NOVEL COINCIDENCE TECHNIQUES, DETECTORS AND CONCEPTS FOR SAFETY, SECURITY AND SAFEGUARDS*

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Research and development (R&D) projects are used to maintain and improve STUK's ability to act and advice other competent authorities correctly in conditions involving nuclear and other radioactive materials. Examples from recent or ongoing in-field and laboratory R&D projects are given. All projects introduced are related to passive detection of alpha particle emitting nuclides or neutron sources.

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1. Introduction

An overview of STUK's (Finnish Radiation and Nuclear Safety Authority) activities along the lines of novel coincidence techniques, detectors and concepts for safety, security and safeguards (3S) during the past few years is given. Instead of a comprehensive list of projects carried out at STUK, certain activities relevant to 3S are highlighted. With these selected examples the author hopes to illustrate the challenges of the 3S field in general. The article starts with in-field applications and finishes off with examples of the laboratory measurement techniques. The article concentrates on the passive detection of neutrons and alpha particle emitting nuclides.

2. In-field neutron detection techniques

Society can be threatened in various ways by RN (radioactive and nuclear) substances. Spreading them out at an urban area, for example, would cause a panic and a lot of economical losses. Growing international crime

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enhances the possibility of illicit trafficking of RN materials and terrorism relying on them. Plutonium can be passively detected via neutrons that are generated in spontaneous fission. Neutrons are also emitted by the neutron-generators (for example, Am/Be and Pu/Be) typically used in industry. Conventionally, in security applications neutron detection has been based on ^{3}He counters. The required ^{3}He gas is a side product of nuclear weapons fabrication. Due to the reduced weapons production and the increased need of neutron detectors for security applications, ^{3}He counters are nowadays expensive. This has triggered a development of novel ways to detect neutrons. Both direct and indirect techniques are developed.

2.1. Neutron detection with high energy gamma-rays

One such indirect novel approach relies on the detection of high-energy ($E > 3.5$ MeV) gamma-radiation emitted by a neutron source or created by the neutrons in the detector or surroundings. Since the natural background above 3.5 MeV is low, high-energy gamma-rays are a sensitive indicator of the presence of neutrons. The same spectrometer can simultaneously be used for the regular gamma-spectrometry ($E < 3.5$ MeV), see Fig. 1. Notice here that there are a lot of gamma-ray detectors in operational field use whose measuring range can easily be extended up to 8 MeV.

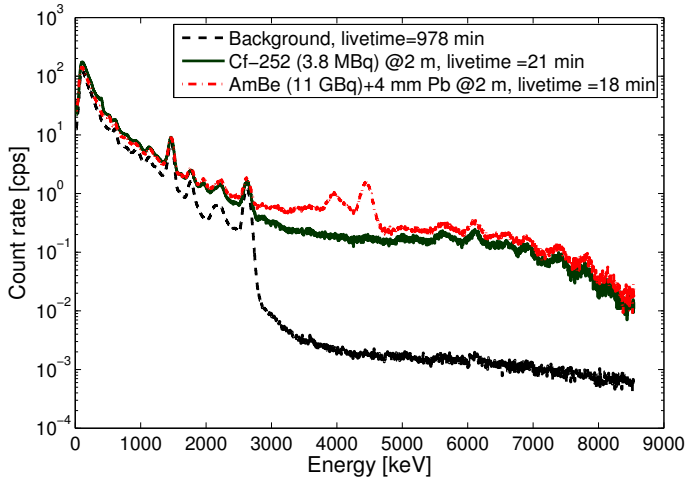

Fig. 1. Spectra recorded using PVC/PE converter/moderator showing the increased count rate at high energies due to the ^{252}Cf and AmBe sources. A source to detector distance was 2 m.

