The Use of Neutron Generators for the Detection of Illicit Materials in the Sea Transportation System

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Abstract. A prototype of portable sealed neutron generator has been recently built to deliver 14 MeV neutron beams tagged by a YAP:Ce α-particle detector. In order to produce simultaneously multiple neutron beams to irradiate complex samples, a study of the position sensitivity of the α-particle detector has been performed. Possible applications in the sea cargo inspections for illicit trafficking and future developments of the Tagged Neutron Inspection System (TNIS) concept are discussed.

1. Introduction

The threat of terrorist use of explosive devices, chemical, biological and radioactive agents has become realistic since the SARIN attack in the Tokyo subway system on March 20, 1995 and after the tragic events of September 11, 2001. The possibility of further attacks against civil populations is one of the most important issues on the international political agenda. A scenario often evocated implies the use of the so called "dirty bombs": a sizeable quantity of radioactive material detonated by conventional explosive and dispersed in the environment. Illicit trafficking of explosives and fissile material through the conventional commercial networks (air, maritime and terrestrial) therefore represents a real challenge to security for the future.

In today's society acts of terrorism must involve in some stages the illicit trafficking either of explosives, chemical agents and nuclear materials. Therefore society must rely on an anti-trafficking infrastructure which encompasses responsible authorities, field personnel and adequate instrumental networks. Manual and visual inspection of large commercial payloads at terrestrial borders (trucks), airports and seaports (containers) would not be a viable solution both from efficiency considerations and for legal reasons. The standoff inspection of cargo by means of imaging and analytical methods in an integrated system based on a sound technology to identify threat materials is then needed.

The key to distinguishing explosives from benign materials is the use of elemental analysis [1,2]. While x-ray or γ-ray based systems (in particular Computerized Tomography) can give good precision density measurements with high-resolution three-dimensional images, these systems provide at best only gross information about the elemental content of the inspected item (low Z vs. high Z). Neutron interrogation, however, offers the possibility of measuring the elemental density of most elements in materials. All neutrons, in particular fast neutrons, are well suited to explore large volume samples because of their high penetration in bulk material. Fast neutrons can be produced efficiently and economically by natural radioactive sources, small accelerators or compact electronic neutron generators, making possible the use of neutron based techniques in field applications. Gamma-rays produced by irradiating the sample with neutrons gives the elemental composition of the material, moreover, knowing the

nuclear cross-sections and estimating the absorption factors in the different materials, it is possible to perform a quantitative analysis of elements in the sample even in depth; this is the most suitable technique for detecting hidden explosives. Furthermore with the use of "tagged" neutrons it is possible to determine the local distribution of elements inside the sample volume, or to inspect a precise element of volume (voxel) that has been identified as suspect [3,4]. Secondary neutrons and γ-rays produced by the irradiation of the sample can be used, by means of multiplicity measurements and/or spectral analysis, to identify the presence of fissile materials in the inspected volume. The list of materials which are subject to inspection with the aim of reducing the acts of terrorism includes explosives and radioactive materials.

The first explosive known was gunpowder, also called black powder. In use by the 13th century, it was the only explosive known for several hundred years. Nitrocellulose and nitro-glycerine, both discovered in 1846, were the first modern explosives. Since then nitrates, nitro compounds, fulminates, and azides have been the chief explosive compounds used alone or in mixtures with fuels or other agents. Today we have a list of some 100 explosive materials. However, the most often used are: Trinitrotoluene (TNT), Tetryl, Tetrytol, Hexatol, PETN and RDX.

There is a real risk that sub-national groups will in the future acquire fissile material – particularly plutonium – and construct a nuclear explosive. Equally disturbing, and perhaps more likely, is the possibility that plutonium may be acquired by a group who will threaten to disperse it, by an explosion, and radioactively contaminate a large urban area. These risks exist because: a large amount of civilian plutonium is being produced and stockpiled; a relatively small amount of such plutonium is needed for a nuclear explosive; the technical information required to fabricate a nuclear device is available in the open literature; and only a small number of competent people are necessary to fabricate a primitive nuclear device. The design of a “first generation” nuclear weapon, such as the bomb that destroyed Nagasaki in 1945, is no longer a secret; a competent nuclear physicist can find the relevant information in the open literature. Several types of these devices are possible: (i) Gun type and (ii) Implosive types. Implosion seems to be a favourite “first try”. For example Iraq’s importation of explosives and electronics suggested development of an implosion-type bomb. For this last type high-explosive charges are required. The amount of high explosive used in a fission weapon has decreased considerably since 1945 – from about 500 kg to about 15 kg or less. Explosive lenses and detonators adequate for an implosion-type atomic bomb are commercially available. Although a sub-national group could choose to use either plutonium or highly enriched uranium as the fissionable material for nuclear explosives, plutonium is increasingly, the more likely option. A sub-national group that in the future decides to manufacture a nuclear explosive device is, therefore, most likely to try to steal or to buy plutonium.

