

## *Chapter 2*

# **Nuclear technologies**

**P. J. Griffin**

**Applied Nuclear Technologies Department, Sandia National Laboratories<sup>\*</sup>,  
P. O. Box 5800, Albuquerque, NM, 87185, USA**

X-ray-based material interrogation methods typically use x-ray interactions with the electrons in a material to determine the material density and sometimes the average atomic number. Nuclear-based interrogation methods involve probing the nucleus of an atom rather than the electron cloud and open the possibility of using the isotope-specific nuclear cross sections, the probability of an interaction, to determine the elemental constituents in a material. Nuclear-based interrogation approaches include but are not restricted to an interrogation with neutrons as the probing radiation. An interaction with an atom in the test material is typically signaled to the detector as either a reduction in the transmission of the probing radiation or by the detection of a secondary radiation associated with the nuclear interaction. A number of nuclear-based interrogation approaches have been investigated with respect to their potential for explosive detection. Some nuclear-based explosive detection algorithms use a combination of detection algorithms to improve the accuracy of the material identification.

The following sections present a summary of the physics that underlies the nuclear detection technologies, a survey of neutron-based detection approaches, and an overview of non-neutron-based nuclear detection technologies.

## **1. PHYSICS UNDERLYING NUCLEAR DETECTION METHODS**

### **1.1. Detection Principles**

The following subsections present background material on the physics and nomenclature used to address the nuclear detection technologies. The subsections are divided into neutron- and non-neutron-based nuclear detection methods.

---

\*. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed-Martin Company, for the United States Department of Energy under contract DE-AC04-94AL85000.

### Neutron-based Detection

Neutrons are uncharged particles, so when they interact with materials they interact by way of nuclear interactions with the neutrons and protons in the nucleus of the atoms of the target material. For most target materials, the neutron cross section, or probability of interaction, is much smaller than that for a photon with the electron cloud surrounding an atom. Thus, the neutrons have a greater penetration range. Because of this ability to penetrate deep into dense materials, neutron interrogation is proposed for explosive detection in small items, such as passenger bags, as well as for large cargo containers. When neutrons interact with materials, they are sensitive to the structure of the nucleus. Thus, they probe not only to the elemental content of the target material, but also the isotopic mixture.

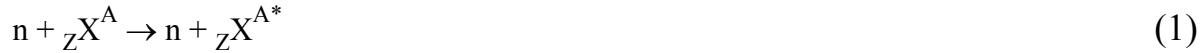
Figure 1 shows the energy dependence of the neutron cross section for representative materials/reactions. Some reactions are seen to exhibit a threshold energy, e.g.  $^{11}\text{B}(\text{n},\alpha)$ . Neutrons with an energy less than the threshold energy are not capable of inducing the specified reaction. Two of the curves in Fig. 1 [ $^{10}\text{B}$  vs.  $^{11}\text{B}(\text{n},\alpha)$  cross section] demonstrate the variability of the neutron cross section even within isotopes of a given element. Whereas photon cross sections are proportional to a power of the atomic number of the target atom<sup>†</sup>, the neutron cross sections are seen to vary between materials but not to have any clearly identifiable rules. Some rules for the magnitude of the cross section for specific reaction channels have been deduced, but these rules are energy-specific and are based upon information on the nuclear structure of the target atoms. The lowest energy threshold for a transmutation reaction in every element has a  $1/v$  energy dependence. There is typically a complex resonance structure in the epithermal energy range for  $(\text{n},\gamma)$  reactions, e.g.  $^{55}\text{Mn}(\text{n},\gamma)$ . Fig. 2 shows that the high energy elastic cross sections for some materials can also exhibit a resonance structure. The curves in Fig. 2 have been offset to avoid an overlap of resonance structures.

Neutrons interactions include scattering as well as transmutation reactions. The target atom is typically notated as  $_{\text{Z}}^{\text{X}}\text{A}$  where X is the symbol for the target element, Z is the atomic number (the number of electrons in the target atom) and A is the atomic weight (the sum of the neutrons and protons in the nucleus). The scattering may be elastic, where the incident neutron energy is transferred to the energy of the outgoing neutron and to the target atom in such a manner that both energy and momentum are conserved, or inelastic, where the outgoing target

---

<sup>†</sup>. For 100 keV photons, the photoelectric cross section is proportional to  $\sim Z^4$ , while Z is the atomic number of the atom. For 3 MeV photons, the photoelectric cross section is proportional to  $\sim Z^{4.6}$ . The Compton photon cross sections are proportional to Z.

atom is excited into a higher energy state. In an inelastic reaction, the excited target atom radiates the energy through emitted gammas and returns to the stable ground state or to a metastable energy state that may subsequently decay with a specified half-life. An inelastic scattering reaction is notated as:



where the “\*” in the outgoing residual atom indicates that the atom is in an excited state. In an elastic reaction, the conservation of energy and momentum imply that the average neutron energy after an elastic collision is given by:

$$\overline{E_{out}} = E_{in} \cdot \frac{1}{2} \cdot \left[ 1 + \frac{(A-1)^2}{(A+1)^2} \right] \quad (2)$$

Thus, for hydrogen with A=1, the incident neutron loses, on average, half of its energy. For a high atomic number target, such as lead ( ${}_{82}^{208} \text{Pb}$  and other isotopes of Pb), the incident neutron loses, on average, only  $\sim 1\%$  of its energy. So, whereas high atomic number materials attenuate photons, the neutrons loose very little energy through scattering by high atomic number materials. It is low hydrogenous materials that rapidly downscatter the neutron energy.

In a transmutation reaction, the incident neutron is absorbed into a compound nucleus that decays so that the residual nucleus is changed and the outgoing channel typically includes two particles. A transmutation reaction can be written as:



In this reaction, a neutron is incident on a target atom with the elemental symbol T (atomic number Z and atomic weight A). The outgoing channel has a light and a heavy particle. The light particle, indicated by an elemental symbol “r” is typically a proton,  ${}_1^1 \text{H}$ ; a deuteron,  ${}_1^2 \text{H}$ ; or an alpha particle,  ${}_2^4 \text{He}$ . The heavy residual atom, notated by the elemental symbol “R”, represents the target atom changed by whatever neutrons and/or protons that were transferred/picked-up from the incident neutron. This reaction is often notated as  ${}_Z^A T(n, {}_Z^{1+a} r) {}_{Z-z}^{A-a} R$  or in a shorthand notation as  $(n, {}_Z^{1+a} r)$ . Sometimes the incident neutron is absorbed. This reaction is called an  $(n,\gamma)$  reaction and is notated as  ${}_Z^A T(n,\gamma) {}_{Z+1}^{A+1} T$ .  $(n,p)$  and  $(n,\alpha)$  reactions, where  $\alpha$  represents an alpha particle,  ${}_2^4 \text{He}$ , are typically threshold

reactions. The  $(n,\gamma)$  reaction does not, typically, have a threshold energy. Thus, the lowest energy reactions is typically a  $(n,\gamma)$  reaction where the incident neutron is absorbed and the target atom is left in a metastable excited state that subsequently decays. The decay is typically accompanied by the emission of prompt gammas. Some elements, such as  $^{48}\text{Cd}^{\text{nat}}$  or  $^{64}\text{Gd}^{\text{nat}}$ , have a very large thermal neutron absorption  $(n,\gamma)$  cross section<sup>‡</sup>. These materials are often used for neutron shielding. For some target isotopes, e.g.  $^{10}\text{B}$  and  $^3\text{Li}^6$ , there is a very large thermal neutron  $(n,\alpha)$  cross section and it extends to higher neutron energies than do typical  $(n,\gamma)$  reactions. These materials (boron and lithium) play a critical role in neutron shielding. Since there are no materials that exhibit a large capture cross section at high neutron energies, high energy neutron shielding is typically accomplished by using an hydrogenous material to downscatter the neutron and boron, lithium, cadmium, or gadolinium to absorb the low energy downscattered neutrons.

Some reactions have a threshold energy, i.e. an energy below which the reaction can not occur due to energetic considerations. Because there can be an exchange of energy between the particle's kinetic energy and the rest mass energy of the atoms, the energy in the reaction outgoing channel can be greater or less than the kinetic energy in the entrance reaction channel. We notate the kinetic energy of a particle as  $K$  and use a subscript to indicate the particle naming convention introduced in Eq. 3. The difference between the kinetic energy of the particles in the exit channel and that in the entrance channel is called the reaction Q-value. For a case where the target atom,  $T$ , is at rest, and the incident neutron has an energy  $E_n$ , the Q-value for the  $(n, {}_z r^{1+a})$  reaction is given by:

$$Q_{(n, {}_z r^{1+a})} = K_R + K_r - K_n \quad (4)$$

### Non-neutron-based Detection

Non-neutron-based detection can be based on probing the atom with other particles, such as photons, protons, muons, or by electromagnetic interrogation. At high energies, typically greater than 8 MeV, an incident photon can include a photonuclear reaction where a neutron or proton is emitted from the target material. At higher energies, multiple nucleons (neutrons or protons) can be emitted. A compendium of photonuclear cross sections can be found in Ref. [1, 2]. Modeling of photonuclear processes is implemented in some of the recent radiation transport codes, such as MCNPX, PICA, and FLUKA. The incident photons

---

<sup>‡</sup>. The superscript “nat” indicates a material with a ratio of isotopes identical to the isotopic ratio for the naturally occurring element.

