Detection of Explosives and Other Illicit Materials by Nanosecond Neutron Analysis

A.V. Evsenin[1], I.Yu. Gorshkov[2], A.V. Kuznetsov[1], O.I. Osetrov[1], D.N. Vakhtin[1]

[1] V.G. Khlopin Radium Institute (KRI), Saint-Petersburg, Russia
[2] Applied Physics Science and Technology Center (APSTEC), Saint-Petersburg, Russia

Email contact: apl@khlopin.ru, info@apstec.ru

Abstract. Experimental results in the detection of explosive and other hazardous materials by Nanosecond Neutron Analysis (NNA) technique are presented. The detecting device SENNA is based on a portable DT neutron generator with built-in segmented detector of associated α-particles, detectors of γ-rays, and fast data collection electronics. Experimental measurement of the response functions of NaI, BGO, and LaBr$_3$-based γ-ray detectors to pure chemical elements is described. Examples of using SENNA to detect explosives in luggage and UXO on a conveyer belt and in the ground are discussed.

1. Introduction

Neutron-based methods of non-destructive analysis are sometimes the only tool to determine the nature of the material that is located behind thick barriers (e.g. a UXO in the ground) or is surrounded by other materials (e.g. in cargo containers) [1]. Among methods that provide the most complete information about the chemical composition of the inspected material is a group of "neutron in, gamma out" methods: TNA, FNA, PFTNA, and NNA/APT.

The main advantages of "neutron in, gamma out" techniques over conventional methods are:
1. High selectivity: chemical composition of the substance can be determined (unlike X-rays or QR-based methods).
2. High penetrating ability of neutrons and γ-rays (much higher than that of X-rays).
3. Sealed containers can be inspected (unlike vapor/trace detectors).
4. Fully automatic data analysis and decision-making of YES/NO type are possible (unlike with X-rays).

The most powerful of these methods is Nanosecond Neutron Analysis / Associated Particle Technique (NNA/APT).

