

MONTE CARLO SIMULATION OF ACTIVE NEUTRON INTERROGATION SYSTEM DEVELOPED FOR DETECTION OF ILLICIT MATERIALS*

A.SH. GEORGADZE

Kiev Institute for Nuclear Research, Kiev, Ukraine
georgadze@kinr.kiev.ua, a.sh.georgadze@gmail.com

(Received September 6, 2017)

Nowadays, the current threat of international terrorism is set to a severe level, demanding worldwide enhanced security. Radioactive materials that could be fashioned into a radiation dispersal device typically emit gamma rays, while fissile materials such as uranium and plutonium emit both neutrons and gamma rays via spontaneous or induced fission. Therefore, the detection and identification of hazardous materials has become increasingly important. We present the results of Geant4 Monte Carlo simulation of an active neutron interrogation system based on highly segmented neutron/gamma-ray detector and pulsed neutron generator. This system is capable of detecting and imaging radioactive and special nuclear materials, explosives and drugs. The segmented scintillation detector works as a scatter camera, allowing selection of a gamma-ray events that undergo multiple interaction in detector blocks for radioactive source localization. The detector consists of blocks made of plastic scintillator which provide scattering and blocks of CsI, used as an absorber, which has to be efficient to detect the characteristic gamma radiation for the identification. Because of this imaging capability, background events can be significantly rejected, decreasing the number of events required for high-confidence detection and thereby greatly improving its sensitivity. A scatter imager for the detection of shielded radioactive materials has been conceptualized, simulated, and refined to maximize sensitivity while minimizing cost.

DOI:10.5506/APhysPolB.48.1683

1. Introduction

Lately, the threat of international terrorism grows constantly, requiring the increased safety in a whole world. Presently, disorganization of state

* Presented at the 2nd Jagiellonian Symposium on Fundamental and Applied Subatomic Physics, Kraków, Poland, June 3–11, 2017.

administration, task of economic and political harm, compulsion of power are the primary purposes of international terrorism to the change of policy. Bomb terrorism is one of the most widespread. Therefore, detection of explosives and threat materials inside luggage of an aircraft passengers has become an essential requirement.

For the search of the hidden treads, systems based on the direct methods on the use of X-rays, vapor detection and sniffer dogs or on the use of nuclear quadrupole resonance have proved their efficiency [1-3]. However, hermetical casing or impenetrable metallic shell make exposure of the hidden hazardous substances impossible and limit the possibility of their detection. In such cases, the use of radiation with high ability to penetrate and detect characteristic signs of interaction with chemical elements of hazardous substances is necessary.

Prompt gamma-ray neutron analysis [4, 5] has been investigated for over six decades as a possible solution offering an on-stream, non-destructive, rapid method for the luggage inspection at the airports. Objective of this type of inspection is to determine small quantity of explosive of the order of 200 g. The basic problem faced by this technique is that the principal elements which constitute explosives and illicit drugs (H, C, N and O) are present also in the common materials, but with different concentrations. High density and somewhat increased concentration of nitrogen is a characterization of explosive substances. In addition, the background associated with gammas, produced by neutron interaction with surrounding materials mask the signals from explosives. So, the spatial methods have to be applied to reduce backgrounds to an acceptable level.

To overcome background, different methods were developed. One approach provides a coded aperture imaging apparatus, and methods for the detection and imaging of radiation which results from nuclear interrogation of a target object. This method allows to localize the object and decrease background but on the expense of gamma-ray signal intensity loss. Additional shortcoming of this method is that changing the coded aperture masks is required, that resulted in time duration increase for an inspection. Another approach is the Associated Particle Imaging Method [6]. In this case, the characteristic gamma radiation is recorded in coincidence with the signal from detector which registers particles from neutron production reaction $d + t \rightarrow \alpha + n$. This method allows tomography of the explosive object. The only shortcoming of this method is a high price of the neutron generator with α -particle detector embedded in.

In this study, we have performed Monte Carlo simulation with the help of Geant4 package [7] neutron interrogation with fast 14.1 MeV neutrons intended to check the luggage of aircraft passenger on presence of explosive materials. For the detailed test of presence of explosives in the luggage, an

electronic collimation method based on the Compton scattering tracking is developed for the reduction of background from gamma rays, produced by neutron interactions with shielding materials.

