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Hazardous Substance Detection in Water Environments using Neutron Beams: the SABAT Project

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Abstract. The ever-increasing risk of terrorist attacks is a stimulus for seeking new, more effective danger detection methods. The article describes new methods of detecting hazardous materials based on the interaction between neutrons and matter. The status of the design of the device for detecting hazardous materials in water environments based on these methods, currently in development at the Jagiellonian University (Kraków, Poland), will be presented as well.
Keywords: nuclear physics, public safety, antiterrorism, Baltic Sea

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1. INTRODUCTION

In the 21st century, the risk of terrorist attacks is growing all across the globe, even in places commonly deemed safe. This fact is a stimulus for seeking new, more effective danger detection and prevention methods. New methods are needed especially for monitoring and protecting ports and coastlines.

Seeking new hazmat detection methods is also necessary for the protection of water environments that have witnessed intense war activities, especially the Baltic Sea. The remnants of war equipment and weapons constitute a serious hazard for sailing, and the toxic substances contained in some projectiles, such as gases used in chemical warfare, are a serious ecological hazard. After World War II, over 25,000 tons of dangerous substances were sunk in the Baltic Sea, especially in the Gotland Deep and the Bornholm Deep [1]. Apart from the official chemical sinking sites, the Baltic also has many unknown weapons resting sites, situated chiefly along the old sea convoy routes. Some of these projectiles are already corroding to such an extent that the gases that they contain, such as mustard gas and tabun, are infiltrating into the sea, causing the contamination of its bottom. There may exist, however, unknown sinking sites, which, situated in shallow locations, may constitute a direct danger, i.e. for fishermen while fishing. Detecting and identifying war remnants at the bottom of the Baltic Sea is pivotal to the continuous endeavours to free it of hazardous substances [2].

The methods of hazmat detection applied presently are based mainly on the use of X-rays, which, interacting with electrons, allow shapes and the density distribution of the examined objects to be determined; they do not, however, permit their identification. In airport safety systems, the analysis of trace amounts of materials is used as well, while the anti-terrorist practice employs devices based on radar methods and induction detectors. Unfortunately, they only allow for detecting the presence of metal elements, or determining the shapes of underground objects. Therefore, the detection of every suspicious item requires additional human verification. Important limitations exist due to the low probability of X-rays' interaction with the light elements making up the majority of hazardous materials, such as nitrogen, oxygen, hydrogen and carbon. Furthermore, modern explosives can be given virtually any shape, rendering their detection practically impossible [3].

In water environments, the main method employed for hazardous chemical detection is sonar; however, it only allows the object's location and shape to be determined, with no indication as to its chemical composition.

The limitations of the commonly-applied methods and the increasing demand for mobile devices allowing for monitoring sea areas and clearing them of mines are a stimulus for seeking new hazmat detection methods. One of the most promising methods is Neutron Activation Analysis (NAA), entailing the measurement of the examined substance's stoichiometry using neutron beams. This method will be described more broadly in Chapter 2 of this article.

Afterwards, the status of the underwater hazmat detection device currently being developed at the Jagiellonian University (Kraków, Poland) within the framework of SABAT collaboration will be presented.

2. USING NEUTRON ACTIVATION ANALYSIS (NAA) IN HAZARDOUS MATERIAL DETECTION

The vast majority of hazardous materials are either organic compounds or mixes thereof. They are mainly made of hydrogen, oxygen, carbon, and nitrogen. This permits, for example, the detection of hazardous materials or narcotic drugs hidden among other substances during stoichiometric analysis of the suspicious items [3]. Such an analysis may be conducted with the use of neutron beams, which, penetrating a given material, interact with the atomic nuclei of the unknown substance, exciting them. As a consequence, neutron irradiation leads to the emission of γ quanta characteristic for a given isotope (Fig. 1).

