TAGGED NEUTRON METHOD FOR DETECTION OF HIGH EXPLOSIVES

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Introduction

The paper is devoted to "Methods and Instruments for Detection of High Explosives".

Active gamma-neutron method - Method of Tagged Neutrons (MTN) [1, 2] - is used in inspection of cargo containers for the purpose of detection of concealed high explosive (HE).

The given method is based on determination of direction of tagged neutron movement by registration of associated alpha-particle. A continuous neutron flux of 14 MeV neutrons, produced by portable neutron generator in $T(d,n)He^4$ reaction is used.

Position-sensitive alpha-detector is measuring time and a coordinate of associated alpha-particle, thus enabling one to specify angle and time of neutron escape ("tag"). Gamma-detectors detect gamma-quanta produced from interaction of tagged neutrons with container material. Scheme of coincidence of alpha-particles with gamma-quanta provides selection of those gamma-quanta, which correspond to object under inspection. Computer processes the obtained data and sends information on elemental composition of container to the terminal. MTN allows one to improve signal-background ratio by more than two orders, as compared to registration of gamma-spectra without coincidence with alpha-particles.

As a result key chemical elements entering HE composition – carbon, nitrogen, oxygen – are determined. This allows one to improve sufficiently the detection probability and decrease false alarm probability, as compared to methods aimed at determination of nitrogen only (for example, TNA method [3] – Thermal Neutron Analysis).

The first prototype system for HE detection in sea containers has been developed in VNIIA during 2006-2008 [4]. The prototype based on MTN provides a selected gamma-energy spectrum from object under inspection. This spectrum is used for determination of O/C and N/C ratios by means of its decomposition to gamma-energy spectra, typical for each chemical element, using the so called template spectra. These O/C and N/C ratio are specific for HE, and this allows one to distinguish HE from not dangerous materials transported in container. But if cargo goods are arranged in front of HE, effects of scattering and moderation of generator 14 MeV neutrons, as well as photon attenuation lead to distortion of gamma-energy response from object under inspection inside a container.
To take this effect into account we suggest a new analytical approach to formation of template spectra, corresponding to each chemical element with allowance made for spectrum distortion inside filled container. Results of system testing on identification of HE simulators are presented as well.

Measuring module

The measuring module of inspection system for HE detection and identification in cargo container represents an assembly of 12 gamma-detectors (BGO) with dimensions of ∅63 x 63 mm, ING-27 neutron generator with built-in 9-pixel Si alpha-detector [4], unit of gamma-detector radiation shielding from generator neutrons, made of borated polyethylene + iron (Fig.1). Overall dimensions of the module: length – 140 cm, width – 80 cm, height – (110÷180) cm. Total mass of the module is about 300 kg. In experiments the measuring module was moved up to the vertical wall of container.

Fig. 1. General view of measuring module

So if the distance from neutron generator target to container wall is equal to 82 cm, projection of tagged neutron beam to this wall is about 40 cm x 40 cm. Region under inspection enlarges proportionally with increase of the distance from neutron source due to angular spread of tagged neutron beam. In the center of real cargo container (about 120 cm from front wall) cross dimensions of such beam from tested measuring module will be about 100 cm x 100 cm.

Analytical approach

Spectrum registered by gamma-detector and composed of contributions of gamma-quanta resulted from nuclear reactions (inelastic scattering mainly) with considered element is usually taken as template gamma-spectrum, when object under inspection is exposed to 14 MeV neutrons. At the same time it is assumed that formed gamma-quanta, falling into detector do not undergo attenuation. When container is filled up and object of interest is arranged behind thick layers of container filler, distortion of the given template spectrum takes place, hence the decomposition of obtained gamma-spectrum to templates is not right.

The above distortions are conditioned by two main factors:

- moderation of neutron beam by container materials, leading to change of quantity of formed gamma-quanta, i.e. neutron interaction cross-section depends on incident neutron energy;
- attenuation of gamma-quanta by container materials.

