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TIIVISTELMÄRAPORTTI (SUMMARY REPORT)

Detection of CBRNE materials using active neutron interrogation – phase 2 (CBRNE-aineiden havaitseminen neutroniherätteen avulla – vaihe 2)

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Abstract

CBRNE substances can be detected using active neutron interrogation. The method is non-destructive and can be used even when the substance is sealed within a package. In the present project, the method was studied empirically in a realistic measurement geometry and light elements were identified. The measurements were performed using grenade shells filled with harmless materials, such as water, salt, fertilizer, and explosives. Californium sources were used as neutron sources. The key result is that the high N/H ratio of the explosives can be detected with thermal neutron interrogation alone whereas fast neutrons are needed for the excitation of C and O, should the device be designed for other security purposes, too.

1. Introduction

CBRNE materials (Chemical, Biological, Radiological, Nuclear and Explosive) are a threat to society if they are available for the adversaries. Identifying the substances is crucial for correct countermeasures. The substances can be inside a closed package and it may be desirable to study the content of the package without opening it. Bomb threats and old warheads with unknown content are examples of such scenarios. Active neutron interrogation, or prompt gamma-ray neutron activation analysis (PGNAA) is a non-destructive method for detecting elements and substances (stochiometry). In the present project, the method was studied empirically with a realistic measurement geometry. Presently, only countries with advanced nuclear technology have the expertise and operative systems for active interrogation.

In PGNAA, gamma-ray signatures of elements are revealed in neutron-induced reactions (absorption and inelastic scattering). Detecting these gamma-ray signatures with a gamma spectrometer gives information on the elemental contents of the unknown target. The choice of a gamma spectrometer and a neutron source or generator depends on the application.

2. Research objectives and accomplishment plan

The project was planned to take two years. In the first year, the suitability of different gamma-ray spectrometers (HPGe, $LaBr_3$ and NaI) was tested with the purpose of demonstrating the capability to detect and identify CBRNE substances. Neutron generators have advantages as compared to neutron sources, providing high energy neutrons, possibility for pulsed neutron beams (observe prompt and thermalized neutrons), radiation safety advantages (emitting neutrons only when in use) and significantly higher flux compared to conventional sources. In the second year (the present project), measurements with HPGe and $LaBr_3$ gamma-ray spectrometers were performed with a realistic measurement geometry using a grenade shell. No neutron generator was available for the present studies. Therefore, the measurements were performed with neutron sources (^{252}Cf and AmBe).

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3. Materials and methods

The laboratory measurements were performed using an empty grenade shell with a caliber of about 150 mm. A 40% HPGe gamma-ray spectrometer was used as a detector. A 3"x3" $LaBr_3$ detector was also used, however, it was deemed useless without a neutron shield[1]. The detector was put inside a lead tube for gamma shielding. The tube shielded the detector from background radiation and provided some collimation in favour of the grenade. See Figure 1 for the measurement geometry where A is the grenade filled with substance under interrogation, B is the plastic shield housing the neutron source and C is the HPGe detector housed inside a lead shield. The target materials put inside the grenade shell were table salt, water and nitrogen-rich fertilizer. In phase 1 of the project, the same materials were used without the grenade shell and a different measurement geometry.

A ^{252}Cf source emitting $1.1 \cdot 10^6$ neutrons per second was used. A plastic moderator with a hole in the front side was used to have both fast and thermal neutrons incident on the target.

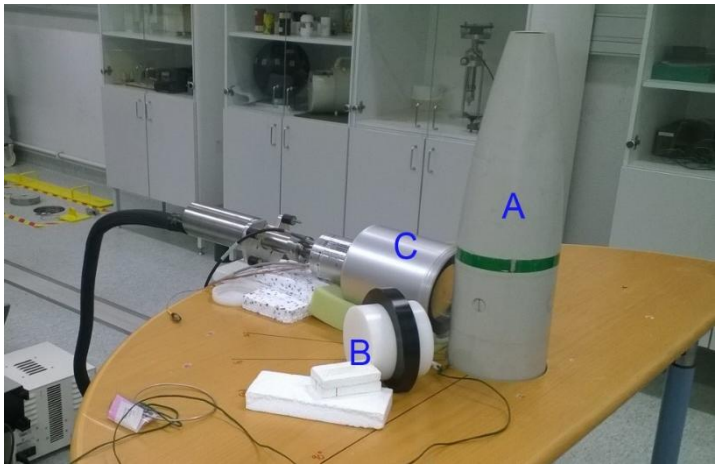

Figure 1. Measurement setup. The grenade shell filled with materials under scrutiny is in the middle (A) partially submerged into the table, the neutron source is inside the white plastic tube supported with black plastic ring (B), the detector system is housed inside a lead casing which is inside aluminium tube (C).

