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Conventional and Non-Conventional Nuclear Material Signatures

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Abstract. The detection and interdiction of concealed special nuclear material (SNM) in all modes of transport is one of the most critical security issues facing the United States and the rest of the world. In principle, detection of nuclear materials is relatively easy because of their unique properties: all of them are radioactive and all emit some characteristic gamma rays. A few emit neutrons as well. These signatures are the basis for passive non-intrusive detection of nuclear materials. The low energy of the radiations necessitates additional means of detection and validation. These are provided by high-energy x-ray radiography and by active inspection based on inducing nuclear reactions in the nuclear materials. Positive confirmation that a nuclear material is present or absent can be provided by interrogation of the inspected object with penetrating probing radiation, such as neutrons and photons. The radiation induces specific reactions in the nuclear material yielding, in turn, penetrating signatures which can be detected outside the inspected object. The "conventional" signatures are first and foremost fission signatures: prompt and delayed neutrons and gamma rays. Their intensity (number per fission) and the fact that they have broad energy (non-discrete, though unique) distributions and certain temporal behaviors are key to their use. The "non- conventional" signatures are not related to the fission process but to the unique nuclear structure of each element or isotope in nature. This can be accessed through the excitation of isotopic nuclear levels (discrete and continuum) by neutron inelastic scattering or gamma resonance fluorescence. Finally there is an atomic signature, namely the high atomic number ($Z > 74$), which obviously includes all the nuclear materials and their possible shielding. The presence of such high-Z elements can be inferred by techniques using high-energy x rays. The conventional signatures have been addressed in another article. Non-conventional signatures and some of their current or potential uses will be discussed here.

Keywords: Nuclear signatures and detection, neutron & photon based fission, high atomic number (Z) detection, homeland security
PACS: 24.75.+i, 25.85.-w, 25.85.Ec, 78.70.-g, 78.70.Ck

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, and in particular ^{235}U and ^{239}Pu. 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, unequivocal and hard-to-defeat detection techniques of concealed nuclear materials are needed and provided by active interrogation. This is achieved by using penetrating radiation of neutrons, photons or other particles to induce signatures which are characteristic of nuclear materials. Fortunately, nuclear materials 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 (constituting the "conventional signatures"). Other useful signatures (the "non-conventional" ones) are nuclear states excited by neutrons via inelastic scattering or by photons via nuclear resonance fluorescence and absorption, as well as atomic number (Z) related signatures.

SNM (Special Nuclear Materials) concealed in loaded cargo containers or trucks can, in principle, be detected by various techniques and multiple

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signatures; some are fission specific and some not. Fission specific conventional signatures are:

- Prompt fission neutrons
- Prompt fission gamma rays
- Delayed fission neutrons
- Delayed fission gamma rays

Non-fission, non-conventional signatures are:

- Isotopic nuclear levels
- High atomic number (Z) manifestations
- 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 actinide nuclides. The non-conventional signatures are not fission-specific but isotope-specific (such as nuclear levels) or atom-specific (like high atomic number Z). The signatures greatly vary in magnitude, level of specificity, ease of excitation and detection, type and intensity of backgrounds, etc. The conventional signatures are reviewed in a companion paper [1]. The non-conventional nuclear material signatures and their measurement methods will be reviewed here.

NUCLEAR LEVELS AS SIGNATURES

The nuclear levels, expressing themselves by the emission of characteristic gamma rays when nuclear transitions are induced by neutron and photon interactions are a unique signature of the specific nuclide. Thus the presence and identity of nuclear and non-nuclear materials can be determined by excitation of these levels. The levels can be discrete lines and/or relatively unique continua.

Inelastic Scattering and Fast Fission Neutron Induced Reactions

One way to excite these signatures is by fast neutron inelastic scattering. This has been demonstrated by the PFNA technique, and gamma ray resonance scattering.