2. Nanosecond Neutron Analysis / Associated Particle Technique (NNA/APT)

NNA/APT is a non-destructive analysis method that allows one to obtain a 3D distribution of chemical elements (isotopes) in the inspected volume [2]. Concentrations of different chemical elements are obtained from the analysis of energy spectra of secondary γ-rays that are induced at different locations ("voxels") of the inspected volume by 14 MeV neutrons. The neutrons are produced in a DT neutron generator equipped with a position-sensitive detector of α-particles that accompany neutrons in the reaction $d + t = \alpha + n$. The location of the material that produced the given γ-quantum is determined from the analysis of γ-α coincidences [2]. Advantages of NNA over other neutron-gamma methods are: a) very good effect-to-background ratio (unlike FNA and PFTNA); b) sensitivity to almost all chemical elements (unlike TNA); c) position sensitivity: the mass of the object can be determined.
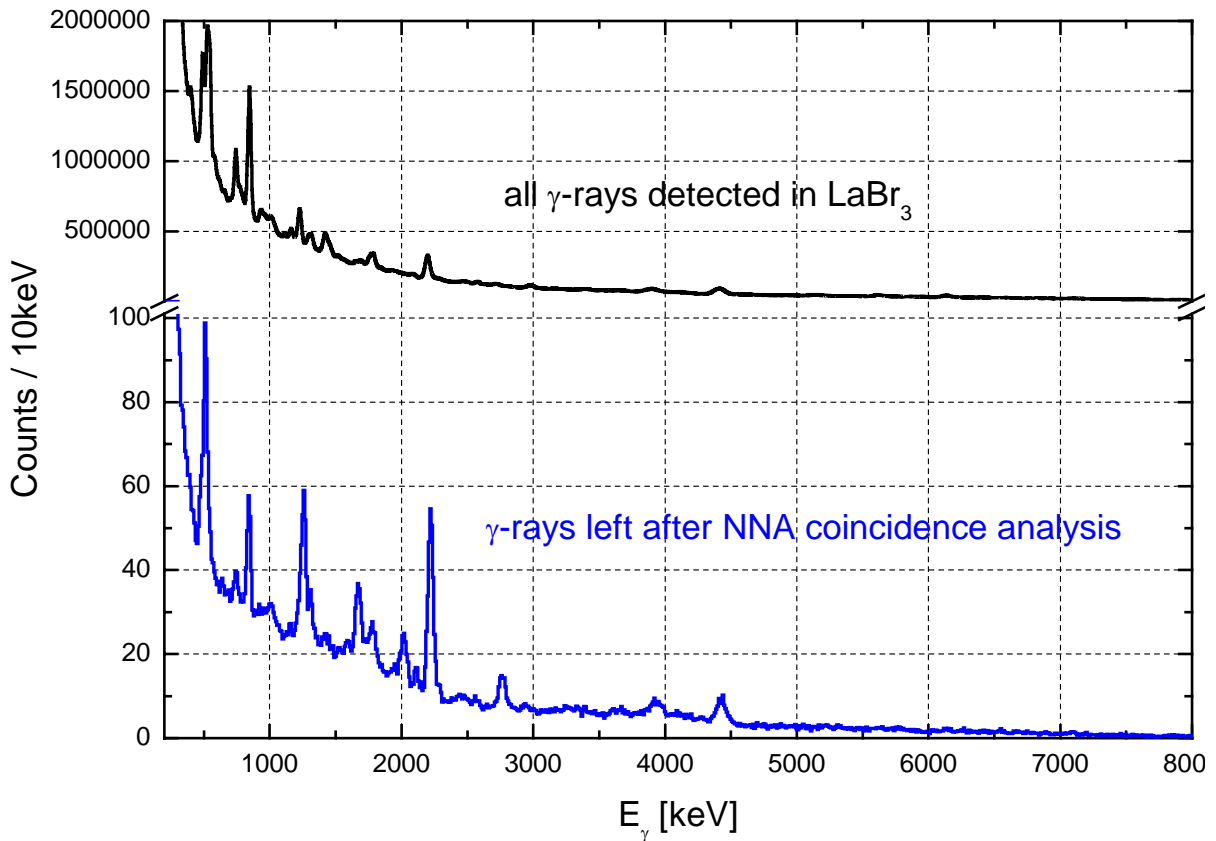
FIG. 1. Experimental energy spectra of γ-rays obtained in a 15 minute-long measurement with 300g of sulfur powder in 44g plastic box. Top line is the spectrum of all γ-rays detected by the LaBr$_3$ detector. Bottom spectrum contains γ-rays selected from all events by the coincidence and TOF analyses.

The improvement of the quality of the γ-ray spectra that can be achieved in NNA/APT is demonstrated on FIG. 1. The top spectrum contains all γ-rays detected in a LaBr$_3$-based detector during 15 minute-long irradiation of 300g sulfur powder in a 44g plastic box. The bottom spectrum is what remains after the proper coincidence and TOF analysis is carried out: the γ-rays that come from the sample are retained, while most of the γ-rays that originate from other sources (reactions of neutrons with the surrounding materials, reactions of neutrons with the γ-ray detector itself, etc.) are suppressed. The suppression of the background achieved in this case is over 4 orders of magnitude. Any attempt to extract the component related to the sample directly from the top (total) spectrum would fail, since the surrounding materials may contain the unknown amount of the same elements as the sample itself.

2.1. NNA/APT hardware

The central piece of any NNA/APT device is a DT neutron generator with built-in detector of associated α-particles. We are currently using ING-27 portable neutron generators equipped with 9-pixel, 36-pixel, or 144-pixel silicon-based α-detectors. These very compact and lightweight neutron generators typically produce $(2 \div 5) \times 10^7$ n/s (10^8 n/s maximum), and the lifetime of the sealed neutron tube is about 1000 hours. Depending on the α-detector type, the share of the neutrons "tagged" by α-particles may be from 1% to 5%.