The spectra presented in Fig. 1 were recorded using a $4'' \times 4'' \times 16''$ NaI detector. The studied detector was surrounded from three sides with a 10 cm thick PVC/PE converter/moderator. PVC and polyethylene slabs were 2 cm thick. This kind of converter/moderator structure more than doubles the high energy signal [1]. A 10 cm thick sandwich converter/moderator was chosen as a compromise between the performance and size. The performance of this detector configuration was compared to the ^{3}He portal monitor (Thermo FisherScientificTUHE2 ^{3}He tube, diameter of 4.9 cm and length of active volume 81.3 cm). Cross-sectional areas of the detectors were comparable but the active volume of the NaI detector was larger by approximately a factor of three. Measurements with both bare and shielded ^{252}Cf and AmBe sources are reported in [1]. It is noteworthy in these studies that the ^{3}He tube has a higher detection efficiency compared to the NaI detector only when measuring unshielded ^{252}Cf source. In all other scenarios, the boosted NaI detector is more efficient. A 4.4 MeV gamma-ray associated to AmBe source [^{9}Be$(\alpha, n)^{12}$C] can be used for the source identification, see Fig. 1.

2.2. Capture-gated neutron spectrometer

Another neutron detector concept recently investigated at STUK is a portable capture-gated neutron spectrometer [2]. The detector is a cylindrical $3'' \times 3''$ borated plastic scintillator — EJ254 with 5% natural boron by weight, Eljen Technology. It is coupled to a photomultiplier and a Canberra Osprey digital MCA. List-mode data acquisition is employed. Analyzing both the singles spectrum and the capture-gated spectrum provides information from the source and the source shield. Developing such a comprehensive analysis approach was the main goal of the project. Testing procedures of this detection system were very similar compared to the NaI detector discussed above. The following indicators for source and source shield characterization were identified in [2]:

1. High-energy part of the singles spectrum.
2. High-energy singles to fast neutron signal ratio.
3. Thermal neutron capture peak area (in the singles spectrum) to fast neutron signal ratio.
4. Slope of the capture-gated neutron spectrum.

As an example, if the neutron source or generator is shielded with light material that will reduce the number of detected thermal and fast neutrons, then it will not influence that much to the high energy part of the singles spectrum (high energy fission or 4.4 MeV AmBe gamma-rays). The shape of the high-energy part of the singles spectrum can also be used to disentangle the type of the source, *i.e.*, it is a fission source or a neutron generator.

3. In-field alpha particle detection techniques

Alpha particle emitting nuclides are very radiotoxic if ingested or inhaled. Therefore, normal procedure in field work involving alpha particle emitting nuclides is first air sampling and then vacuum-based alpha spectrometry for the collected samples. Glass fiber or membrane filters are typically used for air filtration. If the air is found clean then the surfaces will be screened using alpha contamination meters. Swipe samples will be taken from the detected hot spots. The swipes will again be measured with a silicon detector inside a vacuum chamber. In some cases, chemical processing may be required for the collected sample before the alpha analysis becomes possible. In order to perform all these procedures in field, one needs to carry quite a lot of instrumentation along.

3.1. Collimated high resolution alpha spectrometer

To reduce the amount of required instruments, we have developed an integrated surface contamination meter and a high-resolution alpha spectrometer. Surface contamination meter is realized with a silicon detector and high resolution alpha spectrometer by placing a honeycomb collimator in front of it. The use of the collimator leads to high resolution alpha spectrometry but now in ambient air pressure [3]. The transmission of the collimator tested was about 10%. This led to the following performance parameters when the collimator was used: Contamination of 1 Bq/cm^2 at any smooth and flat surface can be detected in approximately 10 s data acquisition time. Obviously, a longer time is needed for radionuclide identification. In these studies, a 2 000 mm^2 Si-detector with an entrance window thickness of less than 2 μm silicon equivalent was used. The next step in this project is a construction of operational hand-held instrument.

3.2. Stand-off detection of alpha particle emitting nuclides

Since the ranges of alpha particles in the air are short (only some cm for 5 MeV alpha particles), the non-destructive alpha particle screening of large areas is difficult and time-consuming using the above discussed conventional technique. Special challenges arise if the alpha screening and analysis are part of the crime scene investigation. In such situation, one needs to take care not to destroy other evidence while doing the work. We have investigated alternative ways to detect alpha particle emitting nuclides. Especially, we have concentrated on the stand-off detection of alpha particle emitting nuclides using the long range UV photons indirectly created by the alpha particles during their thermalization process in the air. One 5 MeV alpha particle creates about 100 UV photons. Most of them are within the wave-

length region of 300 to 420 nm. In addition to crime scene investigation, this so-called UV-imaging has applications in emergency management and decommissioning of nuclear facilities.