To make correct allowance for mentioned factors we used Monte-Carlo MCNP5 code to simulate energy distribution of incident neutrons, formed by 14 MeV neutrons passed through a certain thickness of cargo material. It is assumed that information concerning this material is a priori known (for example, from cargo declaration). So the quantity of formed gamma-quanta from material under inspection, located behind x thickness of cargo material, for each characteristic gamma-line with E_γ energy becomes directly proportional to $\int \sigma(E_\gamma, E_n) \times N(E_n, x) dE_n$, where $N(E_n, x)$ - distribution of incident neutrons over E_n energies, formed by 14 MeV neutrons passed through x thickness of cargo material, $\sigma(E_\gamma, E_n)$ - cross-section of formation of characteristic gamma-quantum with E_γ energy by neutron with E_n energy. Data on cross-sections were taken from ENDF/B-VII library.

Attenuation of gamma-quanta by container materials depends on E_γ gamma-quantum energy, Z charge number, ρ material density, y distance passed by gamma-quantum inside the given material, and calculated as $\exp(-\mu(E_\gamma, Z)\rho y)$, where $\mu(E_\gamma, Z)$ – coefficient of gamma-quantum attenuation in the medium. Resulting coefficient L, taking account of the effects of neutron moderation and gamma-quanta attenuation for each characteristic gamma-line with E_γ energy is equal to:

$$L(E_\gamma, Z, \rho, y, x) = \exp(-\mu(E_\gamma, Z)\rho y) \times \int \sigma(E_\gamma, E_n) \times N(E_n, x) dE_n .$$

Each chemical element has its own characteristic gamma-lines formed in result of various nuclear reactions or inelastic scattering of neutron on nuclei of the given chemical element. Transformed template (modified) energy gamma-spectrum for particular chemical element G^*_{temp} will have the form: $G^*_{temp} = \sum_{i=1}^{n} G^*_i(E_\gamma)$, where n – number of characteristic gamma-lines taken into account for the given chemical element, $G^*_i(E_\gamma) = L_i \times G_i(E_\gamma)$ - energy gamma-spectrum of i-th gamma-line, taking account of neutron moderation and gamma-quanta attenuation for this gamma-line, $G_i(E_\gamma)$ - calculated (non-modified) gamma-spectrum of i-th gamma-line. One gamma-line was taken into account for carbon, 14 lines – for nitrogen and 8 lines – for oxygen. Selected lines are the most intensive ones (σ > 10 mb at E_n=14 MeV) for each chemical element being within the energy range from 2 MeV to 8 MeV.
Stated algorithm was used later on for decomposition of experimental gamma-spectrum and determination of O/C and N/C chemical ratios.

Method testing in experiments

At the first stage of experiments we carried out identification of HE simulators - 1.670 kg of tetryl, 1.800 kg of trotyl and 1.931 kg of hexogen - in the absence of shielding materials. Samples of HE simulators were arranged maximum close to gamma-detectors. Measurement time for each sample was equal to 5 min. at $8x10^7$ neutr./s intensity of neutron generator.

Table 1. Elemental composition of simulators

Name	**Material**	**Simulator**
Hexogen/Octogen	$C_3H_6N_6O_6/C_4H_8N_8O_8$	$C_3H_{8,3}N_{5,7}O_{5,9}/C_4H_{11}N_{7,6}O_{7,9}$
Tetryl	$C_7H_5N_5O_8$	$C_7H_{7,5}N_5O_{7,8}$
Trotyl	$C_7H_5N_3O_6$	$C_7H_5N_3O_6$

Table 2 shows the obtained N/C and O/C ratios in comparison with available theoretical ones for HE simulators and for some not dangerous provoking materials with similar composition: melamine ($C_3H_6N_6$), carbamide (CH_4N_2O) and wood ($C_{22}H_{31}O_{12}$) as shielding material.

