Study of the Associated Particle Imaging technique for the hidden explosives identification

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Abstract

We have investigated main parameters of a laboratory prototype of the device for remote nondestructive identification of explosives, hidden in containers or soil. The device is based on the Associated Particle Imaging (API) technique and was installed on the electrostatic Van de Graaf accelerator. It has been shown that 400 g of TNT shielded by 42 cm of soil are detected within 3 minutes. The sample of 100 g of melamine shielded by 9 cm of soil is detected within 5 minutes. An important distinctive feature of the API method pertains to its sensitivity to chemical properties of the illicit substance. We have demonstrated reliable identification of the explosives on the background of other substances. The unique advantage of the API method is to provide the image of the hidden object in 3-dimensions. It also significantly suppresses the background. A large signal-to-background ratio provided by the API method significantly facilitates the identification of the hidden substances.

Key words: Neutron Probing; Associated Particle Imaging; Remote Identification; Explosives

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1. Introduction

The Associated Particle Imaging (API) is a technique which could provide a 3D imaging and identification of objects hidden in different containers or soil. The technique is non-intrusive and could be used for demining operations, counterterrorist activities, search for illicit substances by customs, as well as for different industry applications. The study of the API method is carried out at the Los Alamos National Laboratory [1], Argonne National Laboratory [2], Bechtel Nevada Special Technologies Laboratory [3], INFN (Italy) [4], JINR [5] as well as at other laboratories (for review, see, [6,7,8]).

This report describes the results of recent experiments on the study of the properties of the laboratory prototype of the device for remote nondestructive identification of illicit substances, based on the API technique, which have been performed at the Joint Institute for Nuclear Research (JINR) in Dubna (Russia).

The API technique uses fast monochromatic neutrons with energy of 14.1 MeV produced in binary reaction $d+t \rightarrow {}^{4}He+n$. In this reaction the α-particle with the energy of 3.5 MeV flights back-to-back with the neutron (in c.m. system). By measuring the α- particle trajectory the direction of the corresponding neutron is determined. These «tagged» neutrons interact with the interrogated object and can produce γ quanta in A(n,γ)A' reactions with energy spectra which are unique for each chemical element in the object. The characteristic γ-spectra could serve as "fingerprints" to identify the hidden substance.

The key feature of the API method is the measurement of the time difference between detection of α particle and γ quantum. It provides the possibility to determine the distance traveled by the neutron before it is inelastically scattered by the nuclei of the interrogated object. It is thus possible to determine a gamma-spectrum from the definite localized region in the object. It strongly suppresses the background. A large signal-to-background ratio provided by the API method significantly facilitates the identification of the hidden substances. The unique

advantage of the API method is to provide the image of the hidden object in 3-dimensions. The use of the fast 14 MeV neutrons significantly increases the probing depth in the investigated object.

An important distinctive feature of the API method pertains to its high sensitivity to chemical composition of the illicit substance. We have performed some tests to verify the ability of distinguishing the explosives among different substances.

2. Experimental setup

2.1. Accelerator and target

The apparatus is installed at the deuteron beam of the electrostatic Van de Graaf accelerator of the Laboratory of Neutron Physics at JINR. The molecular ion beam D^+_2 with the energy of 450 keV per deuteron is used. The beam intensity is about 0.5 μA. The position of the deuteron beam at the target is collimated by two apertures of 4 mm in diameter. The target is a titanium tritide TiT_2. It has been deposited on stainless steel substrate placed at 45^0 angle to the beam axis. The tritium layer density is $\approx 10^{19}$ atom/cm^2. The total activity of the target does not exceed 1.7 Cu. The target and the beam control system are placed in the vacuum chamber (see Fig. 1). The neutron flux generated in the d+t-reaction is $(1\text{-}5)\cdot 10^7$ s^{-1} at the 4π solid angle.