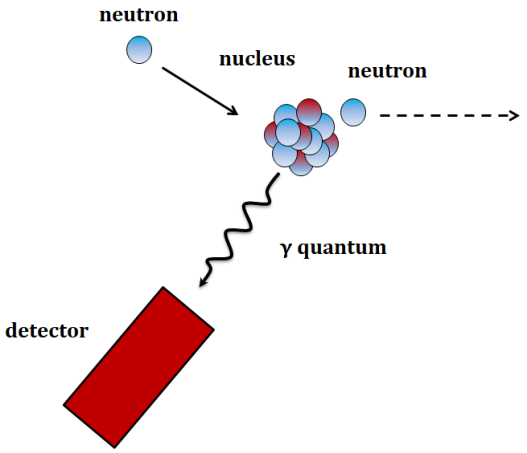

Fig. 1 A schematic representation of the principle of substance stoichiometry examination using neutron beams on the example of non-elastic scattering of those particles on atomic nuclei.

They are registered by the detector, and determining the number and energy of the photons emitted leads to identifying the substance. Considering the probability of the neutrons' interaction with various nuclei and the usually varying effectiveness of the detector in registering gamma quanta possessing a given energy level, one may obtain the numbers of atoms of the particular elements making up a given object and, therefore its stoichiometry [3].

An important advantage of NAA is the high hardness of both the neutrons and gamma quanta, allowing for searching hazardous materials hidden underground, behind an obstacle, *et cetera.*

Furthermore, it is one of the few methods allowing for non-invasive examination of a substance's elemental composition [3].

Out of the many neutron-matter interaction processes, NAA employs non-elastic fast neutron scattering and thermal neutron absorption [5]. Both of these processes lead to excitation of the nuclei of atoms and the emission of the characteristic gamma radiation.

Several NAA-based detection systems are in development in the USA, e.g. by Science Applications International Corporation [6] and CALSEC [7], and in Europe (Sodern [8], the EURITRACK [9] and SWAN [10] projects). In water environments, NAA identification encounters serious problems due to strong neutron absorption in water and the ample background originating from the excited oxygen and hydrogen. The only existing solution is UNCOSS [11]. This device uses neutrons created in mobile generator which undergo moderation in water and reach the examined item with fairly little energy. Due to this, the distance between the sensor and the examined item must be as small and possible, and detection of hazardous materials at the sea bottom is very difficult. Furthermore, the strong neutron and gamma quanta absorption in water markedly increases the time of irradiating the suspicious item and makes interpreting the results difficult. The non-invasive device for detecting hazardous materials in water, currently in development at the Jagiellonian University within the framework of the SABAT project, is free of such limitations [2]. It is characterised by a greater sensitivity and reduced noise (background reduction), owing to which it will allow for more precise detection of hazardous materials, even of those deep at the bottom of the water reservoir, and for determining the density distribution of the hazardous material in the examined object. The description of the detector being designed is presented in the next chapter.

3. SABAT (Stoichiometry Analysis By Activation Techniques)

The diagram of the device being developed within the framework of the SABAT project is presented in Fig. 2. As the source of neutrons, a compact neutron generator accelerating deuterium ions that collide with the tritium found on the generator's disc will be used. As a result of the reaction between those isotopes, a compound nucleus is created which subsequently decays into an α particle and a neutron with an energy of 14.1 MeV, moving in opposite directions. Due to the markedly greater energy released in this reaction as compared with the energy of deuterium, both the alpha particle and the neutron are practically produced isotropically in every spatial direction.

In modern generators, the alpha particle is registered by the detector system situated on the generator's walls (Fig. 2). It may be composed of, i.a., a system of silicon or scintillating detectors, several centimetres in size.

The chosen neutrons move towards the suspicious item within a guide of determined dimensions, filled with air or another gas. This prevents the absorption and slowing of neutrons in water. Moving within the examined substance, the fast neutrons are absorbed and/or scattered non-elastically on the atomic nuclei of the examined item, exciting it.

