

# A novel 3D-imaging and characterisation technique for special nuclear materials in radioactive waste

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Received: 14 May 2022 / Received in final form: 3 October 2022 / Accepted: 10 October 2022

**Abstract.** A novel technique for non-destructive assay (NDA) of radioactive waste called ARCTERIX (Advanced Radwaste Characterisation based on Tomographically Enhanced Radiation Imaging without X-rays) is presented. The concept is based on a 3D-tomographic imaging technique for special nuclear materials – neutron-gamma emission tomography (NGET). ARCTERIX takes the NGET principle from its original application area of nuclear security systems into the realm of radioactive waste assay with its special characteristics and challenges. By adding localisation and imaging of SNM inside shielded waste containers to the array of existing techniques used for radioactive waste characterisation, ARCTERIX complements the state of the art in passive and active NDA interrogation methods. It is aimed primarily at the class of mixed, long-lived radioactive waste that is commonly called “legacy” or “historic” waste which has special safety, security and safeguards concerns due to its mixed composition, commonly poor documentation, and the frequent presence of SNM. The ARCTERIX concept provides rapid imaging and characterisation of nuclear materials in radioactive waste with a high degree of automation and high throughput capabilities, making it possible to quickly scan large radioactive waste inventories for the presence of special nuclear materials with minimal manual intervention. The first ARCTERIX prototype system has demonstrated a high technological readiness for the implementation of the technique in a commercial stand-alone system for rapid assessment of radioactive waste drums or in a system operating in conjunction with established techniques.

## 1 Introduction

The safeguarding of radioactive materials is by necessity a continuous process, starting from its generation to the final decommissioning stage. This is of special importance for nuclear materials for which uninterrupted tracking is needed to minimise the risk of nuclear proliferation and terrorist threats resulting from illegal trafficking. Critical links in the nuclear safeguards and security chains are therefore the implementations of systems for detecting, identifying and localising radioactive materials using radiation sensors and radiation imaging techniques.

There are currently several techniques available for the characterisation of radioactive waste which are either implemented in the state of the art or under development, for a recent review, see, e.g., reference [1]. High-energy photon transmission imaging (radiography, tomography) can reveal essential information on radioactive waste packages, such as density, position, and structure of the waste inside a container, but provides no information on the radionuclide content. Radiological assessment is therefore needed using passive or active non-destructive assay

(NDA) techniques such as gamma-ray spectroscopy, which allows for characterising a wide range of radioactive and nuclear materials. Passive NDA measurements (utilising spontaneous emissions from the radionuclides present in the waste) are often preferred for initial assessment [2] since they typically entail a lower complexity and cost, as well as a lower risk of radiation exposure to personnel. For a complete characterisation, both destructive assay (DA) and NDA measurements are required.

An imaging system for ionising radiation requires either physical or electronic collimation of the incident radiation or otherwise exploiting correlations between detected particles emanating from the same initial physical process, such as a radioactive decay event. Passive tomographic 3D-gamma-ray imaging of radioactive waste is available commercially but suffers from low spatial resolution due to instrument limitations and effects of scattering of gamma rays in the waste matrix and its shielding [3] and is limited to radionuclides with relatively penetrating (high-energy) gamma emissions. Special nuclear materials (SNM) like plutonium, with their notoriously feeble and low-penetrating gamma emissions, are exceedingly difficult to image using passive gamma techniques to meet assay goals. For such

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materials a key signature is neutron emission. Neutron imaging systems have been developed with physical collimators using the traditional transparent channel approach or based on coded apertures, as well as in the form of neutron scatter cameras [4]. Each approach has its drawbacks, such as the tradeoff between angular resolution and detection efficiency in physically collimated systems and neutron scatter cameras. For this reason, passive neutron coincidence counting, used for identifying SNM, does not provide practical means to find its location inside the waste matrix. Active neutron interrogation with the differential die-away technique, or active photon interrogation with high-energy photons (photofission), can be used to achieve a relatively high degree of quantitative accuracy for assessing the presence of nuclear materials in radioactive waste [1,5] but they are cumbersome and costly procedures that also do not provide means of accurate localisation of such materials inside the waste containers.