2.2. Alpha detector

The α particles are detected by 4 decoupled scintillation counters with a size of 10×10 mm^2 each. Different alpha - detector scintillators – ZnS, LSO, GSO, YaP and plastic have been tested. They are located at 75 mm from the target. The scintillators are shielded from deuterons scattered off the target by the 7μm aluminum foil. The typical α- particle counting rate is $\approx 10^4$ s^{-1}. The amplitude spectra of the α-particles from the plastic scintillator are presented in Fig. 2. To control the alpha-detector, we have glued a source of ^{241}Am at the surface of the scintillator. The calibration spectrum of ^{241}Am is shown in Fig.2 multiplied by a factor of $2\cdot 10^3$. A small tail

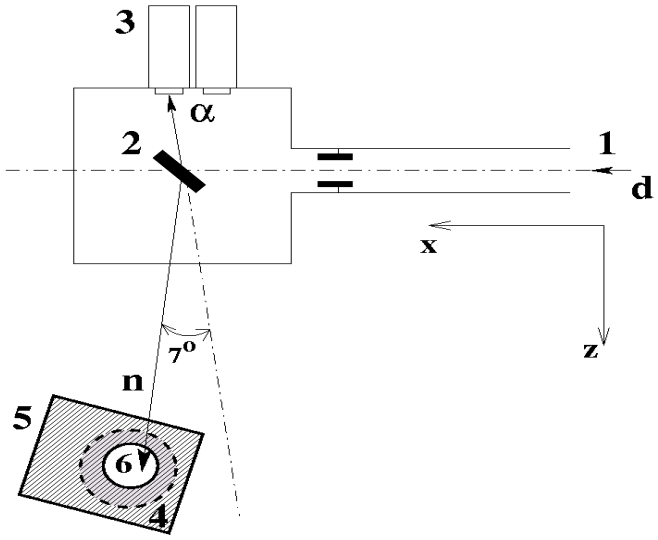

Figure 1. The scheme of the experimental setup.

1. Deuteron beam line of the accelerator.
2. TiT_2 target.
3. Alpha detector.
4. Gamma detector (dashed line).
5. Interrogated object, box with soil
6. Hidden simulant of explosives

can be seen on the left side of the ^{241}Am peak, which is due to residual radioactive background after accelerator operation.

The average energy of the detected α particles from the dt-reaction is about 2.3 MeV. It is less than 3.5 MeV expected from the dt-reaction due to energy losses in the TiT_2 target (~0.4 MeV) and the aluminum foil (~ 0.8 MeV).