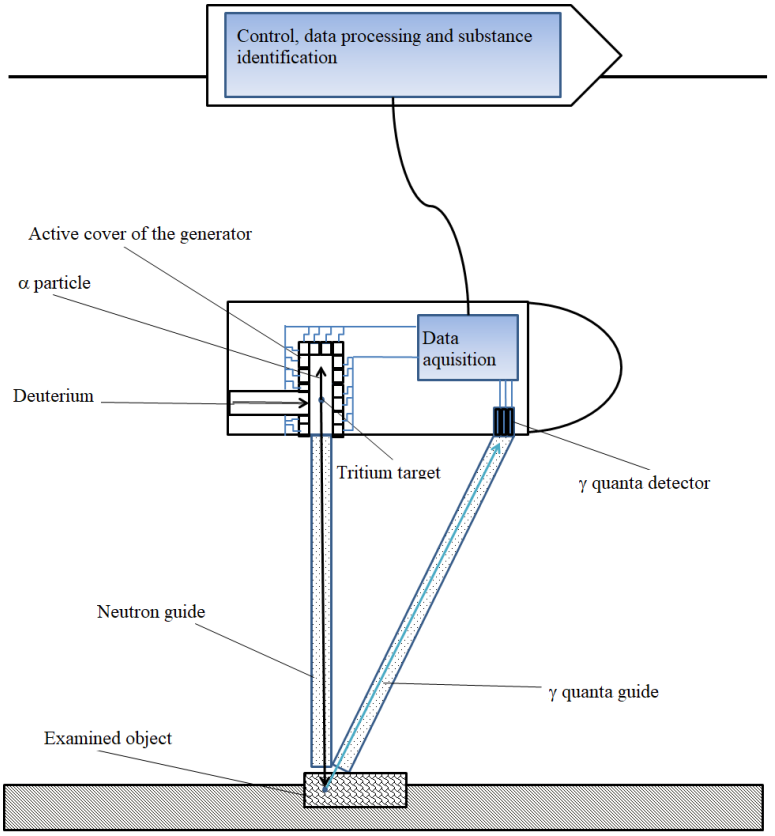

Fig. 2 Diagram of the device being developed within the SABAT project. The underwater drone carrying the compact neutron generator provides a beam that, travelling through the guide, reaches the examined object with no dispersion on the way. Similarly, a fraction of the gamma quanta emitted into the solid angle encompassed by another guide reaches the detector without reacting with the water, facilitating identification. Drawing adapted from [12].

A part of the γ quanta emitted by the excited nuclei while going back to the ground state move towards the detector within another guide. This prevents the absorption and scattering of gamma quanta in the water. The detector conducts a measurement of the energy of the registered quanta.

Additionally, the position of the gamma quantum hit in the detector, as well as the time elapsed between the registered alpha particle and the signal in the detector is recorded.

The measurement of the time and place of the alpha particle and gamma quantum's reaction, along with the known position of the tritium disc and the change of the relative distance and the angle of the gamma-quantum guide with respect to the neutron guide allows the density distribution of the hazardous substance in the examined item to be determined [2].

If the ratio between the guides' diameter and length is sufficiently small (less than 0.14), the depth of the gamma quantum's reaction may be determining by measuring the time elapsed (Δt) from the moment the α particle was registered until that of the signal's registering in the gamma quantum detector. It may be expressed as

$$\Delta t - t_\alpha = t_n + t'_n + t_\gamma + t'_\gamma,$$

where t_α is the time taken by the α particle to travel from the disc to the detector (Fig. 2), t_n and t'_n are respectively the time the neutron takes to travel within the guide starting at the disc on the known, determined path l_n, and the time the neutron takes to travel from the end of the guide to the place of reaction in the given object [2]. Similarly, t_γ stands for the time travelled within the guide at the known and determined length l_γ, and t'_γ is the time taken by the γ quanta to travel from the neutron's place of reaction within the examined object to the guide's inlet. These times may then be expressed by the known particle velocities

$$\Delta t - l_\alpha/v_\alpha = l_n/v_n + x/v_n + l_\gamma/c + y/c.$$

The alpha particle's velocity, as well as that of the neutron, is determined by the known energies of those particles, while gamma quanta travel at the speed of light (c). The path travelled by the neutron (x) from the guide's ending to the place of reaction in the object, as well as the path travelled by the γ quanta from the place of the neutron's reaction to the guide's inlet are connected by the following relationship:

$$x/y = \cos\varphi,$$

where φ is the angle between the guides' axes which can be adjusted.