In order to facilitate compliance with regulatory requirements and to avoid unnecessary radiation exposure to personnel or risk of contamination, the initial characterisation of legacy waste would benefit greatly from high-spatial-resolution 3D imaging of the radionuclide content. This is particularly important for waste containing SNM, which have feeble emissions that make localisation using established techniques exceedingly difficult. Preferably, the imaging task would be carried out with a high degree of automation and without opening sealed and shielded containers. There is currently no established NDA technology for high-spatial-resolution imaging of SNM in radioactive waste.

## 2 Aims and methods

ARCTERIX is based on a novel 3D radiation imaging modality for SNM – neutron-gamma emission tomography (NGET) [6,7], which was inspired by methods developed in fundamental nuclear physics experiments [8]. The method has similarities with emission tomographic techniques used in medical imaging, such as positron emission tomography (PET) [9,10] and its variant time-of-flight PET (TOFPET) [11], which also use the physics of the emission process as a means to locate radioactive sources with high precision. The NGET technology has been recognised by the Royal Swedish Academy of Engineering Sciences (IVA) as one of the top 100 most important Swedish innovative research projects in 2021, aiming at sustainable preparedness for future societal crises [12].

Our main partner for the implementation of the NGET approach to radioactive waste characterisation and one of the end users of the technology is the company AB SVAFO. SVAFO are tasked with decommissioning nuclear facilities in Sweden in a safe, environmentally responsible, and sustainable manner and the management of the Swedish long-lived intermediate-level radioactive waste at the Studsvik nuclear decommissioning and radioactive waste handling site, preparing for the future construction of a final repository called SFL [13]. The operations are conducted on a non-profit basis under the supervision of the Swedish Radiation Safety Authority, SSM. An important part of this task entails characterising the thousands

of legacy waste drums present at the facility with high sensitivity, aiming for imaging capabilities with relatively high spatial resolution. This situation is not unique to Sweden. Orders of magnitude larger quantities of such waste are placed in similar temporary storage worldwide. Such waste, often of unclear composition and shortcomings in the original documentation, requires initial characterisation to accurately determine the inventory before it is processed for final disposal. SVAFO provides the ARCTERIX project access to its nuclear decommissioning and radioactive waste handling facility in Studsvik, around 100 km south of Stockholm, Sweden. The collaboration between KTH and SVAFO, which is funded by the Swedish Foundation for Strategic Research (SSF) [14], has defined the following main goals:

- A high-precision characterisation and imaging system for radioactive waste with a focus on special nuclear materials under safeguards.
- High spatial resolution and image processing in real-time by leveraging recent advances in machine learning.
- An automated approach to waste drum characterisation with minimal human intervention.
- Easily optimised detection geometries and scanning methodologies using a specially developed numerical simulation package.
- Optimised quantitative isotopic characterisation in combination with high- or medium-resolution spectroscopic gamma-ray detectors and 3D-densitometry based on gamma-ray transmission scanning.

While the current ARCTERIX implementation is focused on the class of radioactive waste called “historic” or “legacy” waste, a detection system featuring the NGET imaging modality might also be applied to radioactive waste characterisation in general, potentially including verification of spent nuclear fuel and other types of high-level waste suspected of containing SNM.

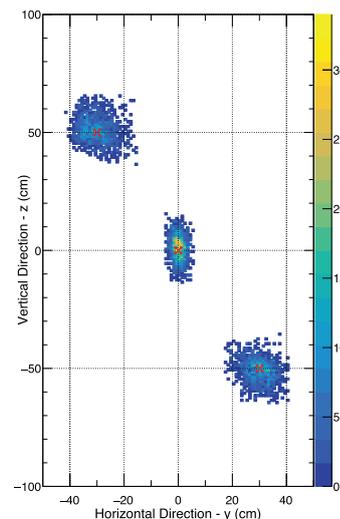
### 2.1 Technical overview

The ARCTERIX prototype system has been developed in an integrated approach combining experimental studies with numerical simulations using the Monte Carlo radiation transport codes Geant4 [15] and MCNP6 [16]. The fission process was modelled using the computational code FREYA [17]. The simulation codes are used to optimise the detection geometry for different types of radioactive waste configurations. The novelty of the ARCTERIX approach to visualising radiation from nuclear materials lies in the combination of rapid and sensitive detection of the radiation that they emit by using measured detailed time and energy correlations between detected particles [6]. The rapid, precise and automatic determination of the location of SNM in the waste matrix leads to key advantages in radioactive waste characterisation. Two alternative image reconstruction methods have been developed, both exploiting the properties of correlated gamma-neutron pairs originating from spontaneous fission events. Since we are primarily concerned with radioactive waste geometries with spatial dimensions of the