We have tested αUV-imaging also at the Institute for Transuranium Elements, Karlsruhe [4]. An EMCCD camera was installed outside a glove box and the imaging was done through a quartz glass window. The glove box contained mixed oxide (MOX) pellets and plutonium nitrate powder in a test tube. The exposure times varied from 1 to 60 s. In one of the measurements, a MOX pellet was imaged when it was placed inside a plastic bag, *i.e.*, the bag contained enough air to produce UV photons that are able to penetrate the plastic. A normal transparent plastic bag is thick enough to absorb all emitted alpha particles. Therefore, if gamma-rays are not emitted or their yields are low, the UV method is the only way to detect that the bag contains alpha particle emitting nuclides without opening it. Capability to detect alpha particle emitting nuclides without opening the bag is important for the sample integrity and also for potential multithread CBRNE (Chemical Biological Radiological Nuclear Explosives) situations. Future work in this project includes: studies related to spectral filtering and background subtraction, and automated screening of wide areas. Screening of wider areas will be realized by installing the camera on a motorized panorama head and by combining multiple images.

3.3. αUV-gamma coincidence spectrometry

Determination of alpha particle emitting radionuclides causing the UV light can be continued with a position-sensitive UV-gated gamma-spectrometry technique, jointly developed by STUK and the Technical University of Tampere, Finland [5, 6]. This measurement is realized by gating the HPGe detector with UV photons. The technique was tested in [6] by measuring simultaneously ^{241}Am and ^{133}Ba sources. In the singles spectrum ^{241}Am 59.5 keV gamma-ray peak is barely visible, while in the UV-gated spectrum it is very well pronounced. Therefore, although the UV gate greatly reduces the photo-peak signal, the reduction in the background is over 500 times more intense. This enhancement leads to significant improvements in performance. As an example, an acquisition time of 40 h is required to reach a minimum detectable activity (MDA) of 3.8 Bq without the UV gate, whereas with the UV gate, the same MDA can be reached in less than 4 h. These figures illustrate the strength of the UV technique while detecting and analyzing alpha particle emitting nuclides in high external gamma-ray background caused by, for example, shorter lived beta-decaying nuclides. These masking nuclides may as well be an integral part of the sample.

The UV-gated gamma-spectrometry technique is more robust compared to the direct *in-situ* alpha spectrometry since the element and nuclide identifications are based on more penetrating X- and gamma-ray radiations. Notice here also the difference in photon and alpha particle interaction mechanisms with matter. On the other hand, low yields of gamma-rays of several alpha particle emitting nuclides reduce the attractiveness of this technique. However, while performing analysis directly from non-ideal surfaces or while analyzing unknown bare samples or samples inside transparent plastic bags, UV-gated gamma-spectrometry is the recommended technique.

3.4. Air filter analysis with alpha–gamma coincidence technique

^{241}Am gamma-ray sources are frequently used in industry. Since ^{241}Am does not emit high-energy gamma-radiation, it is quite difficult to detect if shielded. Consequently, ^{241}Am sources are once in a while accidentally melted in scrap metal processing. In these incidents, approximately 1% of ^{241}Am becomes airborne. That is why the on-line (alarming) and post-incident monitoring of air are important. STUK has developed a conventional alpha–gamma coincidence device for the post-incident *in-situ* air monitoring [7]. In the developed device collected air filter samples are measured in a close geometry between a ZnS alpha particle and NaI gamma-ray detector, see Fig. 2. In order to detect alpha particles, the collection/dirty side of the filter is facing the ZnS detector. The data acquisition of the setup is realized using Ortec digiBASE digital electronics integrally connected to the NaI