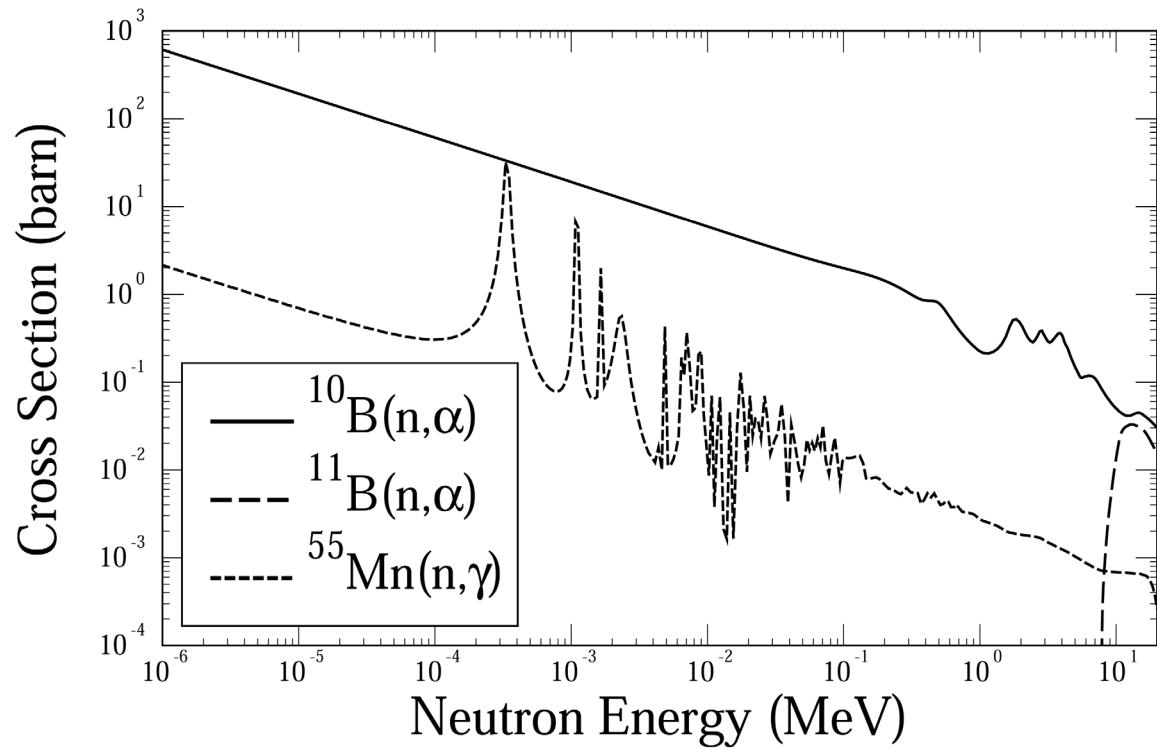


Figure 1: Variability of the Neutron Cross Sections for Different Isotopes

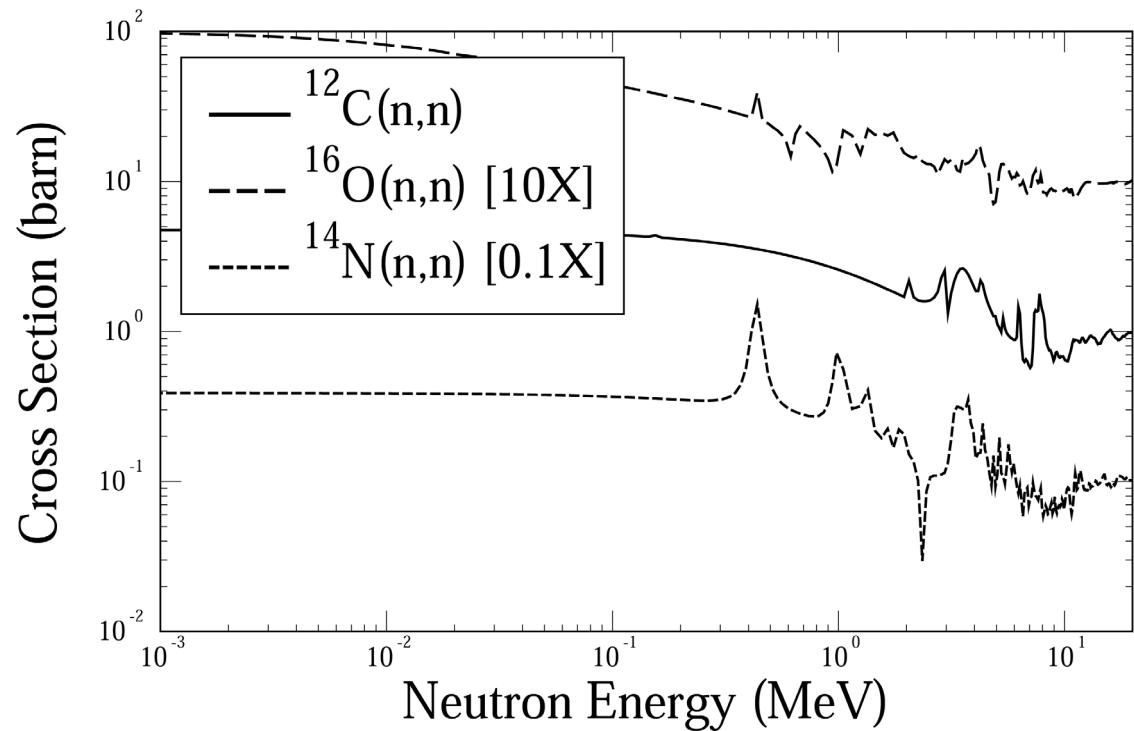


Figure 2: High Energy Resonance Behavior for Some Neutron Cross Sections

used to excite photonuclear reactions are typically a broad energy spectrum generated by bremsstrahlung radiation from high energy electrons produced by linear accelerators and impacting high-Z targets.

Each nucleus has a unique set of excited states. The energy widths of these excited states are very narrow ( $\ll 1$  eV) and they have a small lifetime ( $< 1$  ns). However, when a nucleus is excited into a state above the ground state, it isotropically emits characteristic radiation as it de-excites \*\*. A technique, referred to as nuclear resonance fluorescence (NRF), uses a broad energy photon source (typically a bremsstrahlung source) to excite a target material. The isotropically emitted radiation is then detected by very sensitive high resolution gamma detectors in backward directions that are shielded from the bremsstrahlung source photons.

A giant dipolar resonance (GDR) exists in the majority of photoabsorption and photonuclear reactions. This resonance energy corresponds to the fundamental frequency for absorption of electric dipole radiation by the nucleus acting as a whole. It can be envisioned as an oscillation of neutrons against the protons in a nucleus. The GDR occurs at energies of 20 - 24 MeV in light material and of 13 - 15 MeV in heavy nuclei. A compendium of the GDR parameters is found in Ref. [3].

Protons can also exhibit very strong nuclear resonance reactions in materials. One example is the  $^{13}\text{C}(\text{p},\gamma)^{14}\text{N}^*$  reaction for proton energies of 1.747 MeV. This reaction has a very narrow width, about 122 eV. The resonance reaction produces a photon with a very narrow spread in the emitted energy. In the case of  $^{13}\text{C}$ , the photon is emitted with an energy of 9.17 MeV. The inverse reaction, using a monoenergetic 9.17 MeV photon, can also be used to detect the presence of  $^{14}\text{N}$  in the target material. Nuclear resonance absorption (NRA) or gamma resonance absorption (GRA) are names given to the use of this inverse process for explosive detection purposes. The use of the forward proton-induced reaction has been proposed as a source of monoenergetic photons to be used for the interrogation of nitrogen in a target while the inverse absorption reaction is proposed as a material sensitive detector of the transmitted (unscattered) photons after traversal through the test object. The energy/angle relationship for the photon resonant emission is:

$$\cos\theta_R = (E/Mc^2)/(v/c) = p_\gamma/p \quad (5)$$

---

\*\*.Angular correlations can exist between the emitted photons.

The resonance angle is  $80.7^\circ$  for the  $^{14}\text{N}(\gamma, \text{p})^{13}\text{C}$  reaction. The photon energy in the outgoing channel of the production (inverse) reaction can be affected by doppler shifting of the emitted photons due to the recoil of the residual nucleus. Due to this doppler broadening, only gamma rays emitted in a 0.7-degree wide beam are at the resonance energy.

Alternative resonance reactions with emitted photons resonant with absorption on the constituents of explosives include:

- $^{15}\text{N}(\text{p}, \alpha\gamma)^{12}\text{C}$  that produces 4.44 MeV photons for 2.6 MeV incident protons
- $^{19}\text{F}(\text{p}, \alpha\gamma)^{16}\text{O}$  that produces 6.92 and 7.11 MeV photons for 1.03 MeV incident protons.

For the detection of some materials, even cosmic ray muons have been investigated as a probing radiation. Since cosmic ray muons scatter from heavier elements at larger angles than off those of lighter elements, the trajectories of the cosmic rays has been proposed as a means of determining the location of heavy-weight nuclei. Los Alamos researchers have used the background radiation to acquire images of uranium surrounded by lower atomic number materials.

There are other properties of the nucleus that can be probed by means other than interrogating with an incident neutron. Some nuclei, such as  $^{14}\text{N}$ ,  $^{35}\text{Cl}$ ,  $^{37}\text{Cl}$ , and  $^{17}\text{O}$ , have a nonspherical electric charge distribution and possess a nuclear quadrupole moment. This quadrupole moment is affected by the electronic binding of the atom within the chemical compound. When an atom with a quadrupole moment is within an electric field gradient the quadrupole experience a torque and precesses at a given frequency. The nucleus' precessional frequency is determined by the local quadrupole moment and by the electric field surrounding the atom. A radio frequency (rf) pulse tuned to the quadrupole precessional frequency can be used to disturb the preferred orientation. As the atoms return to the preferred orientation, they release an electromagnetic signal (at one or more frequencies) that can be analyzed. The signal is a decaying voltage picked up in the receiver coil. The signal is Fourier transformed to get a frequency spectrum. The emitted signature is unique to the atom and the local molecular environment. The relaxation time for the excited nucleus is also an important parameter in the nuclear quadrupole resonance (NQR) process. It can depend upon the atom, the compound, and the ambient temperature. No external magnetic field is required for this technique, the field gradient is provided by the molecular composition of the material being interrogated [4, 5].

## 1.2. Neutron Sources

The most difficult part of neutron detection-based explosive detection methods is efficiently generating the required neutron source while protecting the public from radiation exposure. Some approaches desire monoenergetic neutrons while other approaches require a broad energy neutron spectrum. It is very desirable to have a source that can be turned off when the detection process is not in operation. Shielding of neutrons is very difficult, requiring either large volumes for spherical divergence of the neutrons or bulky shielding (typically borated or lithiumated polyethylene) for neutron down-scattering and absorption.

### Isotopic

Some materials have a spontaneous decay process that emits neutrons. Some short-lived fission products are in this class and are responsible for the delayed neutron emission from fission events. Another material in this class is  $^{252}\text{Cf}$  that has a spontaneous fission decay mode.  $^{252}\text{Cf}$  is probably the most useful material to use as a source of neutrons with a broad energy spectrum.

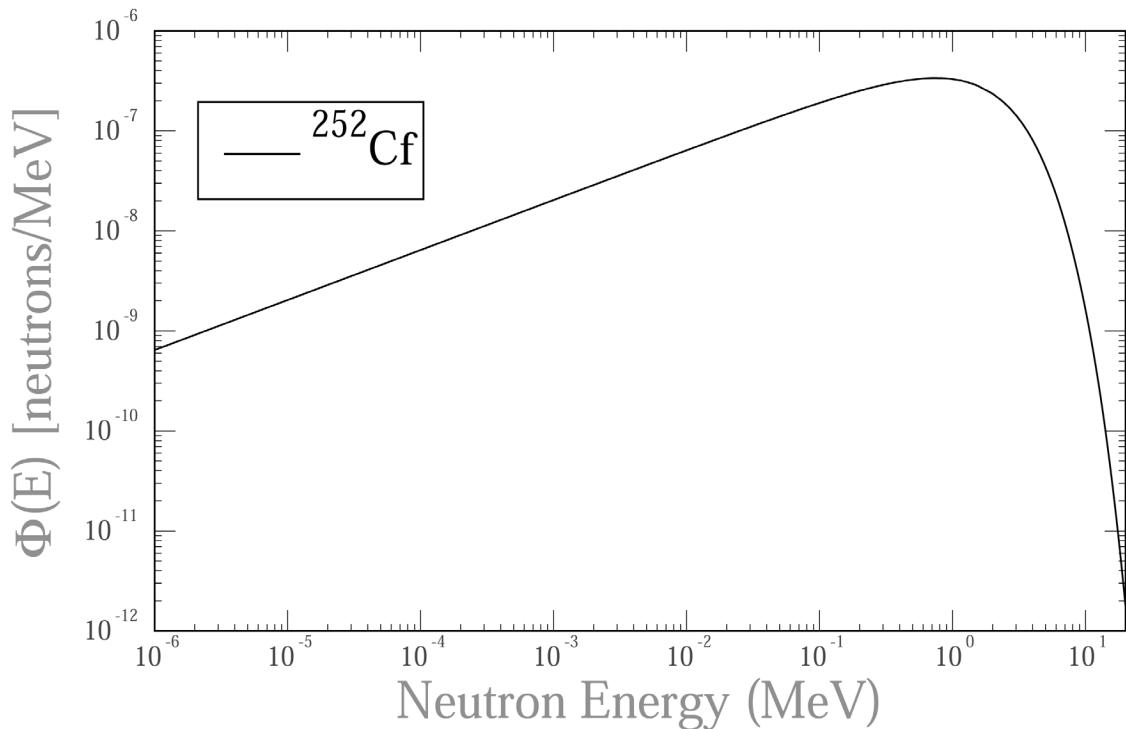
$^{252}\text{Cf}$  spontaneous fissions have a “fast” neutron energy spectrum, shown in Fig. 3, with an average energy of  $\sim 2.2$  MeV. On average, 3.76 neutrons are emitted per spontaneous fission. The neutron emission rate is  $2.34 \times 10^{12}$  n/(sec-gm) or  $4.1 \times 10^9$  n/(sec-Ci) for  $^{252}\text{Cf}$  which has a specific activity of  $5.4 \times 10^2$  Ci/gm.  $^{252}\text{Cf}$  has an alpha decay mode as well as the spontaneous fission decay mode.