This allows the distance from the inlet where the reaction took place to be determined [13]:

$$x = \left(\Delta t - \frac{l_\alpha}{v_\alpha} - \frac{l_n}{v_n} - \frac{l_\gamma}{c}\right)\frac{c v_n \cos\varphi}{c \cdot \cos\varphi + v_n}.$$

If the ratio between the guide diameters is large, performing the Δt measurement permits the determination of the depth of the neutron's reaction (x) by seeking a point within the area encompassed by both guides for which the time of the neutron's travel from the tritium disc to it, and the time of the γ quantas' travel from it to the detector are the closest to the measured time Δt [2]. Additional information on the depth is provided by changing the relative position of the guides and the angle between them. This allows for registering the gamma quanta emitted from various parts of the object - from various depths as well - and determining the density distribution of the hazardous substance in the examined object.

Signals from the alpha-particle and gamma-quantum detectors are relayed to the signal sampling and data acquisition module using the signal lines (Fig. 2). In order to remove the measurement background noise resulting from the reactions of neutrons not emitted towards the examined object, only signals from the γ quantum detector registered in coincidence with the α particles located opposite to the quantum guiderail are treated as γ quanta coming from the examined substance. The data acquisition module then transmits the data via the signal cable or radio to the processing module on the water vessel the detector is controlled from. There, statistical analysis of the collected data is conducted, and the object is determined to contain hazardous substances or not.

Given their excellent energy resolution capabilities, germanium detectors are usually the ones to be used as gamma quantum detectors [14]. They are, however, sensitive to neutron radiation, and age very quickly when exposed to it (radiation damage). Moreover, they are slow and do not allow for precise time measurement. Due to this, they are replaced with scintillation detectors ever more frequently, as they also do not require cooling – a requirement which significantly reduced the mobility of semiconductor detectors.

4. PROJECT STATUS

In order to design and optimise the SABAT sensor, detailed Monte Carlo simulations of the entire system are being conducted. The results of the preliminary simulations are presented below, allowing for determining the geometry and materials required to build the device. To describe the neutron and gamma quantum transport, the MCNP (Monte Carlo N-Particle Transport Code) software package is used [15].

A diagram of the simulated scene is presented in Fig. 3. The 300 x 300 x 50 cm^3 drone (blue rectangle) contains the neutron source (yellow dot) and the gamma quantum detector (red square). The gamma quanta and neutron guides (blue rectangles) are simulated as a 20 x 20 x 10 cm^3 cuboid and a 26 cm tall polyhedron with 20 x 7.56 cm^2 and 16.7 x 20 cm^2 bases. The hazmat container, 194 x 50 x 50 cm^3, (green rectangle) is situated at the sea bottom whose dimensions are 400 x 400 x 151.5 cm^3.

Both the drone and the guides are filled with air at atmospheric pressure [14]. Due to the fact that the sensor must be optimised for the detection of underwater wartime remnants, especially gases used in chemical warfare, the simulated hazardous substance is mustard gas, $C_4H_8Cl_2S$.