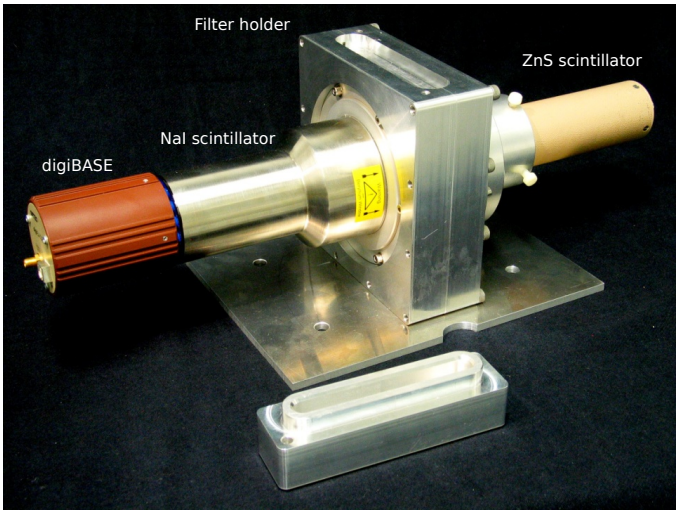

Fig. 2. A setup for the in field detection of ^{241}Am from air filters [7].

detector. Electronics accepts gating signals from the ZnS detector. During 2012, this prototype device has been successfully operated for several months at a steel melting shop. These measurements revealed that in the typical in-field operation mode (1 h sampling — filtration of 10 m^3 air, 15 min cooling of the sample, 1 h data acquisition), low concentrations of ^{241}Am can be detected ($\sim$ 10 mBq/m^3). The experimentally determined in-field performance of the device is comparable to the performance of STUKs multiple times more expensive low background gamma-laboratory (lead-shielded HPGe detectors in a low background environment).

4. Position-sensitive alpha–gamma coincidence measurements in laboratory

Since only a fraction of measurements can be made in field, also laboratory techniques for analyzing the samples need to be developed. Conventional non-destructive techniques for the analysis of samples containing alpha particle emitting nuclides are a low background gamma-ray singles spectrometry, high-resolution alpha spectrometry and X-ray spectrometry. A new technique we have recently studied at STUK is a position-sensitive alpha–gamma coincidence spectrometry using a double-sided silicon strip detector (DSSSD) and a BEGe-detector [8, 9]. The DSSSD used is a BB7 from Micron Semiconductor. It has an active area of 64×64 mm^2. The strip width is 2 mm. The BEGe detector has a diameter of 70 mm and thickness of 21 mm. Samples, such as air filters, are mounted into holders which are installed one at the time to a tip of a linear feedthrough. Feedthrough is used to move the sample between the detectors that are mounted face-to-face inside a vacuum chamber. Source to detector distance is typically few mm. The DSSSD allows position-sensitive alpha particle screening of samples. The resulting alpha particle hitmaps can be used to quickly verify the nature of the sample, *i.e.*, if it contains individual particles or the activity is more evenly spread. Detected particles can, for example, be compared and further characterized by setting position gates to the DSSSD data and by analyzing the resulting alpha and alpha-gated gamma-ray spectra. Notice that the used coincidence technique also allows the accurate determination of detection efficiencies from the actual data.

As shown in [10], where a nuclear bomb particle containing 1.6 ng of plutonium was studied, the position of the particle with respect to the detectors can be determined after a few minutes of data acquisition, see also Fig. 3. Reference [11] presents the nuclear bomb particle data when the acquisition is continued for three weeks. The resulting singles alpha spectrum has a rather poor quality due to the energy loss of alphas to the particle. However, useful information can still be extracted from it by first gating

the alpha spectrum with ^{241}Am 59.5 keV gamma-rays. The resulting gated spectrum provides approximately the alpha peak shape characteristic to the sample. The obtained peak shape can be applied while analyzing the singles alpha spectrum. This analysis allows the determination of $^{239+240}$Pu total activity. By comparing the peak shape of the gated spectrum with the simulations, information from the total mass of the particle can be extracted. This requires assumptions concerning the shape of the particle and even distribution of americium in it.

