [13]. The integrated intensity of these delayed gamma rays over narrow time intervals of interest is given in Table VI, which shows useable intensities of roughly 0.2 per fission in the time interval of 0 to 0.1 s.

V. SUMMARY

The fission signatures that are already playing, or can potentially play, an important role in practical non intrusive inspection systems for detecting SNM were reviewed. Table VII summarizes the approximate intensity, or fission yield, of these signatures. The table covers two orders of magnitude of intensity, different attenuation, ease of detection, signal to background, etc. The optimal choice depends greatly on the application, the concept of operation and also on cost consideration.

TABLE VII
SUMMARY OF FISSION SIGNATURES (APPROXIMATE YIELDS/FISSION)

Fission signature	^{235}U	^{239}Pu	^{238}U	Application [ref #]
Prompt neutrons thermal fission	2.4	2.9	0	[2]
Prompt neutrons from <2.5 MeV n fission or photofission	2.8	3.2	2.9	[8]
Prompt and delayed γ-rays from n or γ fission	6.7	6.7	7.2	[7]
Prompt γ-rays from n or photofission with Eγ > 3 MeV	0.2	0.3	0.2	[7]
Delayed neutrons from n fission	0.015	0.0061	0.044	[2]
Delayed neutrons from photofission	0.01	0.004	0.028	[13]
Delayed γ-rays with E γ > 3 MeV	0.13	0.07	0.11	[10]
Delayed γ-rays with E γ > 4 MeV	0.05	0.02	0.03	[10]

The non fission nuclear material signatures mentioned in Section II covered in a separate paper.

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