2014/2500-0002M

ISSN 1797-3457 (verkkojulkaisu)
ISBN (PDF) 978-951-25-2627-7

TIIVISTELMÄRAPORTTI (SUMMARY REPORT)

Detection of CBRNE materials using active neutron interrogation (CBRNE-aineiden havaitseminen neutroniherätteen avulla)

Harri Toivonen (harri.toivonen@stuk.fi), Philip Holm, Sakari Ihantola, Ari Leppänen, Kari Peräjärvi
Säteilyturvakeskus

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 basics of the interrogation method were studied empirically and with a literature review and simulations. Light elements (H, N, O and Cl) were identified in the measurements carried out with californium and americium/beryllium neutron sources, and then the technical specifications of a measurement system were evaluated. The present project was restricted to everyday substances: however, in a planned continuation of the project, CBRNE substances will be studied.

1. Introduction

CBRNE (Chemical, Biological, Radiological, Nuclear and Explosive) are a threat to society. 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, a measurement system for this purpose was planned and test measurements were performed. Presently, only super powers have the expertise and operative systems for active interrogation.

In PGNAA, gamma-ray signatures of elements are emitted 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 gamma spectrometer and neutron source depends on the application.

2. Research objectives and accomplishment plan

The project is 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. The measurements were performed using neutron sources (^{252}Cf and AmBe) and safe materials, such as fertilizers. Neutron generators have advantages as compared to neutron sources, providing high energy neutrons (enabling neutron reactions requiring high energy), pulsed neutron beams as well as radiation safety advantages (emitting neutrons only when in use) and significantly higher flux compared to conventional sources. One of the objectives was to perform a market and literature review of neutron generators available commercially and identify technical requirements of the neutron generator.

3. Materials and methods

The measurements were performed using gamma-ray spectrometers and neutron sources. The measured targets were water (H_2O), salt (NaCl), fertilizer (containing the elements O, N, K, Cl and H) and liquid nitrogen (N). The source and detector were placed close to or inside the target and the amount of target material used was on the order of tens of kilograms. The gamma spectrometers were a 58 mm x 54 mm HPGe detector, 38 mm x 38 mm and 76 mm x 76 mm $LaBr_3$ detector and a 130 mm x 100 mm NaI detector. Figure 1 shows one of the measurements with the three types of detectors. The target material in this measurement was water, and the measured spectrum is presented in figure 3.

Typically, the energy resolution at 662 keV is ca 6 % for NaI detectors, 3 % for $LaBr_3$ detectors and 0.3 % for HPGe. A good energy resolution is needed for identifying peaks close to each other in the gamma spectrum. NaI detectors have inferior resolution, but are available in large sizes, which correspond to large detection efficiencies. HPGe detectors have superior resolution, but require cooling systems, and Ge does not absorb gamma rays as efficiently as NaI.