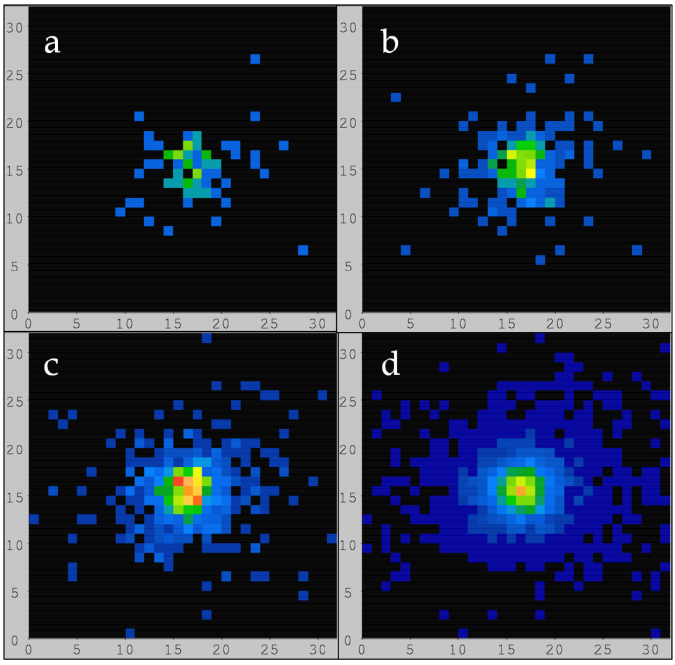

Fig. 3. Alpha particle hitmaps registered with the DSSSD at the start of the measurement of a nuclear bomb particle. The data acquisition times were: (a) 1 min, (b) 3 min, (c) 10 min, and (d) 60 min [10].

The most important data provided by the technique are the alpha-gated gamma-spectrum. As shown in [11], even after three weeks of data acquisition the background at low energies is still close to zero. This allows the detection of low yield transitions of 51.6 keV (^{239}Pu, yield 0.027%) and 45.2 keV (^{240}Pu, yield 0.045%) and furthermore to determine the ^{240}Pu/^{239}Pu isotope ratio. The obtained uncertainty for the isotope ratio is much smaller compared to previous more conventional non-destructive studies of this particle. The results were also in good agreement with the data from the destructive mass spectrometric studies previously performed by other investigators.

Since the coincidence technique is non-destructive, the particle remains intact and its analysis can be continued by other means. One option is nanotomography that provides a 3D density distribution of the particle. This information is important as such. It can also be used to independently calculate the particles total mass, volume, average density and porosity. This information considerably helps to characterize the particle. Obviously, such data can also be used to refine the simulations mentioned above. STUK and the University of Jyväskylä, Finland, have a joint on-going feasibility project on nanotomography.

5. Conversion electron spectrometry in laboratory for chemically prepared thin samples

A sample preparation cannot always be avoided. For example, one may have large quantities of solid or liquid material that contain only a small fraction of alpha or beta particle emitting nuclides. Depending on the case, concentrated samples for the liquid scintillation counting (beta) or high resolution alpha spectrometry can be prepared. Alpha particle emitting nuclides are normally deposited on the surface of a metal planchet. This measurement configuration is not optimal for the alpha-gated gamma-ray spectrometry since the planchet is absorbing a part of the low energy gamma-rays. The BEGe detector is observing the sample from the back side through the metal planchet. In addition, the gamma-ray yields are often low compared to the alpha particle yields. Therefore, these samples are normally mounted into a vacuum chamber and measured with a silicon detector.

Due to the energy resolution of the silicon detector, analysis of samples containing ^{239}Pu (5.157 MeV, 5.144 MeV) and ^{240}Pu (5.168 MeV, 5.124 MeV) is doable but very challenging, see [12]. The samples need to be optimally thin which is not always the case. At STUK, these samples are also currently measured without any protective foils between the source and the detector. This obviously increases the risk of detector contamination. Shiokawa *et al.* [13] presented an alternative, more relaxed (as regards to the sample thickness) method for the plutonium analysis which is based on the analysis of recorded conversion electron and alpha particle spectra. It is also important here that the plutonium electron yields are considerably higher compared to the gamma-ray yields used in the alpha–gamma coincidence technique. In reference [13] a Si(Li) detector, with 0.48 keV resolution (FWHM) for 42 keV electrons, operating at liquid nitrogen temperature was used. While operating a liquid nitrogen cooled detector, one needs to be very careful in order to avoid problems caused by condensation.