Several types of γ-ray detectors are used in different devices: NaI(Tl), BGO, and LaBr$_3$ scintillation crystals. NaI(Tl) crystals are the largest and the cheapest of them all, but they

cannot work at high counting rates due to a large number of low-amplitude pulses that affect the detector's energy resolution. BGO crystals have the highest density (and hence efficiency), and are optimal for use in compact devices to detect explosives and other substances that have limited number of constituting elements (e.g. in explosives they are carbon, nitrogen, and oxygen, all of which have high energy peaks in their γ-spectra that can be well resolved by a BGO-based detector). Detectors of γ-rays based on LaBr$_3$ crystals have excellent energy and time resolution, and are capable of detecting most chemical elements that have low energy lines in their γ-spectra. The price of these crystals is however notably higher than that of NaI and BGO.

The compact data acquisition system (DAQ) that was specially designed for use in NNA/APT devices contains all the components needed to operate the device:
1. detector power supplies;
2. spectrometers of γ-rays;
3. α-particle detection electronics;
4. coincidence analysis module.

The DAQ boards are build right into the respective detectors, and each is connected with the coincidence module by a single cable. The DAQ exists in two versions:
- Ultra-compact DAQ for servicing one neutron generator with any α-detector and up to 4 detectors of γ-rays.
- Universal DAQ for servicing multiple neutron generators with α-detectors and multiple detectors of γ-rays (up to 40 devices in any combination).

2.2. Data analysis and decision-making in NNA/APT

NNA/APT suppresses the background (everything unrelated to the substance of interest) by a factor of 1000 or more (see FIG. 1). Still, the remaining spectra contain "noise" coming from several sources: statistical, "parasitic" material, accidental coincidences. Direct analysis of these spectra by some image-recognition technique would inevitably result in analyzing a lot of irrelevant information contained in the "noise".

Suppression of this "noise" requires a multi-stage data analysis:
- Construction of energy spectra from the "raw" data.
- Determining elemental concentrations by fitting energy spectra of γ-rays with response functions to individual elements.
- Automatic decision-making based on the analysis of elemental concentrations (and any other available information).

At the first stage the "raw" event-by event data are analyzed to construct energy spectra of γ-rays for each "voxel" or the inspected volume. This requires energy and time calibrations that are done automatically using the same experimental data that is being analyzed.

Then, elemental concentrations are determined from spectral regression using modified PLS (Projection to Latent Structures) algorithm. It allow one to fit the spectra collected with poor statistics with more than 30 components that are energy spectra measured for pure chemical elements.

At the last stage the resulting elemental concentrations and their error bars are fed into a "fuzzy" logic decision-making procedure that provides an ALARM / NO_ALARM type answer. If measurement time is used as one of the analyzed parameters, the system can decide to continue the measurement until the desired reliability of the result is reached.

3. Experimental results

Conventional (industrial or military) explosives in luggage and other relatively small packages can be detected by analyzing ratios between carbon, oxygen and nitrogen.

An experiment with SENNA IV device [3] was carried out to determine the possibility to detect explosives in suitcases. Chemical imitators of RDX and ammonium nitrate weighting over 200g were placed in a suitcase among other benign objects typically found in suitcases: books, cotton and woolen clothing, drinks, consumer electronics, toothpaste, deodorants, cocoa butter etc. (see left photo on FIG. 2). The suitcase was rotated to ensure that the sample was inside the sensitive volume of the device.

FIG. 2 shows an example of the measurement, in which the explosive' imitator was detected in the "voxel" corresponding to the central "pixel" of the α-detector at depth 10cm from the front surface of the suitcase.

In these tests all imitators were successfully detected. No false alarms were registered neither in measurements without samples, nor in the "voxels" that contained benign materials.

