

TAGGED NEUTRONS FROM PORTABLE NEUTRON GENERATOR FOR DETECTION OF HIGH EXPLOSIVES AND FISSILE MATERIALS IN CARGO CONTAINERS

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Abstract. Inspection system created in VNIIA and intended for detection of high explosives (HE) in sea cargo containers is based on tagged neutron method. Energy gamma-spectrum obtained from object under inspection is used for HE identification on the basis of oxygen/carbon (O/C) and nitrogen/carbon (N/C) chemical ratios. When container is, however, filled up, moderation of primary 14 MeV neutrons from generator and distortion of gamma-energy response from inspected object inside container make difficulties for correct production of these relations. Analytical approach taking these difficulties into account and tested on experimental data (HE simulator melamine placed in container filled with wood) is presented.

The paper also deals with experimental results on detection of fissile materials (FM), using multiplicity of fission neutrons for different container fillings.

1. Detection of high explosives

1.1. Introduction

Active gamma-neutron method - Method of Tagged Neutrons (MTN) [1, 2] - is used in inspection of large containers for the purpose of detection of concealed HE. Key chemical elements entering HE composition - carbon, nitrogen, oxygen - are determined in result of analysis.

The first prototype system for HE detection in sea containers has been developed in VNIIA during 2006-2008 [3]. 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.

1.2. 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 $\varnothing 63 \times 63$ 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. 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.



Fig. 1. General view of measuring module.

1.3. 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 ($\sigma > 10$ 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.

1.4. Algorithm testing in experiments

Table I shows O/C and N/C ratios, resulted from decomposition of experimental energy gamma-spectra [3] realized by suggested algorithm. The given laboratory measurement was made with described HE detection system for 50 kg of HE simulator (melamine) of 1.07 g/cm³ density, arranged at the distance of 30 cm from wall of container, loaded with wood with density $\rho=0.4$ g/cm³. Measurement time was 30 minutes. Neutron generator intensity - (3-4) $\times 10^7$ n/s. Theoretical O/C ratio for wood (C₂₂H₃₁O₁₂) is 0.545, N/C ratio for melamine (C₃H₆N₆) 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 I 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 mechanical 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 II). 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 \pm 0.2, and this is the same as theoretical value within error.

TABLE I: 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 II: RESULTS OF MEASURED SPECTRA DECOMPOSITION.

Influence of cargo material	Wood at $l = 30$ cm, $O/C_{\text{theor.}} = 0,545$	Melamine at $l = 50$ cm, $N/C_{\text{theor.}} = 2$
	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 (multiply by coefficient, that modified and non-modified carbon spectra coincide with each other). Corresponding oxygen and nitrogen spectra differ significantly for these two cases as are shown in Fig.2.

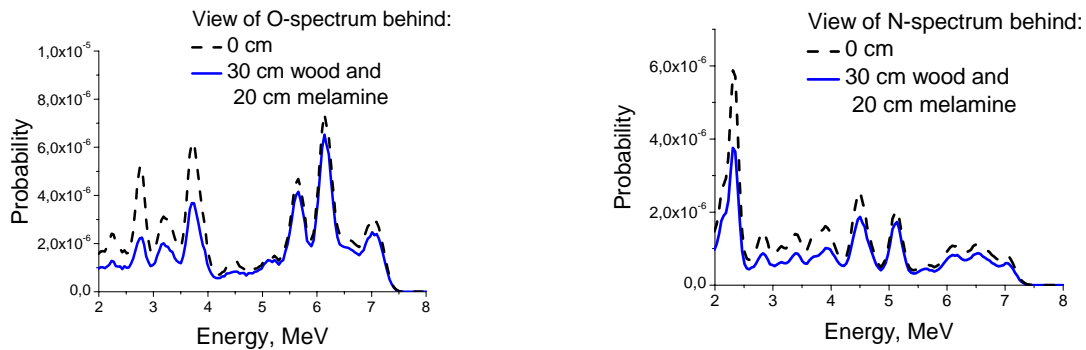


Fig. 2. Change of modified nitrogen and oxygen spectra.