Table 2. Experimental and theoretical ratios of elements

Material	Experiment *		Theory		Exp./ Theor.	
	N/C	O/C	N/C	O/C	N/C	O/C
Hexogen/Octogen	1,82 ± 0,13	2,04 ± 0,12	1,89	1,96	0,96	1,04
Tetryl	0,84 ± 0,09	1,05 ± 0,08	0,71	1,11	1,18	0,94
Trotyl	0,39 ± 0,04	0,79 ± 0,04	0,43	0,85	0,91	0,92
Carbamide	1,79 ± 0,09	0,97 ± 0,05	2	1,00	0,89	0,97
Wood	-	0,53 ± 0,03	-	0,55	-	0,97
Melamine	2,04 ± 0,21	-	2	-	1,02	-

* *Statistical uncertainties presented*

It is convenient to look at obtained results on a curve in C/O to C/N coordinates. Results of correlation of experimental and theoretical values are presented in Fig.2. One can see that experimental results are in good agreement with theoretical values.

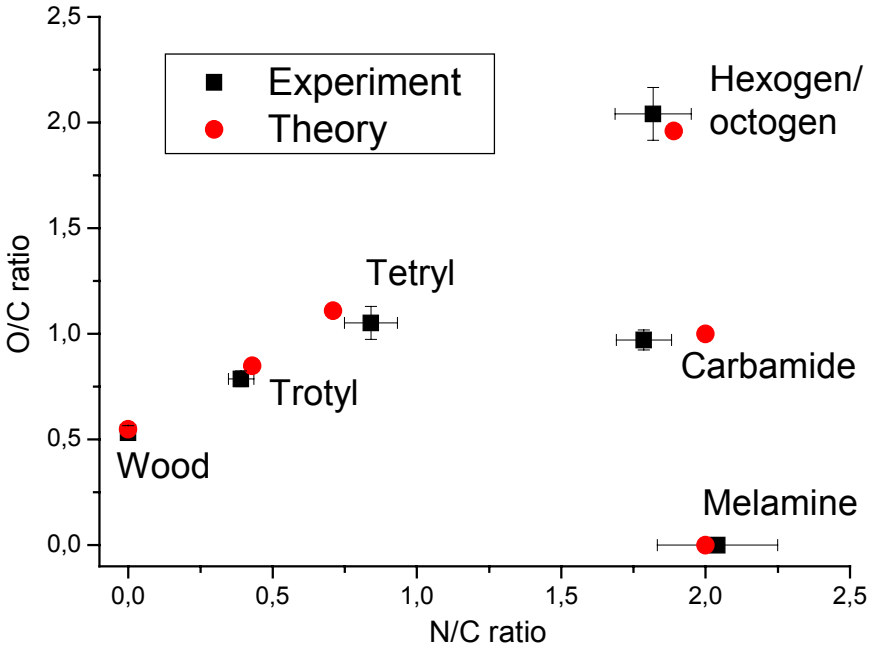


Fig.2. Comparison of experimental and theoretical values in O/C vs N/C coordinates

At the second stage of experiments the suggested method was tested on melamine (HE simulator) identification in the presence of shielding material (wood). Table 3 shows O/C and N/C ratios, resulted from decomposition of experimental energy gamma-spectra [3]. The given laboratory measurement was made with described HE detection system for 50 kg of HE simulator (melamine) of 1.07 g/cm^3 density, arranged at the distance of 30 cm from wall of container, loaded with wood with density ρ=0.4 g/cm^3. Measurement time was 30 minutes. Neutron generator intensity - (3-4)x10^7 n/s. Theoretical O/C ratio for wood ($C_{22}H_{31}O_{12}$) is 0.545, N/C ratio for melamine ($C_3H_6N_6$) is 2. Decomposition was conducted by modified template spectra calculated under the assumption, that container is filled only with wood. One can see from Table 3 that experimental O/C and N/C ratios calculated for front layers of container (for depth of 0 cm, 10 cm, 20 cm and 30 cm) reproduce more correctly theoretical ratios, corresponding to wood layer, as compared to the region, corresponding to melamine. So in the region, corresponding to melamine (depth of 40 cm, 50 cm and 60 cm) the decomposition gives sufficient contribution of oxygen (whereas melamine is free of oxygen), as well as carbon. This is explained by overlapping of gamma-quanta, corresponding to different regions of an object. Due to the absence of additional collimation of gamma-detector and not high enough time resolution of BGO gamma-detector, gamma-quanta from wood fall into time interval corresponding to melamine. Nevertheless the observed behavior of O/C and N/C ratios testifies to the presence of anomaly in the place of melamine arrangement.