### $(\alpha, n)$ Reactions

There are many isotopes that decay by alpha emission. When these isotopes are placed in intimate contact with another material, such as beryllium, the resulting  $(\alpha, n)$  reaction can be used as a neutron source. Beryllium is the target material with the highest neutron yield. Other targets include  $^7\text{Li}$ ,  $^{11}\text{B}$ ,  $^{19}\text{F}$ , and  $^{18}\text{O}$ . Table 1 shows the characteristics of some typical  $(\alpha, n)$  sources.

### $(\gamma, n)$ Reactions

Photoneutron reactions or  $(\gamma, n)$  reactions can also be used to produce neutrons. Most materials have a high binding energy and require incident gamma rays with an energy  $> 10$  MeV to get photodisintegration. Beryllium and deuterium are two exceptions. With these two materials photoneutron emission occurs for incident gammas with energies of 1.666 and 2.226 MeV, respectively. Ra-Be and Sb-Be are two examples of  $(\gamma, n)$  sources based on the decay gammas from activated nuclei rather than photons produced from an accelerator-driven photon reaction. These decay gamma sources have a very large gamma background that is difficult to shield.



**Figure 3:  $^{252}\text{Cf}$  Fission Spectrum**

**Table 1: Isotropic Neutron Sources**

Metric	$^{241}\text{Am-Be}$	$^{239}\text{Pu-Be}$	$^{210}\text{Po-Be}$
yield (n/(sec-Ci))	$2.2 \times 10^6$	$1.7 \times 10^6$	$2.5 \times 10^6$
Alpha Half-life	458 yr	24360 yr	138 d
Specific Activity (Ci/gm)	3.4	6.2E-2	4.5E3
Avg. neutron energy (MeV)	4.5	3.2	4.2

### Accelerator-based

Accelerators can be used to create monoenergetic neutron sources using the reactions:  $^7\text{Li}(\text{p},\text{n})^7\text{Be}$ ,  $^3\text{H}(\text{p},\text{n})^3\text{He}$ ,  $^{12}\text{C}(\text{d},\text{n})^{13}\text{N}$ ,  $^2\text{H}(\text{d},\text{n})^3\text{He}$ , and  $^3\text{He}(\text{d},\text{n})^4\text{He}$ . These reactions are only truly monoenergetic (for a given scattering angle) in specific energy regions [6]. For example, the  $^7\text{Li}(\text{p},\text{n})$  reaction has a threshold energy of 1.881 MeV for the incident proton to produce the (p,n) reaction, but when the incident proton energy exceeds 2.372 MeV, the reaction can result in the residual

$^7\text{Be}$  being in an excited metastable state. At 3.697 MeV a three body breakup channel,  $^7\text{Li}(\text{p},\text{n}^3\text{He})^4\text{He}$ , occurs. In these last two cases with a high incident energy proton, the energy partitioning of the outgoing particles results in the emission of some neutrons (at a given angle) with a lower energy than that for the neutron emission associated with the  $^7\text{Be}$  being left in the ground state.

The range of monoenergetic neutrons that can be produced from these accelerator-driven reactions varies with the specific reaction and the energy of the incident particle. The range of outgoing neutron energies goes from near 0 to 7.7 MeV in addition to the energy region from 11.7-20.4 MeV. Outside of these energy regions in accelerator produced neutron sources, there will be some contamination of the outgoing neutron spectrum with lower energy neutrons.

Electron accelerators can also be used to produce high energy electrons that impact a high-Z target to produce high energy bremsstrahlung radiation. The bremsstrahlung radiation can then be used as a source of neutron production through  $(\gamma,\text{n})$  reactions on materials such as beryllium.

### Reactor-based

Fast fission  $^{235}\text{U}$  metal assemblies or water-moderated reactors with 20% enriched uranium oxide fuel are easy sources of high intensity neutrons [ $> 10^{11} \text{ n}/(\text{cm}^2\text{-sec})$ ]. The major problem with a reactor as a neutron source is that it is not small, portable, nor easily protected in the environment desired for application as part of a typical explosive inspection system.

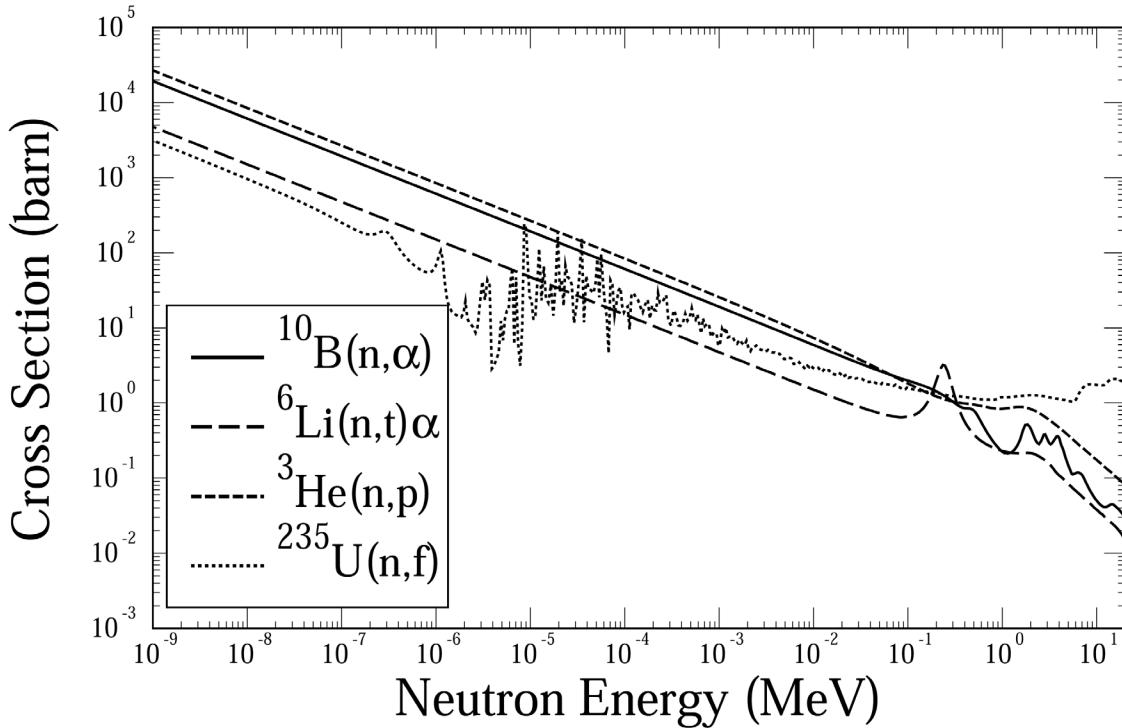
## **1.3. Detectors**

Nuclear explosive detection approaches typically use particles to probe the nucleus of the target material. The interrogation procedure involves the measurement of transmitted/emitted neutron or gamma radiation. The selection of an appropriate radiation detector can be a very important part of a detection technique. Trade-offs in the detector selection involve the sensitivity of the detector, interferent reactions, cost of the detector material, logistic issues such as the need for cryogenic cooling, and possibility of radiation damage to the detector during the measurement process from other types of radiation in the operational environment.

### Neutron Detectors

Neutron detectors are often separated into two types; low energy neutron and fast neutron detectors. Low energy neutron detection typically uses a material/reaction with a large thermal neutron cross section, such as the  $^{10}\text{B}(\text{n},\alpha)$ ,  $^6\text{Li}(\text{n},\alpha)$ ,  $^3\text{He}(\text{n},\text{p})$ , or  $^{235}\text{U}(\text{n},\text{f})$ . The  $(\text{n},\text{f})$  notation indicates a fission reaction where the

exit channel has a light and a heavy fission product. Fig. 4 shows the cross sections for these reactions. The energy deposition of the charged particle in the exit channel is used to register a detection of a low energy neutron interaction. In a neutron pulse-type detector, the ionization of the charged particle is measured. Signal/detector thresholding techniques are used to remove ionization events resulting from gamma interactions.



**Figure 4: High Cross Section Low Energy Reactions**

An example of a low energy neutron detector is a  $\text{BF}_3$  chamber. In this detector, boron trifluoride is the gas in a proportional counter and serves as a target for the neutron. When a reaction occurs, the recoiling alpha and lithium particles produce ionization in the proportional chamber gas. For small  $\text{BF}_3$  chambers (less than the range of the alpha particle or  $\sim 1\text{-cm}$ ), the charged particle can impact the wall before it has deposited all of its recoil energy. This results in a small detected pulse and a “wall effect”. In mixed neutron/gamma fields with a high gamma flux, pulse pileup can result in apparent gamma peaks that have the composite energy from several separate gamma ray interactions.

An alternate approach to this use of boron as a detecting material is a boron-lined ionization chamber. Here a common proportional counting chamber is lined with boron. A proportional counter uses a fill gas that does not exhibit a significant electron attachment coefficient, such as a noble gas. In a proportional counter, the signal is based on the secondary ionization created by collisions between

electrons and the neutral fill gas. Polyatomic gases, such as methane, are often added to preferentially suppress the effect of photon interactions by absorbing the photons in a mode that does not lead to further ionization. One common fill gas is a mixture of 90% argon and 10% methane referred to as P-10 gas.

Boron loaded scintillators are yet a third way of using this high cross section neutron material in a detector. A scintillator converts the kinetic energy of the charged particles into detectable light in such a way that there is a linear relationship between the light yield and the deposited energy. The decay of the luminescence should also be fast so that a distinct light pulse can be generated. A wide range of organic plastic and liquid materials (e.g. stilbene, Eljen EJ-212, Bicron BC-400), as well as inorganic materials (e.g. Na activated cesium iodide, bismuth germanate, yttrium aluminum perovskite) can be used as scintillating materials. These materials are typically loaded with boron in this type of a detector. The scintillator is typically thin (1 - 2 mm) so that it is relatively transparent to its own scintillation light. Scintillators are typically less effective than ionization chambers in discriminating against gamma ray interactions.

A fission chamber is another variation in a ionization chamber designed to detect neutron interactions. Fission chambers have the advantage of a large ionization signal (~160 MeV from the kinetic energy of the fission fragments) that permits discrimination of low neutron fluxes in the presence of high gamma background radiation.

Self powered neutron detectors (SPNDs) use a material such as cadmium that has a high cross section for low energy neutrons and produces copious gammas or betas in the decay of the excited residual nuclei. If the decay particles are gammas, a material surrounds the detector material and converts the gammas into electrons through photoelectric or Compton interactions. The gamma-induced electrons or beta particles produce a current that is then collected and measured directly without any externally supplied bias voltage. SPNDs are always operated in current mode rather than pulsed mode. Some SPNDs have a prompt response (e.g. Cd-based) whereas others have a delayed response due to the half-life of the beta decay reactions.

Fast neutron detection sometimes uses a hydrogenous moderator to slow down the neutrons and then employs a low energy neutron detector as described above. One common fast neutron detector is a Bonner sphere. In this detector, a scintillator is placed in the center of a polyethylene sphere. Radiation transport calculations are used to produce efficiency curves that depend upon the energy of the incident neutron. Another common fast neutron detector is a “long counter”.

This detector uses a slow neutron detector (originally a  $\text{BF}_3$  chamber) at the center of a cylindrical moderator designed so that the detector is only sensitive to neutrons incident from one side of the detector.