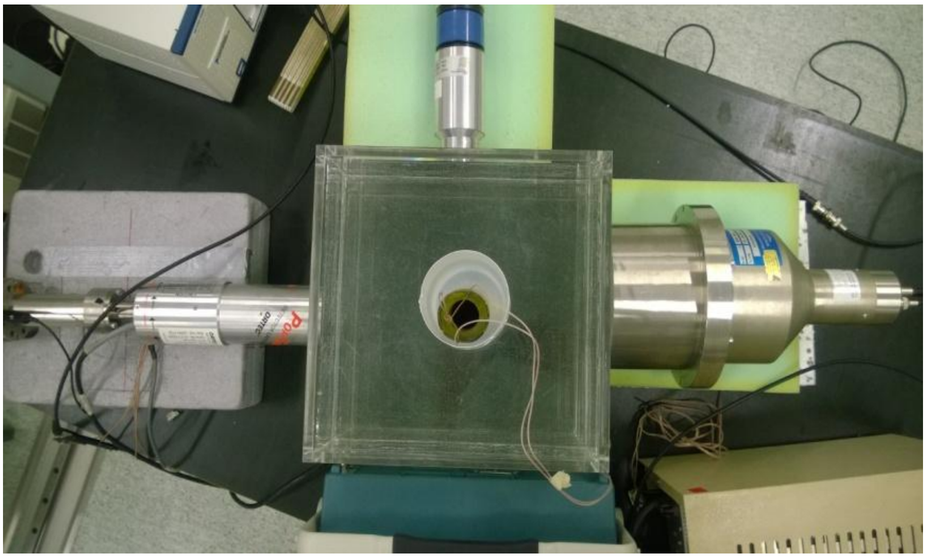

Figure 1. Measurement geometry. HPGe detector on the left, $LaBr_3$ detector on top and NaI detector on the right. In the middle is a neutron source surrounded by the target material, water. The source to detector distance is about 15 cm.

The measured spectra were compared to signatures found in the literature and databases. Monte Carlo simulations were also performed to facilitate the identification of signatures.

4. Results and discussion

All target elements could be identified in the measurements (see Figure 2, 3 and 5 and appendix). The HPGe detector proved to be the most useful due to its superior resolution. In spectra collected with the HPGe detector, the signatures are sharp peaks whereas in other detectors the signatures are larger “bumps”. Better resolution improves detection of small signatures and those signatures which are close in energy. For instance, chlorine and oxygen can be identified by photons with the energy 6111 keV and 6130 keV respectively. Distinguishing these signatures from each other requires good energy resolution.

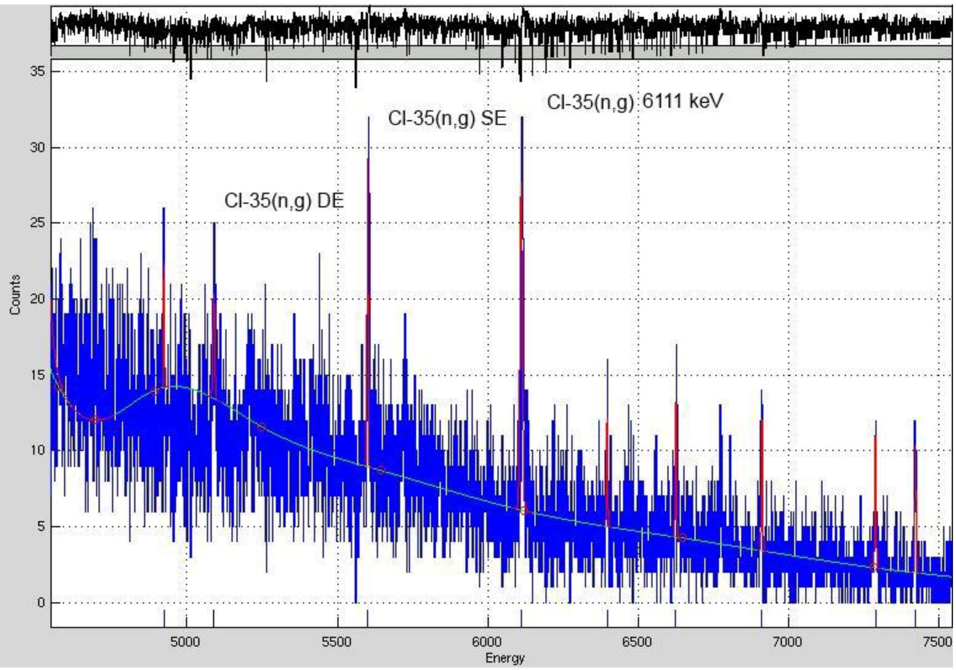

Figure 2. Signature of chlorine in a 10-minute measurement with a 10 GBq AmBe source and HPGe detector.