To remove effect of wood layer in the absence of gamma-detector collimation during melamine identification, one can use wood spectrum obtained for the previous layer (see Table 4). This Table also includes results of spectra decomposition with the use of non-modified samples. One can see that decomposition with modified samples provides much better results. So the N/C ratio for melamine becomes equal to 1.9±0.2, and this is the same as theoretical value within statistical error.

Table 3: Results of spectra decomposition

Material	Depth *l*, cm	O/C		N/C	
		Exp.*	Theor.	Exp.*	Theor.
Wood	0	0,53±0,02	0,545	0	0
	10	0,51±0,01	0,545	0	0
	20	0,55±0,01	0,545	0	0
	30	0,53±0,02	0,545	0	0
Melamine	40	0,46±0,02	0	0,20±0,04	2
	50	0,37±0,02	0	0,45±0,06	2
	60	0,36±0,03	0	0,43±0,08	2

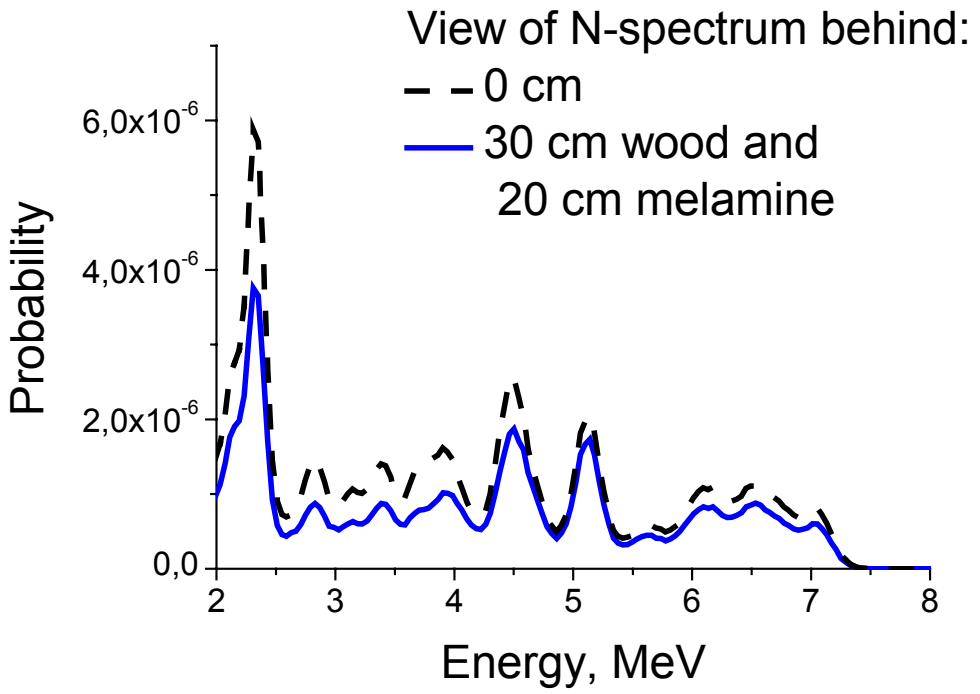
* *Statistical uncertainties presented*

Table 4: Results of measured spectra decomposition

Influence of cargo material	Wood *at l* = 30 cm, $O/C_{theor.}$=0,545	Melamine *at l* = 50 cm, $N/C_{theor.}$=2
	Exp. *	Exp. *
Not taken into account	0,45±0,02	1,2±0,2
Taken into account	0,53±0,02	1,9±0,2

* *Statistical uncertainties presented*

Fig. 2 demonstrates the influence of cargo material on calculation of modified spectra of nitrogen and oxygen. In one case cargo material is absent, in another – gamma-response is from the layer being behind 30 cm thick wood of 0.4 g/cm^3 density and 20 cm thick melamine of 1.07 g/cm^3 density. The spectra are normalized by carbon (multiplied by coefficient at which modified and non-modified carbon spectra coincide with each other). Corresponding oxygen and nitrogen spectra differ significantly for these two cases as shown in Fig.2.