For neutrons with an energy greater than  $\sim 1$  keV, the recoil resulting from elastic interactions can be used as a basis for the detection of ionization from a neutron interaction. Hydrogen is the most common target material for fast neutron detection using the elastic recoil energy. These detectors are often referred to as proton recoil detectors. Whereas low energy neutron detection typically just measures the occurrence of an event or neutron interaction, high energy detectors ( $E_n > 10$  keV) often provide information on the energy of the incident neutron. For non-relativistic neutrons with incoming energy,  $E_n$ , scattering with an angle of  $\Theta$  in the center of mass coordinate system ( $\theta$  for the recoil nucleus in the lab coordinate system) from a target atom with atomic number  $A$ , the energy of the recoiling nucleus is given by:

$$E_R = \frac{2A}{(1+A)^2} \cdot (1 - \cos\Theta) \cdot E_n = \frac{4A}{(1+A)^2} \cdot (\cos\theta^2) \cdot E_n \quad (6)$$

The proton recoil is often measured by hydrogen-containing scintillators. Complications with the use of this type of detector relate to the mixed neutron/gamma response of the scintillating material, nonlinear light output of the scintillator with the deposited energy, loss of energy for events near the edge of the scintillator material due to the range of the proton, multiple scattering effects, pulse shape discrimination against gamma rays, and complications in the deconvolution or unfolding of the energy dependence of the detector response.

### Gamma Detectors

A wide range of methods exist for detecting gamma rays. Methods include a cryogenically cooled high purity germanium (HPGe) detector, ionization chamber, scintillation materials, semiconductor diodes, and photoconductive detectors. A reference such as Ref. [7] should be consulted to get specific information on the various detection approaches. HPGe detectors have excellent energy resolution ( $\sim 0.1\%$ ) but they are expensive and require cryogenic cooling that can present logistic difficulties. HPGe detectors are also easily damaged in neutron environments. HPGe detectors have a signal efficiency about 10X less than a NaI detector. A NaI scintillator has significantly less energy resolution. Figs. 5 and 6 show simulations of the HPGe and NaI detector signal from a nickel sample activated in a water moderated reactor. Bismuth germanate ( $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ , often

notated as BGO) and Cadmium-zinc telluride ( $\text{Cd}_{1-x}\text{Zn}_x\text{Te}$ , often notated as CZT) are two other common gamma detectors. The high atomic number results in a CZT detector showing a high photoelectric absorption cross section and a high efficiency for low energy photons. This detector also has a large bandgap so it can be operated at room temperature. The BGO inorganic scintillator detector has a high density and a high atomic number giving it a good efficiency. BGO is also a fairly rugged detector but it has a low light output. It is often used when high gamma count rates are more important than energy resolution. The same activated nickel gamma spectrum used in Figs. 5 and 6 is used to simulate the gamma response of the BGO and CZT detectors in Figs. 7 and 8.

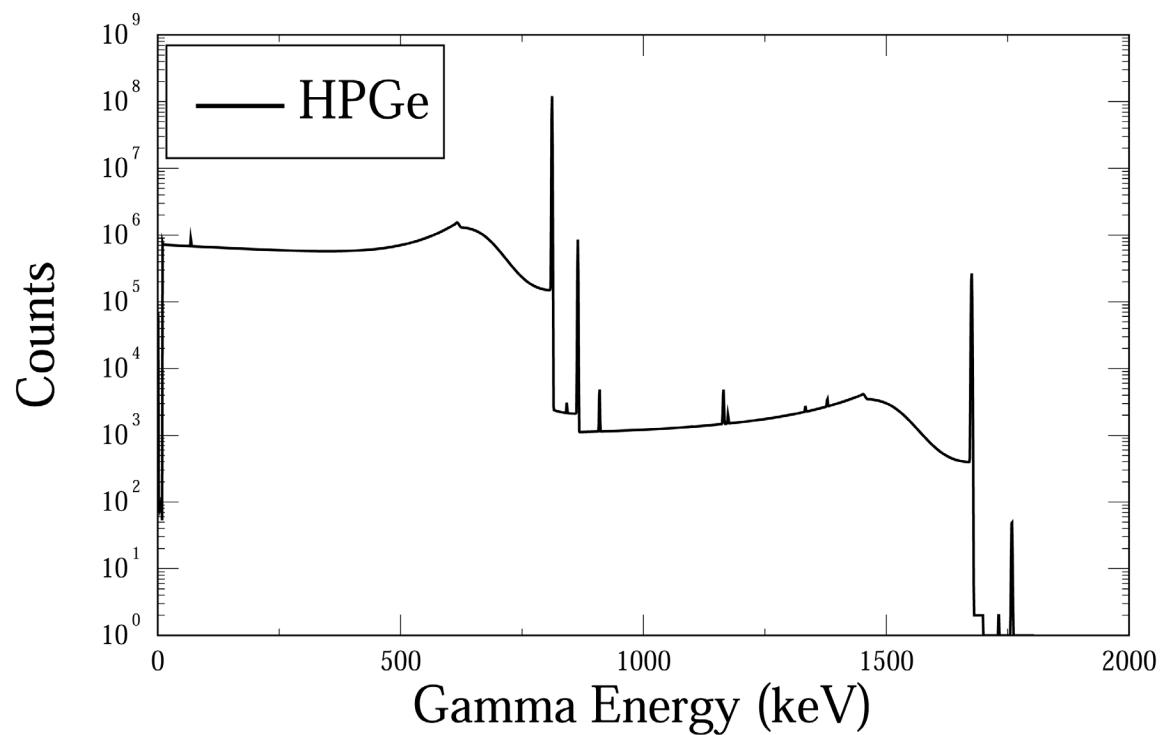
Ionization chambers are very sensitive detectors, but they only report a rad(material) metric rather than providing an energy-dependent spectrum. P-i-N diodes are less sensitive, typically measure rad(Si) in the active area of the detector, and show increased leakage current after being exposed to high levels of neutrons. Diamond photoconductive detectors have a small signal (are less sensitive) but exhibit a very fast response (< nanoseconds) and they can be pre-damaged to stabilize their response in the presence of high levels of neutrons.

## 2. SURVEY OF NEUTRON-BASED DETECTION APPROACHES

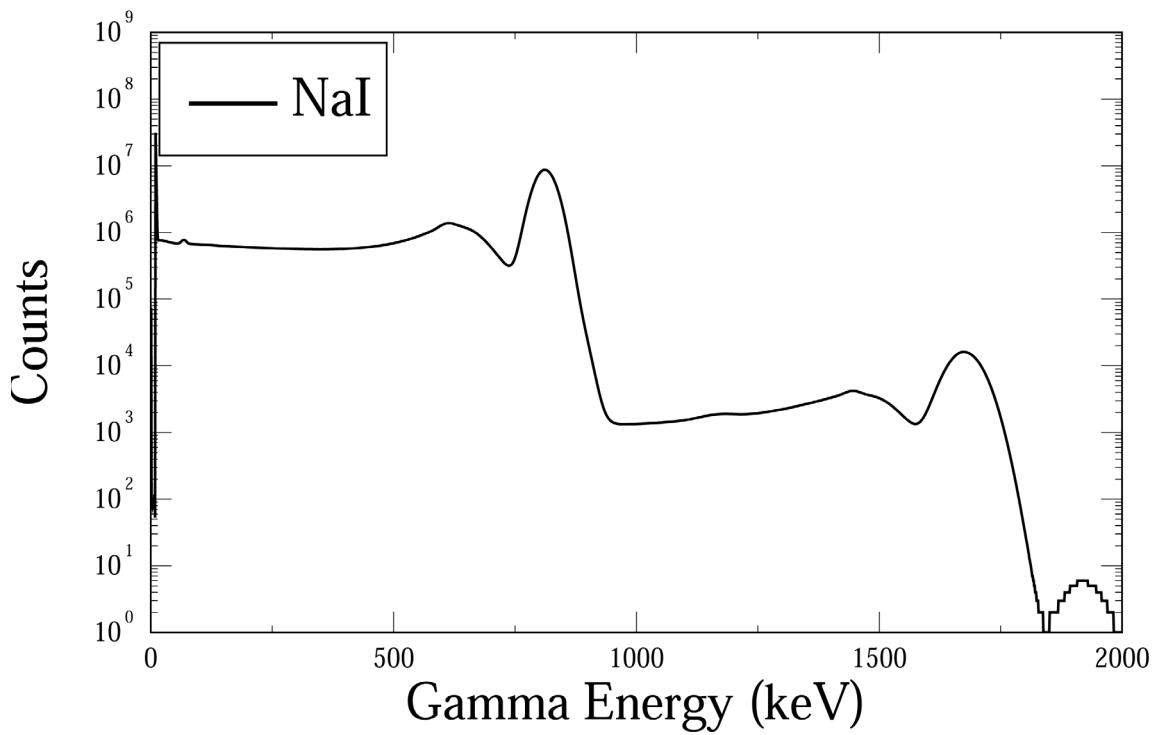
As neutron-based technologies have evolved, the newer approaches are often refinements or extensions of the older approaches. The following survey of approaches is structured to show how the neutron-based detection technology has evolved.

### 2.1. Thermal Neutron Activation (TNA)

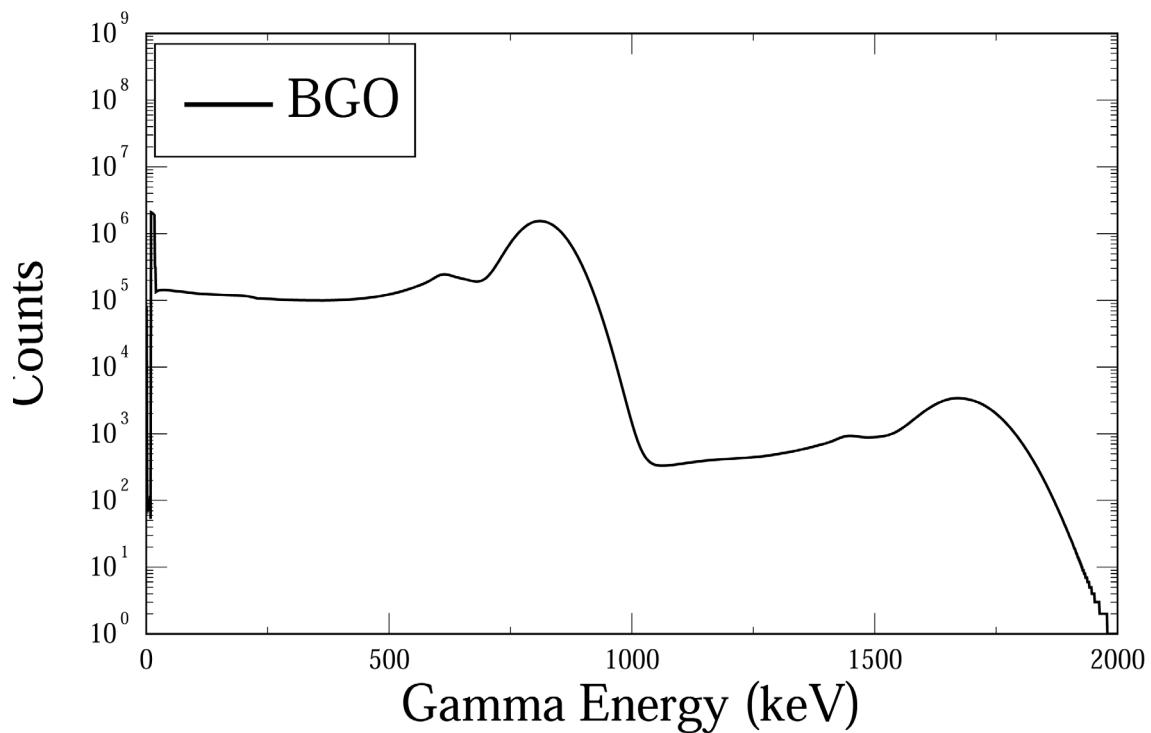
One of the first nuclear technologies [8] proposed for explosive detection was thermal neutron activation. This is also one of the most thoroughly investigated nuclear explosive detection technology [9, 10]. This technologies employs a thermal energy neutron source to interrogate a bag. Since high energy neutrons are easier to generate, implementations of this technologies often employ a scattering material to degrade the source neutron before it is directed onto the bag. When the thermal neutron passes through the material in the bag, there can be capture/absorption reactions with the elements in the bag. Nitrogen has a large thermal capture cross section ( $\sim 75$  mb) relative to other elements found in explosives. The nitrogen capture reaction also emits a very distinctive 10.8292 MeV gamma (along with other gamma rays). The thermal capture and emission of the 10.8292 MeV gamma is delayed by the neutron thermalization time which makes it easier to detect in the presence of many other inelastically scattered gammas. Gamma detectors measure the 10.8292 MeV capture gamma and produce a