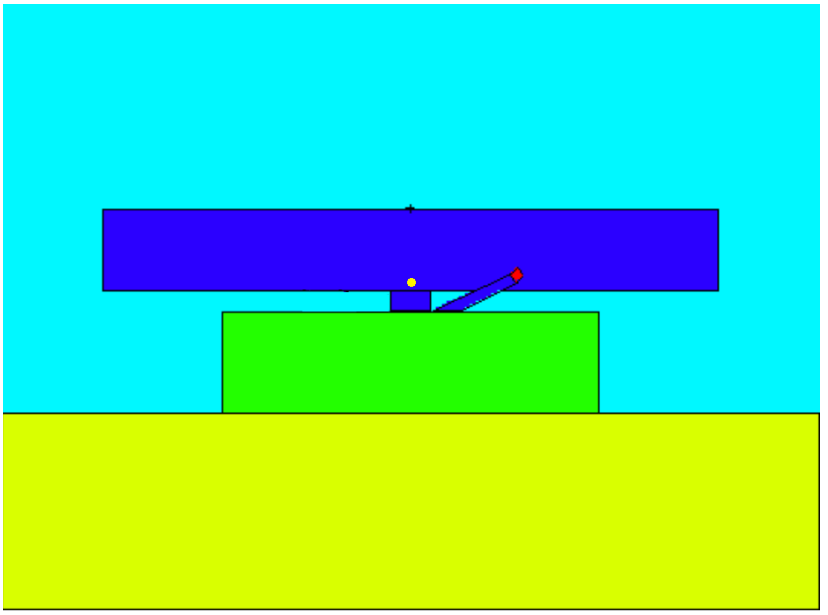

Fig. 3 An example scheme of the simulated geometry of the SABAT sensor. The particular parts of the scene have been described in the text

14.1 MeV neutrons are generated by isotopically, and only a small portion of them pass through the guide to the examined gas container without any interaction. Those neutrons may activate nuclei of the gas elements by non-elastic scattering and neutron absorption, and a part of the gamma radiation emitted as the nuclei de-excite to their basic state reaches the detector, which registers the time and the deposited energy. For initial device geometry optimisation and the materials used, the simulations are performed without considering the registration of α particles.

This also allows for checking whether it is possible to lower the costs of the sensor (generators with α particle detectors are much more expensive than those without them) and reduce its complexity. The use of scintillation crystals as gamma-quanta detectors instead of semiconductor detectors has been adopted. Scintillation hodoscopes are made of a material that produces flashes of light (scintillation) under the influence of radiation, optically connected to a photomultiplier converting light into electric impulses.

Gamma radiation spectroscopy employs, above all, inorganic crystals containing elements with large atomic numbers. They have a relatively large detection efficiency (owing to their large density) and a good energy resolution. Table 1 shows the most popular scintillation materials used for registering gamma radiation. The energy resolution is influenced by the light efficiency, or the average number of photons produced by the scintillator per unit of deposited energy. The temporal properties of scintillation detectors, on the other hand, are associated with the time of emitting the deposited energy as photons.

Table 1. The properties of the most-commonly used scintillation materials [16]

Scintillator	Light efficiency [photons/keV]	Emission time [ns]	Energy resolution (662 keV) [%]	Density [g/cm^3]
Sodium iodide (NaI:Tl)	38	250	9	3.67
Barium fluoride (BaF_2)	1.8	0.7	13	4.88
Bismuth germanate (BGO)	9	300	21	7.13
Lutetium silicate (LSO)	32	40	16	7.4
Lanthanum bromide ($LaBr_3$:Ce)	73	16	3	5.08

As can be seen, the greatest energy resolution can be achieved by constructing a detector using a lanthanum bromide crystal; however, this material is more expensive than the others. Because of this, the simulations of the SABAT sensor have been performed for the said material and for cheap, thallium-activated sodium iodide. The gamma-quantum spectra from the simulated reaction between neutrons and mustard gas are presented in Fig. 4a for the sodium iodide detector, and in Fig. 4b assuming the use of $LaBr_3$:Ce.

The resulting spectra of the simulated gamma quanta contain complicated structures, coming mainly from the oxygen in the water. The 2.12 MeV line, a sulphur signature, is indistinguishable from the hydrogen line (2.2 MeV). Similarly, the most intense ^{12}C (4.4 MeV) carbon line is contaminated with the background gamma quanta, coming mainly from silicon.

Several chlorine lines are visible as well, especially with low energies. The NaI:Tl and $LaBr_3$:Ce spectrum comparison demonstrates that, without additional background noise elimination, identifying substances underwater will be nigh-impossible. Due to this, the scintillation material that has been selected for constructing the detector is lanthanum bromide. This is the scintillator for which the further simulation stages are performed.