In PFNA [2], a high-energy, relatively intense and mono-energetic, collimated beam of neutrons is chopped and bunched to form neutron pulses with a width of the order of 1 ns. This pulsed beam of neutrons is used to irradiate a cargo in a two-dimensional sweep. As each pulse passes through the cargo, the neutrons interact with the elemental makeup of the cargo to produce gamma rays which are sensed by surrounding NaI gamma detectors. Since the pulse is very narrow, the time at which the gamma rays are detected gives the location of the pulse at the time the gamma rays were produced. The energies of the gamma rays are characteristic of the material that produced them. In this way, PFNA provides a three-dimensional map (two dimensions by sweeping the beam and the third by time) of the elemental composition of the cargo.

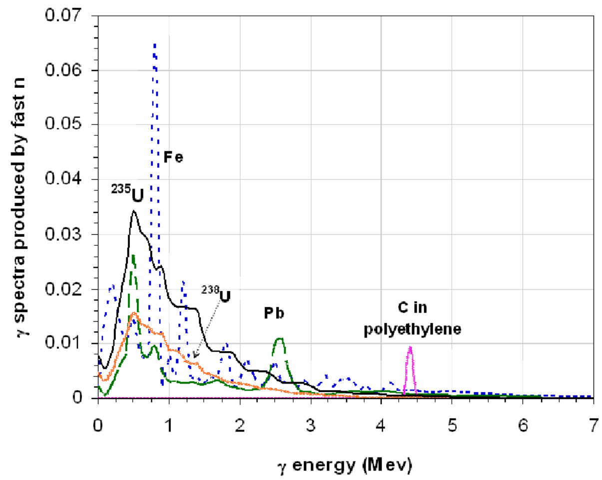


FIGURE 1. MCNP calculation of gamma production spectra by 8.5 MeV neutrons in 10 cm thick polyethylene, iron, lead, ^{238}U and 1 cm thick of ^{235}U.

The elemental composition of the cargo is usually indicated in PFNA by the presence of photons of one or more characteristic energies which produce peaks in the spectra. For example, carbon produces a strong peak at 4.44 MeV. However, SNM isotopes, like many high-Z elements, do not show discrete gamma lines but exhibit continua with unique useable features. This makes identification of SNM isotopes and higher-Z isotopes possible and distinguishable from benign cargo materials such as iron and hydrogenous materials. Figure 1 shows the calculated spectra of gamma rays produced in various materials from 8.5 MeV neutron interactions. The discrete lines in Fe and carbon (in polyethylene) are prominent, but so are also the unique shapes of the spectra in ^{238}U and ^{235}U. In the latter we see the strong enhancement of the low energy part by the fission gamma rays (resulting from the higher fission cross section and neutron multiplication in the SNM), increasing the signal significantly. SNM isotopes, due to the large number of energy states in the few-MeV range, produce gamma-ray spectra with a characteristically sharper high-energy slope than other elements. This is seen even better in the broad resolution spectra shown in Figure 2. This steep slope can be used as a signature for the identification of SNM. In fact PFNA spectrum measurements [3] with graphite, aluminum, iron, tungsten, lead, bismuth and ^{238}U showed a strong discrimination between ^{238}U and all other materials using the ratio of the spectral integral below 3 MeV to the integral above 4 MeV. The good discrimination is retained even if the materials are embedded in cargo of paper. MCNP calculation shows that if the 10 cm of ^{238}U is replaced by 1 cm thick ^{235}U the discrimination ratio increases further by about 20%, due to the increased fission cross section and multiplication.

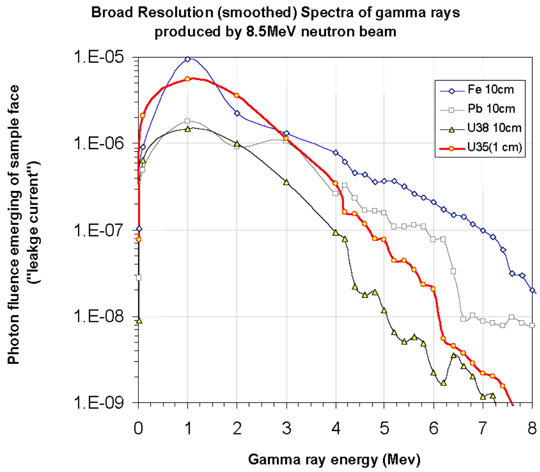


FIGURE 2. Smoothed spectra of gamma rays produced by 8.5 MeV neutrons [3].