Another possible application of NNA/APT is identification of UXO, which may either be empty, or contain conventional explosive charges, poisonous chemicals, or both. Depending on the scenario the UXO may be inspected either at a disposal facility or directly where it was discovered – e.g. lying in the ground at a construction site.

When the UXO is inspected at a disposal facility in controlled environment (e.g. on a conveyer belt), a limited number of chemical elements that may be found in different types of UXO are to be taken into account: Fe, C, N, O, H, S, Cl, As, and P.
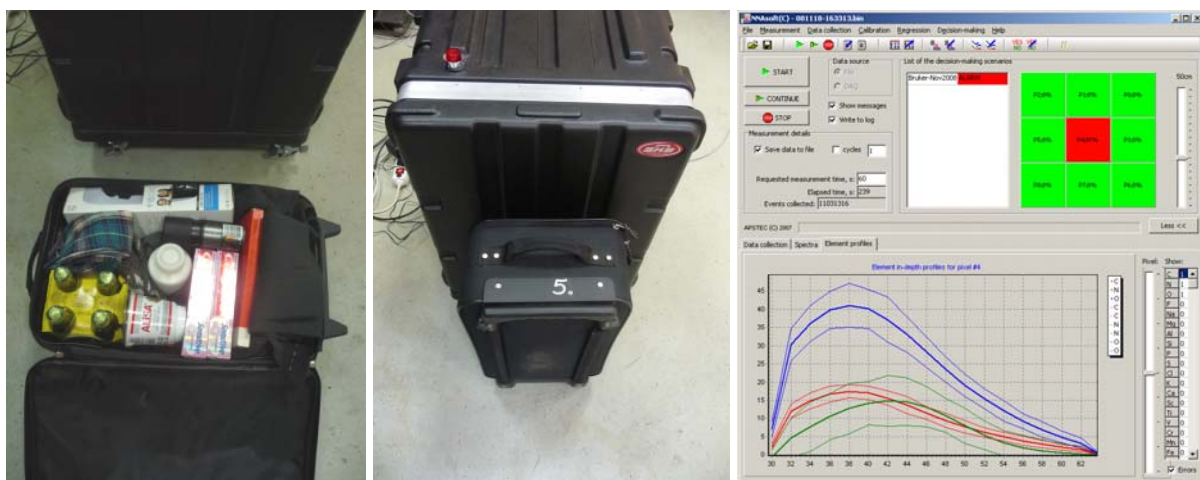

FIG. 2. Automatic detection of explosives' imitators in a suitcase by SENNA IV device. Photo on the left: the sample in the suitcase. Photo in the center: inspection geometry. Screenshot on the right: the result automatically obtained by the device.

The feasibility of using NNA/APT for identification of UXO was experimentally tested, using the device consisting of a neutron generator with 36-pixel detector of associated α-particles, and three detectors of γ-rays based on ∅3"×3" BGO crystals. The imitators of UXO that we used in the experiments are listed in TABLE 1.

TABLE 1. THE IMITATORS OF UXO USED IN THE EXPERIMENTS.

Type	Composition of the actual UXO	Composition of the imitator
"yellow"	50% mustard gas + 50% Lewisite	arsenic oxide, sulfur, salt, graphite, water
"black"	explosive	Melamine, water, graphite
"red"	diphenylcyanoarsine + explosive	arsenic oxide, melamine, water, graphite
"brown"	Prussic acid	Melamine, graphite

The mixtures of the imitating substances were enclosed into 1cm-thick iron containers. The detectors of γ-rays were placed right under the inspected imitator and were shielded from the direct flux of 14 MeV neutrons by a composite shielding (borated polyethylene + lead). The inspection time was 1 minute, and each measurement was repeated 100 times in order to check the reliability of the results. The γ-ray spectrum obtained in each measurement was fitted with the response functions of the device to individual chemical elements, and the resulting elemental concentrations were represented on 2D plots with axes corresponding to different chemical elements. Examples of such plots for "red" and "black" imitators are shown on FIG. 3. Each small dot represents the result of a single 1 minute-long measurement. The color of the dot corresponds to the name of the imitator. Large colored symbols show the calculated location of the corresponding imitator.