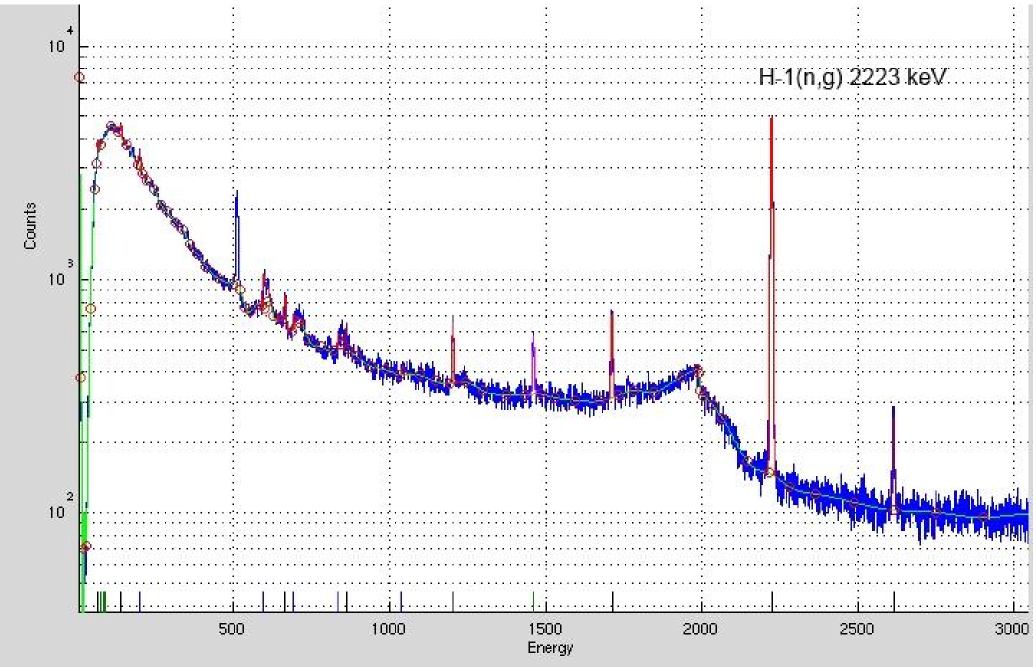

Figure 3. Signature of hydrogen in a 10-minute measurement with a 10 GBq AmBe source and HPGe detector.

Monte Carlo simulations with GEANT4 were used to identify elemental signatures and to study the effects of different neutron spectra on the presence of the signatures. Figure 4 presents a simulated spectrum of photons generated in the target material.

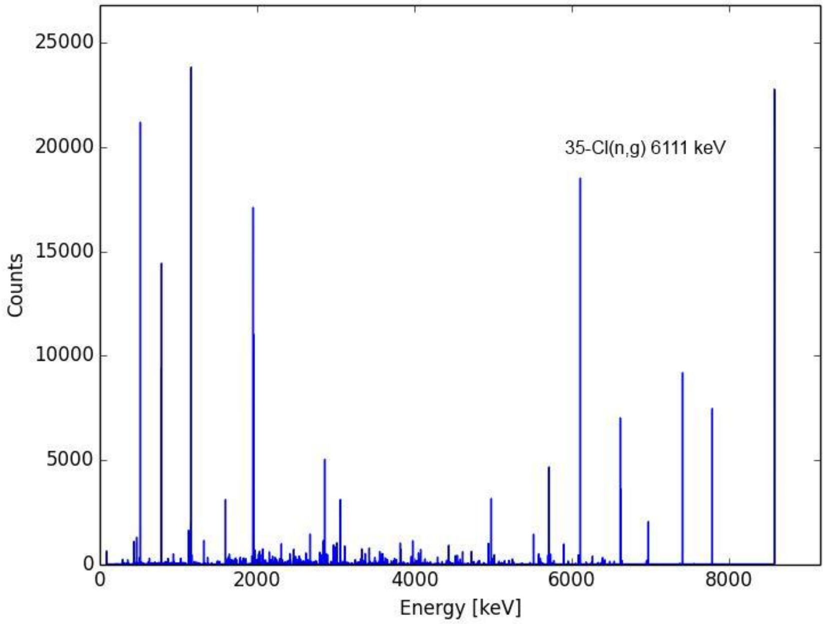

Figure 4. Simulated spectrum of photons generated by thermal neutrons in salt (NaCl).