The measurements were repeated with a hexogen-aluminum grenade. Hexogen ($C_3H_6N_6O_6$) contains 38% nitrogen and can thus be identified as an explosive by studying nitrogen and hydrogen related peaks in the neutron-induced gamma spectrum.

4. Results and discussion

Gamma-ray signatures of nitrogen, chlorine and hydrogen were identified in the analysis. Carbon and oxygen were also present, but were not identified. Carbon and oxygen can only be detected by using fast neutrons for the excitation of the target material. Unfortunately, the ^{252}Cf source does not emit enough fast neutrons for the detection of these el-

[1] This is because of the neutron reactions with the scintillator material cause an unwanted background in the measured gamma spectrum.

ements. Figures 2, 3 and 4 present the measured spectra and characteristic gamma peaks of different elements. The results are summarized in Appendix 1.

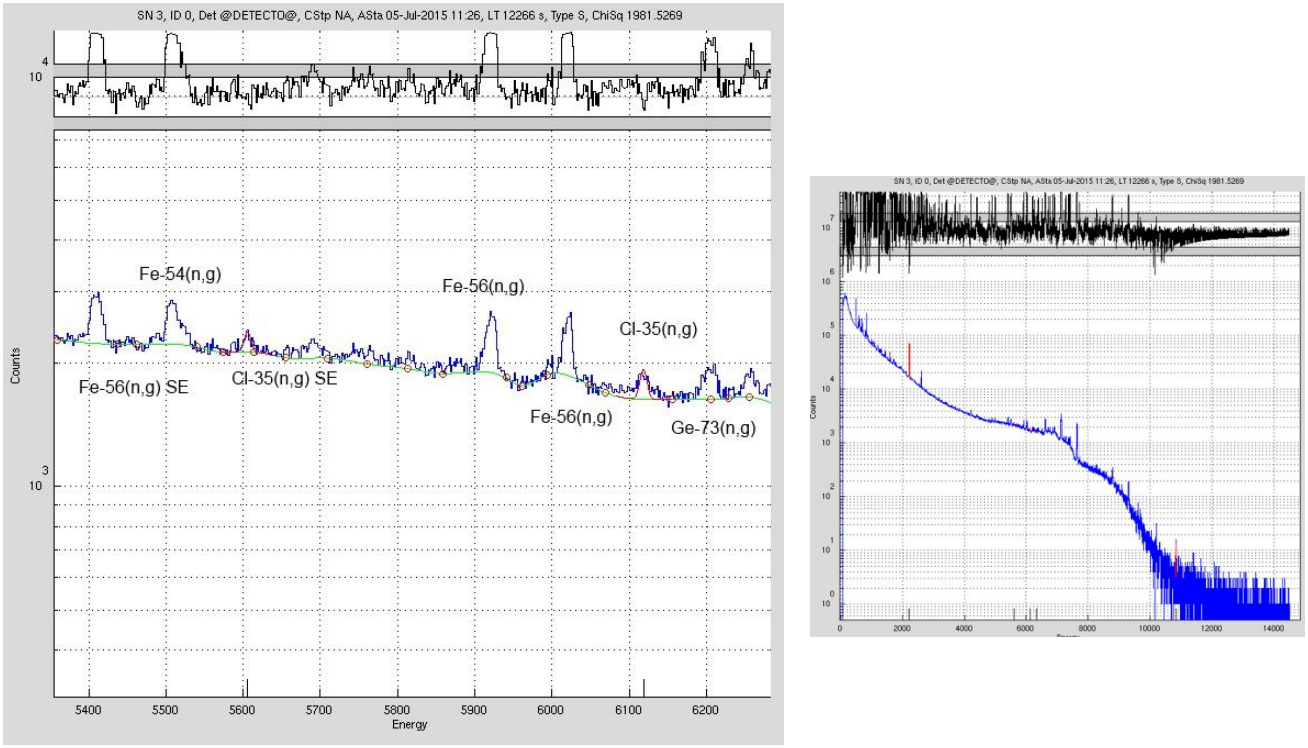

Figure 2. Signature of chlorine in a 3.4 h measurement with a 9.4 MBq Cf-252 source and HPGe detector. Chlorine emits a 6120 keV photon in a neutron capture reaction. The target was the grenade shell filled with nitrogen fertilizer (containing 10 w-% chlorine).