2. Design of detection system

The detector (Fig. 1) consists of plastic scintillation bars with size $5 \times 5 \times 20 \text{ cm}^3$ covered by reflecting film of aluminum-backed Mylar and film that contains gadolinium oxide and bars made of CsI or BGO inorganic scintillator. A layer of air between scintillation bars and Mylar ensures full internal scattering of photons. Bars should be connected into one unit of $15 \times 50 \times 20 \text{ cm}^3$ dimensions consisting of two rows of plastic scintillator bars serving as a scatter, and the last row composed from inorganic scintillator for high-efficiency absorption. Additionally, plastic scintillation bars are covered with a film containing gadolinium oxide paint to allow neutron detection. Such a design will make possible to select gammas which have the Compton scattered in plastic and absorbed in CsI scintillator. Since gamma rays with energy more then 3 MeV are scattered at small angle, the backprojection method allows to reconstruct the source location.

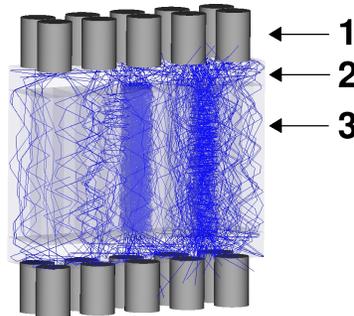


Fig. 1. Gamma-neutron detector scheme (rendered picture): 1 — PMT; 2 — light sharing guide; 3 — scintillation bars.

Scintillation bars are sandwiched on both sides by continuous light guides 5 cm thick. At each side, 10 photomultiplier tubes (PMT) are placed on top of light guides to collect scintillation light. The light sharing guides allow scintillation light produced in one or two bars to be transferred and shared between all 20 PMT almost simultaneously, forming scintillation photon distribution pattern $\{m_1; m_2; \dots; m_{20}\}$. The proposed detector design is based on application of the Anger-logic algorithm which is widely applied in detectors used for the Positron Emission Tomography. The trajectories of the particles under simulation, as well as the simulation geometry, can also be visualized using the VRML visualization driver (see Fig. 2 for illustration).

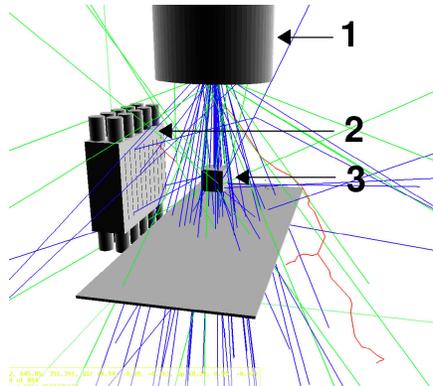


Fig. 2. The 3D – view of neutron interrogation setup: 1 — neutron generator in shield housing from lead and borated polyethylene, 2 — detector, 3 — inspected object.

3. Detector geometry simulation

Full simulations of the detector were performed using *Geant4* package [7]. The scintillation bars were made of PS-923A [8] plastic scintillator produced by the Amcryst company (Ukraine). They were modeled as 95% polished and 5% diffuse (Lambertian). The optical properties of aluminized Mylar film were taken from the *RealSurface1.0* data set [9] of *Geant4* (PolishedVM2000). The data on bulk attenuation length (BAL) of the scintillation 250–450 nm and light output 59% of anthracene, that correspond to 10^4 optical photons/MeV, were taken from [8]. The PMTs were modeled as a 1 mm thick disk with 100% active area, made of borosilicate glass BK7 with refractive index $n = 1.52$. The light detector was placed on outer surface of the PMT glass BK7, and assigned as ideal absorber with 100% active surface and 25% efficiency to detect optical photons.

