LLNL-BOOK-819857
IM-1029000

Lawrence Livermore National Laboratory

Nuclear Techniques to Detect Explosives

Counterterrorist Detection Techniques of Explosives, 2nd Edition

March 1, 2021

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1 Introduction

This chapter is focused on nuclear techniques to interrogate a target to detect explosives. We define nuclear as of or relating to, or constituting the nucleus of, an atom. Nuclear techniques involve reactions with atomic nuclei. Nuclear techniques to detect explosives use nuclear reactions to interrogate, for example, luggage for enclosed explosives. A well-known example of a nuclear technique is TNA—thermal (low-energy) neutron analysis. In TNA, low-energy neutrons are absorbed (captured by), for example, nitrogen atomic nuclei in the luggage can lead to high-energy gamma rays that can be used to determine the amount of nitrogen in the luggage. Since many explosives are rich in nitrogen, high amounts of nitrogen may imply that there is an explosive in the luggage thus leading to explosive detection. However, some non-explosive materials, e.g., salami, are rich in nitrogen, which leads to a false detection. Performance considerations should include probability of detection (P_D), probability of a false alarm (P_{FA}), throughput, weight, size, and cost. The goal is 100% P_D, 0% P_{FA}, hundreds of cargo containers per hour or thousands of bags per hour using small light weight low-cost interrogation systems.

Explosives that need to be detected in countering terrorism include commercial, military, and homemade. [Sun, 2010] Commercial and military explosives are manufactured for applications such as breaking rock, making tunnels, or mining. Examples of commercial explosives are TNT and dynamite. Military explosives are, of course, used for war purposes; examples include C4 and RDX. Homemade explosives (HME) are used by terrorists to damage populated areas, airplanes, etc. HMEs include mixtures of common household items such as large quantities of the ingredients obtained from party poppers, firecrackers, or sparklers, household chemicals, pool cleaning chemicals, or fuels. All three types of explosives have been used by terrorists.

In this chapter, we consider explosives enclosed in luggage, vehicles, and cargo containers. Explosives in such enclosures can be difficult to detect—whether they are hermetically sealed or not—since often vapors and particulates from the explosives are not accessible to the explosive vapor or particle detectors. There are, however, nuclear techniques that use radiation to penetrate these enclosures to excite the atomic nuclei of the explosives and detect their emission signatures. Nuclear techniques for explosives detection are distinguished by the type, energy, and temporal pattern of radiation, e.g., continuous thermal neutrons, used to interrogate the enclosure, and the type, energy, and temporal patterns of radiation detected, e.g., MeV gamma rays. Such techniques for explosives detection have been investigated in which neutrons or, in some cases, high-energy (MeV) photons, are used for interrogating the enclosure. Where possible, we comment on performance of the technologies in terms of P_D, P_{FA}, throughput, weight, size, and cost.

There are several previous works describing nuclear techniques. These include Griffin [Griffin, 2009], Lanza [Lanza, 2007], Schubert and Kuznetsov [Schubert, 2008], Sun [Sun, 2010], Lehnert and Kearfott [Lehnert, 2010], Avtonomov and Kornienko [Avtonomov, 2015], Whetstone and Kearfott [Whetstone, 2014], Buffler and Tichner [Buffler, 2010], and Hussein [Hussein, 1992]. This chapter is an update of ionizing nuclear techniques to detect explosives in enclosures such as luggage, vehicles and cargo containers. We do not discuss nuclear quadrupole resonance or magnetic resonance imaging, since they are covered in other chapters in this book.

In this chapter, we begin with a review of the physical phenomena underlying nuclear techniques We then describe radiation sources and detectors used to measure these phenomena in Sections 3 and 4, respectively. Nuclear techniques that have been researched, developed, and employed to detect explosives are discussed in Section 5. Nuclear techniques include thermal neutron analysis, fast neutron analysis, pulsed fast neutron analysis, pulsed fast neutron transmission spectroscopy, associated particle imaging, nuclear resonance absorption and others.

2 Neutron and High-Energy Photon Interactions with Matter

Because of their ability to penetrate vehicles and cargo containers and to interrogate them for explosives, and the availability of sources and detectors, the most common nuclear inspection methods use neutrons (either "thermal" neutrons at energy 0.025 eV or "fast" neutrons at energies 1 MeV and above) or photons above 1 MeV. We will explore the physics associated with both neutrons and photons in this section.

2.1 Neutron Interactions with Matter

For explosives detection applications, the most important neutron interactions to consider are those involving neutrons and atomic nuclei. Although neutrons have zero electric charge, they have mass and a non-zero magnetic moment that can therefore interact with both atomic nuclei and electrons. The latter magnetic interactions with electrons have not been used for explosives detection, but interactions with atomic nuclei of explosives can elicit responses that can help discriminate them from benign materials.

Neutrons can interact with atomic nuclei in a variety of ways, all of which depend on the energy of the neutron and isotope of the target nucleus. For example, the neutron may simply bounce off the nucleus without changing the internal state of the nucleus (elastic scattering), it may excite the nucleus (inelastic scattering), or it may be completely absorbed by the nucleus. The probability of a specific interaction is also known as a cross section, customarily measured in units of barns (or b, 10^{-24} cm). [Rinard, 1991] The probability of *any* of the aforementioned interactions occurring is called the total cross section. As shown schematically in Figure 1, the total neutron cross section is composed of scattering and absorption parts, and each of these can be further decomposed. There are both elastic and inelastic scattering processes. Neutron absorption (also known as capture) by a nucleus can be followed by electromagnetic (radiative) decay, decay into charged or neutral particles, or fission.

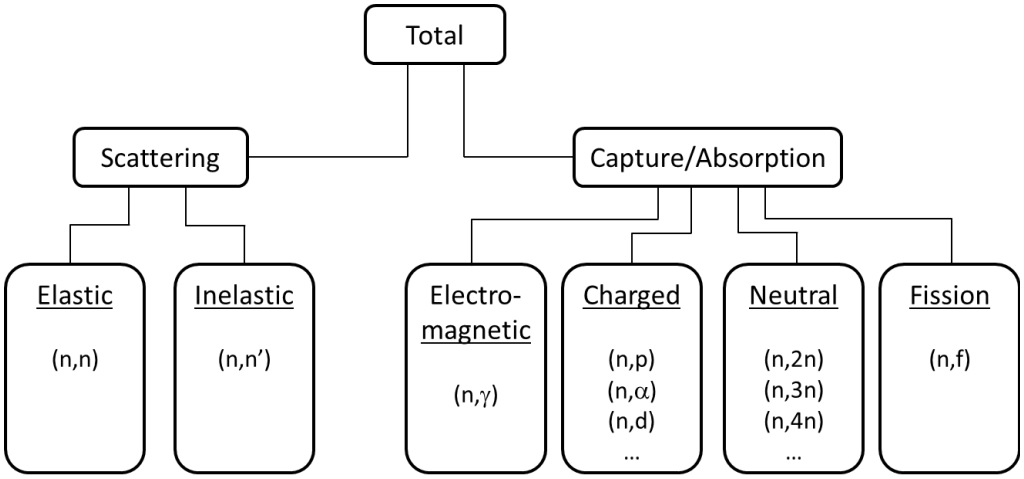


Figure 1. Relationships between neutron cross sections. Here (*n,x*) (see Section 2.1.1) represents an incoming neutron, *n*, that interacts with an atomic nucleus resulting in emission of *x*. *X* can be either a neutron with no change in energy, *n*, with a change of energy, *n'*, a gamma ray, γ, ^{4}He, α, proton, *p*, deuteron, *d*, or fission product(s), *f*. Adapted from Rinard, 1991.

Mathematically, the total cross section can be written

$$\sigma_t = (\sigma_e^s + \sigma_i^s) + (\sigma_{EM}^c + \sigma_{ch}^c + \sigma_n^c + \sigma_f^c)$$

where

σ_e^s is the elastic scattering cross section;

σ_i^s is the inelastic scattering cross section;

σ_{EM}^c is the cross section for capture followed by radiative (electromagnetic) decay;

σ_{ch}^c is the cross section for capture followed by decay into charged particle(s);

σ_n^c is the cross section for capture followed by decay into neutral particle(s);

σ_n^c is the cross section for capture followed by fission of the nucleus.

As shown in Figure 2, the thermal neutron total cross section does not vary monotonically with atomic number, *Z*, and is especially large (~30 b) for hydrogen (*Z*=1). Note also that neutron cross sections are isotopically dependent, e.g., ^{3}He has a much larger thermal neutron cross section (~5000 b) than ^{4}He (~0.9 b). We show both cross sections since ^{3}He is used in a common neutron detector as discussed in Section 4.1 and to show its isotopic dependence.

In contrast to thermal neutrons, the total cross section for 14-MeV (fast) neutrons are generally smaller in magnitude, generally increase and have more monotonic behavior with respect to *Z* than thermal neutrons. As shown in Figure 3, elements pertinent to explosives detection such as carbon, nitrogen, and oxygen exhibit cross section resonances for neutron energies above 300 keV. These resonance phenomena are exploited by techniques like Pulsed Fast Neutron Transmission Spectroscopy (see section 5.1.4) for identifying explosives.

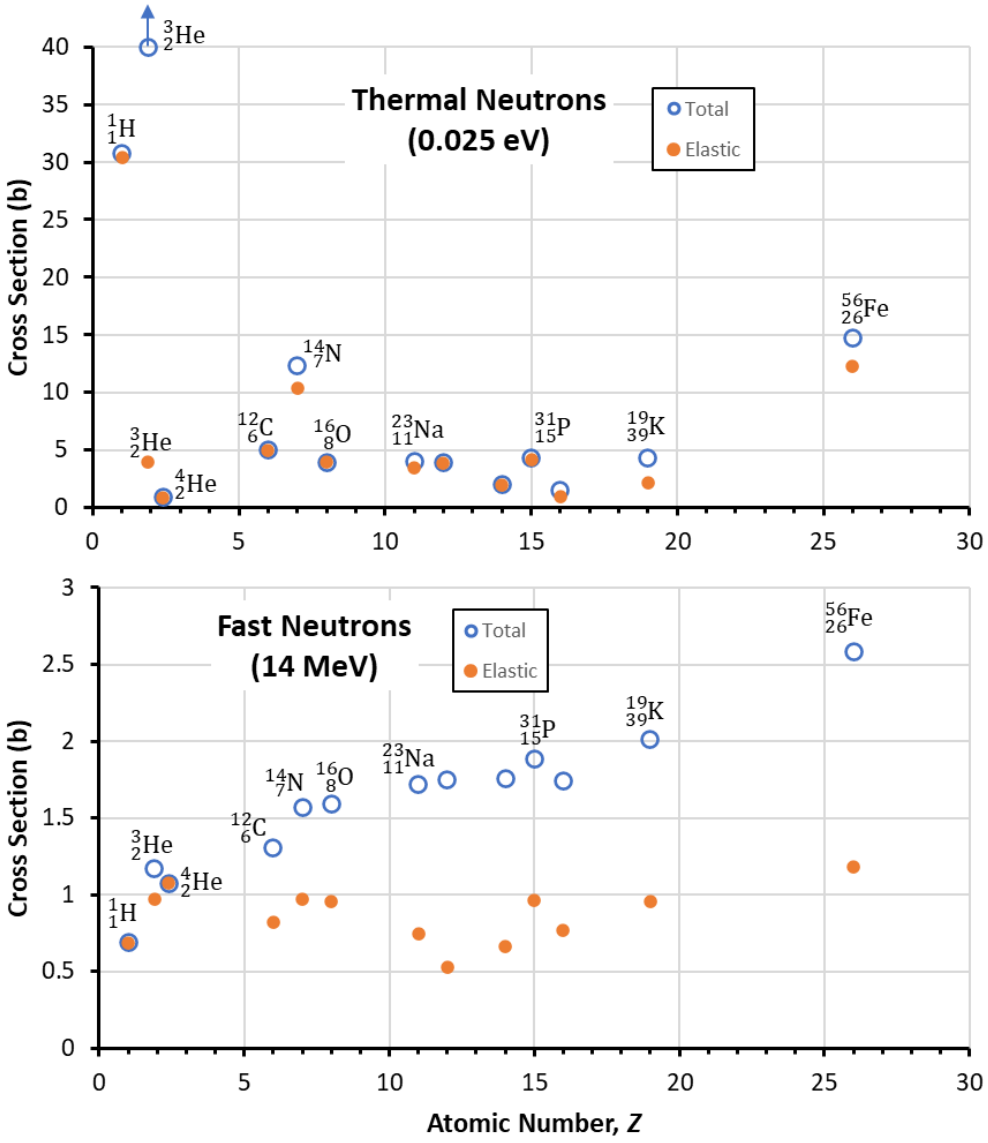


Figure 2. Total cross sections for thermal (top) and fast (bottom) neutron scattering as a function of some elements. For thermal neutrons, the elastic cross is the dominant contribution to the total cross section, except for ^{3}He. For fast neutrons, the elastic cross section is a about half of the total cross section, except for H and ^{4}He. Note that the two plots have different vertical scales. The data used to make these plots are from ENDF/B-VIII [Brown, 2018].