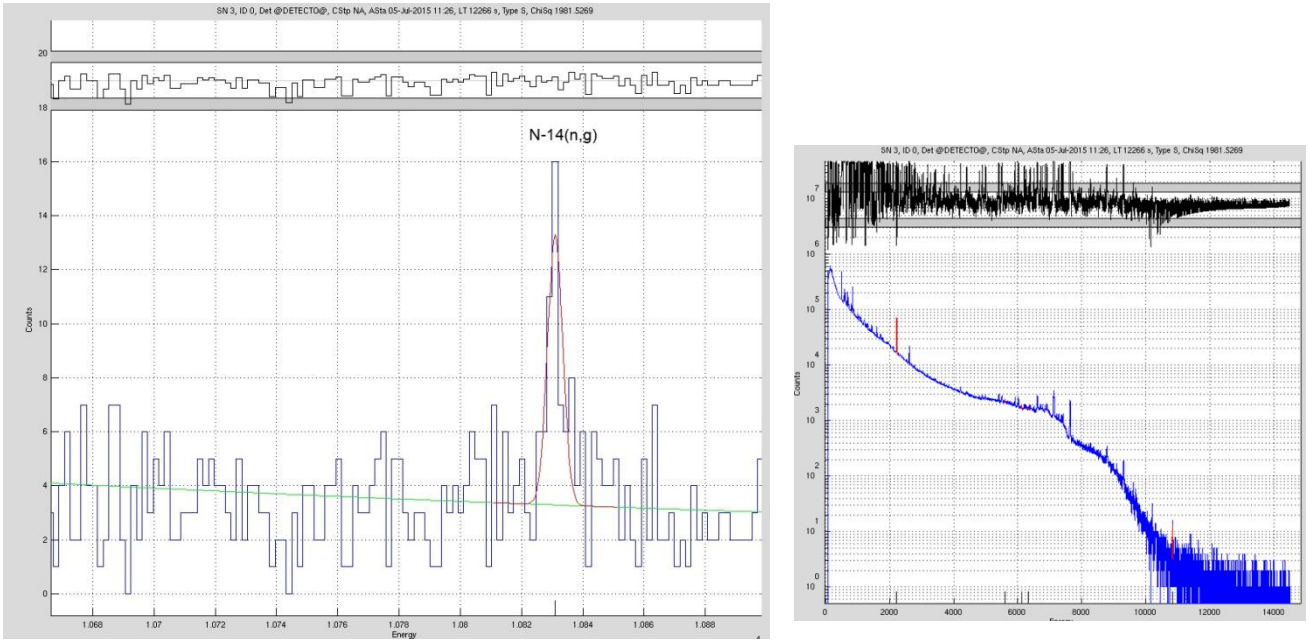

Figure 3. Signature of nitrogen in a 3.4 h measurement with a 9.4 MBq Cf-252 source and HPGe detector. Nitrogen emits a 10.8 MeV photon in a neutron capture reaction. The target was the grenade shell filled with nitrogen fertilizer (containing 27 w-% nitrogen).

