GERITRACK tagged neutron inspection system design

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Abstract. The EURITRACK project aims at developing a non-destructive measurement system, using an associated particle sealed tube neutron generator, to detect explosives or other threat materials concealed in cargo containers. Chemical composition of the suspect item is determined by coincidence measurements between alpha particles and photons resulting from neutron interactions in the inspected voxel of the container. We present the design and the performances of the measurement system obtained by Monte Carlo calculations. Selected gamma detectors are clusters of 5”×5”×10” and 5”×5” sodium iodide scintillators, and a block of 100 kg of TNT located in a container filled with a metallic matrix having a density of 0.2 g/cm³ is shown to be detectable in 10 minutes.

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1. Introduction

Non intrusive inspection of cargo containers has become a key issue in recent years to parry terrorist activities. Controls are mainly based on X- and gamma-ray radiography which cannot be used to determine the chemical composition of the inspected material [1].

Within the 6th Framework Program of the European Union, the EURopeline Illicit TRAfficking Countermeasures Kit (EURITRACK) project aims at developing a non-intrusive measurement system, based on the “neutron-in, gamma-out” method, to detect threat materials (explosives, drugs…) concealed in shipping containers.

The measurement system is named Tagged Neutron Inspection System (TNIS) and uses the Associated Particle Technique (APT) [1]. A neutron tube with a built-in segmented alpha particle detector emits 14 MeV tagged neutrons from Deuteron-Tritium reactions to interrogate a delimited volume of the container. The gamma-ray spectrum induced by neutron inelastic scattering on the threat material characterizes its chemical composition.

In this article, the TNIS is designed by Monte Carlo calculations. Gamma-ray detectors are chosen, the needs for shielding and collimation are assessed, and the performances of the system are calculated. Performances are evaluated for 100 kg of TNT explosive located inside the container filled with (1) a metallic matrix composed of Fe-56 having a density of 0.2 g/cm³, and (2) an averaged organic matrix (C_{2.59}H_{5.05}N_{0.55}O_{1.6}) having a density of 0.5 g/cm³. The metallic matrix is the demonstration case of the EURITRACK project and the organic matrix account for the large portion of containers filled with organic goods.
2. Measurement technique

Explosive and drugs can be sorted out from benign materials by the relative amounts of carbon, oxygen and nitrogen [2]. The proportions of carbon, oxygen and nitrogen can be determined using specific gamma-rays generated by \((n,n'\gamma)\) reactions. Most useful gamma-rays are 4439 keV for carbon, 6130 keV for oxygen, and 5108 keV and 2313 keV for nitrogen.

The Associated Particle Sealed Tube Neutron Generator (ASPTNG) emits \(10^8\) neutrons per second in 4\(\pi\) steradians. Time and position of the associated alpha particles are determined using an 8x8 matrix of 6x6 mm\(^2\) YAP:Ce scintillators with individual read out based on a position sensitive photomultiplier tube. The position of the alpha particle defines the neutron emission angle (because alpha and neutron are emitted almost back-to-back) and the time delay between alpha and gamma detections determines the depth of neutron inelastic scattering. The neutron is tagged by the alpha particle.

Determining the chemical composition of a 40x40x40 cm\(^3\) volume with 14 MeV neutrons, travelling at 5 cm/s, requires nanosecond coincidence measurement between the gamma-rays and the alpha particles.

3. Detector selection

Gamma-ray detectors should have: nanosecond time resolution to define inspection voxel of some tenth of centimeters, high intrinsic efficiency for photons of energy upper than some MeV to reduce the acquisition time, large peak-to-total ratio to ease the useful signal extraction from the gamma spectra, sufficiently good energy resolution (<5% at 4.44 MeV) to separate carbon, oxygen and nitrogen peaks, and cheap enough to buy large quantity. Best candidates are barium fluoride (BaF\(_2\)), bismuth germanate (BGO) and sodium iodide (NaI(Tl)) inorganic scintillators [3, 4].