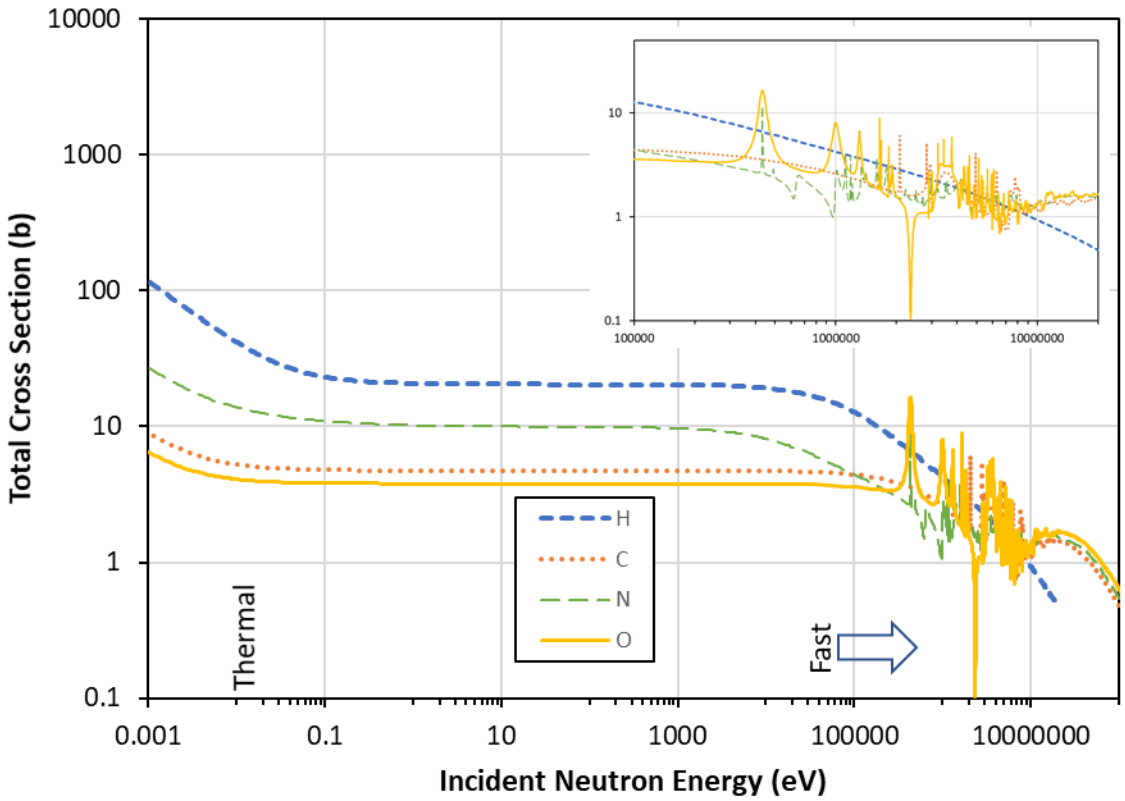


Figure 3. Total cross sections for H, C, N, and O as functions of neutron energy. The inset plot shows resonant features between incident neutron energies of 100,000 eV and 20,000,000 eV (100 keV and 20 MeV). The data used to make these plots are from ENDF/B-VIII [Brown, 2018].

The following subsections discuss the physical phenomena relevant to explosives detection with neutrons. These include elastic scattering, inelastic scattering, and capture (or absorption) reactions.

2.1.1 Elastic Scattering

Elastic neutron scattering occurs when a neutron collides with an atomic nucleus without being absorbed or exciting, i.e., transferring energy to, the nucleus, thereby changing the energy and direction of the neutron. The elastic neutron scattering reaction is written as

$$n + A \rightarrow n + A$$

where n denotes a neutron and A denotes a ground-state nucleus. In the literature, elastic scattering is also written as $A(n,n)A$, $A(n,n)$, or simply (n,n) as in Figure 1.

Elastic scattering is typically the dominant physical interaction for neutrons passing through matter. For slow neutrons elastic scattering is most of the total cross section, while for fast neutrons it is about half the total cross section. A neutron can transfer a significant fraction of its energy to a nucleus if the nucleus has low mass, such as hydrogen or carbon. In this case, the mass of the nucleus can be inferred from measurements of the energy of the scattered neutron and its direction. [Goldstein, 1980] The elastic scattering phenomenon is often used to moderate (slow

down, reduce their energy) fast (energies of 1 MeV and above) neutrons for shielding, and can also be exploited for fast neutron detection. For example, a neutron can be detected through a recoiling charged nucleus resulting from an elastic scatter event by means of light (scintillation) or electrical charge (ionization energy loss).

2.1.2 Inelastic Scattering

The total kinetic energy of the neutron-nucleus system changes during inelastic scattering. One important example is the $A(n,n')A$ reaction in which the neutron, n, transfers energy to an atomic nucleus, A, which subsequently emits a gamma ray, γ:

$$n + A \rightarrow n' + A^* \rightarrow n' + A + \gamma$$

where A denotes a ground-state nucleus, A^* denotes an exited nucleus, and n' denotes an outgoing neutron (possibly different than the incoming neutron) with reduced kinetic energy. Inelastic scattering is much less likely to occur than elastic scattering. Inelastic scattering does not occur if the incoming neutron has less kinetic energy than the energy required to excite the nucleus. Inelastic scattering from a nucleus A is often denoted by $A(n,n')A$, $A(n,n')$, or simply (n,n').

2.1.3 Neutron Capture Reactions

Atomic nuclei can capture or absorb neutrons, possibly resulting in an unstable nucleus. One possibility is production of a gamma ray through radiative decay. If the resulting gamma ray has sufficiently high energy, it can be easily separated from background. Background refers to gamma rays from other processes that are not useful for the detection scheme. An important example is thermal neutron capture by nitrogen:

$$^1n + {}^{14}N \rightarrow {}^{15}N^* \rightarrow {}^{15}N + \gamma\,(10.8\ \text{MeV})$$

The $^{15}N^*$ decays by emitting a 10.8-MeV gamma ray with a 44% probability. [Journey, 1997] The thermal neutron analysis (TNA) technique discussed in Section 5.1.1 exploits this reaction to detect nitrogen, which is also denoted as $^{14}N(n,\gamma)$.

After capturing a neutron, the nucleus may emit both “prompt” (short time scales) and/or “delayed” radiation as it decays to its ground state. The radiation produced by the decay includes not only gamma rays, but also protons, neutrons, electrons, positrons, and alpha particles as shown in Figure 1.

2.2 Photonuclear Interactions with Matter

In addition to neutrons, nuclear techniques for detecting explosives also include those that use high-energy (order MeV) photons to interrogate luggage, vehicles or cargo containers. Nuclear Resonance Fluorescence (NRF) is analogous to atomic fluorescence where photons excite electrons in an atom, except that in this case, the photons are used to excite the nucleus. Each isotope has unique nuclear energy levels and sometimes photons emitted as excited nuclei decay to their ground state at energies characteristic of the isotope in the material being studied. These characteristic emission lines are narrow (10’s of eV’s), which allows NRF photons to be separated from background. [Kniessl, 1996; Jovanovic, 2018]. Cross sections for NRF can be as high as 100s of barns for photons that are near nuclear resonance energies. [Bertozzi, 2005]

Nuclear Resonance Absorption (NRA) is like NRF, except that the excited nucleus does not decay by emitting a photon, but by absorbing the energy. As discussed in Section 5.2.1, an important

example is the $^{14}N(\gamma,p)^{13}C$ reaction involving the resonant absorption of 9.17-MeV photons. If a sample is irradiated with photons near the resonant energy, the presence of ^{14}N can be inferred by a deficit of transmitted photons. [Grodzins, 1992]

3 Neutron and High-Energy Photon Sources

The various nuclear techniques for explosives detection all require means of generating either neutron or high-energy (MeV) photons in the proper energy ranges to interrogate the cargo or luggage. We explore such energy sources here.

Neutron Sources

This section describes neutron sources relevant to explosives detection in order of increasing complexity. For more on neutron sources see [Mank, 2011; Garnett, 2018].

3.1.1 Radioisotopes

Some radioactive materials can be used as neutron sources. These can be compact and do not require a power supply. However, unlike generators or other accelerators of neutrons, radioactive sources that produce neutrons cannot be turned off and thus must be shielded when not in use.

3.1.1.1 Spontaneous fission sources

One of the most common neutron sources is ^{252}Cf, which undergoes spontaneous fission decay:

$$^{252}Cf \rightarrow 2 \text{ fission products} + 3\text{–}4 \text{ neutrons} + \text{gamma rays}$$

with a 3% probability. [Martin, 2000] On average, each resulting neutron has an energy of 2.3 MeV. [Meadows, 1967] A 5-cm long ×1-cm diameter ^{252}Cf source can emit more than 10^{11} neutrons isotopically per second, but it must be replaced periodically since ^{252}Cf has a half-life of 2.6 years.

3.1.1.2 Alpha neutron sources

Neutron sources can also be made by embedding a radioactive material that emits ^4He (alpha or α) particles in some low-Z materials. A common example is an "Am–Be" neutron source consisting of a mixture of radioactive ^{241}Am (432-year half-life) and ^9Be powders. The ^{241}Am decays by α emission into ^{237}Np, then the α interacts with the nearby ^9Be:

$$^9Be + {}^4He \rightarrow {}^{12}C + {}^1n + \gamma$$

to produce neutrons with average energy of about 4 MeV. In this manner, about 2×10^6 neutrons per second can be produced by 1 Curie of ^{241}Am. [Geiger, 1964]

3.1.2 Neutron Generators

Accelerator-based neutron sources or neutron generators are the most common sources of neutrons for explosives detection applications. Neutron generators are neutron source devices that contain compact linear particle accelerators and produce neutrons by fusing isotopes of hydrogen

together. These sources operate by accelerating deuterium ions and directing them onto targets containing deuterium, ^{2}H, or tritium, ^{3}H. Neutrons are produced through the fusion reactions:

$$^{2}H + {}^{2}H \rightarrow {}^{3}He\ (0.82\ MeV) + {}^{1}n\ (2.5\ MeV)$$

and

$$^{2}H + {}^{3}H \rightarrow {}^{4}He\ (3.5\ MeV) + {}^{1}n\ (14.1\ MeV),$$

respectively. These reactions are also referred to as DD and DT reactions, respectively. Neutron generators have also been constructed that accelerate tritium ions and direct them onto targets containing tritium also referred to as the TT reaction:

$$^{3}H + {}^{3}H \rightarrow {}^{4}He + 2\ {}^{1}n\ (0\text{-}9\ MeV),$$

which gives a wider spectrum of energies than the DD or DT reactions. [Reijonen, 2005]

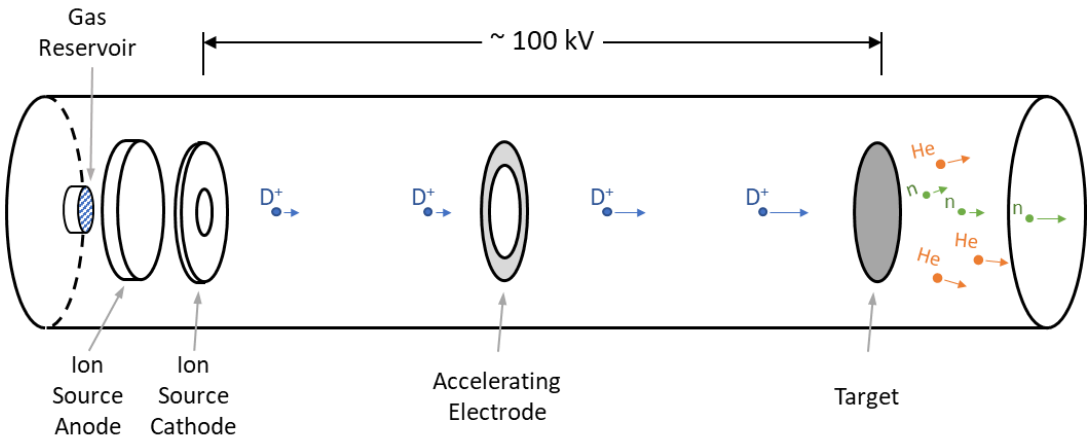


Figure 4: Schematic of a sealed-tube neutron generator. In this example, a source creates, $^{2}H^{+}$ ions, shown as D^{+} which are then accelerated to a target containing deuterium or tritium.