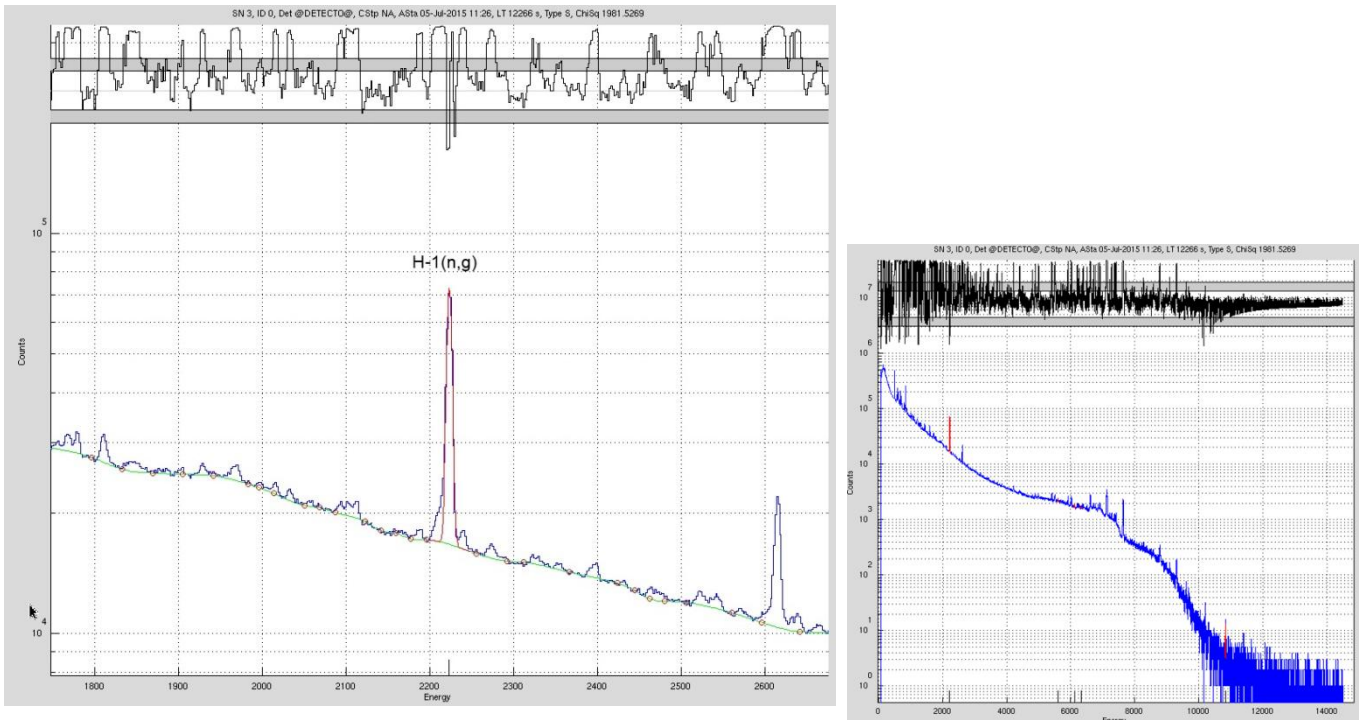

Figure 4. Signature of hydrogen in a 3.4 h measurement with a 9.4 MBq Cf-252 source and HPGe detector. Hydrogen emits a 2223 keV photon in a neutron capture reaction. The target was the grenade shell filled with nitrogen-rich fertilizer (containing 4 w-% hydrogen). Part of the peak area is due to the hydrogen in the neutron source moderator.

The laboratory measurements provided basic understanding on the order of magnitude of the detection efficiency in the given, realistic measurement geometry.

The detector setup failed during the hexogen grenade measurements and thus no conclusions can be drawn from those measurements results. However, there is more nitrogen in the hexogen grenade (1.9 kg) than in the fertilizer measurements (0.95 kg), and the nitrogen is thus in theory detectable with the current setup.

For operational use, the system must be designed to deliver results within a short time span. This can be achieved with a neutron generator which would provide 10^6 neutrons in a pulse with duration of one microsecond, and repeated with a frequency of 100 - 1000 Hz. The short pulses reduce the background significantly, and they separate thermal and fast neutron signatures from each other which is a great advantage in the spectrum analysis. Efficient use of such pulsed neutron sources requires the adoption of digital list-mode data acquisition where detected gamma-rays are stored as time stamped events. Time resolution of the system should be at the sub-µS level. In addition, the same data acquisition system would also record neutron generator's beam on/off duty cycles. All this would be made on a time synchronized manner. The neutron generator also makes neutron imaging possible which is the key requirement for studying unknown objects.

Other design necessities were also identified. A thick gamma shield is required between the neutron source moderator and the detector to absorb all of the photons emitted by neutron capture reactions in the moderator. Otherwise these are indistinguishable from the same reactions in the target material. Also, the detector should be coated with a neutron absorber, such as boron, to minimize neutron reactions inside the detector. Such a neutron shield may make the $LaBr_3$ a valid option.

5. Conclusions

The feasibility of the active neutron interrogation method was demonstrated to detect elements “without opening the package”. The resolution and efficiency of a high-efficiency HPGe detector combined with the advantages of a neutron generator provide the ability to detect CBRNE elements in an operational environment.

6. Scientific publishing and other reports produced by the research project

1. KET-TECOC-2015 ACTI 2015: Active Interrogation of an Ammunition Shell Filled with Innocent Materials, P. Holm, A.-P. Leppänen, K. Peräjärvi, H. Toivonen
2. KET-TECDOC-2015 ACTI 2015: Active Interrogation of Hexogen Grenade, P. Holm, J. Huikari, K. Peräjärvi, A.-P. Leppänen, H. Toivonen

APPENDIX 1: Gamma-ray signatures of the target materials

The following tables present some of the gamma-ray signatures of the elements measured in the present project, as well as information on whether the signature was detected in the measurements or not.