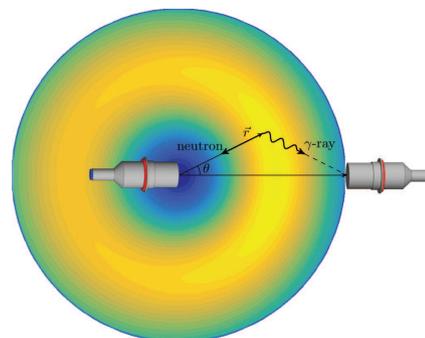
order of 1 m, the detected time differences between correlated gamma rays and fast neutrons are a few to 100 ns. This “prompt” time window is of primary interest for the imaging capabilities.

The rapidly developing field of machine learning (ML), a part of the wider field of artificial intelligence (AI), is based on computer algorithms that can improve and learn automatically through experience and by using data [18]. ML is an analysis approach that is especially well suited for solving multidimensional problems with a variety of applications in radiation detection, including pulse shape discrimination [19], radioisotope identification [20,21], and radiation source localisation based on relative flux measurements [22,23]. In the development of ARCTERIX, it has been found that ML algorithms can favourably be used to analyse the complex correlations in the data obtained from the organic scintillators in the mixed radiation field of neutrons and gamma rays, in particular in combination with the more traditional analytical-statistical approach for radiation imaging using Bayesian methods. One implementation of the image reconstruction employed in ARCTERIX is based on a ML algorithm operating on the time difference distributions (within the prompt time window) recorded in the detector array. This imaging method is a simplified but still very efficient approach to the full NGET event-by-event image reconstruction (see below), providing rapid and accurate 3D localisation of SNM based solely on measuring the accumulated gamma-neutron arrival time difference distributions for different combinations of detector elements. For typical detection geometries relevant for radioactive waste characterisation (see Figs. 4 and 8 below), it may be readily assumed that the photon is detected before the neutron since the mean velocity of neutrons from nuclear fission is roughly an order of magnitude lower than the speed of light. Therefore, this cumulative NGET (CNGET) technique does not require gamma-neutron discrimination capabilities, nor does it require measuring energies. For a system of  $N$  detector elements there are  $N(N - 1)$  unique time difference distributions, taking into account which detector element detected the first in a time-correlated pair of particles emitted from a fission event. The CNGET technique uses this complete set of cumulative time difference distributions to determine the location of SNM in 3D using an artificial neural network (ANN). These time difference distributions are updated and analysed continuously during a measurement, successively improving the accuracy of the localisation. This simplified method can be applied to imaging of single sources or multiple/distributed sources using iterative image reconstruction methods. Results obtained from measurements on a weak, 1.7  $\mu\text{Ci}$ , Cf-252 radioactive source, are shown in Figure 1. A ML algorithm based on a four-layer feed-forward ANN was used to process the data. See reference [6] for details.

The other image reconstruction technique that has been implemented for the first ARCTERIX prototype system uses a deconvolution algorithm based on Bayes’ theorem [24]. The algorithm calculates event-by-event the probability distribution for the point of origin from the measured energy deposited by the neutron, the gamma-fast neutron time difference, and the positions of the detector elements that fired. The result is a spheroidal-like



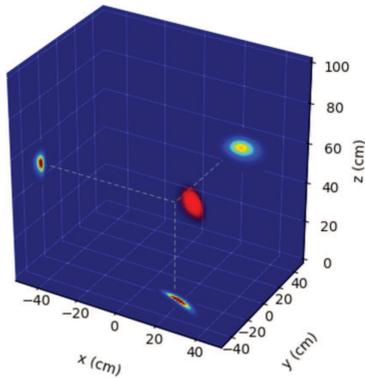
**Fig. 1.** CNGET imaging of a 1.7  $\mu\text{Ci}$  Cf-252 source. The plot shows the source positions calculated by the ANN for a total number of 1440 1-min measurements in each of three different positions (indicated by the red crosses) in the central plane of the detector system. The colour scale on the right indicates the number of measurement results per  $1\text{ cm}^2$  pixel. Taken from reference [6].