Neutron generators are based on mature technology dating from the 1950s, and several commercial vendors currently produce neutron generators, most with yields in the range of 10^{8}–10^{11} *n*/s. [Hawkins, 1960; Chichester, 2007] A diagram of a sealed-tube neutron generator is shown in Figure 4. Traditionally, compact sealed-tube neutron generators use Penning sources for deuterium ions, but more efficient RF-based plasma sources have been developed [Wu, 2009]. In compact sources, deuterons are accelerated electrostatically by biasing the target relative to the source at ~100 kV, resulting in a nearly isotropic fast neutron distribution. [Adams, 2015]

3.1.3 Accelerator-based sources

Some applications require neutron sources with higher intensities and/or neutron energies than neutron radioisotopes and generators can provide. For example, a 300-μA deuteron source with a 6-MeV accelerator was developed to provide fast neutrons via the DD reaction for pulse fast neutron analysis presented in Section 5.1.3. [Brown, 1994; Brown, 1994a] Another neutron-generating approach accelerates deuterons to a ^{9}Be-rich target and the reaction:

$$^{2}\mathrm{D} + {}^{9}\mathrm{Be} \rightarrow {}^{10}\mathrm{B} + {}^{1}n \ ,$$

which is also denoted $^{9}\mathrm{Be}(d,n)^{10}\mathrm{B}$. [Khan, 1994; Micklich, 1996]

Despite their advantages, neutron sources employing accelerators are generally larger, more complex, and more expensive than neutron generators.

3.2 Photon Sources

This section describes photon sources for detecting explosives with nuclear techniques. As discussed in Section 2.2, the nuclear resonance fluorescence (NRF) and nuclear resonance absorption (NRA) approaches require photons with energies very near the nuclear resonance energies of materials of interest. Two approaches are wide-band bremsstrahlung and tunable quasi-monochromatic x-ray sources. For more on photon sources see Chao, 2011; Garnett, 2018.

3.2.1 Wide-band bremsstrahlung photon sources

Since different nuclei have different resonant energies, a source that generates a broad spectrum of energies is advantageous. Electron accelerators with bremsstrahlung targets are the most used sources of photons for NRF and NRA because they produce photons over a continuous range up to the full electron energy. However, since NRF resonances are relatively narrow (10's of eV's), bremsstrahlung fluxes of ~10^{8} photons/s are required to achieve acceptable signal levels within a reasonable amount of time. [Bertozzi, 2007] The source must also be well-collimated to reduce backgrounds caused by irradiating materials outside the regions of interest.

3.2.2 Narrow-band laser Compton scatter photon sources

Over the past decade, there has been considerable interest in the development of tunable, narrow-band (quasi-monochromatic) photon sources for nuclear resonance measurements. [Albert, 2010; Hayakawa, 2009; Kikuzawa, 2009] A main advantage of these sources over bremsstrahlung sources is that objects in the inspection volume are exposed to a much smaller radiation dose since only photons near the energies of interest are used.

Quasi-monochromatic photons are obtained by scattering a laser beam from a relativistic electron beam. The source can be made compact by employing a laser Wakefield accelerator (LWA) for the electron beam. [Geddes, 2015] Restricting the angular divergence of the back-scattered photons by adding a collimator has the added benefit of narrowing the range of photon energies.

4 Radiation Detectors

Nuclear techniques utilizing neutrons or photons need to detect various forms of radiation, including recoiling nuclei, alpha particles, protons, neutrons, electrons, and photons. The following sections describe the two most common types of detectors: ionization detectors and scintillation detectors. Further information can be found in texts such as [Knoll, 2010] and in chapters of texts such as [Pozzi, 2018].

4.1 Ionization Detectors

Ionization detectors work by sensing the electrons and ions produced when charged particles interact with matter. Since neutrons and photons lack electric charge, ionization detection first requires the production of charged particles. For example, neutrons can be detected through ionization caused by either recoiling nuclei moving through material following a scattering event, or ionization due to decay products following a capture event. Similarly, photons can be detected through ionization produced by electrons and/or positrons resulting from photoelectric absorption, Compton scattering, or pair production.

Solids, liquids, and gases can all be used as ionization media. The choice of material affects performance factors such as ionization yield, response time, and resistance to radiation damage, sometimes referred to as hardness. Regardless of which material is used, ionization is generally detected by sensing electrodes. An electric field is generated by two or more electrodes held at different potentials within or across the medium; then drifting charges induce measurable voltage changes on the electrodes. Electronic amplification is often required to raise the signal above noise. An example of an x-ray detector made with a solid ionization medium is shown in Figure 5.

High-purity germanium (HPGe) detectors are commonly used for applications where high-resolution x- and gamma-ray spectroscopic measurements are needed, such as nuclear resonance florescence (NRF, see section 5.2.2). Germanium is a useful detection medium since it enables creation of large (centimeters-thick) sensitive volumes in which photons can deposit their full energy, i.e., little to no escape of secondary radiation outside of the volume. Additionally, techniques have been developed to remove impurities that trap electron-hole pairs and degrade sensitivity. One disadvantage of HPGe detectors is that they need to be cooled, typically to liquid nitrogen temperatures, so that noise due to thermal generation of electron-hole pairs is reduced to an acceptable level. Another disadvantage is they are expensive.

Due to its large thermal neutron capture cross section (~5000 b, see Figure 2), ^{3}He has been commonly used in proportional gas counters. In this case, ionization is caused by the charged products (tritium and hydrogen ions) of the reaction:

$$^{1}n + {}^{3}\mathrm{He} \rightarrow {}^{3}\mathrm{T} + {}^{1}\mathrm{H}$$

Fast neutrons are detected by adding moderating material around the gas volume so that this process has an acceptable cross section. Since ^{3}He has become an increasingly scarce resource, recent attention has focused on alternative neutron detection methods that rely on neutron interactions with boron or lithium. [Polichar, 2007; Ryzhikov, 2018]

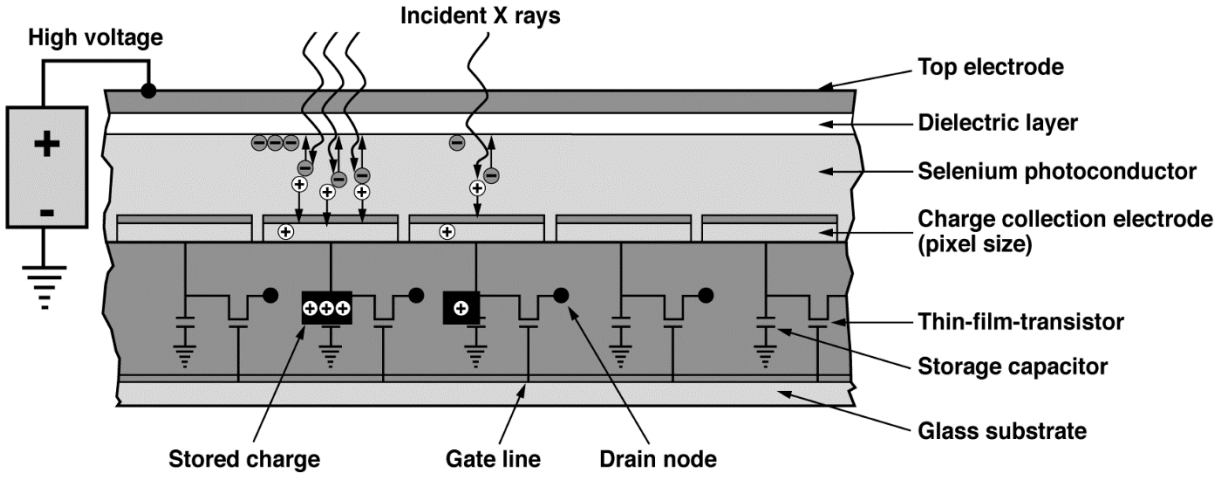


Figure 5. Ionization detector using a solid Selenium ionization medium coupled to readout electronics. From Martz, 2016.

4.2 Scintillation Detectors

In scintillation detectors, radiation is detected by the light it produces as energy is deposited in the scintillating medium. For example, as discussed in as given in Section 5, a commonly used inorganic scintillator is sodium iodide doped with thallium, NaI(Tl). Ionizing radiation impinging on the NaI(Tl) generates electrons that can interact with the crystal lattice and other electrons, creating free carriers in the conduction band that can migrate and become trapped at Tl sites or other defects. Tl sites and other defects trap electron-hole pairs that can recombine and release energy through fluorescence which is captured by a photo detector. [Yanagida, 2018; Getkin, 2017]

A wide variety of organic and inorganic scintillators are in common use, and the optimal material choice depends on the application. The most important scintillator properties include light yield, response time, wavelength of light produced, and resistance to radiation damage. For example, organic scintillators are often used for neutron detection since they contain large amounts of hydrogen, which has a large neutron cross section as shown in Figure 2, making these detectors efficient. Unlike inorganic scintillators, light production in organic scintillators originates from molecular vibrations. Organic scintillators have fast (<1 ns) response times. [Harvey, 1979] Some scintillators have fast enough response time that it is possible to distinguish between photons and neutrons based on the temporal profile of the signals they produce. This technique, known as pulse shape discrimination (PSD), relies on the fact that scintillation light produced by ions recoiling from neutrons takes place over a longer time scale than photon interactions in the scintillator. Using a single detector in conjunction with PSD instead of separate photon and neutron detectors can reduce system complexity and cost.

To make the light produced by scintillation detectors useful for explosives detection applications, it must be converted to an electronic signal. This is usually accomplished by photomultiplier tubes (PMTs), silicon photomultipliers (SiPMs), or, if light levels are high enough, photodiodes.

5 Nuclear Interrogation Techniques for Explosives Detection

In this section, we discuss how researchers and manufacturers integrate nuclear interactions with materials, sources and detectors to interrogate luggage, vehicles or cargo containers for explosives. To detect explosives, nuclear techniques rely on identifying one or more signatures that most explosive materials possess and that differentiate them from benign, i.e., non-explosive, materials. For conventional (i.e., military, and commercial) explosives, the most common differentiating signatures are high-density, high-nitrogen content and, to a lesser extent, characteristic oxygen-to-carbon ratios. [Buffler, 2004; Hussein, 1992] The high density of conventional explosives is mainly what x-ray methods look for [Martz, 2016], while the large amount of nitrogen in conventional explosives is mainly what some neutron-based detection systems look for [Buffler, 2010]. Unfortunately, not all explosives, e.g., homemade explosives, [Menning, 2008; Schubert, 2008, Rettinger, 2019] exhibit high-density and/or high nitrogen content, so novel techniques are required to detect them.

While the physics of neutron and photon interactions with materials are known (see Section 2), the practical implementation of them for explosives detection is complex. Sources and detectors are imperfect and noisy. [Martz, 2016] Source fluxes are low and long times are required to obtain high signal-to-noise data. Detected responses are affected by random quantum effects that will affect performance (probabilities of detection and false alarms as well as throughput). Added to these are difficulties related to the cost, size, weight, and regulatory and safety requirements to generate, shield, and detect neutrons and high-energy x-rays. In addition to the items or cargo being inspected, nuclear interrogation techniques expose operators, bystanders, and the local environment to ionizing radiation. Besides the health and safety concerns for nearby workers, another drawback is the possibility of radiation damage to the luggage or cargo contents and/or making it radioactive, referred to as activation. Still, promising methods have been, and continue to be, introduced, and tested.

This section describes how and why nuclear interrogation techniques have evolved and continue to evolve. We draw on scientific publications, industry reports, private communications, and reviews of photon and neutron techniques such as those published by Runkle et al. [Runkle, 2009], Lehnert et al. [Lehnert, 2010], and Avtonomov et al. [Avtonomov, 2015]. Since the physics is fixed, newer techniques are often refinements or extensions of older techniques for nondestructive evaluation/characterization. As stated by Harmon et al. [Harmon, 2011], "…what has brought innovations in the past 30 years or so is new detectors, sources, algorithms, and computational resources."

A summary of nuclear interrogation techniques is given in Table 1, listed in order of neutron (thermal, fast, thermal/fast combined) and photon techniques, as presented in the next sections. We mainly focus on techniques used to inspect luggage, palletized cargo, cargo containers, or vehicles. We do not discuss techniques developed for use for mail, bottles/liquid containers, people, or land mines.

Table 1. Summary of nuclear interrogation techniques for explosive detection in luggage or cargo containers

Technique	Advantages	Disadvantages	Comments
Thermal Neutron Analysis (TNA)	Detects N, H.	Neutron moderation required. Does not detect C or O. Not able to detect reliably the required aviation security explosives masses. Poor penetration due to high attenuation by organic materials.	Extensive funding by FAA*. Under a contract awarded to SAIC in 1985, the FAA purchased six TNA machines. TNA did not meet the detection requirement.
Fast Neutron Analysis (FNA)	Detects N, C, O.	High neutron background; low-resolution images.	Study funded by FAA*; FNA did not meet the detection requirement.
Pulsed Fast Neutron Analysis (PFNA)	Detects C, N, O, Al, Cl, Fe, Si, P and other elements (produces 3D images).	Large, complex, and expensive system.	Extensive funding by FAA*; PFNA did not meet the detection requirement.
Pulsed Fast Neutron Transmission Spectroscopy (PFNTS)	Sensitive to C, N, O.	Composition of explosives diluted over cargo. Large attenuation in organic cargos. Large and expensive.	Funded by FAA*; PFNTS did not meet the detection requirement.
Associated Particle Imaging (API)	Detects C, N, O. Compact, depth sensitivity.	Low signal rates, short tube lifetime.	Implemented in EURITRACK*** with limited success.
Pulsed Fast/Thermal Neutron Analysis (PFTNA)	Detects H, C, N, O, Cl, Si, P (low fast neutron background for TNA signals).	Low-resolution image; low detection performance. High fast neutron background for FNA signals.	—
Nuclear Resonance Absorption (NRA)	Detects N.	Complexity, large size, large weight, radiation shielding.	Funded by FAA*.
Nuclear Resonance Fluorescence (NRF)	Sensitive to the elemental composition of cargo.	Complexity, large size, large weight, radiation shielding, low signal levels.	Funded by DNDO** for cargo inspection; did not meet detection requirement.
Dual-energy / dual-mode radiography	Potentially more sensitive to density and atomic composition than dual-energy x-ray radiography.	Requires large accelerators or neutron generators. Susceptible to clutter due to material superposition, not enough specificity for explosives.	—

* After 9/11 the security part of the Federal Aviation Administration (FAA) moved to the U.S. Department of Homeland Security (DHS) Transportation Security Administration (TSA) and Science and Technology (S&T) Directorate.