1.5. Summary

New analytical approach to deduce the template spectra corresponding to each chemical element with individual taking account of all gamma-lines is suggested. Testing on experimental data obtained with HE detection system by MTN demonstrated its efficiency. 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.

The improvement of decision-making algorithm we are going to obtain with update system having additional collimation of gamma-detectors. It is also proposed to pay particular attention to effects of anisotropy and Compton scattering of gamma-quanta.

2. Detection of fissile materials

2.1 Introduction

Active methods of FM detection are based on increase of fission rate, when FM is probed by some types of penetrating radiation (neutrons, gamma-quanta). These methods are much sensitive and selective to special nuclear materials, such as uranium-235 or plutonium-239, since a response to active exposure of these materials is unique, and it can't be mixed up with signals from other "inert" materials being present in a standard container.

Prompt neutrons and gamma-quanta of fission, as well as their multiplicities, correlated with probe neutrons, are considered as this specific response.

More than 4 prompt fission neutrons and 8-9 prompt fission gamma-quanta are produced by 14 MeV neutron during induced fission. This property of neutron and gamma-quantum multiplicity is used for FM identification [3,5].

2.2. System description and measurement results

Measuring module of inspection system for FM detection in cargo container represents an assembly of 6 neutron/gamma detectors and ING-27 neutron generator with built-in 9-pixel alpha-detector. To detect secondary fission neutrons (gamma-quanta), produced due to FM irradiation in container, we used large plastic (polystyrene) scintillation detectors (120 cm x 9 cm x 9 cm). Detectors were arranged under container and at the distance of 15 cm from each other. Space between the detectors was filled with lead to prevent detector response to gamma-quantum rescattering.

The detectors like this provide simultaneous detection of fast neutrons and gamma-quanta. Radiation types were separated according to time of flight.

Uranium (depleted uranium U-238, mass 8.3 kg) and lead (mass 10 kg) samples were arranged in turn at the distance of 60 cm from container wall and were measured for different matrices: iron, wood (Fig.3) and empty container. Average matrix density over container was equal to 0.4 g/cm³.



Fig. 3. Container with different matrices.

Figure 4 shows summarized time distributions of double and triple alpha-gamma/neutron coincidences from uranium sample for six gamma/neutron detectors and the central segment of alpha-detector. Measurements are presented with the deduction of appropriate background measurement, i.e. in container free of uranium. Each measurement time was equal to 30 minutes at neutron generator intensity of about 2×10^6 n/s. All results are normalized to the total number of alpha-particles fallen into the central alpha-segment. One can see that results of uranium measurements in empty container and in container loaded with iron are similar enough. When container is filled with wood, neutrons are strongly suppressed, and signal from uranium is evidently weaker.

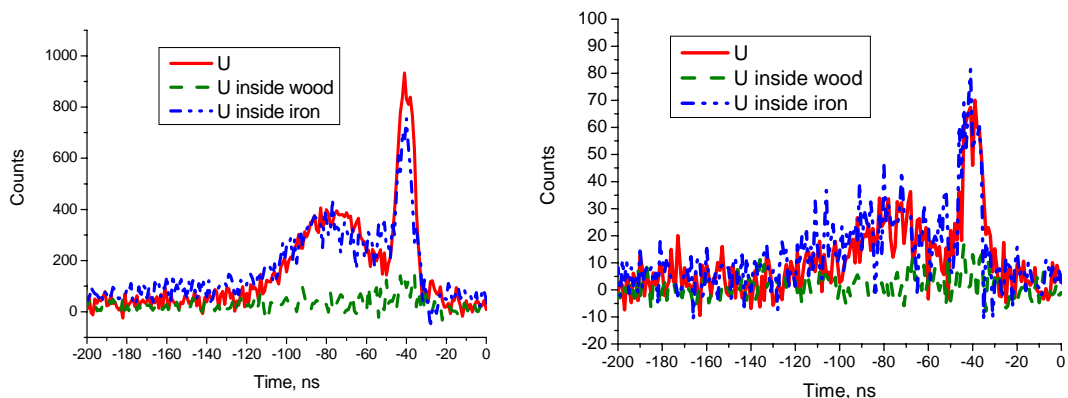


Fig. 4. Total time distribution of net two-fold (left) and net three-fold (right) alpha-neutron/gamma coincidences when uranium is arranged in empty container or loaded with iron or with wood matrix..