The measurements provided understanding of the order of magnitude of the detection efficiency of the test PGNAA measurement system. With tens of kilograms of target material, neutron emission rates around 10^6 neutrons per second, measurement times in the order of 1000 seconds, most signatures were detectable with the HPGe detector (most absolute detection efficiencies were 10^{-5}-10^{-8} counts/neutron). Nitrogen was hard to detect, but was detected by moderating the neutron source, using the more efficient $LaBr_3$ detector and measuring overnight (the absolute detection efficiency for the signature was on the order 10^{-8}-10^{-9} counts/neutron). Figure 5 presents the measured nitrogen signature peak.

For operational use, a significantly better performance must be designed (< 1000 g in 10 s). 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. In addition, the neutron generator also makes neuron imaging possible which is the key requirement for studying unknown objects.

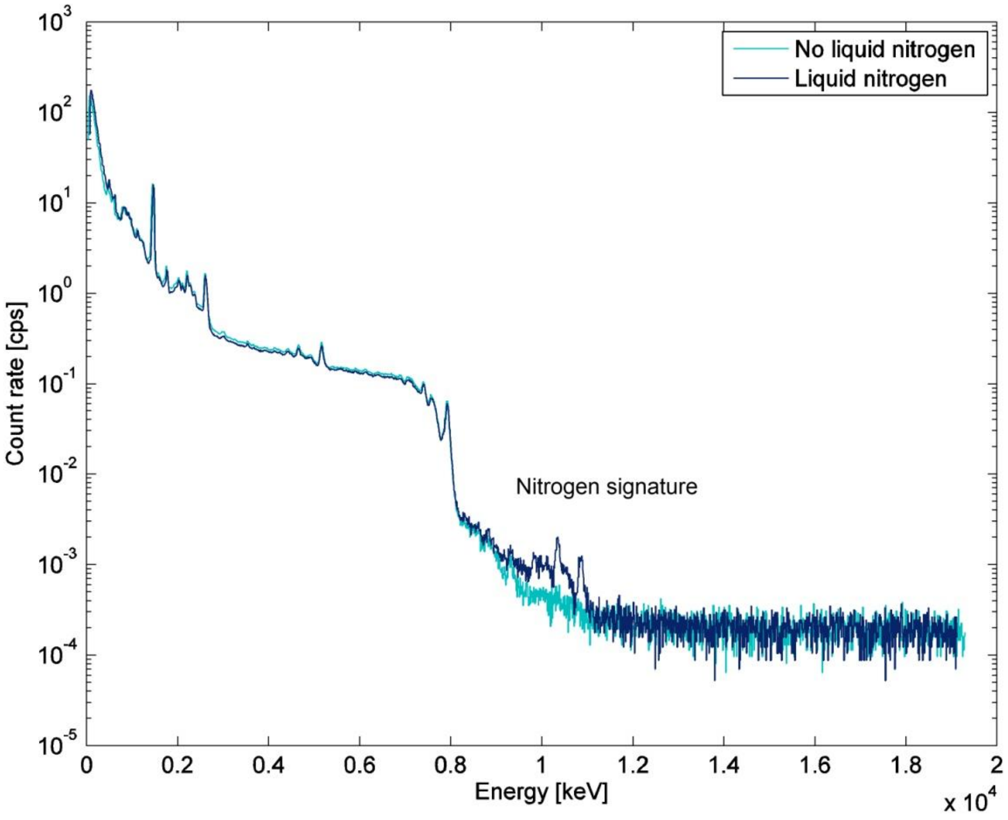

Figure 5. Spectra of the liquid nitrogen. The Cf-252 source activity was 11.2 MBq and the measurement times were 16h and 17 h for nitrogen and background, respectively.