** The Domestic Nuclear Detection Office was reorganized into the Countering Weapons of Mass Destruction (CWMD) office.

*** EURopean Illicit TRAfficing Countermeasures Kit

5.1 Neutron Techniques

There are several different types of neutron interrogation techniques, each with its unique advantages and disadvantages. The various neutron techniques are summarized in Table 1 and described in the sections below.

5.1.1 Thermal Neutron Analysis (TNA)

One of the first nuclear technologies developed by Plessey Nucleonics, Ltd. [Plessey, 1958] for explosives detection was thermal neutron analysis (TNA) using thermal neutrons to detect nitrogen in explosives. This TNA implementation is also one of the most thoroughly investigated nuclear explosives-detection technologies. [Gozani, 1986; Chung, 1993]

With TNA, a thermal (slow at ~2.2 km/s, low energy at ~0.025 eV) neutron striking a nucleus is absorbed with a certain probability and this process is followed by the emission of gamma rays with characteristic energies of the nucleus involved. Although, nitrogen has a low thermal neutron capture cross section, it results in the emission of a 10.8 MeV gamma ray, which is the highest energy of any element, and therefore it sits on a low background. Sometimes a 2.22 MeV gamma ray from hydrogen is also used in detection, but it is of limited use because hydrogen is very common in most materials.

The neutrons are produced either by an electronic neutron generator or a ^{252}Cf radioisotope. A moderator is used to slow down (thermalize) the neutrons before entering the cargo, with some thermalization done by the cargo itself—less so for luggage. An array of energy-resolving gamma-ray detectors, e.g., sodium-iodide, NaI(Tl) [Shea, 1990] are then used to measure the gamma-ray energy spectrum and determine the amount of nitrogen and sometimes hydrogen in cargo or luggage. TNA techniques generate two- (2D) and three-dimensional (3D) images. The 3D nitrogen image is reconstructed from the net nitrogen counts measured in the detectors distributed around the object. All detector counts are combined in a matrix inversion and knowledge of the detectors spatial response to provide a best estimate of the nitrogen density in each volume element (voxel). [Shea, 1990] The 2D or 3D nitrogen image is used to infer if an explosive is present.

Initially, spontaneous fission neutrons from a ^{252}Cf source were the most proposed source [Bartko, 1992; Gozani, 1986] and were employed in a preproduction TNA prototype. Another prototype used a 150 μg 80-mCi ^{252}Cf source, which weighed 28,000 lb, required a 41-m^2 area, cost $1.4 million for fabrication, and had an estimated $705 thousand annual operation cost in 1990 dollars. [Jones, 1990]

Other TNA approaches used antimony-beryllium, radium-beryllium, and polonium-beryllium sources that used an (α,n) reaction (see Section 3.1.1.2) to produce broad-spectrum neutrons that were moderated to thermal energies for the detection of nitrogen. [Gravitte, 1964] These sources proved to have too small a neutron yield for practical development, and attention turned to accelerator sources. Other approaches to implementing TNA systems [Lee, 1992; Hurwitz, 1992] have proposed use of neutrons from ^{9}Be(d,n)^{3}He sources and the accelerator-based deuterium-deuterium sources, represented by neutrons produced by the ^{2}H(d,n)^{3}He reaction. The advantages of accelerator sources over the isotopic ^{252}Cf source for a TNA system relate to a higher neutron yield and the ability to turn off neutron production when it is not needed.

Under a contract awarded to SAIC in 1985, the Federal Aviation Administration (FAA)[1]purchased six TNA systems to detect plastic explosives in luggage. The six TNA

[1] The Aviation and Transportation Security Act (ATSA) enacted into law shortly after the September 11, 2001 terrorist attacks, established TSA and gave the agency responsibility for securing all modes of transportation, including the nation's civil aviation system, which includes air carrier operations (domestic and foreign) to, from, and within the United States. Therefore, the security part of the FAA was moved to the U.S. Department of Homeland Security (DHS) Transportation Security Administration (TSA) and Science and Technology (S&T) Directorate.

machines needed to be combined with an x-ray unit and were called XENIS (X-ray Enhanced Neutron Interrogation System). Of the six systems, four were installed: one each at JFK, Dulles, Miami, and Gatwick airports. The results of these deployments were summarized in a President's Commission on Aviation Security and Terrorism; their final (classified) report was produced on May 15, 1990. The Commission's TNA systems findings and recommendations were as follows:

- 1990 Commission Findings
- TNA technology was not yet refined enough to accurately detect explosive of sizes relevant to aviation security.
- The machines were not fully automated.
- The XENIS machine is massive, weighing close to 14 tons, and a footprint for the TNA alone is about 12 m^2. An additional equivalent area would be needed to add an x-ray system and baggage diverter. TNA weighed 28,000 lbs., required a 41-m^2 footprint, and cost $1.4 M with about $700k operational cost/year (in 1990 dollars).
- For threat masses of concern the false-alarm rates are too high.
- 1990 Commission Recommendations
- The program to require U.S. airlines to purchase and deploy ~150 existing TNA machines should be deferred.
- The FAA should create an R&D program to detect small amounts of plastic explosives.

To this day, a drawback of this method is the high incidence of false alarms, which stems from the inability of this technique to differentiate between the nitrogen in explosives and the nitrogen in benign materials, such as wool or salami. Another drawback is that the 3D image has a resolution lower than required to detect aviation threats. In addition, other important elements that make up explosives, such as oxygen and carbon, are not readily detected with thermal neutron analysis due to their much smaller thermal capture cross sections and with gamma rays that sit on a large background.

A National Research Council (NRC) report in 2002 [NRC, 2002; NRC, 2002a] recommended the following: "Any further development of neutron-based or accelerator-based explosives-detection technologies should concentrate on methods that can also provide quantitative metrics on the presence of other elements, in addition to nitrogen, that are in typical checked bags. The improved detection potential due to quantitative information on the presence of other elements such as carbon, oxygen, and hydrogen in typical bags, is required to offset the weight, volume, and operating-cost disadvantages of neutron-based or accelerator-based approaches." Such recommendations led to the development of practical fast neutron techniques discussed next. TNA has also been used for detection of mines [Cousins, 1998] and large car bombs [Bendahan, 1998] with some success.

A system under development by Bubble Technology Industries funded by CWMD is called MSMAIN and looks for nitrogen gamma-ray signatures from nitrogen-bearing explosives between interrogating pulses of 14.1-MeV neutrons. [Koslowsky, 2020]

5.1.2 Fast Neutron Analysis (FNA)

The fast neutron analysis (FNA) technology was an outgrowth of attempts to obtain more information than nitrogen content from thermal neutron interrogation techniques. In FNA, high-energy neutrons, rather than low-energy thermal neutrons, are used to interrogate, e.g., a bag or cargo container. Gamma ray detectors that surround the container measure gamma rays from neutrons inelastically scattered off oxygen (6.13-, 6.92-, 7.12-MeV gammas from ^{16}O), carbon (4.43-MeV gammas from ^{12}C), or nitrogen (1.64-, 2.31-, 5.11-, MeV gammas from ^{14}N) atomic

nuclei in the bag or container. High-energy neutrons are required to excite these inelastic reactions. In principle, the ratio of these characteristic gamma rays for these elements can be used to help sort conventional explosives from common non-threat cargo items [Gozani, 1994; Buffler, 2004; Whetstone, 2014] see Figure 9 in Section 5.1.4.

A monoenergetic neutron source simplifies the analysis of the inelastic gamma signatures. Most FNA implementations use a DT, $^{3}H(d,n)^{4}He$, reaction to produce nearly monoenergetic ~14-MeV neutrons. The DT reaction is one of the easiest reactions (low incident charged particle energy, typically approximately 150 keV; high cross section) to produce monoenergetic neutrons. One issue with the DT neutron source is that tritium is radioactive, and it comes with safety concerns; however, this issue is mitigated because the tubes are sealed.

The nitrogen inelastic scattered gammas are very weak and hard to detect above background. With low-energy-resolution gamma detectors (such as NaI(Tl)), the 5.11-MeV gamma coalesces with escape peaks from the ^{16}O inelastic lines. Various detector options can be employed to improve the detection of the nitrogen gamma signature, e.g., the use of high-energy-resolution, high-purity-germanium detectors (HPGe) or anti-Compton shields with NaI(Tl) detectors. These detector options raise other considerations, such as detector time resolution and detector cost.

Significant weight, volume, and operating costs are commonly associated with the installation and operation of an accelerator to produce neutrons in an airport environment. However, small, inexpensive sealed-tube neutron DT kHz pulsed accelerators are available [Bach, 1993]. Sealed-tube neutron generators produce a relatively low neutron yield (less than 10^{10} neutrons per second) [Rhodes, 1992; Bach, 1993; Verbeke, 2000] and short tube lifetime (typically less than 2,000 hr at high-flux operation). Some sealed-tube neutron generators also exhibit stability problems during operation. [Khan, 1994]

Even though the accelerator portion of a DT FNA system is small, the 14-MeV neutron shielding requirements are very stressing [Gozani, 1993] and can make insignificant any savings in the total system weight and volume. The 14-MeV neutrons are emitted in a nearly isotropic angular distribution and take much more shielding (on a per neutron basis) than do the fission spectrum neutrons (~2 MeV) from a ^{252}Cf source. Some neutron production reactions (e.g., $^{9}Be(d,n)^{10}B$ as used for PFNTS, see Section 3.1.3) produce a forward-peaked neutron emission that is much more efficiently collimated and focused on the target region of a bag. [Khan, 1994]

FNA can lead to increased detection performance compared to the thermal neutron analysis technique due to the detection of carbon and oxygen in addition to nitrogen. FNA systems only produce a 2D view with no depth profile, i.e., the elemental signatures are produced along the neutron path length, mixing benign and explosives, and diluting the explosives signatures. These systems usually have poor spatial resolution and consequently a low fidelity image. The spatial resolution is related to the size of the collimated incident neutron beam and the spread of the neutron beam while traversing the thickness of the container. [Hall, 1994] The 2D image data produced by this system is a critical limitation, and several approaches have been suggested to address it as discussed in the next few sections.

5.1.3 Pulsed Fast Neutron Analysis (PFNA)

One of the refinements of FNA involves the use of a pulsed neutron source; the resulting technique is called pulsed fast neutron analysis (PFNA). [Gozani, 1995; NRC, 2002a; Gozani, 2007] PFNA is an improvement over the FNA technique. PFNA utilizes a nano-second pulsed

beam of fast neutrons to interrogate the object of interest [Gozani, 2007]. A collimated beam of neutrons passes through cargo and characteristic gamma rays are emitted following inelastic scattering from the cargo. The gamma-rays are detected by an array of energy- and time-resolving detectors, which provide information about the gamma rays and the depth where they were produced, respectively. Since the neutron source is pulsed, some of the gamma backgrounds associated with FNA can be excluded based on signal arrival time at the detector. The energy of the characteristic gamma rays and the depth information provide a 3D image of the elemental content of cargo from which explosives can be discriminated from benign cargo materials. A schematic of a representative PFNA system is given in Figure 6. This 3D elemental image of carbon, nitrogen and oxygen provides an advantage over FNA.

There are many options for the neutron accelerators used for PFNA. One representative approach is the use of a 300-μA ^{2}H- injector with a 6-MeV deuteron accelerator as a DD source for neutrons. [Brown, 1994; Brown, 1994a] All of the PFNA approaches require a very tightly bunched neutron pulse, usually a beam with a 1–2 nanosecond full width at half maximum (FWHM). [Schultz, 1991; Sawa, 1993; Khan, 1994] This nanosecond pulse, when coupled with the neutron time-of-flight, determines the minimum spatial resolution along the direction of the neutrons. For a 6-MeV neutron and a 1-nanosecond neutron pulse, the spatial resolution along the neutron beam direction is limited to about 5 cm.