Nuclide	H-1	
Energy [keV]	Reaction	Detected in measurement
2223	(n,g)	Yes

Nuclide	C-12	
Energy [keV]	Reaction	Detected in measurement
4438	(n,n'g)	No

Nuclide	N-14	
Energy [keV]	Reaction	Detected in measurement
730	(n,n' g)	No
1634	(n,n' g)	No
1885	(n,g)	No
2313	(n,n' g)	No
5269	(n,g)	No
5298	(n,g)	No
10829	(n,g)	Yes

Nuclide	O-16	
Energy [keV]	Reaction	Detected in measurement
5618	(n,n'g)	No
6129	(n,n'g)	No

Nuclide	Cl-35	
Energy [keV]	Reaction	Detected in measurement
788	(n,g)	No
1165	(n,g)	No
1763	(n,n'g)	No
1952	(n,g)	Yes
1959	(n,g)	Yes
6111	(n,g)	Yes
7414	(n,g)	No
7790	(n,g)	No

APPENDIX 2: GEANT4 simulation

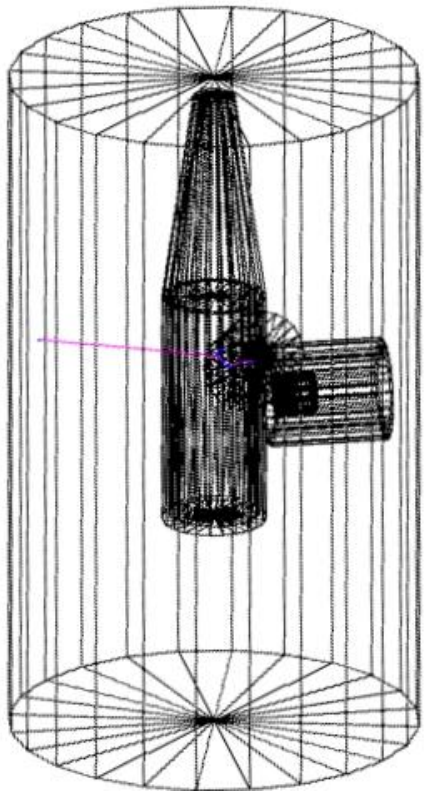

Figure 5. Geant4 simulation geometry with a phlegmatized hexogen-aluminium grenade, HPGe detector and neutron source moderator. The violet line represents a neutron path.

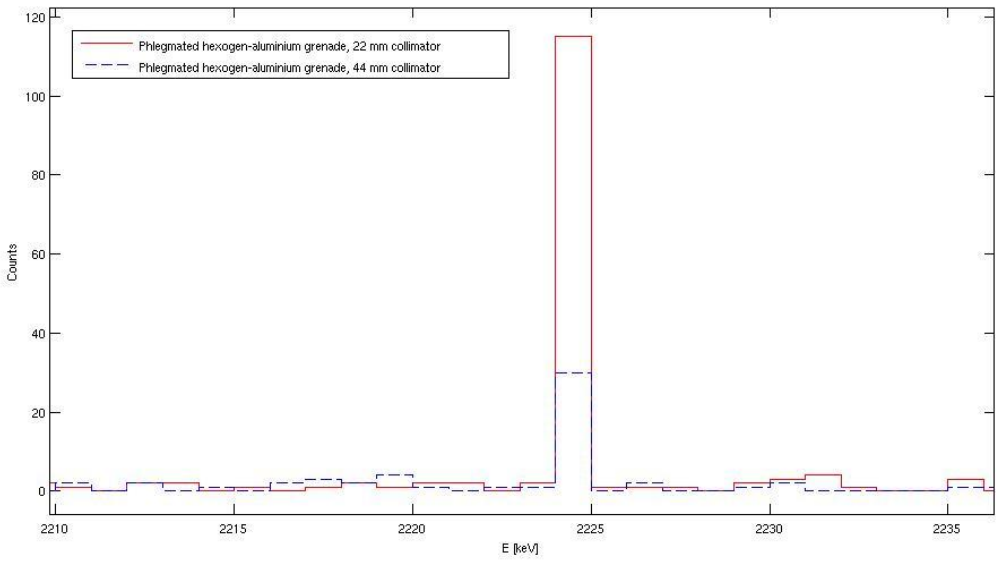

Figure 6. Simulated spectra showing the effect on the 2223 keV photo-peaks detected by the HPGe detector and emitted by hydrogen in a neutron capture reaction. Doubling the detector collimator thickness makes the peak area significantly smaller. This is mostly because the collimator shields the detector from photons emitted by the neutron moderator. Note that the simulated spectrum does not take into account the resolution of the detector