In summary, by using the PFNA technique, it is possible to distinguish SNM from many other materials even when embedded in a variety of cargos including highly attenuating and dense cargo such as paper. It is also possible to distinguish lead and tungsten (along with SNM) from other materials in light cargos such as electronics. If the neutron beam can be localized well enough (reducing voxel size) to separate an object consisting of SNM, lead, tungsten or bismuth from the surrounding cargo, it should be possible to detect and identify these materials even in dense and difficult cargos like paper.

X-ray Based Gamma Nuclear Resonance Fluorescence (NRF) [4]

The process of NRF corresponds to the excitation of a nuclear state by photons and having that state decay to the ground state or an excited state by the emission of a photon. NRF cross sections typically have very large peak values that correspond to hundreds of barns for NRF states with energies in the range of a few MeV. The states are, however, broadened by the zero-point motion of the atom and thermal motion to widths from a fraction of an eV in heavy nuclides to 20 eV for the light nuclei, such as nitrogen and oxygen. The effective cross section (a couple of barns to more than 10 barns) is the same order of magnitude or larger than the usual electromagnetic processes (photoelectric, Compton and pair production) in that energy region.

An example of NRF spectra of ^{239}Pu is shown in Figure 3 [5]. Several specific, though relatively weak, ^{239}Pu lines are clearly shown. This type of measurement requires the use of high resolution (Ge) detectors, otherwise the information will be completely lost. Many detectors, each collimated and defined a cargo voxel, are needed to look at the inspected cargo volume. "Self Indication" techniques are being investigated to reduce the number of detectors [4].

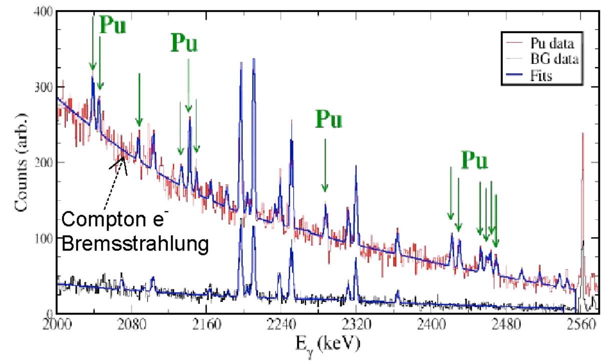


FIGURE 3. NRF spectrum of Pu239 in the energy range of 2.0 to 2.5 MeV [5].

Currently the only practical photon source for NRF that is possibly applicable for nuclear material detection is the broad-energy Bremsstrahlung x-ray spectrum from electrons in the energy range of 3 to 10 MeV. With such a spectrum, the available photon flux even over the broadened resonance is very small, on the order of 10 photons/cm^2/s/µA. Thus high power CW electron-based x-ray sources are required for this signature to be employed.

HIGH Z DETECTION

The techniques falling under this category do not specifically detect nuclear materials as the fission-specific ones or the nuclear-level based techniques are. The techniques here attempt to automatically detect the presence of high-Z materials, generally with Z > 70 in the inspected cargo. All of them are based on x-ray sources.

All the Z discrimination techniques are based on the Z dependence of the total attenuation coefficient as function of the photon energy, see Figure 4. The various techniques attempt to take advantage of the somewhat larger attenuation in the lower energy region (~0.5 MeV to ~1.5 MeV) and the high energy region (> 5 MeV) relevant to large cargo inspection applications.