3.1. Calculations

Characteristics of standard BaF\(_2\), BGO and NaI(Tl) crystals for carbon, oxygen and nitrogen gamma-rays are calculated with MCNP [5] for mono-energetic photons entering the crystal and pulse height tallies [5] to yield the energy deposition spectra. Intrinsic full-energy peak efficiency, “effective” full-energy peak efficiency (intrinsic efficiency times the entering surface), and full-energy-peak-to-total ratio are listed in table 1 for 4439 keV photons.

| Scintillator  | Intrinsic full-energy peak efficiency (count/photon) | Effective full-energy peak efficiency (cm\(^2\)) | Peak-to-total ratio |
|--------------|-----------------------------------------------------|-----------------------------------------------|-------------------|
| 3”x3” BaF\(_2\) | 0.17 | 7.9 | 0.40 |
| 3”x3” BGO | 0.44 | 20 | 1.43 |
| 5”x5” BGO | 0.65 | 83 | 3.02 |
| 3”x3” NaI(Tl) | 0.10 | 4.7 | 0.25 |
| 5”x5” NaI(Tl) | 0.24 | 31 | 0.55 |
| 5”x5”x10” NaI(Tl) | 0.39 | 66 | 0.88 |

Main advantages of BGO crystals are the high peak-to-total ratio and intrinsic efficiency. BGO crystals are however limited in size, have poorer energy resolutions, and are 5 to 10 times more expensive than the widespread NaI(Tl) crystals. BaF\(_2\) crystals are interesting because of their fast scintillation component (0.6 ns) and their full-energy peak efficiency. The light output of BaF\(_2\) is however only 20% of that of NaI(Tl), which leads to poorer energy resolution [3]. BaF\(_2\) detectors are also about 5 times more expensive than NaI(Tl) because of the price of the crystal and the quartz window. NaI(Tl) have intrinsically a lower full-energy peak efficiency and peak-to-total ratio than BaF\(_2\) or BGO. However, using larger NaI(Tl) crystals increases the effective full-energy peak and the peak-to-total ratio by reducing the probability for Compton and annihilation photons to escape the crystal. A market survey showed that the 5”x5”x10” NaI(Tl) detector from Saint-Gobain Crystals &
Detectors has the lowest price per unit volume. It is therefore the best compromise between cost and physical considerations for the EURITRACK project. Additional smaller 5”×5” NaI(Tl) will be used close to the neutron generator because of mechanical limitations and high pile-up rate (section 4).

3.2. Measurements

Timing property and energy resolution of 5”×5” NaI(Tl) crystals were measured for Amcrys-H and Saint-Gobain prototypes mounted with the fast XP4512B Photonis PMT and VD105K/01 voltage divider. Measurements were performed at the Soltan Institute for Nuclear Studies by member of the EURITRACK consortium. Time and energy resolutions for 4439 keV photons are lower than 2 ns and 5%, respectively [6]. Timing property and energy resolution measurement of the 5”×5”×10” NaI(Tl) are currently underway.

Evolution of the energy resolution with irradiation was measured for detectors close to the neutron generator. Measurements were performed at the Institute Ruder Boskovic by member of the EURITRACK consortium. The energy resolution of a 4”×4” NaI(Tl) crystal, located 3 cm away from the neutron generator emitting $10^8$ n/s, was studied after three irradiation steps lasting 4, 8 and 8 hours, respectively. Minutes after the last irradiation step, the total count rate due to activation was approximately 60 kc/s and was mainly due to the 666.3 keV gamma–ray resulting from (n,2n) reaction on I-127, and to the 1368 keV an 2754 keV gamma-rays resulting from (n,γ) reaction on Na-23. This activity will need to be subtracted during the measurements. A month after irradiation, the total activity was divided by ten and mainly originates from the 666.3 keV gamma–ray (the half life of I-126 being 13 days). The 5% energy resolution of the detector at 1173 keV and 133 keV (Co-60) was not impaired by irradiation.