**Fig. 2.** Schematic illustration of the basic principle behind the Bayesian image reconstruction employed in ARCTERIX. The probability density function for the position of a fission event corresponding to the detection of a correlated neutron-gamma pair is mapped event by event. Taken from reference [6].

distribution as illustrated schematically in Figure 2. Differently from neutron scatter imaging (see e.g. Ref. [25] and references therein), only the detection of a single neutron interaction is required. Fast-neutron interactions in organic scintillators are dominated by elastic scattering on hydrogen atoms (protons). The probability distribution for the initial neutron energy can be estimated from the approximately known kinetic energy distribution of fission neutrons above the detected recoil energy, which is the minimum kinetic energy that could be carried by the incident neutron.

There are both similarities and major differences between positron emission tomography, widely used in nuclear medicine, and NGET. PET imaging relies on the fact that 511-keV photon pairs from positron annihilation are strongly correlated in space and time to deduce the



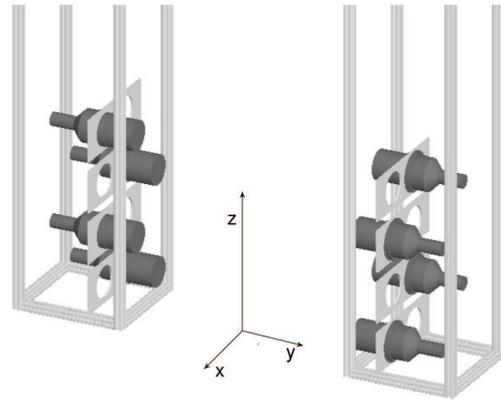
**Fig. 3.** 3D representation of the localisation of a 1.3  $\mu\text{Ci}$  Cf-252 source. The source was placed inside a lead cylinder with a 16 mm radial thickness and 105 mm height at a position  $(x, y, z) = (20, -30, 52)$  cm and measured for 10 s. See text for details.

distribution of positron-emitting isotopes within the field of view. A PET detector system with good enough (subnanosecond) time resolution can, additionally, exploit the relative time-of-flight (TOF) information for the detected photon pairs to improve on the 3D image resolution [11]. Differently from positron annihilation, the physics of the nuclear fission process does not provide easily deduced direct directional correlations between the emitted photons and neutrons. However, neutrons being massive particles have a velocity directly related to their kinetic energy. Furthermore, since neutrons emitted from spontaneous fission of SNM have rather well-established energy distributions, often approximated by a Watt spectrum [26] with parameters depending weakly on the nuclide, it is possible to estimate the probability that a detected neutron had a certain initial velocity based only on a partial energy measurement or even without measuring the energy at all. Using such estimates, the measured relative time-of-flight between the neutron and the photon in a correlated neutron-photon pair can be translated into information about their point of origin, i.e. the position of the radiation source. Similarly to a neutron scatter camera, this technique can be combined with standard image reconstruction techniques. An image obtained from a short measurement of a 1.3  $\mu\text{Ci}$  Cf-252 radioactive point-like source with the ARCTERIX prototype is shown in Figure 3.

As elaborated further below, the ARCTERIX technology can identify and precisely locate in three dimensions the presence of small amounts (grams or less) of SNM inside shielded waste containers.

## 2.2 Laboratory measurements

The ARCTERIX core hardware is a modular organic scintillator-based detection system with an easily modified geometry that can be adapted to different applications. Organic scintillators have high gamma-ray and neutron efficiencies as well as excellent neutron/gamma pulse shape discrimination and timing properties. Several test measure-

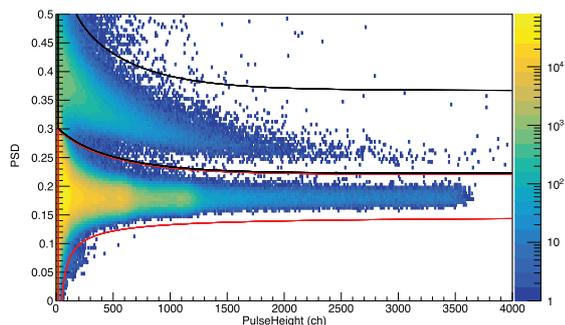


**Fig. 4.** Schematic drawing of the detector configuration in the test set-up employed at AB SVAFO showing the two detection assemblies and the mechanical support structure. The horizontal distance between the front faces of the detection assemblies is 1.0 m. Taken from reference [29].