2. Detection of explosives and fissile materials by “Associated Particle Technique”

Of particular interest in the detection of conventional explosives are the densities of nitrogen, oxygen, carbon and hydrogen and their ratios. Neutron interrogation elemental analysis available till now makes use of radioactive neutron sources (^{252}Cf or AmBe), particle accelerators or compact electronic neutron generators, all these neutron sources produce a nearly isotropic flux of neutrons. The use of detection devices based on such sources could represent a viable solution in applications where the inspection volume is either limited in space or nearly homogeneous in chemical composition; the amount of material of different nature encountered inside a commercial container would severely impair their detection capability. Indiscriminate irradiation of the container itself, of the container load and of the

structures of the inspection system would in fact provide γ-ray spectra representative of the average elemental composition of all the objects in the radiation field.
The technique described in this paper makes use of the "tagged" neutron method which has been the subject of studies performed for the detection of buried landmines in humanitarian de-mining operations.
In the fusion reaction of deuteron with tritium at energies of around 100 KeV the main decay channel produces an α-particle of about 3.5 MeV and a neutron of 14.1 MeV that are emitted nearly back-to-back . These reactions are easily produced in laboratory electrostatic accelerators and are the basis of compact neutron generator systems. The possibility of detecting the α-particle with a fast position sensitive counter of a given small area defines a tagged beam of neutrons emitted in the opposite direction and thus defines a corresponding specific area irradiated by the neutron beam and identified in X-Y coordinates by simple geometric projection. The tagged neutrons of energy 14.1 MeV travel at a fixed speed of about 5 cm/ns, therefore the γ-rays emitted by the elements in the irradiated area can be recorded in time coincidence with the tagged neutron thus defining the third dimension (Z-coordinate) of the irradiated volume.
In this way the region from which the recorded γ radiation originates can be accurately chosen by an "electronic" definition of the volume element (voxel) allowing a significant reduction of the background signals produced in the entire surrounding environment. The definition of the voxel to be inspected is then a crucial parameter in the choice of the appropriate equipment since the lower limit in its size is determined by the characteristic of the "tagging" detectors and of the γ-ray counters.
Radioactive materials can be detected by measuring their decay products by "passive" radiation measurements; however the large volume of a standard container and the shielding of the load to the low energy γ radiation might make their identification very difficult when hidden in containers. Neutron induced β-delayed fission-neutron emission is also a possible way of detection for most fissile material, but the typical long time delay between the primary neutron and the delayed ones makes this technique unreliable for as large inspection volume such as a standard container. The loss of time correlation between neutron-in and neutrons-out would "disperse" the true signal among the background generated by neutron scattering inside the container. The technique described here consists of irradiating a small portion of the inspected volume by means of a tagged neutron beam of 14.1 MeV and measuring in coincidence the shape of the time-of-flight spectrum of neutrons produced by prompt fission decay induced in the fissile material. Such technique would overcome the problem of dealing with a "diffuse" signal of the fissile elements inside a large cargo load making use of a limited number of dedicated neutron counters to measure neutron spectra both by time-of-flight and by amplitude unfolding. Also in this case the third dimension (Z coordinate) of the inspected voxel will be defined by an accurate time requirement between the tagged neutron beam and the secondary out coming neutrons. Use of efficient scintillators like NaI(Tl) crystal allows to measure average multiplicity and spectra of γ-rays emitted in the neutron induced fission events, giving a complementary information on the identification of fissile materials in the cargo.