The typical PFNA DD accelerator pulse repetition rate is 5–15 MHz. These DD accelerators are large and heavy. A pulsed tandem cascade accelerator is typically about 1.5 m in diameter and 8 m long. A deuteron Pelletron Van der Graaf unit weighs about 4500 kg. This system's size and weight are some of the principal issues preventing airport integration of this technology. Approaches have been suggested to reduce the size and weight of the required accelerators [Fazio, 2019] and to ease the airport integration issues. PFNA approaches have been proposed to detect explosives, drugs, and other hazardous materials. [Pentaleri, 1994; Gozani, 2007; Strellis, 2009]

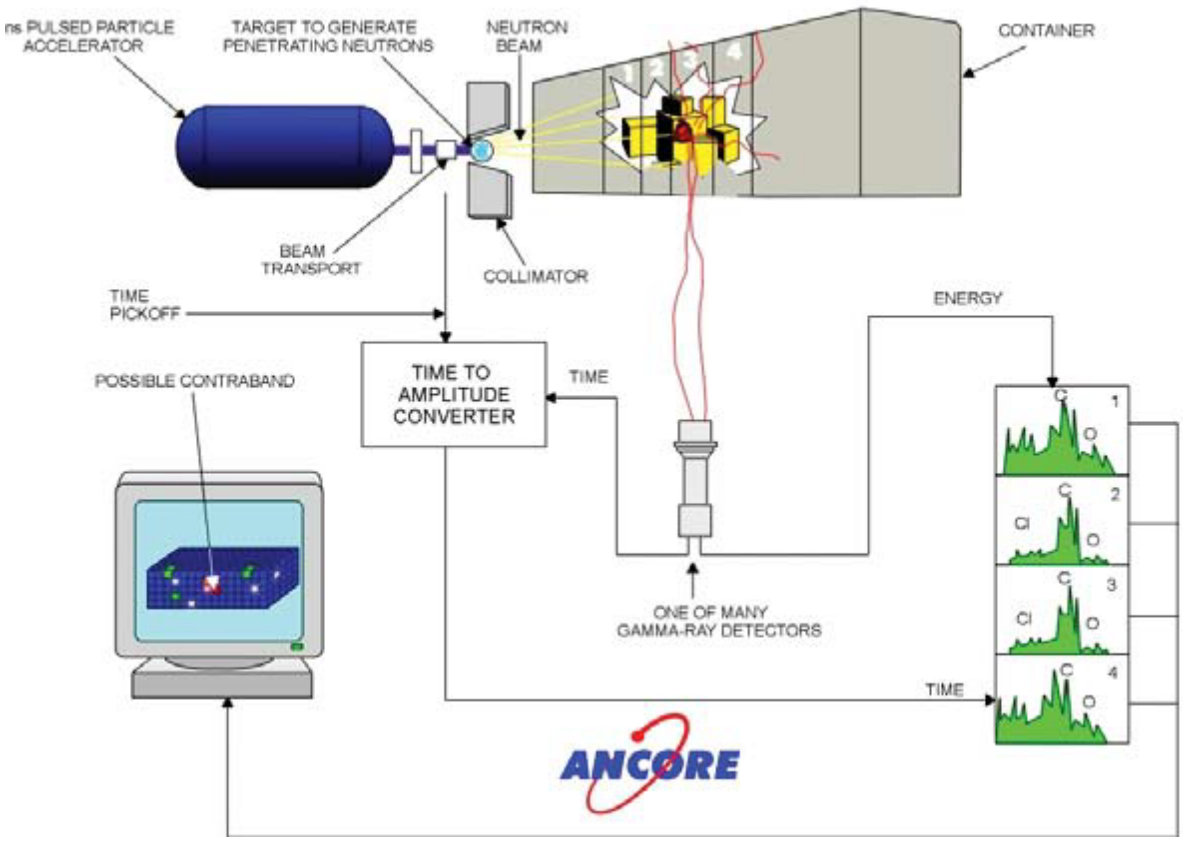


Figure 6. Schematic depicting the principles of PFNA used to produce an elemental image of the inspected object. [Strellis, 2009]

A PFNA system was tested on air cargo inspection at the George Bush Intercontinental Airport in Houston, Texas, USA. [Strellis, 2009] The system utilized a tandem Van de Graaff accelerator operating at 3.5 MHz that produced pulses of deuterons with a FWHM of ~1ns. The deuterons impinged on a deuterium gas target separated from the vacuum by thin havar alloy foil. Neutron pulses are created by the $^{2}H(d,n)^{3}He$ reaction at an energy of around 8.5 MeV with a deuteron beam intensity of up to 140 micro-A. A neutron collimator near the deuteron gas target produced a neutron beam spot 9-cm wide by (typically) 12-cm tall at the center of the container. This neutron beam oscillated vertically by moving a collimator. Translational motion of the air cargo was provided by a constant-velocity conveyor system. The former allowed for spatial resolution in height (y-direction). The latter allowed spatial resolution in the container motion direction (x-direction). The neutron/gamma ray time-of-flight (TOF) allowed for spatial resolution in the neutron beam direction (z-direction).

The inspection volume was surrounded by an array of NaI(Tl) detectors. These detectors were used to collect gamma rays from the neutron inelastic scattering reactions occurring within the system volume. Using the time-of-flight technique to determine where the neutron inelastic scattering reactions occurred, the data acquisition and image reconstruction system produced a 3D image of the cargo contents. The images have a typical volume element size of 6.3-cm wide x 6.3-cm high x 8-cm deep.

Organic plastic scintillator detectors were used in the radiography detector array. These detectors were 2.54-cm diameter and 15.25-cm long. Placed in line with the beam on the far side of the

inspection tunnel, they detected neutrons and gamma rays that emanated from the neutron source. The gamma rays were produced by nuclear reactions of the accelerated deuteron with the foil separating the vacuum from the deuterium gas target. Since the PFNA beam oscillates vertically while the cargo is translated, the detector array configuration was designed to produce optimal radiographic images. Since the neutrons and gamma rays travel at 4.5 and 30 cm/ns, respectively, they formed two groups separated by about 100 ns in time by the time they reached the radiography detectors. This separation allowed the radiography detectors to form distinct neutron and gamma radiographic images of the cargo. These images were used to improve the determination of elemental densities in the PFNA image reconstruction and provided course radiographic gamma-neutron images. [Rynes, 1999]

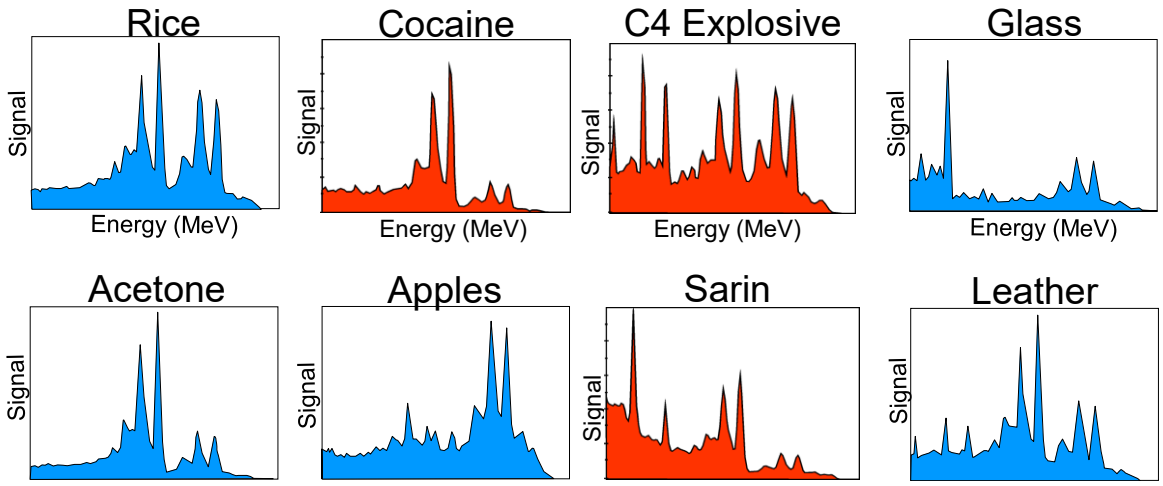


Figure 7. Spectra of gamma rays resulting from inelastic scattering of neutrons from selected materials. Blue spectra are from benign materials and red are from contraband materials. [Bendahan, 1999]

Figure 7 shows example inelastic gamma-ray spectra of C4 and other materials. C4 shows prominent peaks of N, C, and O, in quite different proportions compared to other materials. The spectra were analyzed to extract the quantity of the element and displayed in images. [Bendahan, 1999]. Transmission and 3D elemental images of a car are shown in Figure 8. For example, the carbon image highlights five rubber tires, oil, gas, and explosives. Similarly, iron shows the shell of the car.

The elemental images of a container are utilized by the decision algorithm to determine voxels that have elemental compositions like the threat material. If a threat is detected, the algorithm displays the threat type to the user as a 3D rendered image as shown in Figure 8 top-right. The images were produced by displaying only voxels that exceeded a specified explosive signal threshold.

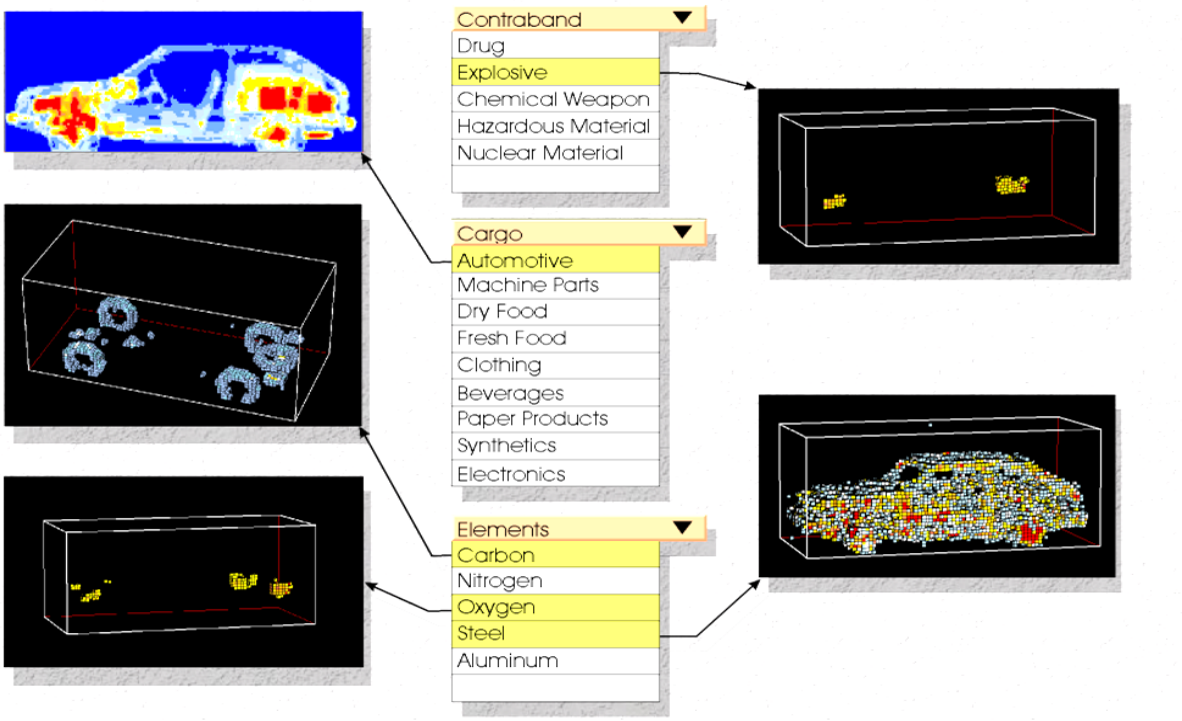


Figure 8. The various images of a car are shown. Transmission image is shown on top-left; carbon, oxygen, and steel images are shown as indicated. Image of explosives as determined by decision algorithm is also shown. [Brown, 1998]

The drawback of this method is that creating a collimated beam of pulsed fast neutrons in a cost-effective way, with a small footprint, and ability to scan cargo at high throughput is not currently feasible. Like FNA, less sensitivity is expected on the opposite side of organic cargo due to fast neutron attenuation by hydrogen in the cargo. This effect is mitigated by rotating the cargo; however, this method still has difficulties penetrating to the center of dense hydrogenous, e.g., organic, cargos.