Because of the operational difficulties related to the liquid nitrogen temperature, we have studied the applicability of silicon drift detector (SDD) operating nearly at room temperature for the conversion electron spectrometry. After finishing a measurement, SDD warms up to room temperature very quickly (in minutes). In [14] it is shown, using a ^{133}Ba source, that the energy resolution of a 10 mm^2 SDD (collimated to 7 mm^2) for 45 keV electrons is 0.50 keV (FWHM). The determined resolution is comparable to the Si(Li) detector. The approximate thickness of the dead layer was determined to be 140 ± 20 nm Si equivalent. The relative efficiency of the detector is approximately constant in the energy range of 17–75 keV. This is concordant with the high transparency of the thin dead layer and the sufficient thickness of the detector (450 μm) to stop the electrons. A flat detector efficiency curve for electrons simplifies the data analysis process.

In reference [14], we also investigated the performance of the SDD using chemically prepared plutonium samples. In these studies, it was experimentally verified that the electron spectrometry preserves better than alpha spectrometry its capability to determine the ^{240}Pu/^{239}Pu isotopic ratio as a function of sample thickness. Based on all these results, the SDD-assisted conversion electron spectrometry can be considered as a promising new technique for the 3S field. Notice that it is complementary to existing methods. Figure 4 presents a ^{239}Pu X-ray and conversion electron spectrum measured with the SDD. The activity of the source is 2 Bq, source-to-detector surface distance is approximately 4 mm and the acquisition time 10.8 d. All significant ^{239}Pu peaks are clearly visible.

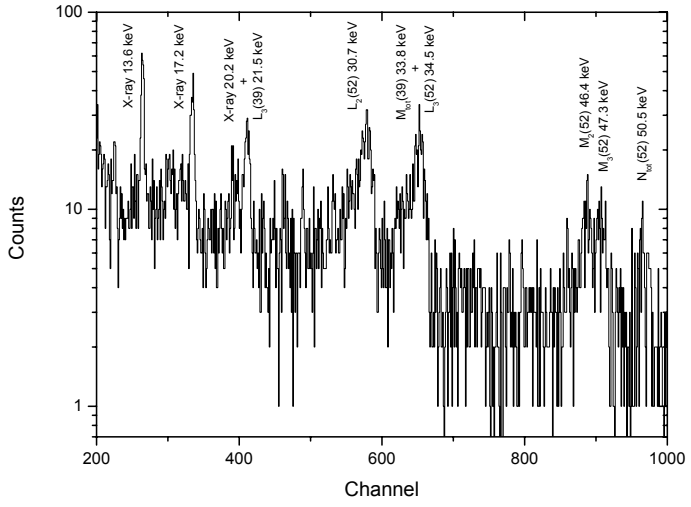

Fig. 4. A combined ^{239}Pu X-ray and conversion electron spectrum measured with the silicon-drift-detector.

6. Summary

Presented research and development activities are used to maintain and improve STUKs ability to measure alpha particle emitting nuclides and/or neutron sources. In general, the 3S field can be developed in multiple ways (broad projects may include components from several topics listed below):

1. Novel detectors and/or technical concepts for the 3S field are introduced.
2. Conventional detectors and concepts of the 3S field remain to be used but data acquisition is brought up-to-date (list-mode).
3. Analysis algorithms and/or measurement and sample geometries are improved.
4. Special calibration sources are produced.

The presented examples belong primarily to topics 1 and 3. The examples were selected so that they also familiarize the Reader with the problematics and challenges often encountered in the 3S field.

The basic research community has a lot to give for the R&D of 3S via technology and knowhow transfer. The aim is that in the future the connection between these fields could be made stronger. Notice here also that the radioactive and nuclear materials can be totally shielded requiring the use of active interrogation techniques for the detection of material itself and/or its shield. Active interrogation techniques are intensively developed for the 3S applications. They have a lot of common with the accelerator-based nuclear physics basic research, *i.e.*, they also rely on the synchronized use of sophisticated accelerators and detectors.

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