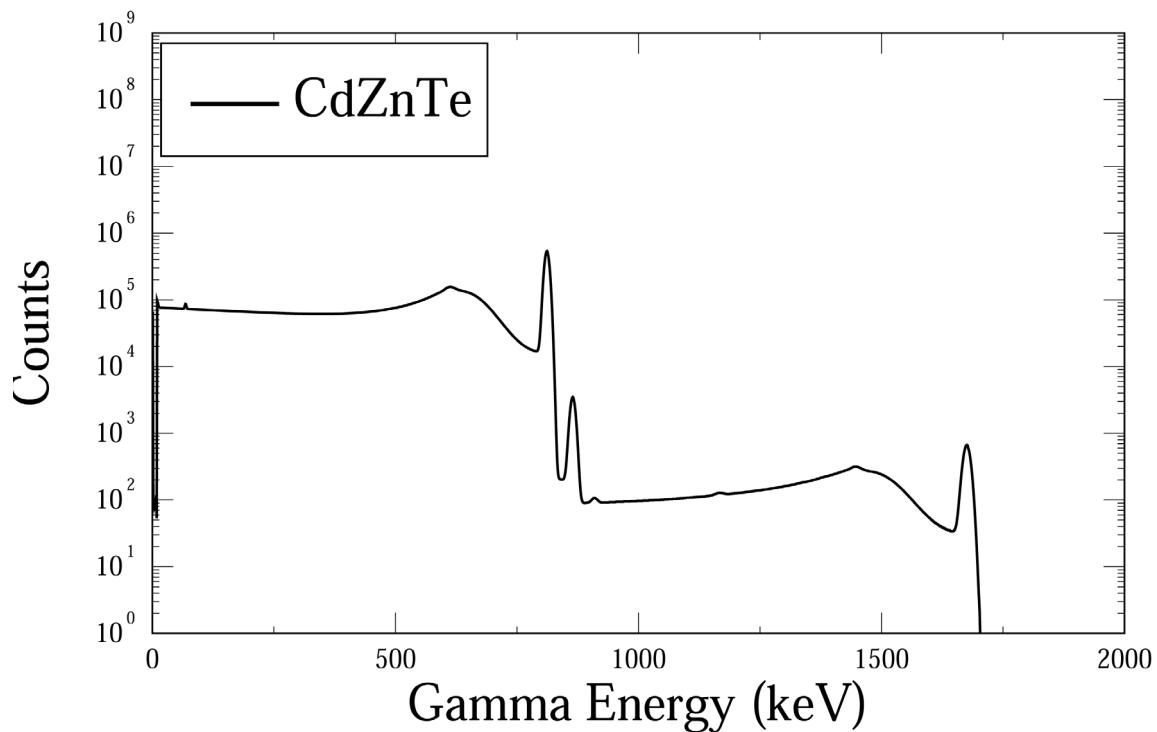
**Figure 5: HPGe Response Activated Nickel**



**Figure 6: NaI Response to Activated Nickel**



**Figure 7: BGO Response Activated Nickel**



**Figure 8: CZT Response Activated Nickel**

measure of the nitrogen content in the section of the bag being interrogated by the neutron beam. In practice, the neutron beam is collimated and scanned over the bag. The resulting nitrogen map is used to determine the possible presence of explosives.

Various neutron sources have been proposed for use with this detection technology [9]. The TNA approach uses a thermal capture reaction, so it does not require an accelerator to produce a monoenergetic neutron source. Spontaneous fission neutrons from a  $^{252}\text{Cf}$  source is the most commonly proposed source [11, 9] and was employed in a pre-production prototype of TNA. This prototype used a 150  $\mu\text{g}$  80 mCi source. One implementation of TNA weighted 28,000 lb, required a 41  $\text{m}^2$  area, cost \$1.4M for fabrication, and had an estimated \$0.705M annual operation cost [12]. Early thermal neutron work [13] proposed the use of antimony-beryllium, radium-beryllium, and polonium-beryllium sources that used an  $(\alpha, n)$  reaction to produce the broad spectrum neutrons. These sources proved to have too small a neutron yield for practical development and attention was turned to accelerator sources. Neutrons from the accelerator-based  $^2\text{H}(\text{d},\text{n})^3\text{He}$  and  $^9\text{Be}(\text{d},\text{n})^3\text{He}$  sources have been proposed for more recent implementations of TNA [14, 15]. The advantages of the accelerator sources for TNA relate to a higher neutron yield and the ability to turn off the neutron production.

The major advantage of the TNA technology is that it can produce a nitrogen map and many important explosives have a high nitrogen content. The neutrons employed by this technology will also penetrate metal screens that may shield conventional x-ray methods. The most important problem with this approach is that nitrogen content, by itself, is not distinctive enough to prevent a large false alarm rate. Because of this, attempts have been made to combine the TNA nitrogen map with the density map from conventional x-ray methods. One example of this combined technology is the XENIX system [14]. The detection probability and false alarm rate of this combined technology did not meet the then current FAA explosive detection system (EDS) certification specifications.

Other disadvantages of this system include its large weight, high cost, and the presence of a radioactive source. The fielding of TNA prototypes demonstrated that none of these issues preclude the application of the technology, they just suggest that other technologies are preferable. Shielding and collimation of the thermalized neutron beam are significant challenges to this technology. Refinements to the TNA prototype have been proposed which address decreases in the size and weight.

Pre-production TNA prototypes exist, but there is no obvious path for the development of a stand-alone TNA system that meets FAA/TSA EDS certification requirements for large checked baggage. The TNA technology development resulted in advances in several areas and highlighted the importance of including bag clutter in determining detection performance [16]. A comparison of the potential of the TNA technology for checked baggage with the detection performance demonstrated by current tomographic x-ray systems suggests that a neutron-based or accelerator-based technology that can only detect nitrogen is not a good candidate for further development by the FAA. Further development of neutron-based or accelerator-based explosive detection technologies has been concentrated on methods that can also provide quantitative metrics on the presence of other elements, in addition to nitrogen, that are in explosives and in typical checked bags. The better detection potential that comes from having quantitative information on the presence of other elements in typical bags, such as carbon, oxygen, and hydrogen, is required to offset the weight, volume, and operating cost disadvantages of neutron-based or accelerator-based approaches.

## 2.2. Fast Neutron Activation (FNA)

The fast neutron activation technology was an outgrowth of attempts to get more information than nitrogen content from neutron interrogation approaches. In FNA, high energy neutrons, rather than thermalized neutrons, are used to scan the contents of a container. Gammas from neutrons inelastically scattered on oxygen (6.13 MeV gammas from  $^{16}\text{O}$ ), carbon (4.44 MeV gammas from  $^{12}\text{C}$ ) and nitrogen (5.11, 2.31, 1.63 MeV gammas from  $^{14}\text{N}$ ) atoms in the container are measured by detectors that surround the container. High energy neutrons are required to excite these inelastic reactions. A monoenergetic neutron source simplifies the analysis of the inelastic gamma signatures. Most FNA implementations use a deuterium-tritium  $^3\text{H}(\text{d},\text{n})^4\text{He}$  reaction (DT) to produce a monoenergetic  $\sim$ 14-MeV neutron source. The DT reaction is one of the easiest reactions (low incident particle energy, typically  $\sim$ 150 keV deuteron, onto a tritiated target; high cross section) to produce monoenergetic neutrons. Another reaction used is the deuterium-deuterium  $^2\text{H}(\text{d},\text{n})^3\text{He}$  reaction (DD) which can produce monoenergetic neutrons with energies between 2.45 and 7.71 MeV [17].

The inelastic scattered gammas from nitrogen are very weak and hard to detect in the detector pulse continuum. With low energy resolution gamma detectors (such as NaI) the 5.11 MeV gamma coalesces with escape peaks from the  $^{16}\text{O}$  inelastic lines. Various detector options can be employed to improve the detection of the nitrogen gamma signature, such as the use of high energy resolution high purity germanium detectors (HPGe) or the use of anti-Compton shields with

NaI detectors. These detector options raise other considerations such as the detector time resolution and the cost of the detectors. In response to this difficulty in using FNA to measure the nitrogen content, variants of the FNA approach use a microsecond ( $\mu$ s) or faster pulsed neutron source and a time-dependent detection of the neutron capture signatures from nitrogen. A later section will discuss this variant in more detail. The neutrons that are thermalized in a target being analyzed are used to measure the nitrogen content with the same capture reaction that was used in TNA. In a typical airport passenger checked bag, only a small percentage ( $\sim 1\%$ ) of the fast (14 MeV) neutrons will be thermalized and the neutron thermalization time in a hydrocarbon, such as plastic, is about 0.2 ms.

One FNA approach is referred to as Pulsed Interrogation Neutron and Gamma (PING) [18]. In this approach, an accelerator is used to produce 14 MeV neutrons from the deuterium-tritium (DT) reaction. The accelerator is pulsed at about 8 kHz with a 7  $\mu$ s wide deuteron pulse. In addition to nitrogen, sulfur (5.42 MeV gammas from  $^{32}\text{S}$ ) and chlorine (6.111 MeV gammas from  $^{35}\text{Cl}$ ) 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 and in the detection of cocaine. Sulfur can be important in the detection of some chemical warfare agents.

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 [19]. The issues with these sealed tube neutron generators is their relatively low neutron yield (typically about  $10^{10}$  n/s) [20, 19] and short tube life-time (typically less than 2000 hr. at high flux operation). Sealed tube neutron generators also exhibit stability problems during operation [21]. Recent advances in sealed tube sources is addressing some of these limitations.

Even though the accelerator portion of a DT system is small, the 14-MeV neutron shielding requirements are very stressing 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 ( $\sim 2$  MeV) from a  $^{252}\text{Cf}$  source. Some neutron production reactions (e.g.  $^9\text{Be}(\text{d},\text{n})^{10}\text{B}$  as used for PFNTS discussed in a Section 2.4) use a higher incident energy particle and produce a more forward-peaked neutron emission that is much more efficiently collimated and focused on the target region of a bag [21].

FNA systems only produce a 2D view with no depth profile. The systems usually have large a pixel size and subsequently a poor image. The pixel size 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.

The 2D view produced by this system is a critical limitation. Several approaches have been suggested to address this limitation. One such approach, discussed in the next section, is to use the associated particle method to determine the direction and timing of the source neutrons. Others have suggested the application of imaging processing techniques that can vary the location of a focal plane to provide a 3D image while using an isotropic uncollimated and continuous neutron source. The most popular refinement to FNA is the use of a pulsed neutron source. The pulsed fast neutron analysis (PFNA) method, addressed in a section 2.5, can provide the needed 3D information.

### **2.3. Fast Neutron Associated Particle (FNAP)**

This technology is a refinement of the sealed-tube DT neutron source FNA approach. In FNAP, the detection of the associated alpha particle recoil is used to specify the time and direction of the neutron emission. The DT neutron source uses the reaction  $^3\text{H}(\text{d},\text{n})^4\text{He}$ . The reaction product consists of a 14-MeV neutron and a 3.5-MeV alpha particle. In associated particle imaging (API) [22], the alpha particle is detected by a position sensitive alpha detector. The site 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 velocity of the neutron, and the velocity of the photons, to estimate the depth in the bag where the inelastic reaction took place. Thus, with associated particle detection, the inexpensive sealed tube DT application of the FNA approach can be used to produce a 3D image.