This technology has been studied extensively by the Government Accounting Office (GAO) [GAO, 1999], the NRC [NRC, 1999; NRC, 2002]; the second NRC report is an unclassified summary of a more complete Official-Use-Only report. [NRC, 2002a] These reports found that, while PFNA can discriminate conventional (commercial and military) explosives from non-explosive materials and drugs hidden in cargo, it also has several practical limitations. These limitations include its large size and weight, the need for radiation shielding, difficulty in penetrating hydrogenous materials, and regulatory and safety issues associated with nuclear-based technologies. It is also useful to point out that PFNA, as well as several other nuclear techniques, have not been tested for their usefulness in detection of homemade explosives. An update on aviation cargo security is given in a 2011 GAO testimony. [GAO, 2011]

A compact version of PFNA has been proposed [Bendahan, 2013], which could potentially mitigate some of these issues. The system is based on a pulsed DT neutron generator that does not require accelerating the deuterons to high energy, avoiding a large accelerator, which is the main contributor to the size, weight, and system complexity. Future implementation could use lasers to induce high-energy DD reactions and produce subnano-second neutron pulses in a compact and

more affordable way, see, for example, Hilscher et al., [Hilscher, 2001] Chichester et al., [Chichester, 2007] and Bolton.;[Bolton, 2018]

5.1.4 Pulsed Fast Neutron Transmission Spectroscopy (PFNTS)

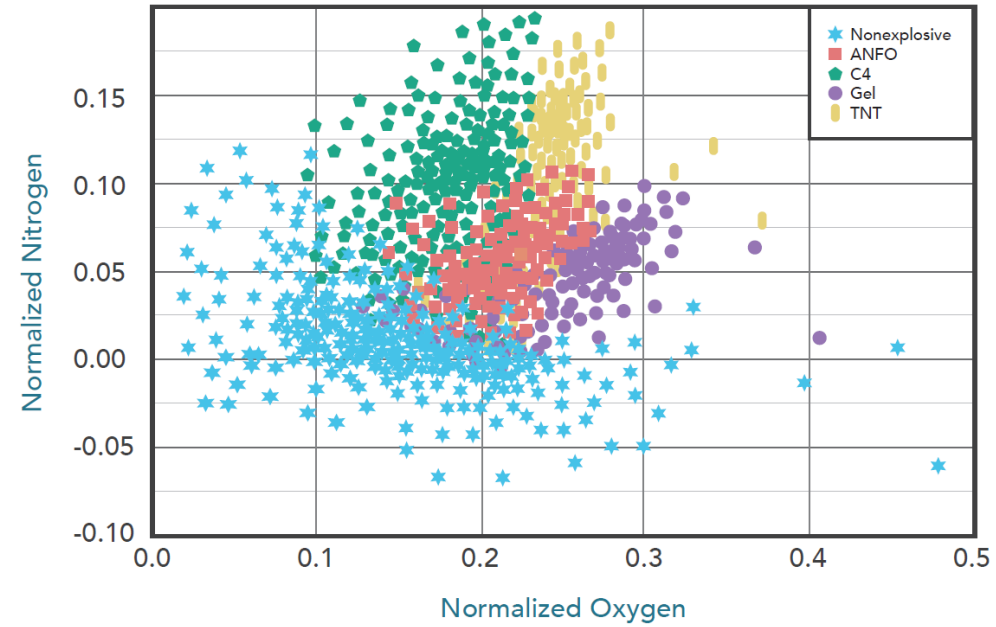
Pulsed Fast Neutron Transmission Spectroscopy (PFNTS) is an imaging method that exploits the characteristic neutron cross-section structure (resonances) of different isotopes in the neutron energy-range of 1-10 MeV. Recently, this method was referred to as Neutron Resonance Transmission Analysis (NRTA) [Schillebeeckx, 2014] and Fast Neutron Resonance Radiography (FNRR) [Mor, 2014].

The inspected cargo is irradiated with 1-10 MeV neutrons of a broad spectral distribution and the energy-dependent neutron transmission is measured. By comparing the energy-dependent attenuation to the incident neutron spectrum, the ratios of oxygen, carbon, and nitrogen for volume elements (or voxels) in the bag or container can be estimated. [Overley, 1985] The hydrogen cross section has no resonances, but it can be inferred from the "missing" attenuation. If the cargo contains elements that exhibit sharp cross-section resonances, the transmitted neutron energy spectrum will exhibit dips and peaks at specific energies corresponding to these resonances. Thus, the transmission spectrum carries information about the elemental composition of the cargo, from which the presence of explosives can be inferred. The energy spectrum is measured using neutron TOF spectroscopy, requiring pulsed neutron beams and imaging systems with capability for TOF measurements.

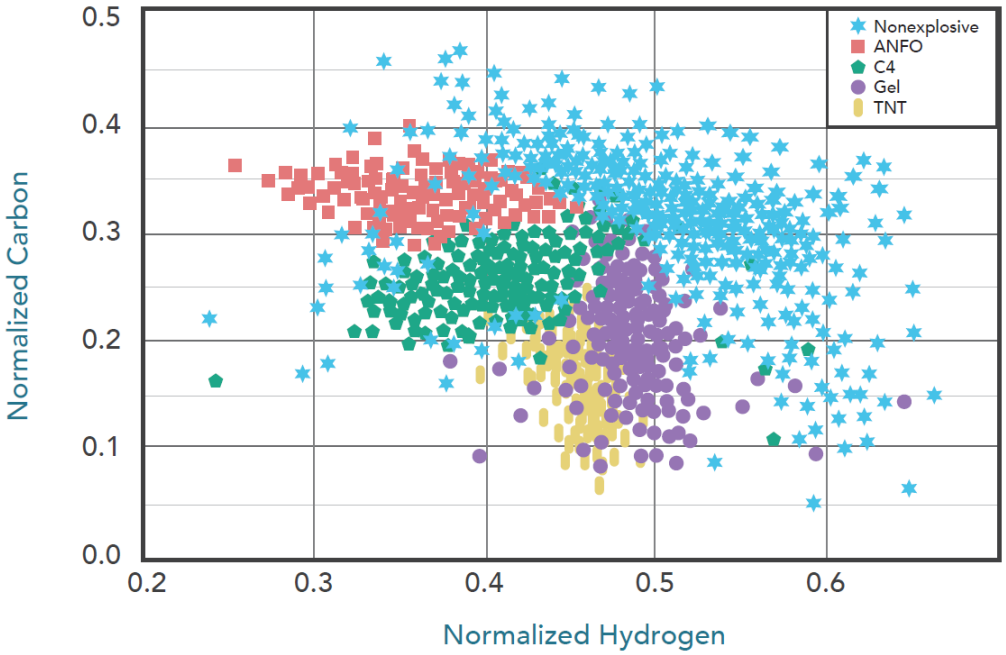
Since the PFNTS technique examines energy-dependent neutron attenuation, it requires employment of a broad-energy, tightly-bunched, pulsed neutron source. This rules out $^{2}H(d,n)^{3}He$ and $^{2}H(t,n)^{4}He$ sources whose energy spectra are too restricted. To produce a reasonable neutron flux for high-energy neutrons (up to 8 or 10 MeV), accelerators are generally required. The $^{9}Be(d,n)^{10}B$ or $^{9}Be(p,n)^{9}B$ reactions are prime candidate neutron sources. [Micklich, 1996] Most researchers in laboratory-based PFNTS experiments have utilized a deuteron accelerator and the $^{9}Be(d,n)^{10}B$ reaction with a time-averaged current of 1–2 μA. [Lefevre, 1998] The accelerators required to exploit these neutron-producing reactions need a high current (~50 μA time-averaged current, nanosecond pulse width, and millisecond pulse repetition interval) and a fairly high-energy deuteron source (> 4 MeV).

Time-of-flight is used to determine the energy of the transmitted neutrons. The width of the initial neutron pulse and the time resolution of the time-of-flight measurement limit the energy resolution of the transmitted spectrum. Flight paths of 4–10 m are commonly used. The narrow peaks in the interaction cross sections will not be seen in a typical PFNTS measurement due to the limited energy resolution of the detectors [Miller, 1992]. Thus, element identification in this approach depends on the broad energy-dependent structures in the cross sections, not on the narrow resonances.

Scatter plots for four of the five elemental dimensions can be used to distinguish the presence of explosive and possibly identify the type of explosive as shown in Figure 9. [Chmelik, 1997; Lefevre, 1998] The points in Figure 9 represent 38,000 measurements from actual airline suitcases, with and without explosives. The scatter in the plotted points for explosives and non-explosives in Figure 9(a) indicates why path-integrated nitrogen-only detection schemes have a very difficult task. The overlap of the distribution of non-explosive data (stars) and explosive data in Figure 9 show how difficult it is to eliminate the possibility of a false alarm without impacting the probability of detecting explosives.



(a) Nitrogen/oxygen distribution



(b) Carbon/hydrogen distribution

Figure 9. PFNTS elemental distributions for explosives and non-explosives in air passenger bags. The four explosives shown are nitrogen-based explosives. ANFO is ammonium nitrate and fuel oil, C4 is composition 4, a plastic RDX(Royal Demolition eXplosive, aka cyclonite or hexogen), Gel is an explosive gel, and TNT is trinitrotoluene. No homemade explosives are shown since they were not common at the time. Adapted from NRC, 1999.

In some applications of the PFNTS method, a five-dimensional representation of the elemental composition and the spatial distributions of "potentially explosive" adjacent pixels are used in the

detection algorithm. Algorithms can be used to reduce this five-dimensional elemental information into an "explosive potential" metric for a single pixel. Variations in the detection algorithm can increase the basis set beyond these five elements. For example, the Tensor algorithm [Tensor, 1998] considers another element, Y, which is changed from element to element within a specified set of cross sections during a regression calculation until a best fit is obtained. Different spatial correlation algorithms can be used to further reduce the map of explosive potential metrics into a yes or no decision on the presence of an explosive.

The University of Oregon team [Overley, 1994] detection algorithm uses an "explosive potential" metric based on comparison with the explosives/non-explosives probability seen in a simulation database. Separate comparisons are maintained for each explosive class that the algorithm is designed to detect. The Tensor work uses a neural network trained on a set of explosives and non-explosives bags.

PFNTS was evaluated by the National Research Council [NAP, 1999][2] for detection of explosives in luggage, which arrived at the following conclusions:

- Some PFNTS tests demonstrated detection levels consistent with the FAA's (now TSA) EDS (Explosive Detection System) baggage scanners certification standards, but deficiencies were revealed.
- PFNTS did not demonstrate an ability to detect Class A explosives, an important class of explosives that most alternative technologies also have problems detecting.
- PFNTS, when used with a 2D area neutron detector, had a higher false-alarm rate than the FAA's EDS certification criteria allow.
- In the area of Class B explosives, the system did not show a significant performance advantage over x-ray CT-based EDSs.
- Due to difficulties in deployment and integration of a PFNTS-based device, including size, weight, and safety issues, airlines were advised in 1999 to choose currently available x-ray CT-based EDSs.

These issues are exacerbated for more attenuating high-density cargo because the production of the 2D elemental image along the neutron path dilutes the elemental composition of explosives by benign cargo reducing detection. Also, the high attenuation of organic cargos prevents neutron penetration for such cargo. Even if the detection performance was reasonable, the complexity of generating nano-second-pulsed neutrons results in a large and expensive system not compatible with most inspection facilities.

Recent advances are given in Mor et al. [Mor, 2009; Mor, 2015] where they describe development of a high-spatial-resolution fast-neutron imaging system with multiple-energy TOF-spectrometer.

5.1.5 Associated Particle Imaging

The associated particle imaging (API) [Ussery, 1988] technology is a refinement of the sealed-tube DT neutron source FNA approach. API is also referred to as fast neutron associated particle (FNAP, associated-particle sealed-tube neutron generator (APSTNG) [Rhodes, 1992], and tagged neutrons [Carasco, 2008; Bishnoi, 2014; Batyaev, 2015; Bishnoi, 2020]. In API, the detection of

[2] Note that the NRC report was completed in 1999, before homemade explosives became a method used by terrorists. It is not clear how well this technique would perform with homemade explosives. Their findings still apply today.

the associated alpha particle recoil DT, reaction $^{3}H(d,n)^{4}He$, neutron source is used to specify the neutron time of emission and direction. . The reaction product consists of a 14-MeV neutron and a 3.5-MeV alpha 'associated' particle, which travels in the opposite direction of the fast neutron due to conservation of momentum. Neutron DT generators with internal alpha particle detectors are commercially available. One advantage of API is the system's small source volume. Initial sources had a mean-time-to-failure at a neutron flux of 10^{7} neutrons per second of about 200 hr. [Rhodes, 1993; Wu, 2009] Newer sources can run at 10^{8} neutrons per second for ~2000 hours [Adelphi].

In API, the alpha particle is detected by a position-sensitive alpha detector. The detected location of the alpha particle can be combined with simple kinematic considerations to determine the direction of the emitted neutron. The timing between the alpha detection and any subsequent gamma from neutron inelastic scattering can be combined with the geometry, the speed of the neutron, and the speed of the photons, to estimate the depth in the bag at which the inelastic reaction took place. Thus, API can be used to produce a 3D image.

Because the neutron direction is known, the API approach does not require the use of collimators to focus the incident beam, and there is no need to pulse the source. However, since the neutrons are emitted in an essentially isotropic distribution, many neutrons fail to impact the target bag or container, and neutron shielding is needed in all directions to surround the source and bag or container regions. In addition, the scattering of the neutrons in the shielded material, along with the resulting inelastic and capture gammas produces a significant time-uncorrelated background that may interfere with the detection of the prompt gammas from the inelastic events produced in the target material.