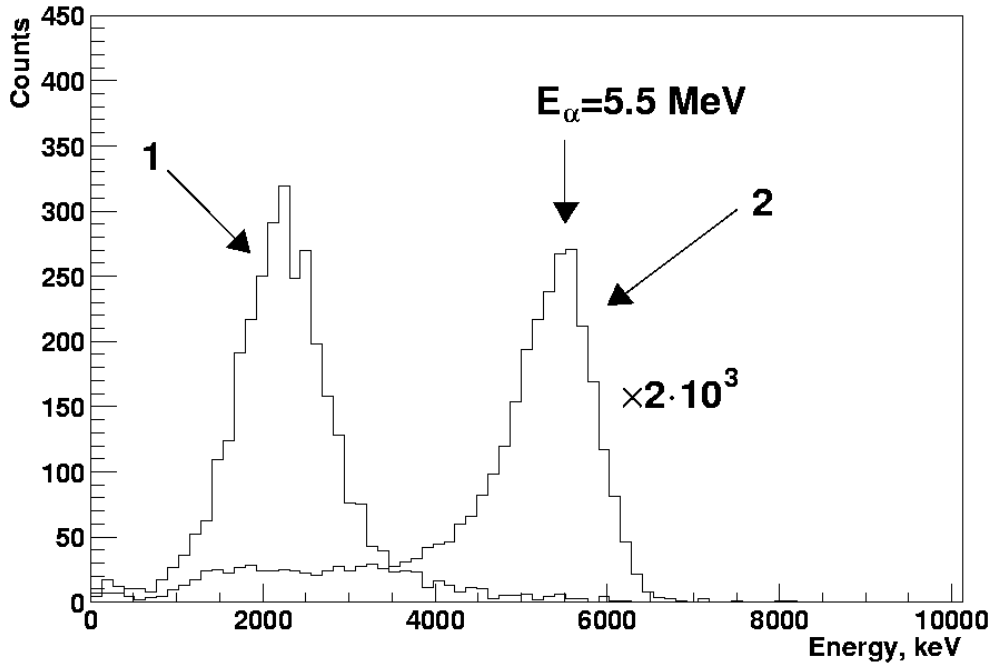

Figure 2. The energy spectra of the α-particles (1). The calibration spectrum of ^{241}Am is shown multiplied by factor $2 \cdot 10^3$ (2).

2.3. Gamma -detector

The γ-rays from (n,γ)-reactions are registered by the NaI(Tl) detector with the scintillator of ∅ 150 mm and thickness 100 mm. The energy resolution of the gamma – detector is 8% at E_γ =1.33 MeV. The time resolution of the α - γ system is determined in the experiments with tagged neutron beam irradiating of 3 cm graphite plate. The time resolution of the α - γ coincidence is Γ=2.93±0.09 ns. It determines mainly by the NaI(Tl) as the slowest element. The spread in the time of flight of α-particles caused by the different ionization energy losses in different points of the TiT_2 target is estimated as 0.4 ns.

By using a plastic scintillator as a tagged neutron detector, it has been measured that the α-n time resolution is Γ=2.71±0.08 ns.

The NaI (Tl) detector is shielded by 10 cm of lead. A block of 35 cm thick made of boron-doped polyethylene is placed between the neutron source and the γ-ray detector. The distance between the interrogated object and gamma-detector is varied within the range of 30-50 cm.

2.4. Tagged neutron beam

To determine the position of the tagged neutron beam, a movable plastic scintillation counter of 4 x 4 x 0.5 cm^3 was used. In the laboratory frame the center of mass of the d+t system moves along the X-axis (see Fig.1). It slightly breaks the back-to-back correlation between the α-particle and neutron trajectories, existing in the c.m.s. It has been found that each tagged neutron beam axis deviates from the normal to the accelerator beam line at an angle of $\approx 10^0$. This value is in agreement with the calculated value, based on the kinematics of the dt - reaction. The distance between the tritium target and the interrogated object was varied in the 70-130 cm interval. The velocity of the 14.1 MeV neutron from the dt-reaction is 5 cm/ns.

2.5. DAQ system

The data acquisition system is assembled in the CAMAC standard. The PMT's signals from α and γ detectors are fed to the constant fraction discriminators and to the time shapers, and further - to the α - γ coincidence block together with the signal ” busy” generated by the controller. The controller is equipped with the processor. The α - γ coincidence block generates a signal of interruption for the processor and strobe for the analog-to-digital converter (ADC). The signals from PMT pass through amplifiers and fed to the ADC, which generates the amplitude spectra. To perform the time measurement, the signal from the α-detector passes the shaper and fed to the time-to-digital converter (TDC) start, the γ-detector signal is fed to the TDC's stop. The selection of the α - γ coincidences is performed within 80 ns gate. The crate controller with

build-in PC performs the data readout from the converters and the data acquisition control. The information is stored on the build-in hard disk and transferred via the local net.

3.Results

3.1. Time distribution and background suppression

To illustrate the principal advantages of the API method let us consider the time and energy distributions of characteristic gamma radiation from the interrogated substances, registered by the NaI(Tl) detector. A typical time-of-flight spectrum is shown in Fig.3a. It has been collected for a sample of carbon 10x10x10 cm^3, placed at 115 cm from the tritium target. The start of the time-of-flight measurement is given by the α particle signal and the stop is given by the γ signal. If the γ signal is absent, then the compulsory stop signal is generated within 80 ns after the arrival of the α signal.