This approach is often referred to as associated particle sealed tube neutron generator (APSTNG) [20]. Some proposals [23] have been made to combine the APSTNG time-correlated 3D image information with the “not time-correlated” and “not image-related” slow neutron capture gammas. These proposals also suggest that an array of neutron detectors can be added so that neutron transmission spectroscopy can be performed. Unfortunately, these proposals have failed to provide details on how these detection approaches can be applied with any degree of synergy, rather than in a strictly additive sense.

Since the neutron direction is known, the FNAP 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 still fail to impact the target bag and neutron shielding is needed in all directions surrounding the source and bag regions. In addition, the scattering of the neutrons in the shielded material along with the resulting inelastic and capture gammas produces a significant 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 ns. 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  $\sim 5$  cm  $\times$  5 cm  $\times$  5 cm.

Dead time considerations in the alpha particle detection limit the count rate, and hence limit the neutron flux that can be used with this approach. This means that large scan times will be required with this approach.

The incorporation of associated alpha particle detection in a sealed tube neutron generator (STNG) appears to severely aggravate the concerns over the limited neutron flux and tube lifetime previously detailed for STNG FNA approaches. A mean-time-to-failure of some APSTNGs at a neutron flux of  $10^7$  n/s is about 200 hr [24]. Work is continuing to improve this mean-time-to-failure.

Signal-to-noise considerations make most neutron-based explosive detection approaches very difficult to implement. The basis for combining multiple detection approaches (FNA, along with thermal gamma detection and neutron transmission spectroscopy) in a FNAP application that preserves the small volume advantage of a APSTNG remains to be established. There are distinct advantages associated with the API approach, but the concomitant reductions in available neutron flux, issues of tube life-time, and the intrinsic poor spatial resolution must be taken into consideration in potential applications.

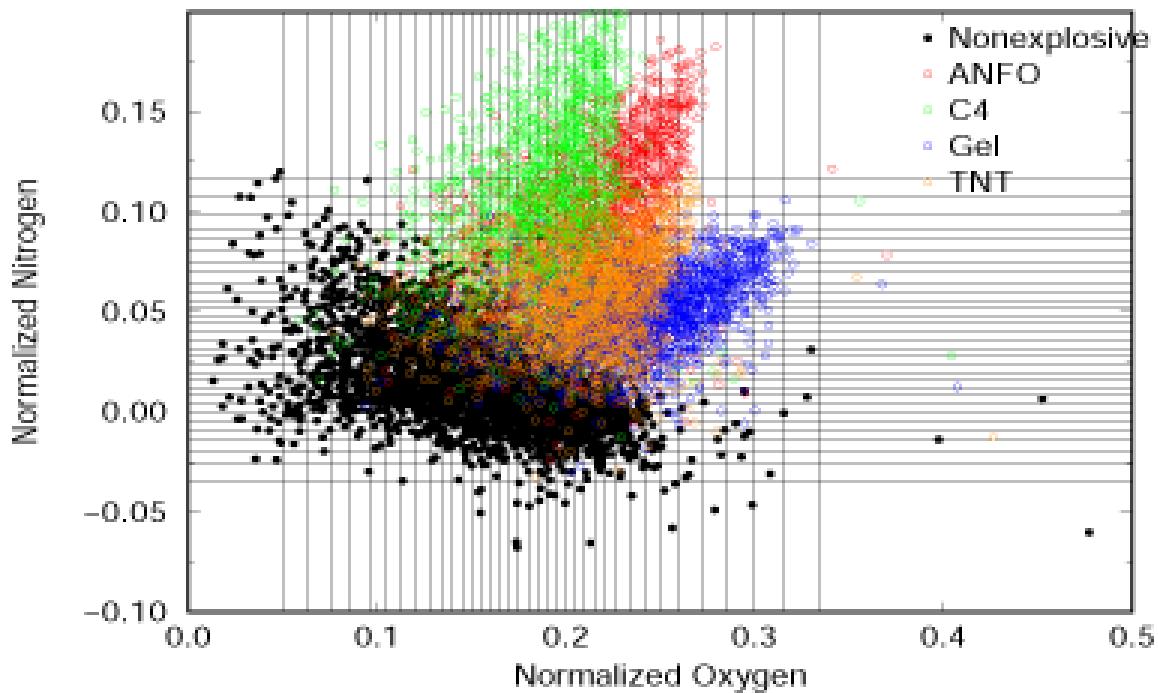
#### **2.4. Pulsed Fast Neutron Transmission Spectroscopy (PFNTS)**

In the pulsed fast neutron transmission spectroscopy method, a collimated broad-energy (0.5 - 8 MeV) or “white neutron” neutron beam is passed through the material being examined. The energy-dependent neutron transmission is measured. By comparing the energy-dependent attenuation of the source neutron spectrum, the ratios of hydrogen, oxygen, carbon, and nitrogen in the bag volume elements can be determined [25]. A fictitious element “X” with a smooth energy dependent cross section is often considered [26] to help normalize the transmitted

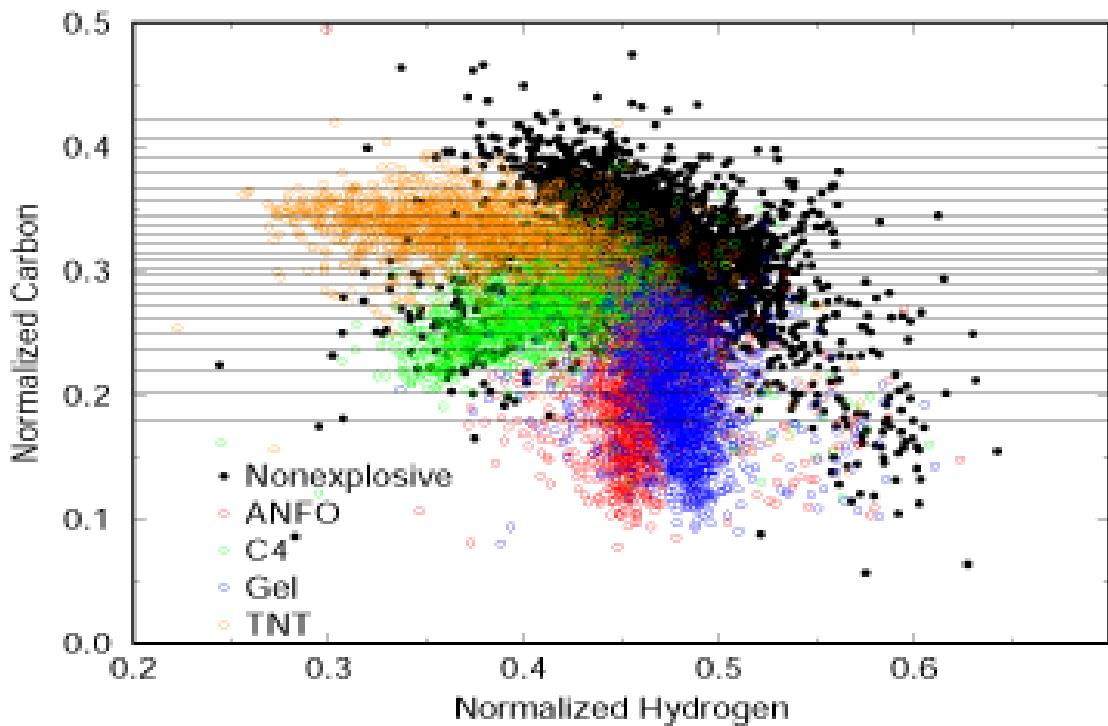
number density. This element “X” is intended to represent a smooth neutron attenuation which can be attributed to elements not specifically represented in the hydrogen/oxygen/carbon/nitrogen decomposition. For every pixel in the target, the energy dependence in the transmitted neutron spectrum is used to unfold the relative amounts of these five elements (hydrogen, oxygen, carbon, nitrogen, and element “X”). Figs. 9 and 10 [27] show how projections in these five dimensions can be used to distinguish the presence of explosive and, often ( $\sim 72\%$  of the time [28]) even to identify the type of explosive. The points in the figures represent 38,000 measurements from actual airline suitcases, with and without explosives. The scatter in the plotted points for explosive and non-explosive paths in Fig. 9 indicates why projected-path nitrogen-only detection schemes have a very difficult task. The outliers in the distribution of “nonexplosive” black points in Figs. 9 and 10 show how difficult it is to eliminate the possibility of a false alarm without impacting the probability of detecting the explosive.

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 to support the detection algorithm. Various algorithms can be used to reduce this five-dimensional elemental information into an “explosive potential” for a single pixel. Variations in the detection algorithm can increase the basis set beyond the nominal 5-elements. The Tensor algorithm 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/no decision on the presence of an explosive in the test article. The University of Oregon refers to their detection algorithm as a “B-matrix” and bases their “explosive potential” metric upon comparison with the explosive/non-explosive probability seen in a simulation database. Separate “B-matrices” are maintained for each explosive class which the algorithm is designed to detect. The Tensor work uses a neural network trained on a set of explosive and non-explosive bags.

The PFNTS approach requires the use of a tightly bunched, pulsed neutron source. Time-of-flight is used to determine the energy of the transmitted neutrons. This width of the initial neutron pulse and the time resolution of the time-of-flight measurement, limits the energy resolution of the transmitted spectrum. Flight paths of 4 to 10 meters are commonly used. The narrow peaks in the



**Figure 9: Nitrogen/Oxygen Distribution in Explosive and Non-explosive Paths in Cluttered Bags. Source: University of Oregon.**



**Figure 10: Carbon/Hydrogen Distribution in Explosive and Non-explosive Paths in Cluttered Bags. Source: University of Oregon.**

interaction cross sections will not be seen in a typical PFNTS measurement, but will be smeared out by the energy resolution of the detectors [29]. Thus, the element identification in this approach depends upon the broad energy-dependent structures in the cross sections, not the narrow resonances.

Since this method examines the energy-dependent neutron attenuation, it is critical that a broad energy neutron source be employed. This rules out  $^2\text{H}(^2\text{H},\text{n})^3\text{He}$  (DD reaction) and  $^2\text{H}(^3\text{H},\text{n})^4\text{He}$  (DT reaction) sources which have too restricted an energy spectrum. In order to get a reasonable neutron flux for high energy neutrons (up to 8 MeV), accelerators are generally required.  $^9\text{Be}(\text{d},\text{n})^{10}\text{B}$  or  $^9\text{Be}(\text{p},\text{n})^9\text{B}$  reactions are candidate neutron sources [30]. The accelerators required to exploit these neutron-producing reactions need a high current ( $\sim 10$  mA peak current, ns pulse width, and ms repetition frequency) and a fairly high energy deuteron source ( $> 4$  MeV). Most researchers in laboratory PFNTS experiments have utilized a deuteron accelerator and the  $^9\text{Be}(\text{d},\text{n})^{10}\text{B}$  reaction.

## 2.5. Pulsed Fast Neutron Analysis (PFNA)

### General Concept

One of the most popular refinements of FNA involves the use of a pulsed neutron source. There are many options for the neutron accelerators used for PFNA. One representative approach is the use of a 300  $\mu\text{A}$   $^2\text{H}^-$  injector with a 6 MeV deuteron accelerator as a DD source for neutrons [31, 32]. All of the PFNA approaches require a very tightly bunched neutron pulse, usually a beam with a 1-2 ns full width at half maximum (FWHM) [18, 33, 21]. This nanosecond pulse width represents the current state-of-the-art, and, when coupled with the neutron time-of-flight, determines the minimum spatial resolution within the depth of the bag. For a 6 MeV neutron and a 1 ns neutron pulse, the spatial resolution is limited to about  $1.4 \times 10^6 \times \sqrt{E_n(\text{eV})} \times \Delta t(\text{sec}) = 3.43 \text{ cm}$ .