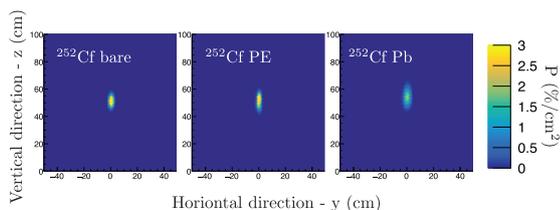
ments have been carried out using the detection system geometry illustrated schematically in Figure 4. The geometry of the first ARCTERIX prototype system was adapted from the ANSI N42.35-2016 [27] industry standard for radiation portal monitors and consisted of two pillars, each holding four 127 mm diameter by 127 mm length cylindrical cells forming a zig-zag pattern (Fig. 4). Each detector cell, containing approximately 1.6 l of EJ-309 scintillator [28], was optically coupled to a Hamamatsu R1250 photomultiplier tube (PMT) [9]. Since the detection system is modular it is easily scalable. The support structure is designed to accommodate up to 20 horizontally oriented detector cells in each pillar and, additionally, ten detector cells at the top of the structure, making up a total of 50 detector cells. The current version of the prototype system, which is being integrated with an automatic waste drum scanning system, includes 12 additional 95 mm diameter by 76 mm length cylindrical cells.

The PMT anode pulses were read out by an 8-channel digitiser board featuring 14-bit resolution and 500 MHz sampling rate. The digitiser has pulse shape discrimination (PSD) capabilities for distinguishing between neutron and gamma-ray interactions in the scintillators based on field programmable gate arrays (FPGA). For further details concerning the detection assembly and readout electronics, see Reference [6]. The digitised signals (“traces”) from the detector modules are processed in real time within the digitiser’s FPGAs to extract charge integrals, pulse shape information and timing information. The PSD algorithm for distinguishing gamma-ray interactions from neutron interactions is based on the charge comparison method (Fig. 5), whereas sharp time stamp extraction is performed using a digital constant fraction discrimination algorithm. Energy information for each trace is extracted using a moving window deconvolution algorithm.

Figures 6 and 7 show the results of measurements carried out using an encapsulated californium-252 (Cf-252) radioactive source in different shielding configurations. The source had an activity of 1.3  $\mu\text{Ci}$  (mass  $2.4 \times 10^{-9}$  g,



**Fig. 5.** Pulse shape discrimination between gamma rays and neutrons employed in the ARCTERIX prototype. The plot shows the distribution of tail integrals (taken over the last 73% of the signals) divided by the total pulse integrals (“PSD” parameter) vs the pulse height of the PMT signals. The vertical colour scale is indicated on the right. Neutrons and gamma rays were selected by choosing events within the regions indicated by the black and red lines, respectively. The signals saturate at around  $6.5 \text{ MeV}_{ee}$ .



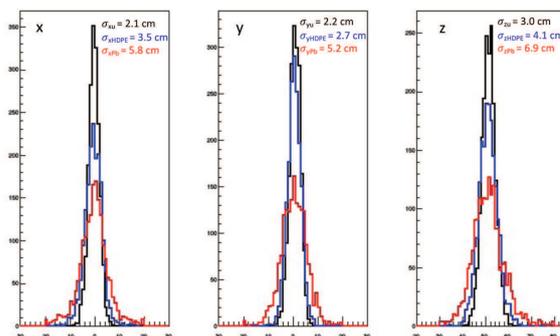
**Fig. 6.** Source localisation results based on Bayesian inference applied to event-by-event data. Each panel corresponds to a 10 s measurement of the  $1.25 \mu\text{Ci}$  Cf-252 source placed at  $(x, y, z) = (0, 0, 52) \text{ cm}$ . The deduced projected probability density distributions in the  $y-z$  plane are shown for the bare source (left), inside the PE1000 shielding (middle), and inside the lead shielding (right). The calculated probability of finding the source per  $\text{cm}^2$  pixel is indicated by the colour scale on the right. Taken from reference [29].

neutron emission rate  $5400 \text{ s}^{-1}$ ), around 27% of the ANSI N42.35-2016 standard Cf-252 source [28].