The two-peaks structure of the time spectrum superimposed on the flat background is clearly visible in Fig. 3a. The flat part of the spectrum corresponds to the accidental α – γ-coincidences. The first peak corresponds to the detection of the prompt γ produced in the reactions $A(n, n'\gamma)A$ for the interaction of the tagged neutrons with the sample. The second peak is caused by the neutrons scattered in the sample and detected by the NaI(Tl) counter later. The delay time of the second peak is about 10 ns. It corresponds to the time which is needed for a neutron to pass the distance of 50 cm between the sample and the detector with the velocity of 5 cm/ns. The clear separation of two peaks significantly improves the discrimination of the signal from the background.

The Fig. 3b shows the energy spectrum of the γ-quanta for the time gate indicated in Fig.3a by the shadow region around 35 ns. One can see a clear twin peak feature from the carbon line of 4.43 MeV with its single escape (SE). The Fig. 3c shows the energy spectrum for the time gate indicated in Fig.3a by the shadow region around 45 ns. This spectrum is smooth, without any

trace of carbon line at 4.43 MeV. This distribution is typical for the delayed neutrons rescattered in the graphite cube.

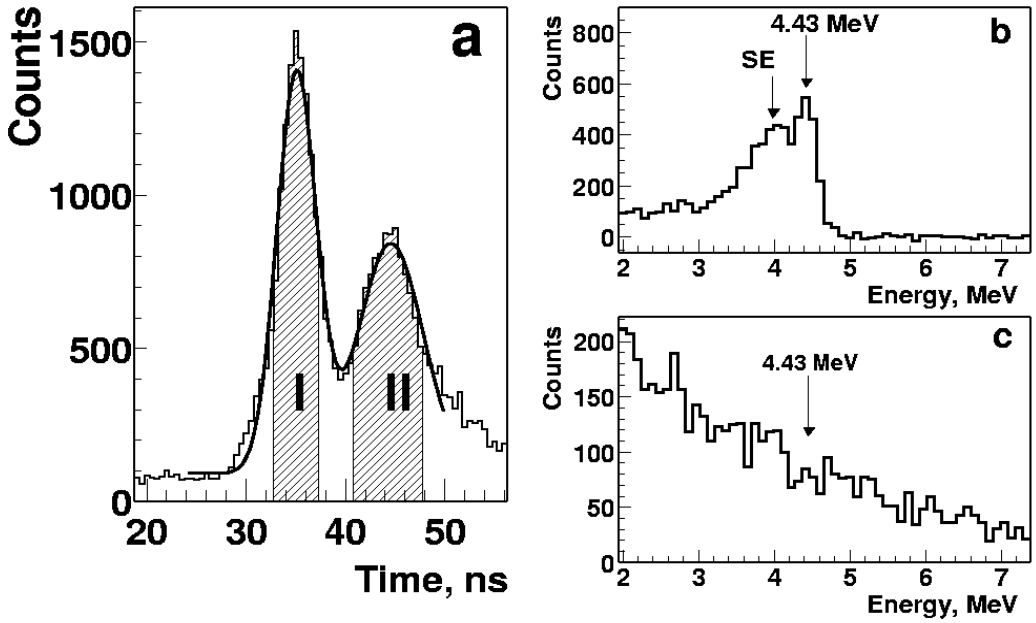

Figure 3. The time-of-flight spectrum of α – γ-coincidences for 10 cm cube of ^{12}C (a). The energy spectrum of γ quanta for the time interval indicated by the shadow region around 35 ns (b) and 45 ns (c). The carbon line at 4.43 MeV and its single escape (SE) is clearly seen at (b). The width of the first time-of-flight peak in (a) is Γ=4.3±0.5 ns.