The typical PFNA accelerator pulse repetition rate is 5-15 MHz with DD sources and 8 kHz with DT sources. These accelerators are large and heavy. A pulsed tandem cascade accelerator is typically about 5 feet in diameter, 25 feet long, and 6 feet high. A deuteron Pelletron Van de Graff weighs about 4,500 kg. The size and weight are the principle issues in the airport integration of this technology. Approaches have been suggested to reduce the size and weight of the required accelerators and to ease the airport integration issues.

A prototype of a PFNA system is being tested at the Ysleta Port of Entry on truck cargo containers. A proposal has been made for a PFNA-based system designed

to detect illicit materials for air cargo inspection (ACI). This system is referred to as PFNA ACI. The PFNA ACI is a proprietary design that utilizes a tandem Van de Graff accelerator to accelerate deuterons. The neutron beam is collimated in a scan arm and focused on the front surface of the container. Testing has been conducted on aircraft LD3 container as well as truck cargo trailers.

The first step in the detection algorithm is to use the detected gammas to produce “image files” containing the basic scan carbon, oxygen, and nitrogen density in each voxel. The radiographic neutron attenuation and the elemental density images are then used to derive a hydrogen image file. All elements that scatter but do not absorb neutrons are grouped into the hydrogen image file. The image files are then optimized using smoothing, contrasting, and artifact removal. The PFNA prototype systems have used a discriminant analysis, a neural net algorithm, and the input elemental density images to determine if each voxel might constitute an alarm area. Each voxel can be tagged as either “benign”, “explosive”, or “opaque”. The parameters in the discriminant analysis include various elemental densities as well as more complicated features represented by sums, products and ratios of elemental densities. Thresholds for the discriminants (densities, density ratios, and estimated statistical significance) are considered in the analysis algorithms. The analysis algorithms also contains a “blob” algorithm which can place connectivity requirements on elemental features in adjacent voxels before an algorithm-based region alarm is raised. The “blob” algorithm includes features (threshold and discriminants) that are averaged over the blob. As the volume-dependent cargo composition is approximated by the radiographic measurements, this information is fed back into the algorithm to refine the estimates for the elemental densities in the voxels. A typical scan produces a large quantity of data.

Filters can be derived from the test cargo calibration runs in order to optimize the explosive detection. The PFNA detection algorithm can be totally automated but previous demonstrations (prior to the upcoming 2004 Ysleta Port of Entry testing) have used operator intervention to evaluate the basic scan alarm regions before a directed scan is made of the selected basic scan alarm regions.

### **3. SURVEY OF NON-NEUTRON-BASED NUCLEAR DETECTION METHODS**

The following sections detail some of the nuclear detection technologies that are not based on the use of neutrons. Most of these detection technologies are based upon material interrogation using electromagnetic radiation that interacts with atoms and gives off a signature according to the properties of the target nucleus.

### 3.1. Nuclear Resonance Absorption (NRA)

The nuclear resonance absorption (NRA) explosive detection approach is quite different from the neutron-based detection technologies discussed in the previous sections. The NRA method uses a high energy photon to interrogate the nitrogen content in the target material (cargo) through the  $^{14}\text{N}(\gamma, \text{p})^{13}\text{C}$  reaction. Because this approach uses an incident gamma it is sometimes referred to the gamma resonance absorption (GRA) method. NRA has been considered for the detection of explosives in cargo because the high energy photon has the penetration needed for container inspection systems. The original application of NRA to explosive detection was done by Triumph [34]. Work on NRA has been conducted by Soreq [35] and Los Alamos National Laboratory [36]. Grumman has also been involved in NRA development activities. 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 \$12.1M [37] on the development of a NRA system for explosive detection.

The NRA detection method is based on the 122-eV wide giant resonance in the  $^{14}\text{N}(\gamma, \text{p})^{13}\text{C}$  reaction at an incident photon energy of 9.17 MeV. This resonance reaction has a peak resonance cross section of  $\sim 200$  mb and an energy- integrated resonance cross section of  $\sim 4$  b. Even at the resonance energy, the 9.17 MeV photons are very penetrating (7-cm of liquid nitrogen provides only 10% beam attenuation). The generation of the high intensity incident 9.17 MeV photon beam is one of the principle 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 explosive detection is to use the  $^{13}\text{C}(\text{p}, \gamma)^{14}\text{N}$  inverse reaction to produce the photons. Due to the doppler shift of the recoiling  $^{14}\text{N}$ , only the gamma rays emitted in an 0.7-degree wide beam at  $80.7^\circ$  are at the resonance energy. The accelerator concepts proposed for this inverse reaction with a 1.745 MeV incident proton include a radio-frequency quadrupole (RFQ) pulsed linac and an electrostatic CW accelerator [38, 39]. A representative proposal for cargo explosive detection is for a very high current, 10 mA, accelerator with a 1-cm beam spot, a 25 keV beam spread, a 600  $\mu\text{s}$  pulse width, and a 10 Hz pulse repetition frequency. 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.

Bismuth germanate (BGO) scintillator detectors (temperature stabilized and with a 15% average energy resolution) have been used in NRA detection systems [40] but interference from other proton-induced gamma rays provides a high background signal. In order to exploit the selective absorption of the 9.17 MeV photons in the nitrogen contained in explosives in cargo containers, one would like a gamma detector that is particularly sensitive to the transmitted 9.17 MeV photons. Soreq [41] has developed a nitrogen-rich (31% by weight)  $^{14}\text{N}$ -based ionization detector liquid scintillator based on di-methyl-tetrazole that is resonance sensitive (24% efficient for resonant photons) and is well suited for the explosive detection application.

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 have used the 6.13, 6.9, and 7.1 MeV gammas from the  $^{19}\text{F}(\text{p},\alpha\gamma)^{16}\text{O}$  reaction produced by barium fluoride in the  $^{13}\text{C}$  target 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 explosive detection at the FAA requirement levels (high  $P_d$ , low  $P_{fa}$ , small explosive quantity). Other resonance reactions on  $^{12}\text{C}$  and  $^{16}\text{O}$  have been suggested but they have not been demonstrated to be feasible for the explosive detection due 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 due to the 3-body breakup inverse reaction that has no defined resonance angle.

Another disadvantage with NRA is the requirement for very advanced high current accelerator. This accelerator will have all of the disadvantages (large size, large weight, radiation shielding) associated with the neutron-based accelerators.

### **3.2. Nuclear Quadrupole Resonance (NQR)**

NQR is an electromagnetic interrogation technique that probes with radiation in the MHz frequency range. Unlike nuclear magnetic resonance (NMR) techniques that require a large static magnetic field to orient the nuclei in the target material, NQR does not require an external magnetic field. This technique makes use of the splitting of the nuclear spin states by the electromagnetic radiation interaction with the nuclear charge density in the interrogated material. The coupling between existing nuclear quadrupole moments and the gradient of the electric field determines the NQR signal. NQR provides a chemical specificity since

the signal is related to the particular molecular configuration of the nuclei possessing the quadrupole moment. The NQR detection is restricted to crystalline solids; amorphous materials and liquids are not detected. The physics behind this detection technique was addressed in Section 1.1.

In NQR, the electromagnetic radiation flips the spin of a nucleus with a quadrupole moment. As the nucleus relaxes, it emits a unique signal. This approach has been investigated for use in the detection of landmines and for small explosive masses in mail [42]. The chemical specificity of this approach and its ability to detect small threat quantities of some explosives, such as RDX<sup>††</sup>, are attributes that make it useful in some explosive detection scenarios. The relaxation time for the material determines how rapidly the electromagnetic pulse sequence can be repeated. Some materials, such as PETN<sup>‡‡</sup>, have a long relaxation time making multiple pulse interrogation approaches more time consuming. There is a temperature sensitivity to the relaxation time. Thus the interrogation process typically uses pulse sequences parameters that are effective over the probable temperature range for the inspected materials [43].

The major drawbacks of the NQR approach are: 1) not all explosives have a NQR signal (either nitrogen and chlorine are not present or they are not in a position where there is a gradient in the electric field); 2) material can be shielded from the radiofrequency interrogation by thin metallic containers; 3) there can be interference from AM band radiofrequency signals. Another important limiting consideration for this interrogation is the thermally generated internal noise. This noise can increase the time required to acquire the signal. When metallic shielding is present, this presence is clearly indicated and a shield alarm can be raised rather than a false positive or false negative. The presence of piezo-electric and ferro-magnetic materials in an inspected container can also give rise to spurious signals that can mask the true NQR signals. The effect of these spurious responses can be reduced by using specially designed pulse sequences. Because of the first two limitations of this approach (metal shielding and limited scope of explosive detection materials), NQR will probably have a restricted role as a stand alone explosive detection approach. But, because of its very good chemical specificity and sensitivity, it can play a significant role in: 1) in “systems approaches” where the sensor fusion is used, and 2) in the alarm resolution of

---

<sup>††</sup>.RDX stands for Royal Demolition eXplosive. This material has the formal chemical name Hexahydro-1,3,5-trinitro-1,3,5-triazine and is also called cyclotrimethylenetrinitramine, cyclonite, or hexogen.

<sup>‡‡</sup>.PETN is an explosive with the chemical formula  $C_5H_8N_4O_{12}$ , and it is named as Penterythritol tetranitrate.

detections from other explosive detection systems.

### **3.3. Nuclear Resonance Fluorescence (NRF)**

The physics foundations for NRF were presented in Section 1.1. This technique uses photons to excite the nucleus that then emits characteristics gamma radiation as it de-excited. The de-excitation structure is unique to the target nucleus so this technique provides very good elemental/isotopic identification. However, the detection of the high energy (2-10 MeV) narrow line gamma emissions will require a high resolution detector, such as an HPGe detector, that is expensive and probably\*\*\* requires cryogenic cooling. The source of the incident photons is typically bremsstrahlung radiation. This is a very inefficient photon source and presents significant radiation shielding issues.

The NRF explosive detection technology is much more immature than are the NRA, PFNA, and NQR approaches. Whereas PFNA and NQR have prototype systems and are examining engineering trade-offs for specific applications, NRF is at the stage where basic physics experiments are being performed at science laboratories.

## **4. Problems with the Use of Nuclear Techniques for Explosive Detection**

The previous sections detailed the performance of nuclear detection technologies. The reader will see that these technologies have some important advantages, including their potential for elemental identification and ability to penetrate deep into cargo containers. Unfortunately, nuclear technologies also have some important disadvantages. The following sections briefly describe some of these disadvantages.

### **4.1. Field Deployment of Neutron Sources**

Nuclear detection approaches that use radioactive isotopic sources (e.g.  $^{252}\text{Cf}$  for spontaneous neutron emission or  $^{60}\text{Co}$  for gamma emission) will have to obtain state and federal licenses to field the equipment and abide by applicable health and safety regulations. The licensing process takes some time to put into place and may restrict the easy movement of the detection equipment to new locations. This impacts the ability to rapidly re-locate equipment based upon intelligence estimates of the behavior of smugglers. The use of fixed pre-licensed sites can help to some extent.

---

\*\*\*. HPGe detectors can be operated at temperatures higher than that of liquid nitrogen, 77 K, but with a decrease in the energy resolution.

The presence of nuclear material at a site increases the threat from terrorists who may attempt to steal the material or to explosively disperse it. In the post September 11<sup>th</sup> environment, the requirements for the protection of radioactive material are being significantly increased. This results in increased security costs and risks.

#### **4.2. Health Hazards Due to Radiation**

The regulations associated with the use of radioactive source material will typically require that access to the source material be restricted from untrained individuals. This is usually accomplished by postings, interlocks, and physical barriers. In addition, the operations and maintenance personnel that may come into close proximity to the radioactive source material may be required to use personal dosimeters to monitor their yearly exposure to radiation. This monitoring has logistic costs.

#### **4.3. Material Activation**

When neutrons are involved in the detection process, neutron activation of the inspected materials will occur. The logistics of the operation of the detection system must be designed to ensure that no irradiated material can be activated to a degree that it requires material control measures or presents a hazard to the owner of the material. Bounding analysis of existing TNA and PFNA systems have shown that these systems can easily meet these requirements without impacting the operations protocol. Other neutron detection technologies will have to be assessed with respect to their activation potential, but this is not expected to be a significant issue for currently envisioned applications of neutron-based explosive detection systems.