4. Simulation of explosive detection

The Monte Carlo *Geant4* transport code has been used for the present simulations. A D–T pulsed neutron generator operating at 10^8 neutrons per second has been simulated as fixed point isotropic neutron source. The neutron source was located inside the shield made of lead and borated polyethylene with opening in a form of cone, which serves as neutron flux collimator to ensure that all area of $50 \times 50 \text{ cm}^2$ in size where luggage is located will be covered. The emitted 14.1 MeV fast neutrons produce high-energy gamma-ray signals as a result of the inelastic interaction of fast neutrons with carbon, oxygen and nitrogen nuclei present in an explosive material. The radiation shield made of lead and concrete is surrounding the detector and volume where the luggage inspection takes place.

Simulations were implemented in several stages. In the first stage, the iron was considered as a shielding material. But in this case, large continuous background does not allow to resolve gamma lines. Then lead was used as a shield. After careful choosing of shielding geometry, the gamma lines from C, N and O have become possible to resolve.

In Figs. 3 and 4, we presented gamma spectra of simulation of two second exposure to 14.1 MeV neutron flux of explosive RDX ($C_3H_6N_6O_5$), and an explosive simulant melamin ($C_3H_6N_6$) of 5 cm³ in volume. To take into account an impact of common harmless materials on explosive detection, we have simulated the explosive placed inside the bag of 20 × 20 × 30 cm³ in size, filled with clothes simulated as a cotton of 0.2 g/cm³ density, composed from cellulose cellophane and air, but no considerable impact on detected spectra is found.

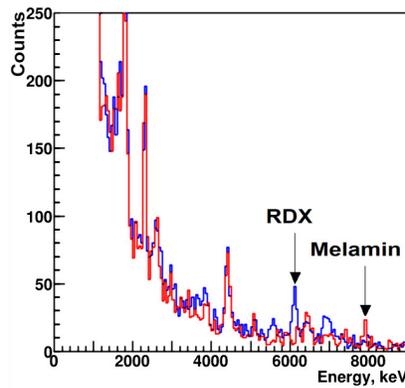


Fig. 3. (Color online) The energy spectra of the RDX (black/blue) and Melamin (gray/red).

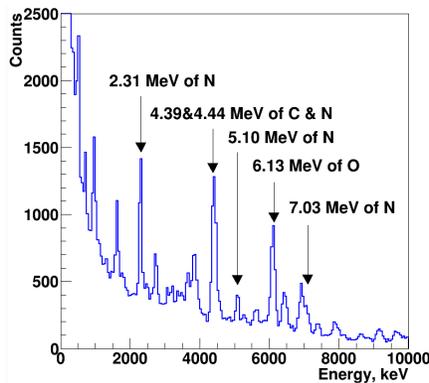


Fig. 4. The energy spectra of the RDX after 120 seconds of testing with spatial selection of gammas.

As seen from Fig. 3, the energy distribution is characterized by three peaks in the spectrum: a peak in the total gamma energy absorption from the line $E_\gamma = 4,43$ MeV from ^{12}C , a peak with $E_\gamma = 5.10$ MeV from ^{14}N and peak with $E_\gamma = 6.13$ MeV from ^{16}O . The RDX spectrum (in black/blue) has characteristic gamma with $E_\gamma = 6.13$ MeV from ^{16}O , but there is no such a line present in the gamma spectrum of explosive simulant — melamin (in gray/red). Another explosive materials used for the simulation are: ammonium nitrate ($\text{H}_4\text{N}_2\text{O}_3$), TNT ($\text{C}_7\text{H}_5\text{N}_3\text{O}_6$) and C4 ($\text{C}_4\text{H}_6\text{N}_6\text{O}_6$).

5. Electronic collimation

In short inspections during several second when the luggage is moving through the detection area, the precise chemical composition cannot be retrieved. In the case of suspicion that luggage is containing illicit material, the luggage can be stopped at the inspection area and additional testing can be performed during several minutes with applied electronic collimation for background suppression.

The simulation of gamma-ray transport in the detection system results in conclusion that approximately 16% of gammas produced by neutrons interactions with inspected object are scattered first in plastic scintillation bar and absorbed in CsI scintillator allowing reconstruction of source location. This is due to the fact that high-energy gammas of several MeVs are scattered mainly on small angles. Due to segmented design, the effect of Compton scattering can be applied to select gammas coming from only inspected area. For the source localization, only coincidence events are selected. Coincidence events are defined as events with non-zero energy deposit in exactly one cell in either one of the scatter plastic scintillation bar, and exactly one in the absorber layer. This ensures the events have gone through a Compton scatter but also minimizes the chance of multiple-scattering events.