This picture illustrates two main advantages of the API method. First, the knowledge of the time-of-flight provides the location of the spatial region under investigation. Second, the selection of the α – γ-coincidences within a certain time interval drastically suppresses the background. Without the time selection the resulting γ-spectrum is superposition of the energy spectra from the interrogated substance (Fig.3b) and the background from the scattered neutrons,

like that shown in Fig.3c. The background suppression is essential for enhancing the sensitivity needed to identify small mass objects.

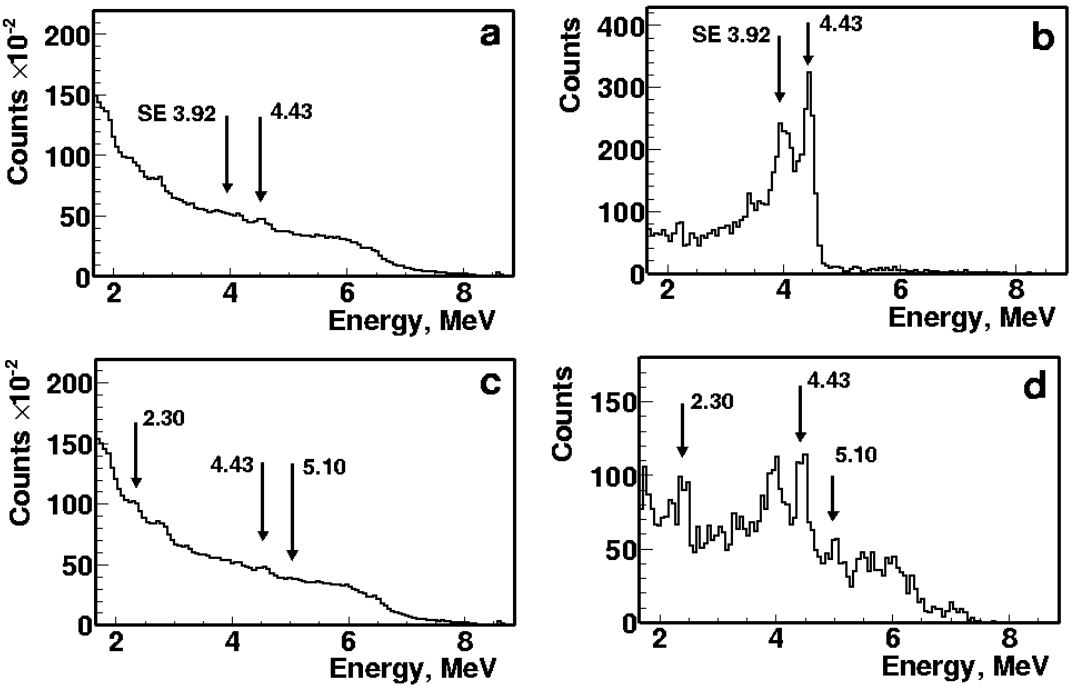

Figure 4. The γ- energy spectra of carbon block (upper part) and the melamine (bottom) without time selection (left) and with time selection (right).

Large value of the signal-to-background ratio is an important advantage of the API technique. In Fig.4 we compare the energy spectrum of the γ-quanta obtained by the traditional method of irradiation of 14 MeV neutrons without α – γ-coincidences (left column), with the spectrum of the same samples obtained by the API method (right column), selecting α – γ-coincidences belonging to the first peak of time distribution as in Fig. 3a. The upper row shows the spectra of carbon block (2.4 kg) and the spectra on the bottom row of Fig.4 are of the explosives simulant of melamine ($C_3H_6N_6$). The spectra obtained with the API method

demonstrate clear γ-lines of ^{12}C at 4.43 MeV, of ^{14}N at 5.10 and 2.30 MeV as well as corresponding single escape (SE) lines. In case of ordinary neutron irradiation these peaks are sitting on the large background. These experiments show that the ratio signal-to-background for the API method is larger than for the ordinary neutron irradiation by factor of 230-240.