Dual Energy X-ray

In this technique, as applied to large conveyances, radiography is typically conducted alternately with two energies: 6 MV (or for less dense objects 4.5 MV) and 9 MV (or 6 MV for less dense objects). Inserting selective attenuators, such as high Z material to modify the x-ray spectrum, is similar to the dual energy approach. The ratio (or other functional relationship) between the two x-ray readings is used to infer the presence of higher Z materials because of the higher absorption in the high energy region as a result of the increased pair-production effect. The x-ray detector signal is an integral equation of, among others, the attenuation coefficient that contains the Z information. It cannot be solved to provide this information without major assumptions, such as that the material in the beam is "binary," i.e., made of a low-Z and a high-Z material. Additional assumptions are made regarding the material, its shape, size, density etc. [6]. The performance of the dual energy x-ray system depends how close to reality are the many assumptions made in the detection algorithm. It obviously depends on the type of cargo and its density clutter and would be prone to false alarms.

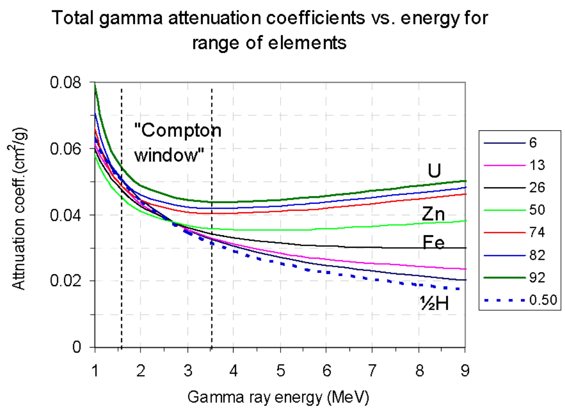


FIGURE 4. Total gamma ray attenuation coefficients vs. photon energy for wide range of elements.

High Energy Bremsstrahlung Backscattering

The normal backscattering spectrum of bremsstrahlung radiation is made up of low-energy photons with a maximum energy of about 0.5 MeV (scattering at 90^0, see Figure 5). A 0.511 peak of variable intensity also appears in the backscattering spectra but it originates from annihilation radiation in the surrounding materials. However there exists a small but noticeable high-energy component in the backscattered spectrum that is significantly stronger for high-Z scatterers like tungsten and uranium, and can provide a strong indication for their presence. This is the result of the bremsstrahlung of high-energy electrons (see Figure 5) which are generated, preferentially in the x-ray beam direction by backscattered Compton photons, and to a smaller extent, by the electron-positron pairs created by high-energy photons. Electrons lose part of their energy by the Bremsstrahlung process. The effect is much stronger in high-Z materials (it behaves as Z^2). The Bremsstrahlung radiation is also forward peaked, however a few of the electrons change their direction of motion to the back angles by multiple collisions with the atomic electrons and/or one or more collisions with the material nuclei. The bremsstrahlung radiation in these angles can then be detected by energy sensitive detectors (such as NaI) placed in the backward angles ($>90^0$). The intensity of the backscattering signal above 1MeV for tungsten in air is estimated to be 10^{-12} to 10^{-11} photons/cm^2/electron.

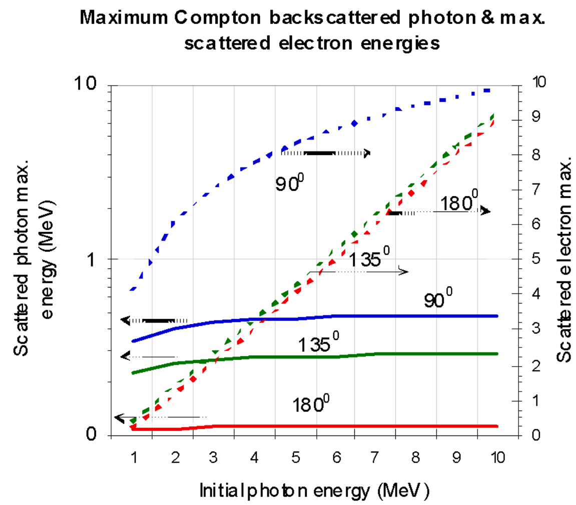


FIGURE 5. Maximum Energy of Backscattered Photons.