The device was able to correctly identify the imitator in 100% cases by comparing the distances (in four dimensions: O, S, N, As) from the experimental dot to the expected locations of all imitators and choosing the closest one.

Such a simple data analysis procedure worked, because the system had a very limited number of classification options. However, when inspecting a totally unknown UXO that is lying on the ground, one has to additionally take into account a large number of chemical elements that can be found in the soil around the UXO: H, O, Si, Al, Ca, Mg, K, Na, Ti, and many others. Practically speaking over 30 abundant chemical elements must be taken into account, since all of them may contribute to the measured spectrum of γ-rays.
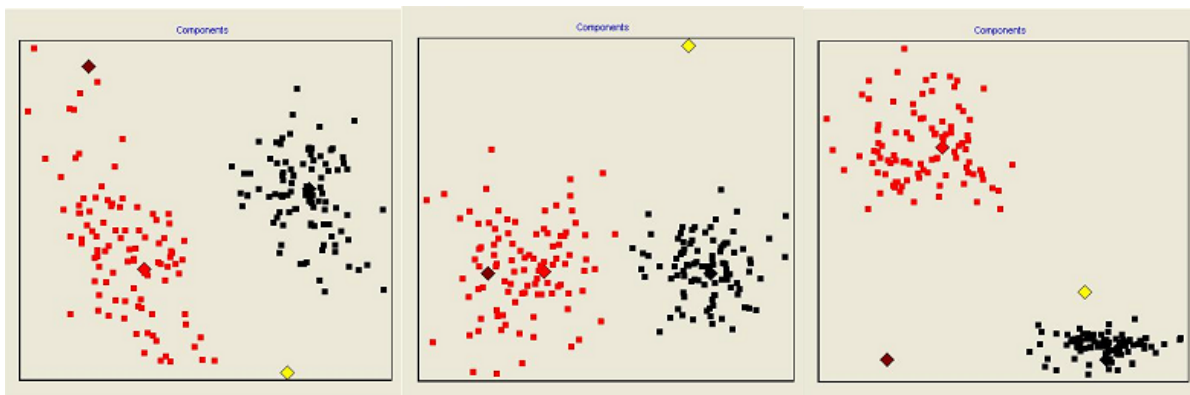

FIG. 3. Distribution of measurements with "red" and "black" UXO imitators in coordinates N-O (left), S-O (center), and As-O (right). Large symbols show the positions of the "red", "black", "yellow", and "brown" imitators calculated from their known chemical composition.

The energy resolution of BGO-based detectors of γ-rays is too rough to allow the spectral fitting procedure to reliably extract contributions of over 30 chemical elements from the experimental spectrum that is usually measured with poor statistics. Among other available types of γ-ray detectors $LaBr_3$ scintillation crystals have the best energy resolution, time resolution, and efficiency.

3.1. Response functions for different types of γ-ray detectors

In order to effectively use the spectral fitting procedure one must have correct response functions to individual chemical elements. The response functions used so far for BGO-based detectors were calculated using MCNP[1] code. For such elements as carbon, nitrogen, oxygen, iron, aluminum and others that are important for detection of explosives the results given by MCNP coincide with the experiment. For other elements (e.g. sulfur) the MCNP results are wrong. The $LaBr_3$-based detectors produce spectra with much more detailed peak structures, and the exact knowledge of the response functions becomes critical.

Experiments were carried out to measure the response functions of the detectors based on NaI, BGO, and $LaBr_3$ crystals to the most abundant chemical elements. The experimental setup consisted of a neutron generator with a narrow (24° opening angle) beam of "tagged" neutrons, and three detectors of γ-rays (∅3"×3" $LaBr_3$, ∅3"×3" BGO, and ∅6"×4" NaI) located at different sides from of the sample. Samples were pure chemical elements (where practical), oxides, hydroxides, or acids placed in lightweight plastic containers. The masses of the samples were such that each contained 300g of the pure element of interest. Duration of measurements with each sample was 1 hour, intensity of the neutron generator was $2×10^7$ n/s.