It significantly facilitates the detection of the small quantities of the hidden substance

3.2. Identification of the hidden illicit substances

To identify the hidden illicit substances the experiments are done in three steps. First, the measurement of the background conditions in the laboratory is performed. Second, the measurement of the reference spectrum of the not-shielded illicit substance is carried out. Third, the trial with the shielded investigated sample is performed. The process of identification is based on the comparison of the spectra of the investigated substances and the reference one. Various decision taking algorithms have been tested.

To demonstrate the potential of the API method, we present here the results of the experiments on identification of the TNT explosive ($C_7H_5N_3O_6$). The sample of 400 g TNT is buried in the box with soil. The sample is shielded by the 25 cm of soil in front of the box and 9 cm of soil in front of NaI(Tl). We have measured spectra of TNT buried in the box and the box of the soil without TNT. The results are shown in Fig. 5.

The time distribution of γ quanta for the TNT buried in the soil is shown in the first plot (upper row, left) by the solid line, the time distribution of γ quanta for the box without TNT sample is shown by the dashed line. The distributions are normalized on equal number of the tagged neutrons. One can see that these two distributions are different. In the next plot (upper row, right) the difference between these time distributions is shown. The picture shows a large

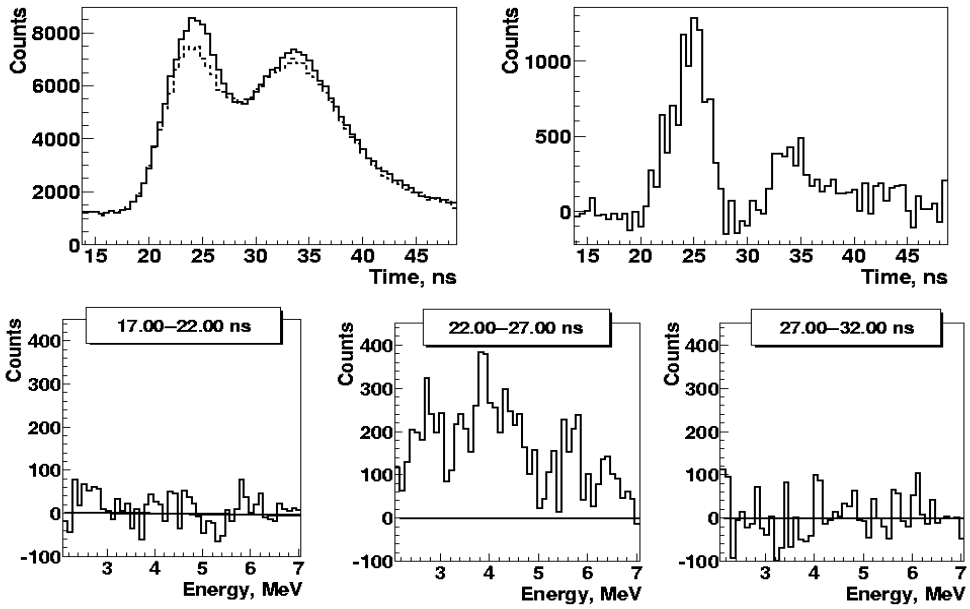

Figure 5. Time and energy spectra of γ quanta for the TNT buried in the box with soil. The upper row, left - the time distribution of γ quanta for the TNT buried in the soil is shown by the solid line, the time distribution of the box without TNT sample is shown by the dashed line. The upper row, right – the difference between the time distribution with and without TNT sample. The bottom row - the difference of the energy spectra of γ quanta for tests with and without a TNT sample.

bump at the time of flights interval 22-27 ns. It indicates that something is hidden there.

These two plots demonstrate an important advantage of the API method, its possibility to localize the place of the illicit substance along the direction of the neutron beam. It is important to note that the sharp bump at the difference spectrum of Fig. 5 (upper, right) is a “fingerprint” of the hidden TNT. For other substances (as wood, or stones) the difference spectrum exhibits a

deep rather than the bump. In general, the form of the difference spectrum depends on the difference between the chemical content of the hidden substance and the chemical content of the soil.