3. Tests of "Associated Particle" performances

A YAP:Ce scintillator [5] of 4 cm diameter and 0.5 mm thickness has been selected for the present application. The YAP:Ce exhibits, indeed, several characteristics that are very well suited for this task: excellent mechanical and chemical properties, radiation hardness, fast response and high light output [5]. The scintillator has been mounted on a standard stainless steel CF63 flange equipped with a sapphire window (3 mm thick, 48 mm diameter)

without any optical grease, as required in the sealed neutron generator manufacturing procedure [15]. The surface of the YAP:Ce crystal was coated with a layer of 1 mg/cm^2 of metallic silver to maximize the light collection, stop the elastically scattered deuterons and protect the crystal from the UV glow inside the neutron generator. Since the counting rate capability and the time resolution are the major goals of the present application, we used a small diameter fast PMT Hamamatsu R1450, without light guide. The tagging system (i.e. the YAP:Ce mounted on the optical flange with the PMT and the associated front-end electronics) has been tested with neutrons produced in the D+T reaction making use of the 150 kV electrostatic neutron generator at the Institute Ruder Boskovic (IRB) in Zagreb.
The experimental system used at IRB is shown in Fig. 1. The neutron tagging detector is placed inside the reaction chamber at approximately 8.8 cm from the target at 90^o degrees with respect to the beam direction.

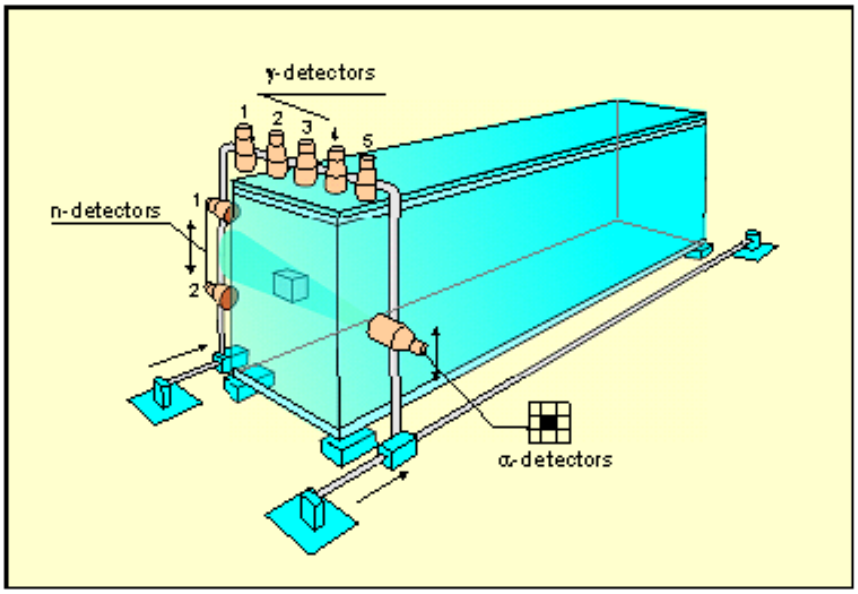

Fig. 1

For this test a 5.5 mm diameter collimator has been positioned in front of the YAP:Ce detector in order to limit the size of the tagged neutron beam at the inspection plane which is defined by the gamma detector. As a test to measure the effectiveness of the method on the signal-to-noise ratio we have irradiated a graphite sample of 5x5x5 cm^3 located at about 90 cm from the source of neutrons. The detection of the 4.4 MeV γ-rays emitted by the ^{12}C first excited level populated by neutron inelastic scattering by means of a 4"x 4" NaI(Ce) scintillation detector have been used to determine the improvement of the spectrum quality. The results of the irradiations is shown in Fig. 2: on the left panel is shown the γ-ray spectrum detected without requiring a strict coincidence between the alpha particle and the associated neutron hitting the sample. One can see a peak between 5 and 6 MeV due to neutrons hitting directly the NaI counter, and the 4.4 MeV and "first escape" peaks associated to the decay of ^{12}C in the sample. On the right-hand side is shown the same spectrum but with a strict condition (of about 2-3 ns) on the alpha-gamma coincidence time.