The time resolution of the alpha- and gamma-correlated detection is limited to about 1 nanosecond. This results in a spatial depth resolution for the inelastic reaction of about 5 cm. The edge-smearing from the deuteron spot size and neutron scattering within the bag similarly limit the resolution in the x and y directions. Thus, this approach typically has used a detection voxel of ~5 cm × 5 cm × 5 cm at best.

In alpha particle detection, dead time considerations limit the count rate and hence limit the neutron flux that can be used with this approach to about 10^{6}–10^{8} neutrons per second. Thus, large scan times are required for this approach. New alpha detectors and faster gamma-ray detectors are expected to improve the spatial resolution and reduce the dead-time limitations.

Signal-to-noise considerations make most neutron-based explosives-detection approaches very difficult to implement. The neutron beam may be collimated into a small cone beam to reduce background and needs to be scanned (by moving the generator or the cargo), which results in a slow throughput. The signal statistics are poor due to the high background produced by random coincidences. [Harmon, 2011] Penetration depth can be limited due to high attenuation of organic (hydrogenous) cargos, which can be mitigated by scanning both sides with loss of throughput. Applications of API are limited by its disadvantages such as low neutron flux, issues of tube lifetime, and the intrinsic poor spatial resolution.

5.1.6 Combined Thermal and Fast Neutron Techniques

Pulsed Fast/Thermal Neutron Analysis (PFTNA) combines aspects of PFNA and TNA. [Whetstone, 2014] One technique uses 14.1-MeV neutrons in ~10 µs pulses that are separated by ~90 µs. [Vourvopoulos, 2001] While the source is emitting, a time-gated detector system records

the energies of prompt gamma rays that result from inelastic scattering of 14.1-MeV neutrons off of nuclei such as carbon and oxygen. Between pulses, the neutrons thermalize and are captured, resulting in gamma rays that are distinguished by their delayed arrival times. The time-gated detector system is then activated to record the gamma rays resulting from neutron capture, which gives an estimate of the relative amounts of hydrogen, nitrogen, chlorine, and iron present. Finally, after several pulse cycles, the source is turned off, and the detector is used to record gamma-rays emitted from decaying activated nuclei with half-lives longer than 100 μs, such as silicon and phosphorous. Analysis of gamma-ray emissions collected at all three-time scales—prompt (from fast neutrons), short (from thermal neutrons), and long (activation) yields estimates of several atomic ratios, which are then used to infer the presence or absence of explosives. Two-dimensional "color" imaging of atomic species is possible with PFTNA by employing a segmented detector array in conjunction with a coded aperture. [Evans, 1999] This technique is employed in a variety of applications such as bulk coal analysis, contraband detection, and detection of explosives.

Another combined fast/thermal neutron technique arose in response to the difficulty in using FNA to measure nitrogen content. Variants of the FNA approach use a microsecond or faster pulsed neutron source and a time-dependent detection of the neutron capture signatures from nitrogen. The neutrons that are thermalized in the bag or cargo are used to measure nitrogen content with the same capture reaction used in TNA. In a typical airport passenger-checked bag, only a small percentage (typically ~1 percent) of the fast (14.1-MeV) neutrons will be thermalized, and the neutron thermalization time in a hydrocarbon, such as plastic, is about 0.2 ms.

One such modified FNA approach is referred to as pulsed interrogation neutron and gamma (PING). [Schultz, 1991]. In this approach, an accelerator is used to produce 14.1-MeV neutrons from the deuterium-tritium reaction. The accelerator is pulsed at about 8 kHz with a 7-μs-wide deuteron pulse. In addition to nitrogen, sulfur (5.42-MeV gammas from ^{32}S) and chlorine (6.111-MeV gammas from ^{35}Cl) have been detected with PING using the thermal capture gamma signature. Chlorine detection may be important in the detection of some non-nitrogen-based explosives such as potassium chlorate mixtures [Rettinger, 2019] and in the detection of cocaine. Sulfur can be important in the detection of some chemical warfare agents.

Note that the 6.11-MeV ^{35}Cl thermal neutron capture gamma is only separated by 20 keV from the ^{16}O inelastic scattered gamma at 6.13 MeV. It would be very difficult to discriminate these gammas based on their energy. In the PING system [Schultz, 1991] a fast-pulsed 14.1-MeV neutron source is used and the ^{16}O and ^{35}Cl gamma signatures are differentiated by the gamma emission time. In a PING 14.1-MeV neutron pulse, the fast neutrons interact with the ^{16}O by inelastic scattering, and the neutron pulse is shut off after 7 μs, after which the neutrons down scatter in the material. Only then do the thermalized neutrons react with the ^{35}Cl. Thus, the ^{16}O and ^{35}Cl isotopes are discriminated based on the arrival time of the gammas in the detector. Other gammas with different energies from inelastic scattering in ^{35,37}Cl can also be used in FNA detection methods. [Sawa, 1992]

5.2 Photonuclear Techniques

Nuclear resonance techniques initiated by high-energy x-ray photons include Nuclear Resonance Absorption (NRA) and Nuclear Resonance Fluorescence (NRF), both of which exploit absorption and emission of high-energy photons by atomic nuclei at specific energies. Historically, NRA and NRF have been used to perform fundamental nuclear science. By 1960, NRA and NRF were

established techniques to identify new excited states in stable nuclei and measure their properties. [Metzger, 1959] NRA and NRF have been identified as potentially powerful tools to identify and characterize illicit material, such as nuclear material and conventional explosives. Explosives may be identified by the isotopic ratios of carbon, nitrogen, and oxygen in the scanned material. The NRA and NRF physical phenomena have been investigated thoroughly over many decades and have yielded measurements of properties for many isotopes as noted in [Booth, 1964] and [Runkle, 2009]. Since the mid-1980s, researchers have proposed variations of these techniques to detect both nuclear and explosive threats and have even performed several proof-of-concept demonstrations [Vartsky, 1994; Bertozzi, 2007; Caggiano, 2007; Pruet, 2007]. Reviews of photon and neutron techniques that discuss NRF and NRA in the context of inspecting air cargo for explosives can be found in [Runkle, 2009], [Lehnert, 2010] and [Avtonomov, 2015]. Most recently, Passport Systems, Inc., developed a cargo inspection system for CWMD's Nuclear and Radiological Imaging Platform (NRIP) program that aimed to use NRF for contraband detection [NRIP, 2019]. The types of contraband tested during NRIP included not only explosives, but also explosive precursors, drugs, ammunition, and currency. Passport's system did not do well at detecting these items.

5.2.1 Nuclear Resonance Absorption (NRA)

The NRA explosives-detection approach is quite different from other nuclear-based detection technologies. NRA uses a high-energy (9.17-MeV) photon to interrogate the nitrogen content in luggage or cargo container through the $^{14}N(\gamma,p)^{13}C$ reaction. Because this approach uses an incident gamma ray, it is sometimes referred to as the gamma resonance absorption (GRA). [Vartsky, 2004] NRA has been considered for the detection of explosives in luggage and cargo containers, and it has the high-energy photon penetration needed for container inspection systems.

Work on the application of NRA to explosives detection was done at Canada's TRIUMF accelerator at the National Laboratory for Particle and Nuclear Physics [Schmor, 1994], at Israel's Soreq Nuclear Research Center [Vartsky, 1994], and at the Los Alamos National Laboratory [Morgado, 1994a]. Soreq developed nitrogen resonance detectors for use with NRA in explosives detection. In addition, Soreq collaborated with Science Research Laboratory (SRL) and the Massachusetts Institute of Technology (MIT), to explore the role of NRA in drugs detection. [Flusberg, 1995; Goldberg, 1996] Others have also been involved in NRA development activities, for example [Grumman, 1995] and [Sredniawski, 1995]. The Grumman work emphasized the advances in the accelerator design (high target current, target cooling) required for the application of NRA to cargo. Between 1978 and 1996, the FAA spent about $12.1 million [GAO, 1996] on the development of an NRA system for explosives detection.

The NRA detection method, depicted in Figure 10 [Kuznetsov, 2009; Grodzins, 1992], is based on the large 121-eV-wide resonance [Vartsky, 1990] in the $^{14}N(\gamma,p)^{13}C$ reaction at an incident photon energy of 9.17 MeV. This resonance reaction has a peak resonance cross-section of ~200 mb and an energy-integrated resonance cross-section of ~4 b. Even at the resonance energy, the 9.17-MeV photons are very penetrating. The generation of the high-intensity incident 9.17 MeV photon beam is one of the principal challenges associated with this NRA approach. One approach is to use a filtered bremsstrahlung source, but this option has major signal-to-noise issues.

The approach used in all previous demonstrations of NRA for explosives detection is to use the $^{13}C(p,\gamma)^{14}N$ inverse reaction to produce the resonant energy photons. Due to the Doppler shift of the recoiling ^{14}N, only the gamma rays emitted in an 0.7-degree-cone beam at 80.7° are at the

resonance energy. The accelerator concepts proposed for this inverse reaction with a 1.745-MeV incident proton are a radio-frequency quadrupole (RFQ) pulsed linear accelerator (LINAC) or an electrostatic continuous wave (CW) accelerator. [Todd, 1992; Schmor, 1992; Grumman, 1995] A representative proposal for cargo explosives detection is for a very high-current 10-mA accelerator with a 1-cm beam spot, a 25-keV beam spread, a 600-μs pulse width, and a 10-Hz pulse repetition frequency [Grumman, 1995]. NRA prototypes have used a 0.5 mA beam current. The major challenges for the design of this accelerator are the proton heating of the target, the associated potential target degradation, and detector interference from gammas produced in the accelerator target materials (often copper). New developments in producing 9.17 MeV gamma rays are given in [Onischenko, 2004; Kuznetsov, 2009].

Bismuth germanate (BGO) scintillator detectors (temperature-stabilized and with a 15 percent average energy resolution) have been used in NRA detection systems [Morgado, 1994b], but interference from other proton-induced gamma rays provides a high background signal. To exploit the selective absorption of the 9.17-MeV photons in nitrogen, one would like a gamma detector that is particularly sensitive to the transmitted 9.17-MeV photons. Soreq [Vartsky, 1993] has developed a resonance ionization detector using a nitrogen-rich ^{14}N-based (31 percent by weight) liquid scintillator based on di-methyl-tetrazole that is resonance-sensitive (24 percent efficient for resonant photons) and is well suited for the explosives-detection application. Vartsky et al. analyzed and compared the response of these two detector-types to the resonant radiation. [Vartsky, 2004] Resonant detectors give rise to much higher nitrogen-contrast sensitivity in the radiographic image than their non-resonant detector counterparts and furthermore, do not require proton beams of high energy-resolution. By comparison, the non-resonant BGO detectors have higher γ-detection efficiency, but their contrast sensitivity is very sensitive to the quality of the accelerator beam.

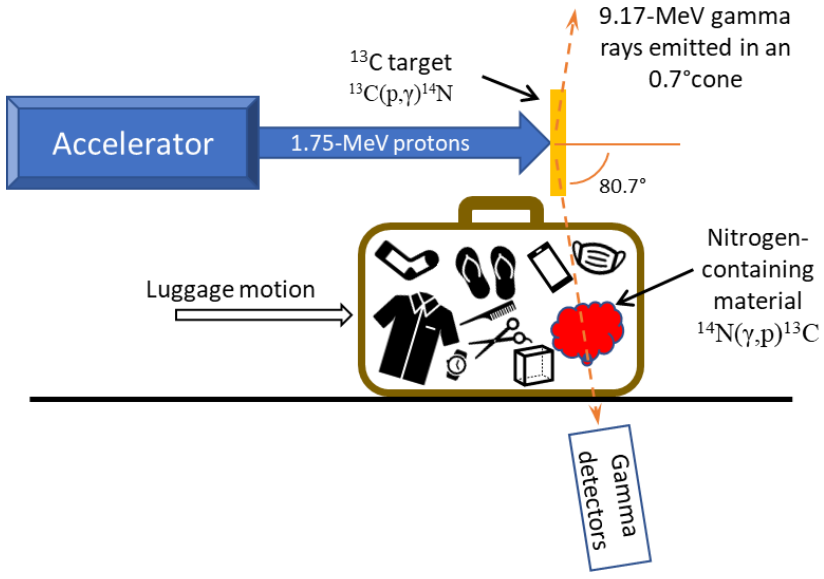


Figure 10. Schematic of the NRA detection method. Adapted from Grodzins, 1992 and Kuznetsov, 2009.

In addition to the 9.17-MeV photon transmission, the NRA approach needs a background gamma measurement to assist in compensating for the mass-dependent attenuation (non-resonant

absorption) of the photons in the cargo. Prototype NRA systems [Vartsky, 2004] have used a composite target containing barium fluoride to produce 6.13-, 6.9-, and 7.1-MeV gammas from the $^{19}F(p,\alpha\gamma)^{16}O$ in addition to 9.17-MeV gammas from ^{13}C for this purpose.