The effect of electronic collimation is illustrated in Fig. 5. Gamma quanta coming from different directions resulted in considerably different scintillation photon distribution patterns. To be able to reconstruct the position of the gamma source in the inspection volume, the detector response has to be calibrated as a function of gamma source position. This 3D simulated calibration set was then used to select gammas coming from inside the inspected area by Maximum Likelihood Estimation (MLE).

The energy spectra of the RDX after 120 seconds of additional testing with spatial selection of gammas is presented in Fig. 4. One can see that all gamma peaks from C, N and O are resolved and their intensities can be used for chemical analysis of inspected substances.

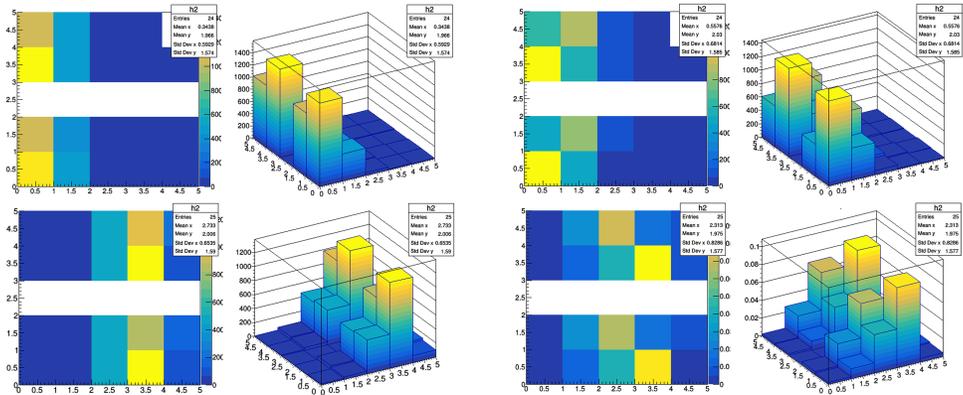


Fig. 5. Photoelectron distribution patterns $\{m_1; m_2; \dots; m_{20}\}$. The left column indicates gamma-ray orthogonal to detector surface and right column indicates gamma-ray beam hitting the detector surface under the angle of 10° .

6. Method of delayed coincidences

The detection of concealed Special Nuclear Material (SNM) in the airport passengers luggage is an important goal for prevention of a nuclear terrorism. Fast neutrons emitted in the neutron-induced fission on SNM can be detected with a plastic scintillator combined with Gd lined neutron–gamma converter. Neutron detection and neutron–gamma discrimination can be done with delayed-coincidence signature of two time correlated pulses, the so-called capture-gated method [13]. The prompt signal is produced by fast neutron scattering on protons in hydrogen rich plastic scintillator material, and delayed signal is produced by capture of thermalized incident neutron on gadolinium nucleus. The latter reaction leads to a cascade of 2–3 gamma rays emitted with total energies of 8.46 MeV (^{155}Gd) and 7.87 MeV (^{157}Gd) that are essentially bigger than energy of events from natural radioactive background with upper limit of 3 MeV. Gd in natural abundance can be used as active $n\text{-}\gamma$ converter due to high abundance of isotopes ^{157}Gd (15.7%) and ^{155}Gd (14.7%).

Another benefit of using this isotopes is the highest thermal neutron capture cross sections among all known nuclides, $\sigma_{\text{cap}} = 254000$ b and $\sigma_{\text{cap}} = 61000$ b, respectively. In simulations, we have considered neutron–gamma converter as a thin film placed in between scintillation bars and made of acrylic (PMMA) mixed with gadolinium oxide (Gd_2O_3) with a Gd/scintillator weight ratio of 0.5%.