The Cf-252 material was embedded in a ceramic cylinder with dimensions 4.6 mm (diam.) by 6 mm and encapsulated in a double-welded stainless-steel cylinder with outer dimensions 7.8 mm (diam.)  $\times$  10.0 mm (ANSI classification code C66544) [30]. High-density polyethylene (HDPE) plastic and lead shielding was used to investigate the effect of fast-neutron and gamma-ray attenuation on the imaging performance, respectively. The obtained spatial resolution of around two up to a few cm ( $\sigma$ ) is remarkable, considering the much larger dimensions of the detector cells. The results also indicate a robustness of the NGET imaging technique against the presence of moderate amounts of shielding materials of different types.

### 3 Measurements at the Studsvik nuclear decommissioning site

Radioactive waste in general and legacy waste in particular is commonly stored in drums with various passive



**Fig. 7.** Projections of measured source position distributions on the  $x$ -,  $y$ - and  $z$ -axis for different shielding conditions. A total number of 1800 10-s measurements were performed for each case. Black: bare source, blue: source inside a 4-cm radial thickness HDPE cylinder, red: source inside a 1.6 cm radial thickness lead cylinder. The Cf-252 source was placed at  $(x, y, z) = (0, 0, 52) \text{ cm}$  in the coordinate system indicated in Figure 4. Taken from reference [29].

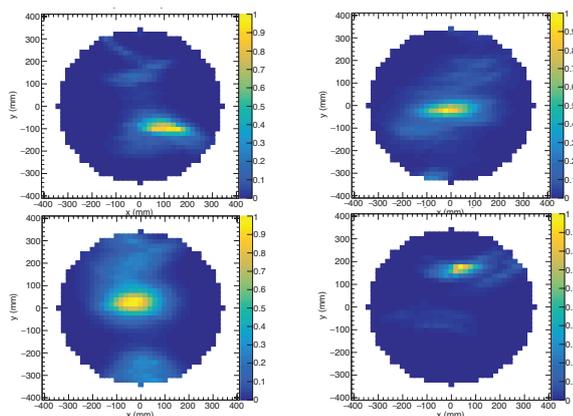


**Fig. 8.** Set-up used in the test measurements on legacy waste at AB SVAFO. Photo by Fredrik Ekenborg.

shielding materials. The emitted radiation is also subject to intrinsic shielding from the radioactive materials themselves. Therefore, the performance in realistic conditions required further investigation, which was the purpose of the first ARCTERIX prototype system. At AB SVAFO, most of the radioactive waste is stored in drums of 200 to 300 l volume enclosing an inner drum with the waste form and typically a 5 cm concrete lining.

#### 3.1 Preliminary results

Test measurements on radioactive waste drums were carried out at AB SVAFO in May 2021 using two detector assemblies. A photo of the measurement set-up is shown in Figure 8. Figure 9 shows horizontal slices through the 3D images of four of the investigated waste drums, each measured for approximately one hour. The slices were taken at the vertical position with maximum spontaneous fission activity in each drum. Estimates of their Pu-mass content were made by comparing the number of registered gamma/neutron-coincidences with Geant4 [15] Monte-Carlo simulations. In a typical case, the resulting estimate of the effective mass of Pu-240 in the drum was 0.9 g with a relative statistical uncertainty of 6%,



**Fig. 9.** Examples of projected 2-cm thick horizontal slices of the full 3D image for four different legacy waste drums at the SVAFO facility in Studsvik, Sweden. Each slice is applied at the vertical position of the maximum detected concentration of spontaneous fission activity. See text for details and reference [31].

in agreement with the rough assessment of the contents of the waste drum by AB SVAFO based on available documentation. The detailed sensitivity of the method, in particular when applied to other types of radioactive waste with different structure and characteristics, requires further studies. The systematic uncertainties of the quantitative estimates are complex and difficult to assess from the emission tomographic data. However, they can be significantly reduced by combining NGET imaging with standard transmission tomographic techniques (3D densitometry).