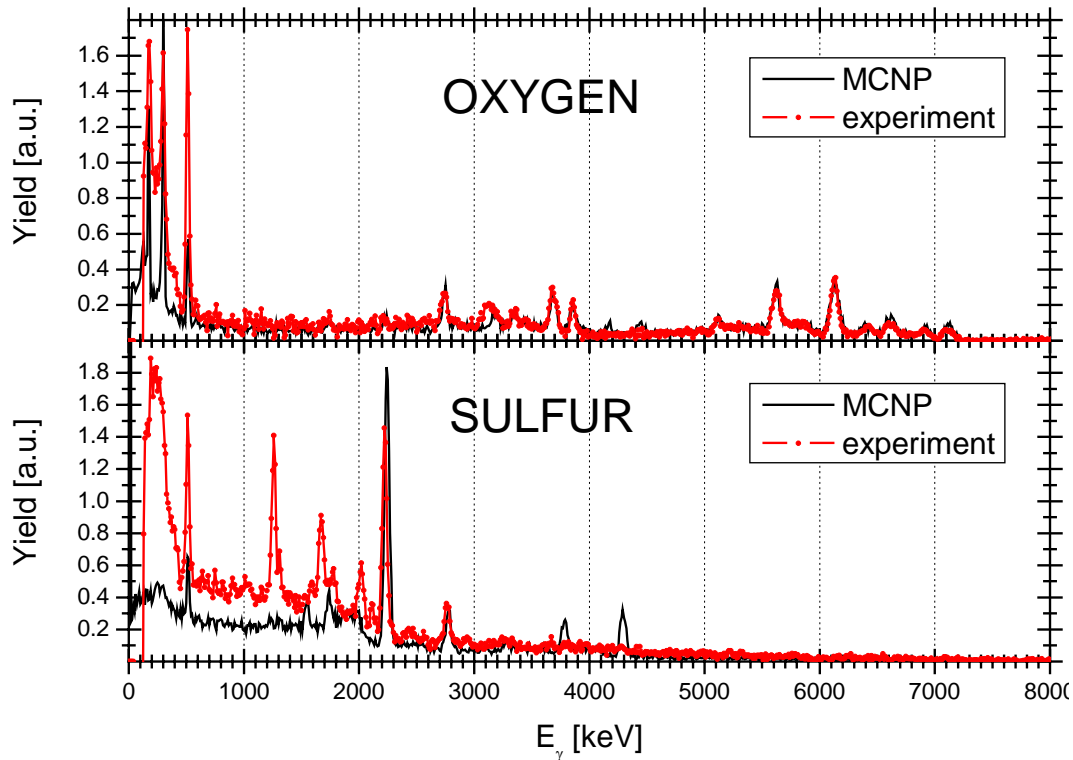*FIG. 4. Experimental and calculated (MCNP) response functions of $LaBr_3$-based γ-ray detector to oxygen and sulfur.*

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[1] MCNP – A General Monte Carlo N-Particle Transport Code, Version 5, LA-UR-03-1987.