4. Design

The TNIS is designed to detect a threat material in a suspect volume previously identified by a classical X-ray radiography of the container. Gamma-ray detectors are grouped in reflection, top and transmission arrays (figure 1). Top and transmission arrays are each composed of four 5”×5”×10” NaI(Tl) detectors to maximize efficiency, whereas the reflection array is composed of four 5”×5” NaI(Tl) detectors to find a compromise between efficiency and pile-up at high count rates (section 6). Depending on the location of the 40×40×40 cm³ inspected voxel, reflection and transmission detectors are movable with the neutron generator along the Z-axis and top detectors along the X-axis.

The neutron generator is modelled in MCNP by a source point located 30 cm away from the container wall. The tagged neutron beam is conic, has a diameter of 40 cm in the centre of the container, and strength of $1.8\times10^7$ n/s. The reflection detectors are 50 cm away from the neutron source and 20 cm away from the container wall. The top and transmission detectors are 5 cm away from the container wall. The four 5”×5”×10” NaI(Tl) are separated and surrounded by 5 cm of lead. The lead shields are 50 cm long and can be used as collimators by moving the detectors away from the container.
5. Collimator

Gamma-ray spectra due to tagged neutron interactions in the metallic matrix and an organic material such as the TNT differ completely. Photons resulting from inelastic scattering on the carbon, oxygen and nitrogen nuclei of the TNT are easily sorted out by their energies and collimation of gamma detectors is not required. However, when the TNT is located in an organic matrix, the location of the tagged neutron interactions is mandatory to single out photons coming from the TNT. The location is primarily controlled by the tagged neutron beam aperture and the time delay between alpha particle and gamma-ray detections. Collimation is an additional mean to improve the location of the detected interactions.

The time distribution of all 4439 keV photons created by tagged neutrons emitted at the initial time is calculated for the TNT block located in the center of the organic matrix. The ideal useful signal due to 4439 keV photons coming from interactions in the TNT only is extracted using the PIKMT card of MCNP [5]. Total and TNT contributions entering the top detectors are shown in figure 2(a) and 2(b) without and with collimator, respectively. The signal-to-background ratio (SBR), defined as the ratio of the TNT to total contribution integrated during the interval time $\Delta t = [30\text{ns}, 40\text{ns}]$, increases from 0.2 to 0.7 when adding the collimators. Collimators for the top and transmission detectors are mandatory to improve SBR for measurement in the organic matrix. The 0.5 m length is a compromise between SBR and geometrical efficiency.
Figure 2. 4439 keV photons flux entering the top detector without collimation (a) and with collimation (b). All photons (solid), photons generated in the TNT (dashed).

6. Shielding
The calculated count rate of each 5"×5" sodium iodide reflection detector exceeds 200 kc/s without shielding. Count rate is mainly due to 14 MeV neutron inelastic scatterings on Na-23 and I-127 [7] and count losses due to pile-up and processing dead time in electronics are expected to be significant.

To shield the detector, we simulate 20 cm thick blocks composed of iron and lead plates around the neutron generator. The iron layer mainly scatters the source neutrons out of the solid angle of the detector. The lead layer absorbs photons created by inelastic scattering in the iron layer and scatters out the remaining neutrons.

The intrinsic properties of the shield blocks are calculated without modeling the container. Plates of 15 cm of iron and 5 cm of lead reduce the total source neutron flux approximately 9 times. 30% of the remaining neutrons have energies lower than 2 MeV and can be removed by a lower energy threshold. Five centimetres of lead is enough to absorb about 2/3 of the gamma-rays generated by the iron layer.