#### **4.4. Neutron Shielding**

Shielding of the scattered neutrons is a significant issue, but one that is confidently modeled and easily verified by measurement. Neutrons are not easily shielded. They tend to scatter without attenuation from high atomic number materials and only lose part of their energy when they scatter off low atomic number materials. The neutron fluence drops off as the square of the distance from a point source. Shielding is typically accomplished by a combination of distance and by using borated hydrogenous material. Implementation of shielding techniques will probably require that the explosive detection system be operated in a separate building and requires a significant footprint within an inspection facility (e.g. an airport or port of entry).

#### **4.5. Public Perception of Radiation**

The most significant issue with the fielding of a neutron-based explosive detection technology is public perception. Most people have difficulty putting into perspective low probability high consequence events. This has resulted in the word “nuclear” being associated with health risks and environmental issues even when detailed analysis shows that there is no issue.

### **5. SUMMARY**

This paper has detailed the physical principles underlying the use of nuclear technologies that have been proposed for the detection of explosives. It has also recapitulated the current stage of development for nuclear-based explosive detection systems. Nuclear technologies are shown to have some strong advantages, but they also come with some distinct logistic issues. The future of nuclear detection technologies will depend upon the details of the threat detection (set of materials and threat quantity) requirements for a particular application. Nuclear technologies have a high penetration capability and a material specificity that makes them very promising options for small threat quantities in dense cargo containers.

### **REFERENCES**

- [1] B.L. Berman, Atlas of Photoneutron Cross Sections Obtained with Monoenergetic Photons, Lawrence Livermore National Laboratory, report UCRL 78482, 1976.
- [2] S. S. Dietrich and B. L. Berman, Atlas of Photoneutron Cross Sections Obtained with Monoenergetic Photons, At. Data Nucl. Data Tables 38 (1988) 199.
- [3] A. V. Varlamov, V. V. Varlamov, D. S. Rudenko, M. E. Stepanov, Atlas of Photonuclear Reactions: Table of GDR Parameters and Graphs of Cross Sections, International Atomic Energy Agency, report INDC(NDS)-394, January 1999.
- [4] T. P. Das, E. L. Hahn, “Nuclear Quadrupole Resonance” chapter in Solid State Physics, Academic Press (1958).
- [5] A. N. Garroway, J. B. Miller, M. L. Buess, “Explosive Detection by Pure  $^{14}\text{N}$  NQR” in Proceedings of the 1st International Symposium on Explosive Detection Technology, November 13-15 in Atlantic City (1991).
- [6] J. Csikai, CRC Handbook of Fast Neutron Generators, Volume 1, Boca Raton, CRC Press Inc., 1987.
- [7] G. F. Knoll, Radiation Detection and Measurements, Third Edition, John Wiley & Sons, New York, 2000.
- [8] Plessey Nucleonics, Ltd., Mine Detection and Auxiliary Equipment, Progress Report No. 1, 31 December 1958.

- [9] T. Gozani, R. Morgado, C. Seher, "Nuclear-based Techniques for Explosive Detection," *Journal of Energetic Materials*, 4 (1986) 377.
- [10] C. Chung, S. Liu, J. Chao, C. Chan, "Feasibility Study of Explosive Detection for Airport Security Using a Neutron Source," *Applied Radiation and Isotopes A*, V44 (1993) 1425.
- [11] J. Bartko, F. H. Ruddy, "A Review of the Development of a Luggage Explosive Detection System," *Proceedings of the Seventh ASTM-Euratom Symposium on Reactor Dosimetry*, ed. G. Tsotridis, R. Dierckx, P. D'Hondt, Kluwer Academic Press, Dordrecht, The Netherlands, 1992.
- [12] Environment Assessment of the Thermal Neutron Activation Explosive Detection System for Concourse Use at U.S. Airports, U. S. Nuclear Regulatory Commission, NUREG-1396, February 1990.
- [13] D. L. Gravitt, Status of Mine Detection by Nuclear Techniques, U.S. Army Engineer Research and Development Laboratories, Fort Belvoir, Virginia, Technical Report 1786-TR, report ERDL-TR-1786, 31 August 1964.
- [14] V. Leung, P. Shea, F. Liu, M. Sivakumar, "Fusion of the Nuclear and Different X-ray Technologies for Explosive Detection," *Proceedings of the First International Symposium on Explosive Detection Technology*, pp. 311-318, November 13-15, 1991. U.S. Department of Transportation, February 1992.
- [15] M. J. Hurwitz, W. P. Noronha, T. A. Atwell, "Airport Testing of a New Thermal Neutron Analysis Explosives Detection System," *Proceedings of the First International Symposium on Explosive Detection Technology*, pp. 388-395, November 13-15, 1991. U.S. Department of Transportation, February 1992.
- [16] S. B. Buchsbaum, D. Knize, L. Feinstein, J. Bendahan, P. Shea, "An Approach to Improving TNA EDS," *Proceedings of the First International Symposium on Explosive Detection Technology*, pp. 70-81, November 13-15, 1991. U.S. Department of Transportation, February 1992.
- [17] M. Drosig, "Sources of Variable Energy Monoenergetic Neutrons for Fusion-Related Applications," *Nuclear Science and Engineering*, 106 (1990) 279.
- [18] F. J. Schultz, D. C. Hensley, D. E. Coffey, D. J. A. Chapman, B. A. Caylor, R. D. Bailey, "The Pulsed Interrogation Neutron and Gamma (PING) System," CONF-9110168, December 1992.
- [19] P. Bach, M. Jatteau, J. L. Ma, C. Lambermont, "Industrial Analysis Possibilities Using Long-Life Sealed-Tube Neutron Generators," *Journal of Radioanalytical and Nuclear Chemistry*, 168 (1993) 393.
- [20] E. Rhodes, C. W. Peters, "APSTNG: Neutron Interrogation for Detection of Explosives and Drugs and Nuclear and CW Materials," *SPIE Vol. 1737 Neutrons, X-Rays, and Gamma Rays* (1992) 160.
- [21] S. Khan, "Review of Neutron-based Technologies for the Inspection of Cargo Containers," *SPIE Vol. 2276 Cargo Inspection Technologies* (1994) 294.

[22] L. E. Ussery, C. L. Hollas, K. B. Butterfield, R. E. Morgado, Three-Dimensional Imaging Using Tagged 14.7-MeV Neutrons, Los Alamos National Laboratory, Los Alamos, NM, report LA-11423-MS, October 1988.

[23] V. I. Mostovoi, A. N. Rumyantsev, G. V. Yakovlev, E. A. Gomin, L. V. Mayorov, A. V. Marin, Y. Tarabrin, A. DeVolpi, V. Minkov, "Detection of Hidden Explosives and Drugs," Proceedings of the Second Explosive Detection Technology Symposium & Aviation Security Technology Conference, pp. 148-153, November 12-15, 1996. U.S. Department of Transportation, 1997.

[24] E. Rhodes, C. E. Dickerman, C. W. Peters, "Associated-Particle Sealed-Tube Neutron Probe for Characterization of Materials," ANL/RE/CP-78949, Active Probe Technologies Conference on International Symposium on Substance Identification Technologies, 4-8 October 1993, Innsbruck, Austria.

[25] J. C. Overley, "Determination of H, C, N, O content of bulk materials from attenuation measurements," International Journal of Applied Radiation and Isotopes, 36 (1985) 185.

[26] J. C. Overley, M. S. Chmelik, R. J. Rasmussen, R. M. S. Schofield, H. W. Lefevre, Detection of Explosives through Fast-Neutron Time-of-Flight Attenuation Measurements, Final Report, FAA Technical Center, report DOT/FAA/CT-94/103, August, 1994.

[27] M. S. Chmelik, R. J. Rasmussen, R. M. S. Schofield, G. E. Sieger, H. W. Lefevre, J. C. Overley, C. J. Bell, "Analysis of Blind Tests for Explosives in Luggage Through Fast-Neutron Transmission Spectroscopy (FNTS)," University of Oregon, Eugene, OR, January 1997.

[28] H. W. Lefevre, J. C. Overley, Detection of Explosives through Fast Neutron Time-of-Flight Attenuation Measurements, Final Report for FAA Grant #94-G-020, 1998.

[29] T. G. Miller, W. H. Makky, "Application of Fast neutron Spectroscopy/Radiography (FNS/R) to Airport Security," SPIE Vol. 1737, Neutrons, X-Rays, and Gamma Rays (1992) 184.

[30] B. J. Micklich, C. L. Fink, L. Sagalovsky, F. L. Smith, T. J. Yule, Feasibility of Explosive Detection Using Fast-Neutron Transmission Spectroscopy, Report No. DOT/FAA/CT-96/XX, 1996.

[31] D. R. Brown, "Cargo Inspection System Based on Pulsed Fast Neutron Analysis: An Update," SPIE Vol. 2276, Cargo Inspection Technologies (1994) 449.

[32] D. R. Brown, T. Gozani, R. Loveman, J. Bendahan, P. Ryge, J. Stevenson, F. Liu, M. Sivakumar, "Application of Pulsed Fast Neutrons Analysis to Cargo Inspection," Nucl. Instr. Methods A353 (1994) 684.

[33] Z. Sawa, "PFN GASCA Technique for Detection of Explosives and Drugs," Nucl. Instr. Methods. B79 (1993) 593.

[34] Report of the Project Definition Study on the Nitrogen Detector System, TRIUMF,

Vancouver, B.C. Canada, July 1992.

[35] The Development of a Prototype High-Explosive Detection System Based on Nuclear Resonance Absorption (Project OPERA), Summary report, Soreq Nuclear Research Center., December 1993.

[36] Morgado et al., "The Effects of Proton Beam Quality on the Production of Gamma Rays For Nuclear Resonance Absorption in Nitrogen," in Substance Detection Systems, Harding, Lanza, Myers, Editors, Proc. SPIE 2092, (1994) 503.

[37] Terrorism and Drug Trafficking: Testing Status and Views on Operational Viability of Pulsed Fast Neutron Analysis Technology, Report to the Subcommittee on Treasury and General Government, Committee on Appropriations, U.S. Senate, GAO/ GGD-99-54, April 1999.

[38] A. M. M. Todd, C. C. Paulson, J. W. Rathke, M. F. Reusch, O. A. Anderson, K. N. Leung, "Design of a High Current Proton Accelerator," Proceedings of the First International Symposium on Explosive Detection Technology, November 13-15, 1991, ed. S. M. Khan, February 1992.

[39] P. W. Schmor, L. Buchmann, "An H<sup>+</sup> Ion Source for Resonant Gamma Production," Proceedings of the First International Symposium on Explosive Detection Technology, November 13-15, 1991, ed. S. M. Khan, Feb. 1992.

[40] R. E. Morgado, G. Arnone, C.C. Cappiello, S.D. Gardner, C.L. Hollas, L.E. Ussery, J.M. White, J.D. Zahrt, R. A. Krauss, Prototype Explosives-Detection System Based on Nuclear Resonance Absorption in Nitrogen, Los Alamos National Laboratory, Report LA-12776-MS, Los Alamos, NM, June 1994.

[41] D. Vartsky, G. Engler, M.B. Goldberg, R. A. Krauss, A Method for Detection of Explosives Based on Nuclear Resonance Absorption of Gamma Rays in 14N, SPIE, Vol. 2092, Substance Detection Systems (1993) 307.

[42] M. L. Buess, J. B. Miller, A. N. Garroway, Detection of Explosives and Narcotics by Nuclear Quadrupole Resonance, U.S. Patent 5233300.

[43] R. A. Marion, S. M. Klainer, "Multiple Spin Echos in Pure Quadrupole Resonance," Journal of Chemical Physics, 67 (1977) 3388.