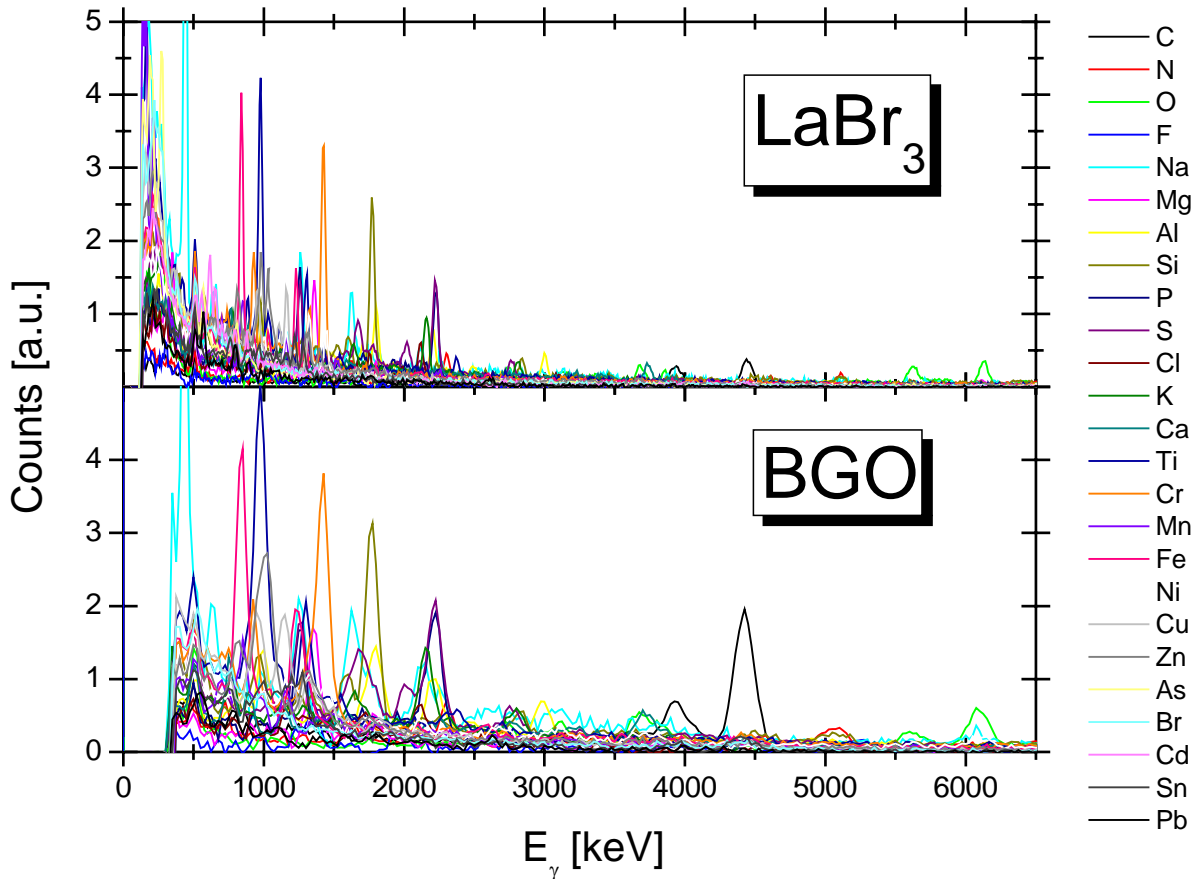*FIG. 5. Response functions for BGO-based and LaBr₃-based detectors.*

Experimental data were analyzed to produce spectra of γ-rays for the 3D "voxel" containing the sample. These spectra were normalized to the actual number of the detected α-particles to get rid of the variations of the intensity of the neutron generator. Pre-measured contributions from the background, plastic container, and "parasitic" elements (e.g. oxygen in oxides) were then subtracted from the spectra.

As one can see from FIG. 4, the quality of the MCNP simulation is different for different elements. While for oxygen the response function calculated by MCNP is very close to the experimental one, for sulfur the standard MCNP5 cross-section library describing (n,n'γ) reactions is definitely wrong.

The superior energy resolution and high light output of γ-ray detectors based on LaBr₃ crystals allow one to get response functions with rich peak structure and in wide dynamic range – in our case from 100 keV to 10 MeV. Response functions measured with BGO and LaBr₃ are shown at FIG. 5.