The above effect was modeled with photon/electron Monte Carlo transport codes (such as EGS and MCNP) and reliably measured. Figure 6 shows the EGS calculation for a 9 MV x-ray source with and without the bremsstrahlung process. Switching off the process completely eliminates the high-energy part of the spectrum. The presence of a high-energy continuum can be seen in Figure 3, as sample related background for the discrete gamma lines. Figure 7 shows the high-energy backscattering spectra from tungsten, iron and polyethylene scatterers, as measured with NaI detector. The source was a commercial 6 MV pulsed linac x-ray source. The strong Z dependence garnered from this run is shown in Figure 8. The Z information gleaned by this method is stronger at 9

MV, as the high energy flux of the bremsstrahlung is richer with high energy photons.

The presence of cargo reduces the signal due to the x-ray beam attenuation through the cargo going to the sample as well as the scattered beam attenuation from the sample to the external detector. In addition the size of the detector collimation determines the degree of debasing of the high Z scattering signal with the benign cargo surrounding it. The tighter collimation the better, but the signal strength suffers and the number of detectors required to view the cargo rapidly increasing.

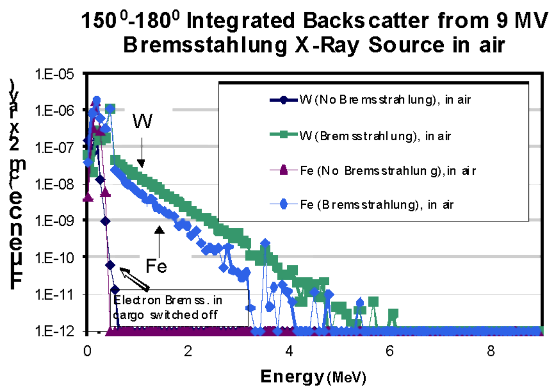


FIGURE 6. High Energy Bremsstrahlung Backscattering Spectra.

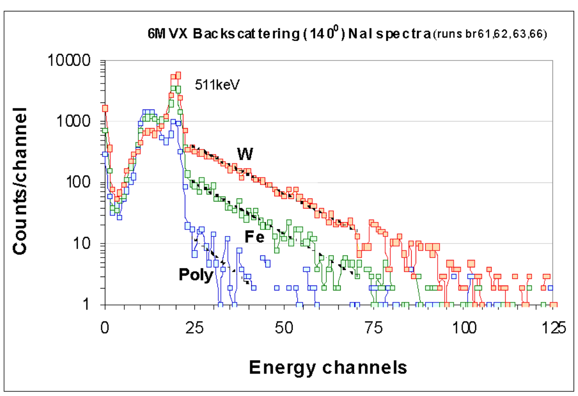


FIGURE 7. Backscattered photon spectra from tungsten, iron and polyethylene. Source: 6MV pulsed linac (Rapiscan P6000 system).

Dual Species Radiography

The Z or elemental information obtained can be greatly augmented if the regular x-ray radiography is supplemented with fast-neutron radiography. The main reason for this is that the two species, namely photons and fast neutrons, have a very different behavior vs. Z (see Figure 9). The feasibility and applicability of this approach to the inspection of full-sized cargo was shown several years ago [7]. The low-resolution gamma radiograph and the gamma-ray-to-neutron ratio image are shown in Figure 10.

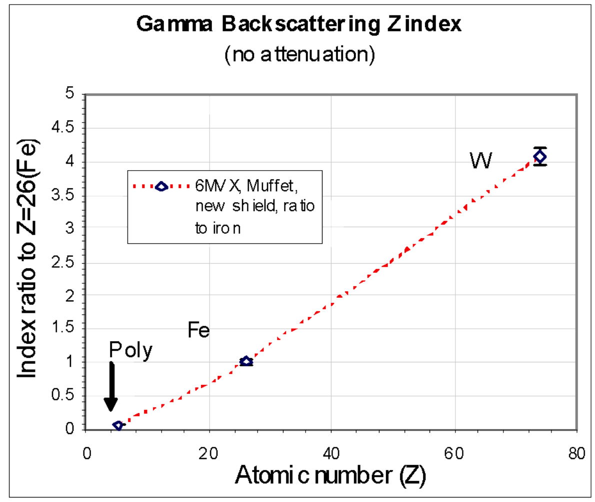


FIGURE 8. Integrated backscattered spectra as Z index.