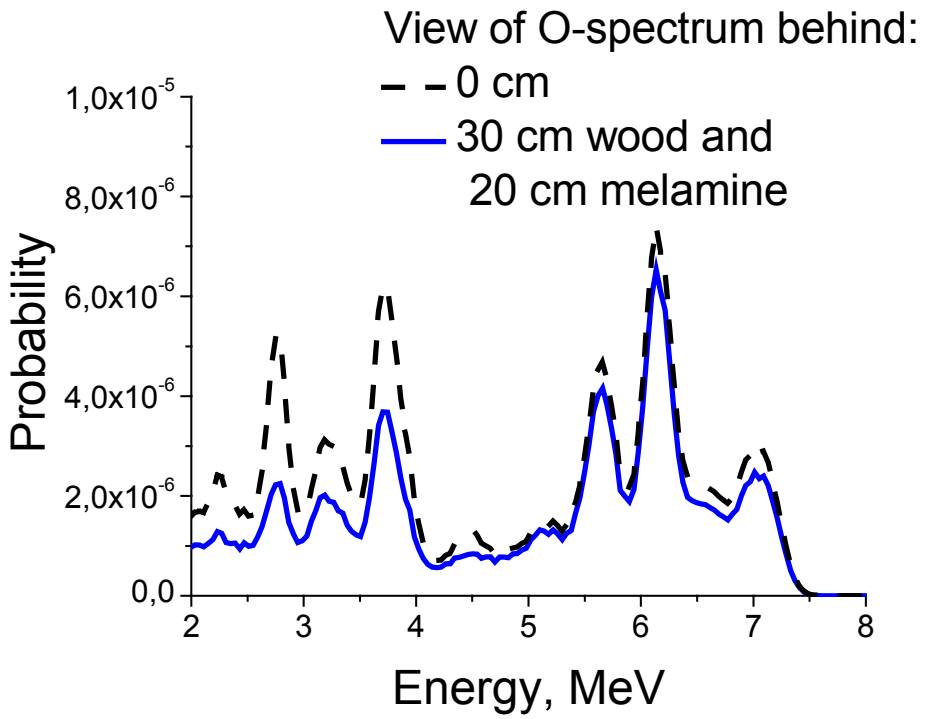


Fig. 3. Change of modified nitrogen and oxygen spectra in the presence of shielding material

Conclusion

A new analytical approach of HE detection and identification behind thick layer of shielding material is suggested. Testing on experimental data obtained with HE detection system by the Method of Tagged Neutrons demonstrated its efficiency. Results of identification of HE simulators and other chemical substances (melamine, carbamide, wood) according to O/C and N/C ratios and in the absence of shielding materials have demonstrated good agreement of experimental and estimated values. Presence of 30 cm thick wood layer in front of identified object, absence of collimators on gamma-detectors, the system geometry (neutron generator and gamma-detector arranged on one side from exposed object) and time resolution of gamma-detectors (3.7 ns) lead, however, to sufficient overlapping of gamma-responses from different layers of container. So in this case the method is able to recognize just the presence of anomaly in the place under inspection (occurrence of nitrogen, change of O/C ratio typical for wood). To provide further identification of an object by decomposition of experimental gamma-spectrum it is advisable to use gamma-spectrum of cargo material, obtained for the previous layer.
To improve the performance of module, both in HE and FM detection, it is proposed to pay particular attention to secondary radiation detectors with better characteristics and their collimation protection.

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