As a result of these measurements, libraries of experimental response functions of BGO and LaBr₃-based detectors were created. These libraries were used in the analysis of the spectra measured for UXO imitators (listed in TABLE 1) lying on the surface of sand. In this experiment the masses of the UXO constituents were determined, and then compared to the known chemical composition of the imitators (FIG. 6).
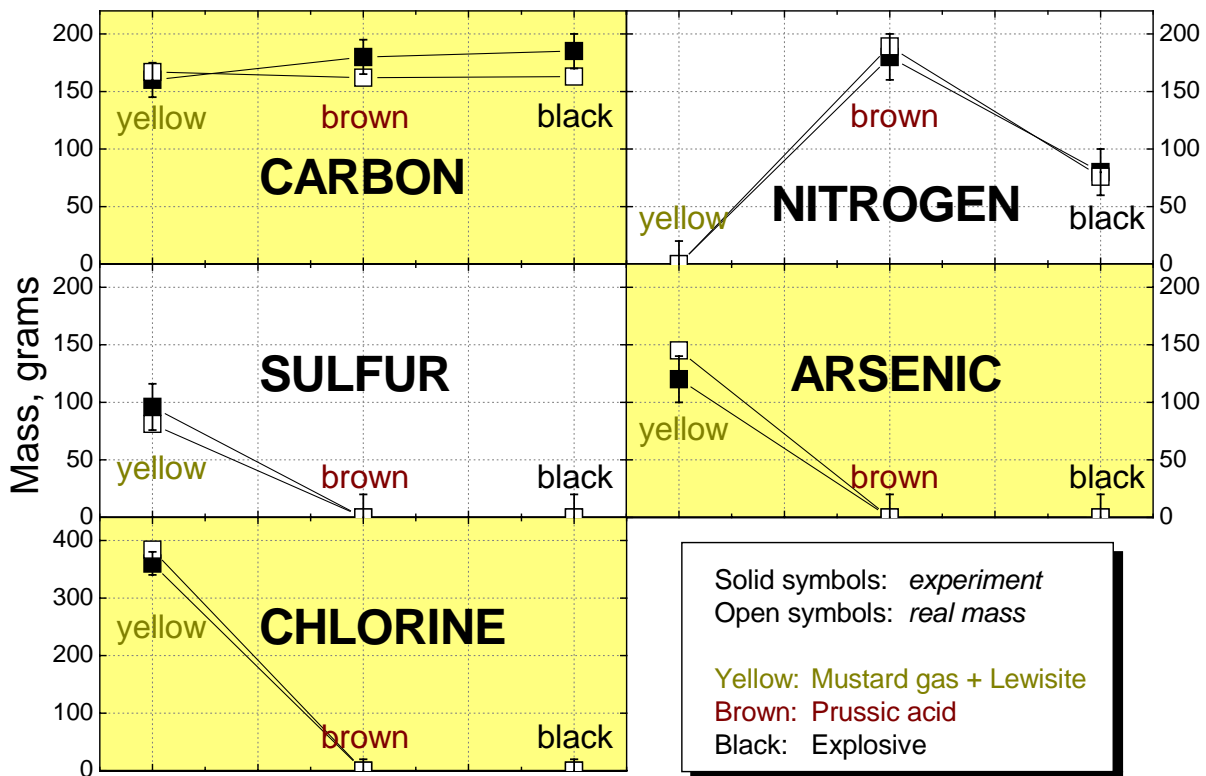
FIG. 6. Experimental determination of masses of chemical elements in UXO from spectra of γ-rays measured with LaBr₃ detector.

One can see, that masses of carbon, nitrogen, sulfur, arsenic, and chlorine were reproduced with good precision. The mass of oxygen cannot be used for identification since the unknown amount of oxygen from sand contributes to the measured spectra.

4. Conclusions

Devices based on Nanosecond Neutron Analysis have been successfully used to automatically identify concealed explosives, hazardous chemicals, and UXO both on a conveyer and in the ground. Superior characteristics of the LaBr₃-based γ-ray detector make it a perfect candidate for use in NNA/APT devices, the only drawback being its high price.

5. References

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