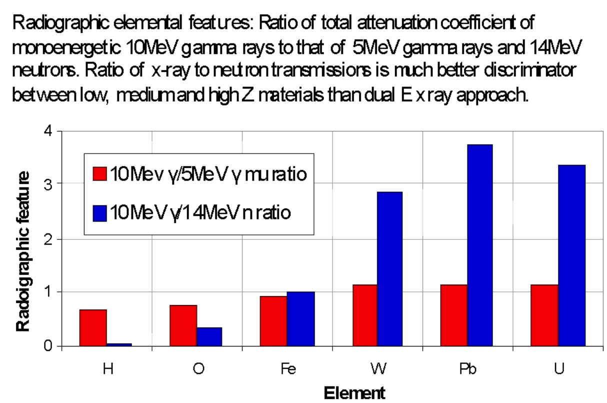
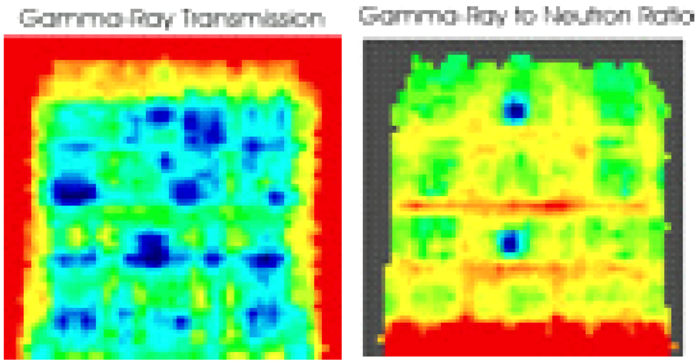


FIGURE 9. Ratio of total cross sections between 14 MeV neutrons and gamma rays.

SUMMARY

Nuclear material signatures which are not based on the fission process, labeled here as "non-conventional", have been reviewed. Their brief overall description is given in Table 1. Their sensitivity, specificity (or ability to discriminate between isotopes), ease and cost of application are widely different. The use of these techniques, as part of nuclear material inspection systems, like the use of the fission based techniques (described elsewhere), requires a thorough trade-off study. It is hoped that the brief review provided here will help in such studies.



Anomalous ("threat") areas in γ radiography to be inspected by fast n-radiography

The ratio of the gamma ray to neutron transmission clearly highlights the nuclear material

FIGURE 10. Two DU samples in 8' cargo (2 pallets) of microwave ovens inspected simultaneously by high energy gamma rays and 8.5MeV neutrons with low spatial resolution [7].

TABLE 1. Summary of key features of non-fission signatures of nuclear materials

Basis	Source	Technique	Reaction type	Specificity (discrimination)	Sensitivity (signal strength & S/B)
Nuclear levels	Monoenergetic fast neutrons	γ spectroscopy & TOF (PFNA)	(n,n'γ)	high	med.- high
Nuclear levels	CW x-ray Bremsstrahlung	NRF	(γ,γ)	V. high	med.-low
Atomic # Z	Linac	Dual energy (e.g. 6/9MV)	Atomic (Compton & pair production)	low	high
Atomic # Z	X-ray Bremsstrahlung; CW or linac	High energy backscattering spectra	Atomic (Compton & pair production) & electron Bremsstrahlung	high	med.
Atomic # Z	γ rays or x-ray Bremsstrahlung plus14MeV ENG	Dual species radiography	Atomic+ total (n,n)	med.-high	med.

ACKNOWLEDGMENTS

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