Table III includes results of multiple alpha-gamma/neutron coincidences from uranium and lead samples measured in time window, corresponding to the moment of arrival of prompt fission neutrons for container loaded with wood and iron. In both cases the excess of double and triple coincidences from uranium over corresponding background is observed. Object under inspection was placed in one tagged neutron beam, and the data on another tagged neutron beam, where object was absent, were taken as a background. So the information concerning signal and background values was withdrawn from one measurement cycle. One can see that statistics of four-fold coincidences is practically absent.

TABLE III: MULTIPLE ALPHA-GAMMA/NEUTRON COINCIDENCES IN TIME WINDOW CORRESPONDING TO PROMPT FISSION NEUTRONS.

Coincidences	Signal	Bg	Signal/Bg	Signal	Bg	Signal/Bg
	U inside iron			Pb inside iron		
$\alpha + n + n$	22261	14020	1,59	21450	13806	1,55
$\alpha + n + n + n$	461	198	2,33	333	194	1,72
$\alpha + n + n + n + n$	15	4	3,75	2	3	0,67
	U inside wood			Pb inside wood		
$\alpha + n + n$	17512	16121	1,09	15404	14487	1,06
$\alpha + n + n + n$	219	198	1,11	161	156	1,03
$\alpha + n + n + n + n$	6	5	1,20	3	0	-

Results presented in the Table III testify to the fact that the number of coincidences from uranium exceeds the number of coincidences from lead for all multiplicities. As multiplicity grows, signal/background ratio increases and achieves the maximum for four-fold coincidences, when 4 $n(\gamma)$ events fall on one alpha-particle. So it was possible to distinguish uranium from lead in performed experiments, but in view of 4-fold events, observed in measurements with lead we can not say unambiguously, that the presence of four-fold coincidences is an attribute of fissile material only. This is conditioned by coincidences, resulted from registration of one and the same neutron or gamma-quantum by two and more detectors, as well as by non-correlated random coincidences.

Results of using time window corresponding to prompt fission gamma-quanta appeared to be ambiguous. Conducted investigation [3] demonstrated the presence of a large number of alpha-gamma/neutron coincidences of high multiplicity from iron sample in time window, corresponding to prompt fission gamma-quanta, therefore we couldn't identify events from uranium.

2.3. Summary

Results of measuring uranium and lead in container filled with wood and iron are presented in the form of multiple alpha-neutron/gamma coincidences of different multiplicity for time window corresponding to the moment of arrival of prompt fission neutrons.

The results testify to the presence of background alpha-neutron/gamma coincidences of high multiplicity (2, 3), therefore existence of events of high multiplicity can not be unambiguously considered as an attribute of FM presence in container.

It seems that this problem can be solved by adding to the number of detectors in order to increase statistics and to get alpha-neutron/gamma coincidences with multiplicity of 4 and above. Detector shield must be improved to provide maximum possible elimination of probability of detection of one and the same gamma-quantum or neutron by two and more detectors.

3. Conclusion

Measuring module for HE and FM detection in cargo containers with the use of tagged neutron method was tested. New analytical approach to deduce the template spectra used by decision-making algorithm for HE detection and identification is suggested. This analytical model realizes individual taking account of all main gamma-lines for each chemical element composing HE. This approach provides more correct account of neutron and gamma-quantum distortion by container materials, as compared to one presented in [6].

It is shown that detection of both HE and FM using the same processing electronics and neutron generator is possible.

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.

4. References

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