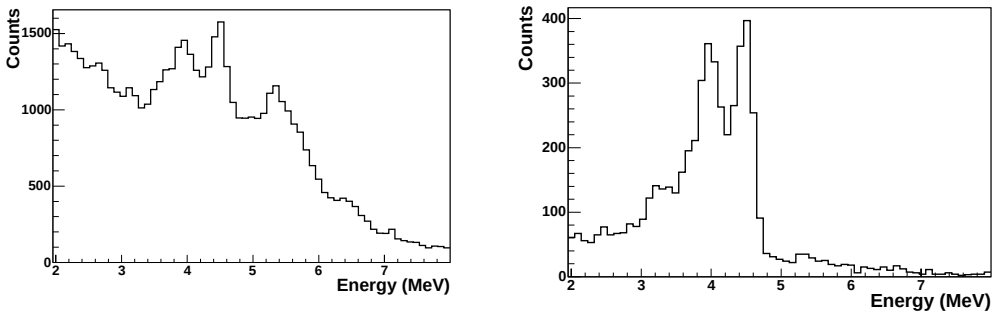

Fig. 2

One can notice a dramatic improvement of the signal-to-noise ratio due to both the "geometrical" and "timing" related reduction of the background contribution.

4. A position sensitive alpha particle detector for "Associated Particle Imaging"

In order to produce multiple neutron beams of variable sizes, to possibly mitigate the loss in signal rate associated with small beam sizes and maintain the ability to inspect complex samples, a study of the position sensitive α-particle detector has been performed. Two systems have been built and tested with the 40 mm diameter YAP:Ce crystal: the first uses 3 fast PMT's Hamamatsu R4141 (13.5 mm diameter) and the second one employs a 2×2 multi-anode PMT Hamamatsu R5900U-00-M4 (18×18 mm^2).
In both cases, the PMT photocathode diameter is smaller than the YAP:Ce crystal. Consequently, the light collection efficiency depends on the relative position of the α-particles on the detector surface with respect to the PMT. Such effect has been exploited in order to link the measured pulse height values to the distance r between the α-particle and the PMT center. The pulse height distributions have been studied for different positions of an α-particle source (^{241}Am) placed behind a mask with several 1 mm diameter holes. The pulse height distributions have been fitted by a Gaussian function determining average amplitude and width values. Using such experimental data, average amplitude and width values have been parameterized as a function of the distances r. Consequently, each point of the YAP:Ce crystal corresponds to a set of three (for the 3 PMT's) or four (for the 2x2 multi-anode PMT) calculated average amplitude (A_i^{calc}) and sigma (σ_i^{calc}) values. When an α-particle hits the YAP:Ce crystal in an unknown point, the measured amplitudes (A_i^{exp}) are compared with those calculated for all (x,y) positions evaluating the function:

$$f(x,y) = \sum_{i=1}^{3} \left(\frac{A_i^{\exp} - A_i^{calc}(x,y)}{\sigma_i^{calc}(x,y)} \right)^2$$

The impact position of the α-particle is then reconstructed with a search for the coordinates that minimize the *f(x,y)* function. The reconstruction algorithm has been tested for both configurations (3 PMT's and multi-anode PMT) by means of the above mentioned mask. As an example of this position reconstruction, the comparison between the "true" positions of the holes (circles) and the reconstructed ones (dots with error bars) is shown in Fig. 3 for the case of the multi-anode PMT. The central positions of the 4 PMT sectors are also marked. The RMS values of the reconstructed position distribution vary from 0.5 mm in the center of the PMT to 1.2 mm in the periphery, thus defining the smallest neutron beam achievable with this technique.

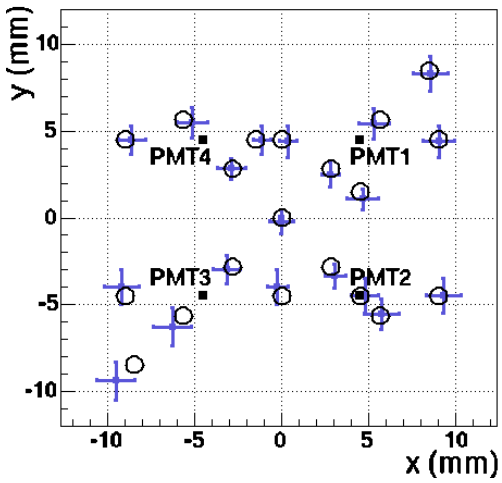

Fig. 3

A test with a collimator with a cross-shaped aperture of 7 ×1 mm^2 placed in front of the YAP:Ce detector was performed obtaining the reconstructed positions shown in Fig. 4. On the left panel is shown a 2D image of the collimator, the grey cross indicates the real dimension of the cross, in the right panel is shown the 3D image of the same collimator.