## 4 Conclusions and outlook

Using the recently developed neutron-gamma emission tomographic technique (NGET), ARCTERIX provides rapid and accurate localisation of SNM, enhancing current capabilities in nuclear security, safeguards, and radioactive waste management applications. The NGET imaging technique has so far been demonstrated using detectors based on large organic liquid scintillator cells, achieving a typical spatial resolution of a few cm in laboratory conditions, significantly less than the dimensions of the radiation sensors themselves. Even better spatial resolution than was obtained with the first ARCTERIX prototype system should therefore be possible by reducing the size of the detector cells. This is addressed in the current version of the prototype system which features twelve 0.5 l scintillator cells, in addition to the eight 1.6 l cells used in the first tests. The prototype system currently under development also features a high-resolution HPGe-based gamma-ray emission tomographic scanner and a transmission tomographic gamma-ray scanner for integrated 3D densitometry. Solid organic scintillators such as stilbene or plastic might also be preferable to liquid-scintillator-based systems in some applications since they are more robust and less sensitive to temperature variations. New developments in scintillation plastics have led to performance approaching or even exceeding the level of commercially available scintillation fluids [32], and we, therefore, are

investigating the possibility of replacing or complementing the liquid scintillators with plastic scintillators in the next prototype version of ARCTERIX.

The development of a new generation of fast inorganic scintillators with moderate energy resolution for gamma rays, sensitivity to both thermal and fast neutrons, and PSD capabilities to discriminate between the two types of radiation is also promising for future developments. Examples are CLLBC [33] and CLYC [34,35]. Although these and similar inorganic scintillators have reduced cross sections for fast neutrons compared with organic scintillators, the energy transfer mechanism (primarily due to  $^{35}\text{Cl}(n, p)$  and  $^{35}\text{Cl}(n, \alpha)$  reactions) also enables neutron spectroscopic measurements [36]. The intermediate-resolution gamma-spectrometry made possible by these detector materials enables isotopic analysis of radioactive waste components, in particular for plutonium and uranium isotopic analysis [37,38]. An additional development direction we are following is to optimise the ARCTERIX detection system for Compton scatter imaging of pure gamma-emitting radioactive nuclides, in addition to the crude intensity-based localisation capabilities that are already present due to the granularity of the detector system.

## Conflict of interests

The author is the inventor of patent applications related to this work filed by KTH Holding AB, KTH Royal Institute of Technology (Nos. US 17/255143, EP 3811121, CN 201980043247.2).

## Acknowledgements

This work has received funding from the Swedish Radiation Safety Authority under contract nos. SSM2016-3954, SSM2018-4393, and SSM2018-4972, the Carl Trygger Foundation for Scientific Research under contract no. CTS14:89, and The Swedish Foundation for Strategic Research under contract no. ID21-0095. The author is the inventor of patent applications related to this work filed by KTH Holding AB, KTH Royal Institute of Technology (Nos. US 17/255143, EP 3811121, CN 201980043247.2). Contributions from J. Vasiljević, A. Göök, and the SVAFO team led by A. Puranen and F. Ekenborg are gratefully acknowledged.

## Funding

This work has received funding from the Swedish Radiation Safety Authority under contract nos. SSM2016-3954, SSM2018-4393, and SSM2018-4972, the Carl Trygger Foundation for Scientific Research under contract no. CTS14:89, and the Swedish Foundation for Strategic Research under contract no. ID21-0095.

## Data availability statement

Data used for generating the figures in the paper can be made available upon request.

## Author contribution statement

The project was conceived and supervised by the author. Contributions from J. Vasiljević, A. Göök, and the SVAFO team led by A. Puranen and F. Ekenborg are gratefully acknowledged.

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**Cite this article as:** Bo Cederwall. A novel 3D-imaging and characterisation technique for special nuclear materials in radioactive waste, EPJ Nuclear Sci. Technol. **9**, 8 (2023)