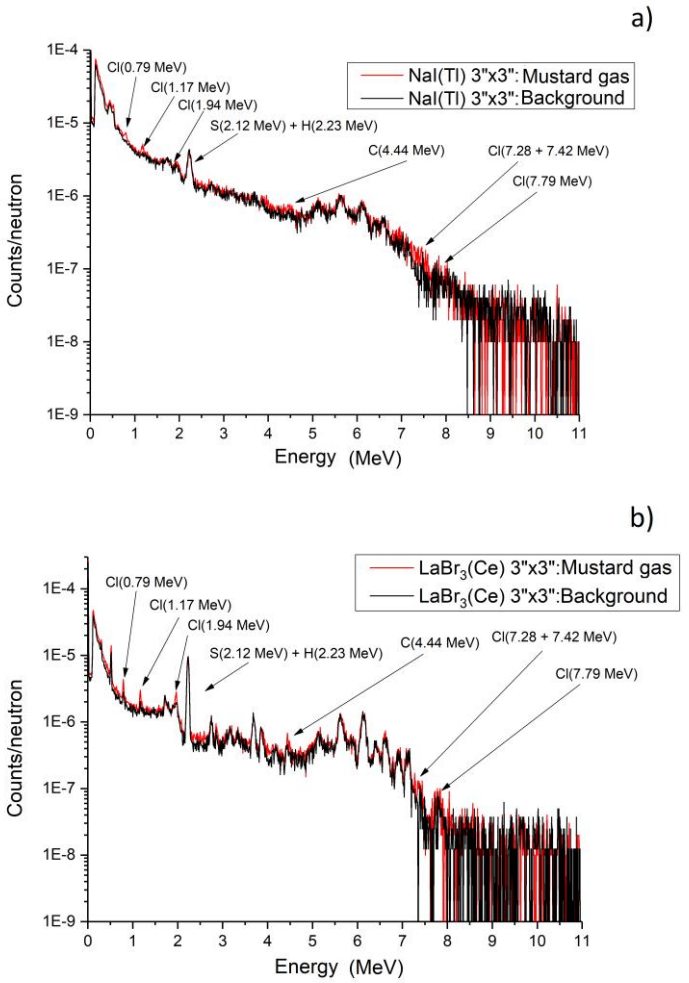

Fig. 4 The energy spectra of the γ quanta simulated with the assumption that the detector is made of thallium-activated sodium iodide (a) and cerium-activated lanthanum bromide (b). The red curve shows the simulations using mustard gas, while the black one represents the background noise coming from the surroundings. The distributions have been adapted from [14].

5. CONCLUSIONS

The ever-increasing risk of terrorist attacks is a stimulus for seeking new, more effective danger detection methods. New methods are needed especially for monitoring and protecting ports and coastlines. Employing Neutron Activation Analysis may prove an important complement to the currently-used methods.

It allows for quick, non-invasive hazmat detection by way of measuring the gamma quanta created when neutrons react with the matter of the examined object. An NAA-based device that will permit tomographic stoichiometry imaging of suspicious substances is being developed at the Jagiellonian University. The preliminary simulations of the SABAT sensor demonstrate that, notwithstanding the large amount of noise created by the interaction between neutrons and water, it is possible to detect the presence of mustard gas at the bottom of the sea using a gamma-quantum detector made of lanthanum bromide. Further information is supplied by taking into consideration the gamma-quantum registration time in relation to the time spent on generating the neutron. This allows for the rejection of a part of the background for the water, dividing the spectrum of the quanta generated during non-elastic scattering, and absorbing neutrons. Such an operation splits the sulphur and hydrogen lines, improving and accelerating their identification [14].

We are working within the framework of the SABAT project to create the first detector prototype within several years.

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Wykrywanie niebezpiecznych substancji w środowisku wodnym za pomocą wiązek neutronów: projekt SABAT

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Streszczenie. Wciąż rosnące ryzyko ataków terrorystycznych skłania do poszukiwania nowych, efektywniejszych metod wykrywania zagrożeń. W artykule opisane zostaną nowe metody wykrywania materiałów niebezpiecznych, których podstawę stanowi oddziaływanie neutronów z materią. Przedstawiony zostanie również status projektu urządzenia do wykrywania materiałów niebezpiecznych w środowisku wodnym opartego na tych metodach, które jest rozwijane na Uniwersytecie Jagiellońskim.
Słowa kluczowe: fizyka jądrowa, bezpieczeństwo publiczne, antyterroryzm, Morze Bałtyckie