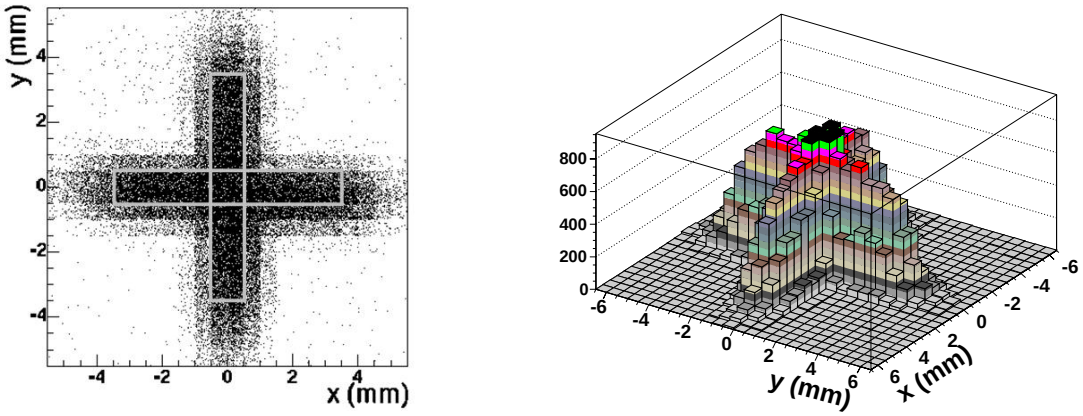

Fig. 4

The system with the 4 sector PMT has also been tested at IRB by detecting alpha-gamma coincidences produced in the irradiation of two 5×5×5 cm^3 graphite samples with a cubic 10×10×10 cm^3 NaI(Tl) scintillator. The tritium target was at 8.8 cm from the YAP:Ce detector and at 88 cm from the plane defined by the two graphite samples. The average distance between the two samples and the center of the NaI(Tl) detector was about 25 cm. The NaI(Tl) was protected from direct neutrons by a heavy metal shadow bar. The position distribution of the alpha-gamma coincidences on the YAP:Ce detector is shown in Fig. 5 as obtained by setting windows on the time of flight of the neutrons hitting the graphite samples and on the NaI(Tl) energy corresponding to the 4.4 MeV γ-ray produced by (n,n'γ) reactions on ^{12}C nuclei.

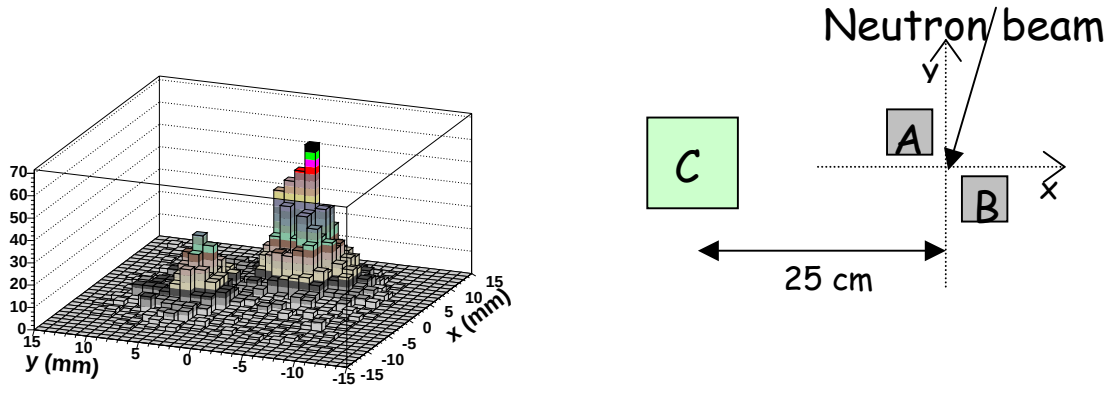

Fig. 5

On the right panel is shown the position of the NaI(Tl) detector with respect to the two graphite samples and the neutron beam axis. The distance between the corners of the samples A and B is of 4 cm, on the left panel is the 3D image of the α-particles hits in coincidence with the 4.4 MeV γ-rays from the graphite samples.
Two well defined structures in the reconstructed YAP:Ce (x,y) positions are clearly separated thus providing a direct test of the imaging capability of our prototype. The different yields of the two structures are due to the difference in the solid angles subtended by the two samples with respect to the NaI(Tl) detector.