One serious disadvantage of the NRA method is its limitation to detecting just nitrogen. The work with TNA has suggested that a nitrogen-only detection method is not a good candidate for explosives detection at the TSA requirement levels (high probability of detection, low probability of false alarm, small explosive material quantity). Other resonance reactions on ^{12}C and ^{16}O have been suggested [Flusberg, 1995; Goldberg, 1996] but they have not been demonstrated to be feasible for explosives detection owing to the narrow width of the proposed resonances and the inability to exploit the inverse reaction for efficient production of the required resonance energy photons. Requirements for NRA to use a monoenergetic photon source and highly selective detector makes this a very complex system. Another disadvantage with NRA is the requirement for a very advanced high-current accelerator. This accelerator will have all the disadvantages (large size, large weight, radiation shielding) associated with the neutron-based accelerators.

5.2.2 Nuclear Resonance Fluorescence (NRF)

NRF can non-intrusively interrogate luggage, vehicles, or cargo containers and measure the isotopic content for any element with atomic number greater than that of helium. The NRF technique involves exposing material to a continuous energy distribution of photons and detecting the nuclear fluorescence photons unique to its isotopic composition. [Goldhaber, 1958; Mossbauer, 1958; Bertozzi, 2005; Bertozzi, 2007; Caggiano, 2007] Interrogating photons range from 2 to 8 MeV, a range that includes many nuclear resonances and happens to be high enough to penetrate cargo containers. Determination of the isotopic components of the material occupying luggage or cargo containers enables the identification of threats such as explosives, fissile materials, toxic materials, and weapons of mass destruction. For example, as illustrated in Figure 11, the NRF signatures of an explosive simulant, melamine, and water are readily distinguished from one another. [Bertozzi, 2007] In the late 2010s, Passport Systems, Inc., developed a Performance Test Unit (PTU) for the Nuclear and Radiological Imaging Platform (NRIP) program sponsored by the Countering Weapons of Mass Destruction (CWMD)[3] office of DHS. The Passport NRIP PTU was sited at the Port of Boston's Conley Container Terminal in Massachusetts. This enabled the PTU to scan stream-of-commerce (SOC) cargo containers. [NRIP, 2019]

The PTU was designed to perform both initial and prolonged inspections of vehicles and cargo conveyances using five measurement subsystems [Ledoux, 2015]. A 9-MeV high-flux electron beam could be steered to any one of 64 ports where a pencil-shaped beam of bremsstrahlung x-rays was generated. During initial inspection, backscatter imaging was accomplished by rapidly rastering the beam between x-ray ports while cargo passed underneath on a moving platform. Suspect regions of interest (ROIs) identified during initial inspection were then adjudicated during prolonged inspection, during which the NRF subsystem was used to find materials such as explosives, drugs, and currency. A diagram of the Passport NRIP NRF subsystem is shown in Figure 12.

[3] Formerly known as Domestic Nuclear Detection Office (DNDO).

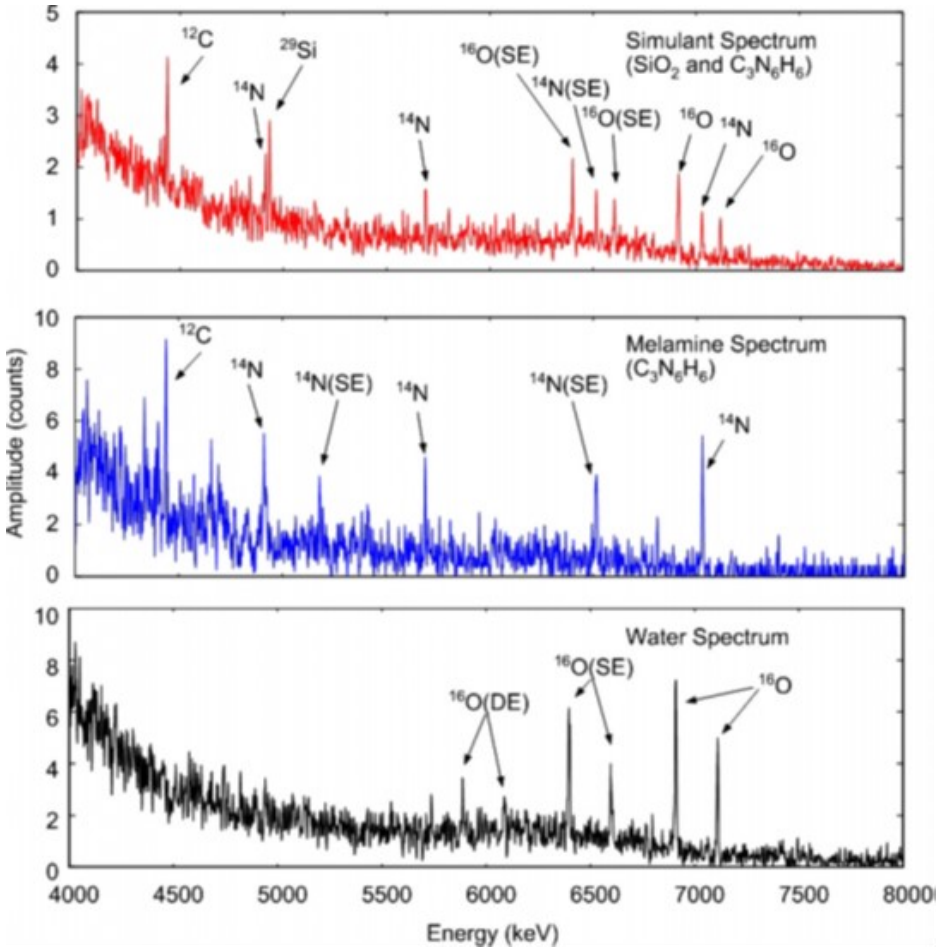


Figure 11. Nuclear resonance fluorescence spectral signatures of an explosive simulant (top), melamine (middle), and water (bottom) in air, i.e., no clutter. From Bertozzi, 2007.

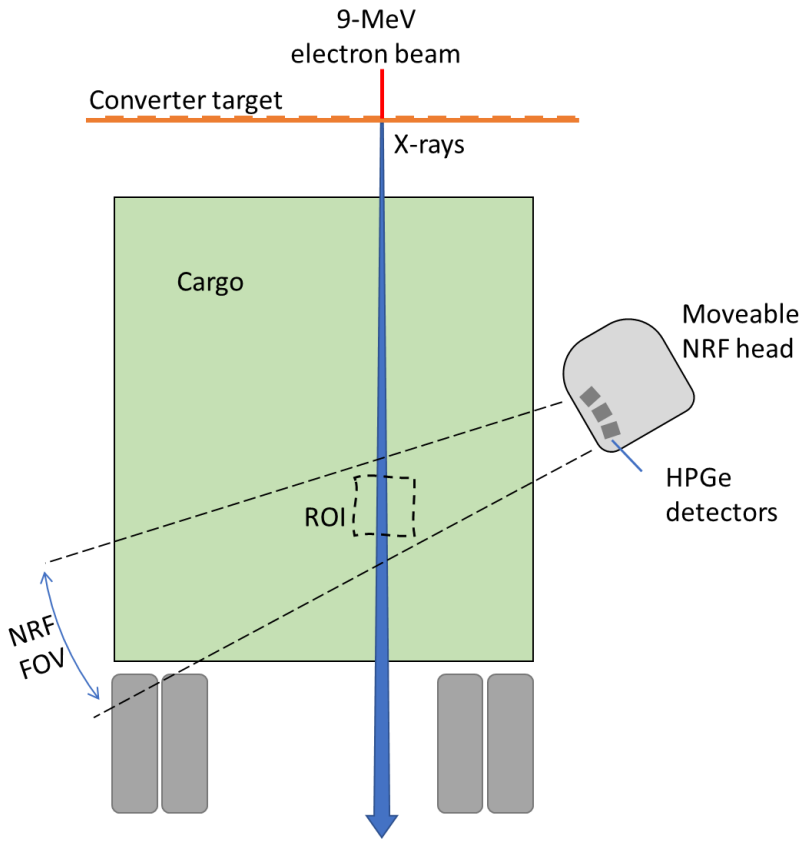


Figure 12. Diagram showing the components of the Passport NRIP PTU involved in NRF as viewed from the end of the inspection tunnel. A 9-MeVelectron beam was steered to one of 64 ports to give a pencil beam of x-rays. An array of HPGe detectors was positioned to accept NRF photons within the field of view (FOV) surrounding a region of interest (ROI) that was exposed to the x-ray beam.

The NRF detector subassembly consisted of an array of nine mechanically cooled HPGe detectors (~80% efficiency, see also section 4.1) that could be translated and tilted (typically 30 degrees below horizontal) to best receive NRF photons emitted from each ROI. Configurable collimators in the detector array were used to limit the field of view and reduce background from areas above and below the ROI that were intersected by the x-ray beam. Materials (elemental composition) within a ROI were identified by comparing the measured NRF signature to a collection of signatures gathered during system calibration.

Although the NRF technique is potentially both selective and sensitive for detecting explosives, there are several obstacles that need to be overcome before it is practical. The high-energy photon source and required shielding mean NRF-based systems will have a large system footprint, high complexity, and high cost. Since nuclear resonances are narrow, large doses of wide-band x-rays are needed to detect signals above background, which may not only be harmful to the objects under inspection but may take a long time (hours) to accumulate unless a very high flux source is used. This limits the NRF approach to secondary inspection, where a faster but less specific system first identifies suspect areas. If the primary system has a high false-alarm rate, this approach may not be suitable for explosive detection operations. In addition, a high-power x-ray source and energy-resolving detectors are expensive and complex to operate.

5.3 Radiography performed with nuclear reactions

There are several implementations of radiography performed with particles produced by nuclear reactions. One of the first cargo inspection systems that fused neutron and gamma-ray radiography data was based on high-energy DD neutrons and gamma rays as part of the PFNA system (see section 5.1.3). [Rynes, 1999] The gamma rays were produced by nuclear reactions of the accelerated deuteron with the foil separating the vacuum from the deuterium gas target. Since the PFNA beam oscillated vertically while the cargo is translated, the detector array configuration was designed to produce optimal radiographic images. In this case, the main detection system was PFNA and the radiography provided information for the image reconstruction and for the decision algorithm. This would be a very expensive method to produce neutrons and gamma rays for radiography.

More recently, a dual-mode radiography system for scanning air cargo was developed to use a beam of fast neutrons in addition to high-energy, dual-energy x-rays [Sowerby, 2009]. For compactness and high-flux, a DT neutron generator is used to produce 14.1-MeV neutrons. A product for scanning air-cargo containers based on this approach has been developed by CSIRO and Nuctech under the name AC6000XN. However, while material discrimination could be enhanced, spatial resolution and contrast issues may make the detection of explosives in cluttered cargos difficult.

New algorithms are becoming available for more accurately determining bulk density and effective atomic number of cargo contents, which can benefit threat identification. However, clutter and the existence of materials with similar characteristics as some explosives prevent these methods from achieving high detection performance.

6 Summary

A wide range of neutron and high-energy (>1 MeV) photon nuclear interactions have been exploited to interrogate luggage, vehicles, and cargo containers for explosives. These nuclear techniques, summarized in Table 1, have been shown to detect explosives with varying degrees of performance, where performance includes detection probability, false-alarm rate, throughput, weight, size, and cost. Some of the neutron-based techniques that mainly detect nitrogen result in high false alarm rates since nitrogen is found in many benign materials. Techniques such as PFNA and API that detect multiple elements, e.g., carbon, nitrogen, and oxygen, and provide 3D images have been shown to have better performance, but challenges need to be overcome before such systems are competitive with x-ray techniques. Of the two photonuclear techniques discussed here, NRF is the most promising, yet it currently has shortcomings with respect to cost (both capital and operational) and detection performance that need to be overcome before it is ready for deployment. Nuclear techniques are currently not feasible—due to poor detection, high numbers of false alarms, large size, weight, costs, and shielding requirements—but there is potential for detecting conventional (commercial and military) explosives with relevant threat masses in cargo and luggage, even in high-density cargo.

7 Future Possibilities

Future opportunities include advances in higher-flux compact neutron generating sources, more efficient detector technologies, and advanced automated detection algorithms. Potential new sources include laser-based technologies to generate sub-nanosecond pulsed neutrons in a compact form factor. [Bolton, 2018] Emerging detectors incorporating new scintillators such as CLYK—Cs_2LiYCl_6 are efficient and can detect and discriminate between both gammas and neutrons based on their pulse shapes. [Pozzi, 2018] Advances in machine learning and deep learning methods may advance automated threat detection of explosives. [Hossny, 2020] There remain, however, issues related to penetration of hydrogenous cargo, large footprint required for shielding, safety, and the possibility of radiation damage or activation of the cargo contents and equipment by neutrons. However, it remains to be seen if the nuclear techniques can detect homemade explosives with low false alarms. This needs to be explored further.

8 Acknowledgements

We want to thank Jimmie Oxley for asking us to write this chapter. We appreciate all the technical edits by Joseph Bendahan, and other edits by Steve Azevedo, Carl Crawford, and JC Ross. We thank Karen Roland for helping obtain all the figure permissions.

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