The shield properties are also calculated in realistic conditions, i.e. with the container filled with the organic matrix. The photon and neutron fluxes and count rates are listed in table 2. Neutron and photon fluxes are calculated in a first step with MCNP [5]. The neutron count rate of the 5"×5" sodium iodide detector is then deduced from the neutron flux using an envelope efficiency of 0.3, extrapolated from the efficiency of a 3”×3” sodium iodide crystal [8]. The photon count rate is determined with a pulse height tally [5] in a second MCNP calculation where the source is the photon flux calculated in the first step.
Table 2. Neutron and photon fluxes and count rates for 20 cm thick shields – Organic matrix.

| Shield                  | Fluxes [cm⁻²] | Count rate [kc/s] |
|-------------------------|---------------|-------------------|
|                         | Total (10⁻⁶)  | Neutron/Photon     |
|                         | < 2MeV        | Photon             |
| No shield               | 37.1 / 7.08   | 11% / 59%          |
| 20 cm Fe                | 11.3 / 8.99   | 60% / 66%          |
| 15 cm Fe / 5 cm Pb      | 11.5 / 8.25   | 58% / 65%          |
| 10 cm Fe / 10 cm Pb     | 12.0 / 8.13   | 55% / 65%          |

Comparing to calculations without the container, the neutron and photon fluxes are multiplied approximately 5 and 50 times, respectively. They are dominated by neutrons and photons respectively scattered and created in the organic matrix. Photon spectra are similar for all shields of table 2 and, as previously, 5 cm of lead is adequate to absorb photons created in the iron plate. In realistic conditions, shielding a 5”×5” sodium iodide reflection detector by a 15 cm iron / 5 cm lead block reduces the total count rate from approximately 220 kc/s to 140 kc/s.

7. Useful signal

Useful signal for carbon and oxygen is defined respectively as the count rate due to 4439 keV and 6130 keV photons resulting from interaction of tagged neutrons (section 2) with carbon and oxygen nuclei of the explosive, which deposited all their energy in the detector. Count rates are calculated from photon fluxes using the detector effective efficiencies (section 3).

Theoretically, the useful signal can be defined as the count rate due to all photons coming from interactions of tagged neutrons in the TNT, whatever the energy deposited in the detector. Practically, carbon, nitrogen and oxygen spectra can be extracted by deconvolution analysis. To determine the design of the TNIS, however, the useful signal restricted to specific gamma-rays depositing all their energy in the detector is meaningful and easier to compute.

The detected photon flux of 4439 keV and 6130 keV due to neutron inelastic scattering on the carbon and oxygen nuclei of the TNT explosive, and the associated useful signals, are listed in table 3 when the TNT is located in the centre of the metallic matrix.

Table 3. Useful signal for the metallic matrix – TNT in the centre.

| Detector Location | Detector Type | Fluxes [cm⁻²] | Effective Efficiency [cm²] | Useful Signal [count/s] |
|-------------------|---------------|---------------|--------------------------|------------------------|
| Top               | Four 5”×5”×10” | 10.1 / 6.06   | 264 / 228                | 4.80 / 2.49            |
| Transmission      | Four 5”×5”×10” | 6.10 / 3.79   | 264 / 228                | 2.90 / 1.55            |
| Reflection        | One 5”×5”     | 12.2 / 7.04   | 30.4 / 25.4              | 0.67 / 0.32            |

If the background can be subtracted (section 8) and pile-up effects are negligible, carbon-to-oxygen ratio can be estimated in 10 minutes with a statistical uncertainty lower than 3%, not accounting for background subtraction.

When the TNT is located in the last voxel of the metallic matrix (figure 1), the flux entering the transmission detector is largely predominant. The calculated carbon useful signal is approximately 45 c/s, which is 4 to 5 times higher than when the TNT is in the central position.

For the organic matrix, the useful signal and signal-to-background ratio (SBR) are reported in table 4 when the TNT is located in the first and second voxels of the matrix. Reflection detectors are not collimated but are located close to the TNT whereas top detectors are collimated.
Table 4. Carbon useful signal for the organic matrix.

| TNT location | Detector location | Detector Type | SBR (counts/s) | Useful Signal (counts/s) |
|--------------|-------------------|---------------|----------------|-------------------------|
| Top 1st voxel | Reflection | Four 5”x5” x10” | 19 | 1.43 |
| Top 2nd voxel | Reflection | Four 5”x5” | 12 | 31.3 |
| Top 2nd voxel | Reflection | Four 5”x5” x10” | 3.70 | 0.44 |
| Top 2nd voxel | Reflection | Four 5”x5” | 0.81 | 2.78 |

When the TNT explosive is located in the first voxel, all tagged neutrons first enter the TNT and the background is negligible. The useful signal of the reflection detectors is sufficient to detect the explosive. When the TNT explosive is located in the second voxel, the SBR drops significantly for both detectors but best SBR values are obtained for the top detector because of the collimator. The carbon useful signal is about 3 c/s and is enough to yield acceptable statistical uncertainty within 10 minutes.