The bottom row of Fig.5 shows the difference of the energy spectra of γ quanta for tests with and without TNT sample. This difference should be zero for the part of the soil box without TNT.

Indeed, the difference of the energy spectra is practically zero for the time slice of t=17-22 ns (bottom row, left) and for the time slice of t=27-32 ns (bottom row, right). However in the region of the bump in the time difference spectrum, at the t=22-27 ns, the difference of the energy spectra also exhibits a spectrum with a number of events. This spectrum is similar but does not coincide with the spectrum of unshielded TNT. That is due to admixture of the γ-quanta from the soil. To extract the resulting pure γ-spectrum of the TNT, we need to enhance the spatial resolution to the size less than the dimensions of the TNT sample, which is a cylinder of 6 cm in diameter and 6 cm high.

To demonstrate the potential of the method for identification of the explosives placed in between other objects, the sample of 400 g TNT was placed in a standard suitcase of 33x45x15 cm^3 situated at 70 cm from the tritium target. The width of the case along the neutron beam is 33 cm. The case is divided on 4 equal compartments. Two tagged neutron beams irradiate the case simultaneously. Each beam passes through 2 compartments of the case. The sample is placed in one compartment, whereas the other compartments are filled by different background substances. The identification code should determine in which compartment the illicit substance is placed.

At first the sample of 400 g TNT was placed in one compartment and three other compartments were filled with carbon blocks of 2.5 kg each. The correct identification of TNT occurred after 60 s in 4 trials placing the TNT sample in different compartments. The neutron beam intensity was $2 \cdot 10^7$ n/s. Then we try to recognize the position of the sample of melamine of 1 kg among the samples of salt, washing powder and soil of the same mass. The samples were

shielded by the 5 mm steel sheet placed in front of them and 10 mm steel sheet placed between them and the NaI(Tl)-detector. The duration of each trial was 300 s. In 27 from 30 trials the identification was successful.

To obtain some typical characteristics for identification of explosives, we put in the case samples of melamine and soil of 100 g each and shielded them with 9 cm of soil in front of the case and 9 cm of soil in front of the NaI(Tl) detector. The neutron beam intensity was $4 \cdot 10^7$ n/s. Correct identification was obtained after 300 s. Additional shielding of 4 cm layer of water did not change the identification result. Indeed, the fast neutrons are not too sensitive to the humidity of the soil, in comparison with the mine detection technologies which use the thermal neutron probing [7,8,9].

In another trial 400 g of TNT was shielded by a 42 cm layer of soil placed in front of the case and by a 30 cm layer of soil placed in front of the NaI(Tl) detector. The neutron beam intensity was $4 \cdot 10^7$ n/s. Correct identification occurred after 180 s. This result demonstrates the high potential of the API for detecting deeply buried explosives.

4.Conclusions

We have investigated the main parameters of the laboratory operational prototype based on the Associated Particle Imaging (API) technique for the remote nondestructive identification of explosives, hidden in containers or soil. The prototype was constructed on the base of JINR Van-de-Graaf accelerator.

Based on the studies fulfilled and their analysis, the following conclusions could be made:

1. The API method is capable to identify explosives hidden in the containers or soils. In our experiments with neutron intensity of 4×10^7 s^{-1} we were able to detect 400g TNT shielded by 42 cm of soil within 3 minutes. The 100 g of melamine shielded by 9 cm of soil was detected within 5 minutes.

2. An important distinctive feature of the API method is its sensitivity to the chemical composition of the illicit substance. We have demonstrated that it is possible to discriminate and identify explosives placed in between other innocuous objects.

3. The experiments have demonstrated that the API method is superior compared with other techniques that use probing neutron irradiation, due to the facts that API has a higher signal-to-background ratio (on factor ~200) and therefore higher sensitivity. Only the API method can deliver a 3D image of the hidden object. Both advantages in combination significantly facilitate the identification of the illicit substance.

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