5. First measurement on Depleted Uranium (DU) samples

A first test of the prompt fission neutron production from irradiation of a sample of DU has been recently performed at IRB. A block of about 10 kg of DU has been placed inside a maritime container in the direction of a neutron beam defined by the tagging of our α particle detector. An NE213 liquid scintillator based 4"x 2" neutron counter has been placed on the side of the DU sample at a distance of 75 cm as shown in fig. 6.

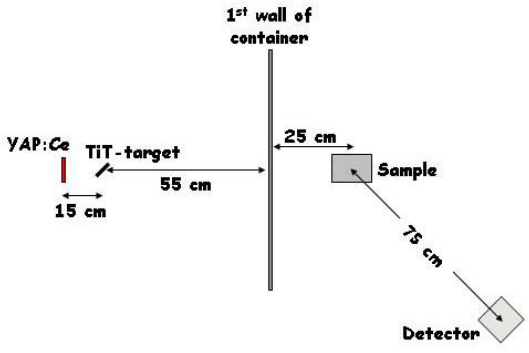

Fig. 6

A time-of-flight (TOF) measurement between the α particle and the neutrons detected has been used to have indications on the possibility of identifying fissile materials by direct prompt neutron measurements. Two samples, one of lead and the second of DU, both of about the same mass (10 kg) were irradiated in two separate runs.

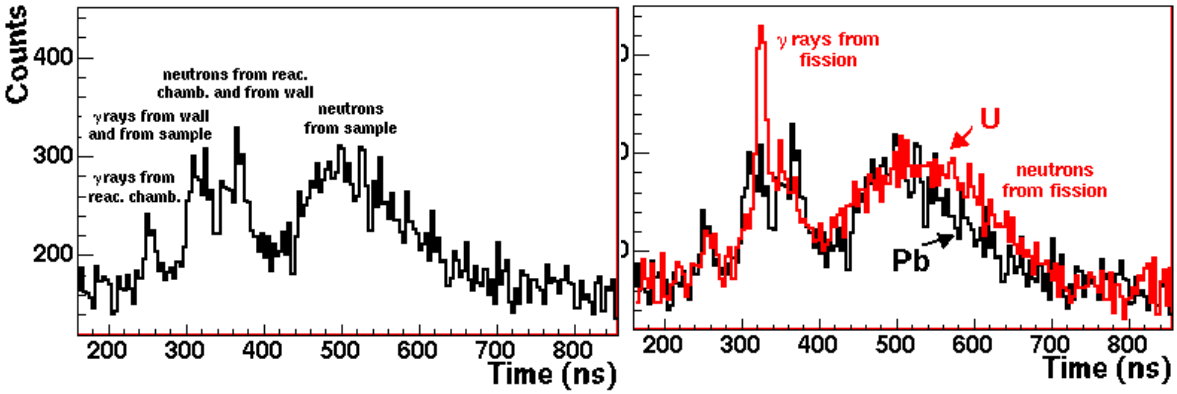

Fig. 7

In the left panel of Fig. 7 is shown the TOF spectrum relative to the Pb irradiation, one can notice all the peaks related to neutron scattered as well as γ-rays produced on various points of the set-up. In the right panel is shown the TOF spectrum relative to the DU irradiation (red histogram) compared to the previous one. One can notice that there is an extra yield of neutrons on the low energy side (high TOF) that are probably due to the fission events induced in the DU sample. One also notices an even more sizeable increase in the γ-ray yield due to the fission events compared to the lead irradiation. In fact the average γ-ray multiplicity per fission in DU is about 7.2 compared to a fission neutron multiplicity of about 2.4.
More experimental work is foreseen using an array of 4 NaI(Tl) gamma ray detectors in order to verify the possibility of detecting fissile materials by simultaneous measurements of γ-ray and neutron multiplicities.

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