Scintillation bars geometry impacts considerably neutron detection efficiency. With larger cross section of scintillation bar, there is the high probability of neutron to be captured by hydrogen with emitting 2.2 MeV gamma quantum which is difficult to detect due to high counting rate in this energy region from the natural background detection. The smaller cross section results in increased light absorption and energy resolution degradation. For chosen geometric dimensions of scintillation bars, simulation results in probability of neutron capture on gadolinium of 0.88 and of an average time of neutron capture — 26 μ s. The efficiency of neutron detection is estimated to be about 10–15%.

7. Enriched uranium detection

Modeling of detector function response to neutrons and gammas with Monte Carlo method shows that high effectiveness for gammas and neutrons detection at the same time can be reached and discrimination between these two particle types can be performed by application of capture-gated method based on delayed coincidence of neutron scattering at protons of plastic scintillator and reaction of neutrons capture by ^{155}Gd and ^{157}Gd nuclei.

Discrimination between gammas and neutrons can be reached due to different quantities of scintillation bars that detected events dealing with interaction of secondary particles with detector material. Preliminary modeling of detector response with application of software code **Geant4** showed that detection effectiveness of neutrons from SNM will be about 10%. Although most fissile materials naturally emit neutrons and/or γ rays, the intensity of the spontaneous radiation is low, and the energies of the γ rays are fairly low in most cases. The active neutron interrogation approach involves bombarding the sample with external neutron source and thereby inducing additional fission in the sample and counting the emitted neutrons due to fission.

To test sensitivity of the system to presence of fissile materials, the enriched uranium sample consisting of 10% of ^{238}U and 90% of ^{235}U with the mass of 200 g was placed in the center of system inside the bag filled with clothes. The clothes were simulated as 0.2 g/cm³ cellulose cellophane material taken from **Geant4** material library mixed with air. The sample inside the luggage was irradiated with 10^8 of 14.1 MeV neutrons, placed inside a shield collimator. Presently, Differential Die-Away (DDA) active neutron interrogation technique was not used for fissile content determination. Instead of that the considerable growing of integral counting rate in the whole energy range with characteristic peaks of full absorption of gammas from neutron capture on gadolinium is an indication of presence of enriched uranium.

8. Conclusions

According to preliminary modeling results of detector parameters, it was determined that there is a possibility to improve explosive detection effectiveness at the account of electronic collimation for source localization in space resulting in essential background reduction. For the neutron generator, intensity of 10^8 n/s, the 400 g of explosives hidden in a luggage can produce an alarm in the presence of suspicious material. Within a few minutes of the additional test by applying the electronic collimation, the chemical composition of the object can be established. Additionally, 200 g of enriched uranium sample can be found with the help of active neutron interrogation.

REFERENCES

- [1] R. Bermbach, S. Vey, *Proc. Int. Soc. Opt. Eng.* **1824**, 199 (1993).
- [2] P.Z. Jankowski *et al.*, *Proc. Int. Soc. Opt. Eng.* **1824**, 13 (1993).
- [3] S. Singh, M. Singh, *Signal Process* **83**, 31 (2003).
- [4] S. Pesente *et al.*, *Nucl. Instrum. Methods Phys. Res. A* **531**, 657 (2004).
- [5] A. Buffler, *Radiat. Phys. Chem.* **71**, 853 (2004).
- [6] J.P. Hurley, The Twelfth International Conference on the Application of Accelerators in Research and Industry, University of North Texas, Denton, Texas, November 2–5, 1992.
- [7] S. Agostinelli *et al.*, *Nucl. Instrum. Methods Phys. Res. A* **506**, 250 (2003).
- [8] A. Artikov *et al.*, *Nucl. Instrum. Methods Phys. Res. A* **555**, 125 (2005); <http://www.detecsciences.com>
- [9] M. Janecek, W.W. Moses, *IEEE Trans. Nucl. Sci.* **55**, 2432 (2008).
- [10] I.A. Pawelczak, *et al.*, *Nucl. Instrum. Methods Phys. Res. A* **629**, 230 (2011).
- [11] E. Fanchini, *IEEE Trans. Nucl. Sci.* **63**, 392 (2016).
- [12] S. Normand, *et al.*, *IEEE Trans. Nucl. Sci.* **49**, 1603 (2002).
- [13] G.F. Knoll, *Radiation Detection and Measurement*, Wiley, 2010.