8. Random and tagged backgrounds

Useful signal must be extracted from random and tagged backgrounds. Random background is due to accidental coincidences between an alpha particle and photons or neutrons originating from source neutrons not tagged by this alpha particle. Coincidences between an alpha particle and particles originating from the source neutron tagged by this alpha particle are labelled true coincidences. If the particle measured in true coincidence with an alpha particle is a 4439 keV or a 6130 keV photon resulting from inelastic scattering on the carbon or oxygen nuclei of the TNT and if the photon deposits all its energy in the detector, then it is labelled useful signal, else it is labelled tagged background.

The random background during the coincidence measurement is estimated from the total count rate and the fraction of time during which the coincidence time window Δt is open. Total count rate is calculated using the methodology presented in section 6 for the reflection detectors. The fraction of time during which the coincidence measurement is performed is the rate of tagged neutron times the coincidence time window Δt, i.e. $1.8 \times 10^7$ n/s $\times$ 8 ns = 0.15 %. The true-coincidence-to-random-background ratio, calculated with a $10^7$ n/s neutron source, varies from 1 to 3 for different detector positions and matrixes. The random background is proportional to the square of the source strength whereas the true coincidences (useful signal plus tagged background) are only proportional to the source strength [3]. The random background is predominant for higher source strength. Increasing the source strength to enhance the useful signal must therefore be considered cautiously.

Calculated true coincidence pulse height spectra due to all photons (solid line) and to photons generated in the TNT (dashed) are shown in figure 3 for the top detector and the TNT located in the centre of the organic matrix. The tagged background is the difference between the two spectra. Experimentally, the suspect voxel containing the TNT is inspected by the tagged neutron beam, whose angle is defined using a central cluster of pixels of the 8x8 pixels alpha detector (section 2). Other pixels of the alpha detector are used to evaluate the tagged background out of the inspected voxel. The tagged background is then subtracted from the true coincidence to yield the useful signal.
Figure 3. Calculated true coincidence pulse height spectra for the container filled with the organic matrix. All true coincidences (solid), true coincidences due to photons generated in the TNT (dashed).

9. Conclusions and prospects
The EURITRACK project aims at developing a non-intrusive measurement system to detect illicit materials inside shipping containers using the associated particle technique. Design of the TNIS detection system was studied using MCNP calculations. Appropriate gamma-ray detectors were selected and the TNIS performances were evaluated in terms of useful signal and signal-to-background ratio.

The selected TNIS design is composed of two clusters of four 5"×5"×10" sodium iodide detectors located in the top and transmission positions and four 5"×5" sodium iodide detectors in the reflection position. To quicker inspect multiple positions of the TNT inside the container, a financial outlay was granted to buy three additional top clusters of four 5"×5"×10" sodium iodide detectors. The reflection detectors need to be efficiently shielded to limit the number of incoming 14 MeV source neutrons, and the top and transmission detectors need to be collimated to extract the useful signal when the container is filled with organic goods.

For the reference case of the EURITRACK project, which is a 100 kg block of TNT explosive located in the centre of a metallic matrix of density 0.2 g/cm$^3$, calculations show that the TNT explosive can be detected in less than 10 minutes.

With the organic matrix, detection of 100 kg of TNT explosive within the first 80 cm of the container can be achieved in approximately 10 minutes if count losses in the reflection detectors are not too stringent because of the high incoming count rates. Full inspection of the container can be achieved using a second neutron generator on the other side.

Calculation results will be experimentally validated during year 2005 at the Neutron Laboratory facility of the Institute Ruder Boskovic (IRB), in Zagreb, Croatia.

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