The market review of the neutron generators showed that many portable, pulsed neutron generators have a neutron emission rate on the order of 10^8 – 10^9 neutrons/second, or about 100 – 100 times larger than the emission rate of the sources used in the present measurement. The price of such a neutron generator is around 100 000 €.

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. Further research is needed to understand the identification of CBRNE substances, as opposed to just elements. The literature and market review found several prototype applications and some commercial systems, showing that the method can be successfully used for CBRNE substance identification. The present project was restricted to measuring only safe materials. A continuation of the project is planned for 2015 with the aim to study properties of CBRNE substances.

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

1. TTL-TECDOC-2014-002 Data tables for material analysis with neutrons, P. Holm, S. Ihantola, A. Leppänen, K. Peräjärvi, H. Toivonen, STUK, 2014.

    This report contains data (cross sections and energy levels) needed for material analysis using neutron reactions such as neutron capture and inelastic scattering.

2. TTL-TECDOC-2014-003 Active neutron interrogation: Measurement campaign 1, S. Ihantola, P. Holm, A. Leppänen, K. Peräjärvi, H. Toivonen, STUK, 2014.

    Neutron-induced gamma radiation of different materials was studied. The targets were irradiated both with AmBe and Cf-252 neutron sources. The target materials included water (H_2O), salt (NaCl) and fertilizer. To measure the gamma radiation, HPGe, LaBr and NaI detectors were used.

3. TTL-TECDOC-2014-006 Active neutron interrogation: GEANT4 simulations, P. Holm, S. Ihantola, A. Leppänen, K. Peräjärvi, H. Toivonen, STUK, 2014.

    The measurements were simulated with a GEANT4 Monte Carlo model. The simulations were performed with different neutron energy spectra (Cf-252, AmBe, 14.1 MeV and 2.5 MeV neutrons). The target materials of the measurements were simulated (H_2O, NaCl, fertilizer (O, N, K, Cl and H) and blank (air).

4. TTL-TECDOC-2014-010 Active neutron interrogation: Liquid nitrogen measurement, P. Holm, S. Ihantola, A. Leppänen, K. Peräjärvi, H. Toivonen, STUK, 2014.

    The 10.8 MeV nitrogen neutron capture signature was detected using a moderated Cf-252 source, liquid nitrogen and a 3”x3” $LaBr_3$ detector.

5. TTL-TECDOC-2014-011 Active neutron interrogation: Summary of signatures, P. Holm, S. Ihantola, A. Leppänen, K. Peräjärvi, H. Toivonen, STUK, 2014.

    Summary of PGNAA signatures from the literature and MATINE measurements.

6. TTL-TECDOC-2014-012 Active neutron interrogation: a market review, A. Leppänen, STUK, 2014.

    An overview of the available commercial neutron generators. The aim was to find a generator which produces high energy neutrons at 10^6 neutrons/pulse intensity in 1 µs interval at 1kHz rate.

7. TTL-TECDOC-2014-013 Associated Particle Imaging (API), A. Leppänen, STUK, 2014.

    An overview of the technology which provides neutron imaging of the target.

APPENDIX: 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)	Yes
5298	(n,g)	Yes
10318	(n,g)	Indistinguishable from SE.
10829	(n,g)	Yes

Nuclide	O-16	
Energy [keV]	Reaction	Detected in measurement
5618	(n,n’g)	Indistinguishable from SE.
6129	(n,n’g)	Yes

Nuclide	Cl-35	
Energy [keV]	Reaction	Detected in measurement
788	(n,g)	No
1165	(n,g)	Yes
1763	(n,n’g)	Yes
1951	(n,g)	Yes
1959	(n,g)	Yes
6111	(n,g)	Yes
7414	(n,g)	Yes
7790	(n,g)	Yes