Correlated fast-neutron-gamma emission tomography for rapid localisation of special nuclear materials in legacy waste drums

Vivian Peters[1,2], Jana Vasiljević[1], Anders Puranen[2], Giacomo Manessi[3], Ihor Tavrovskyi[1], Sarabjot Kaur[1], and Bo Cederwall[1]

[1]*Department of Physics, KTH Royal Institute of Technology, SE-10691 Stockholm, Sweden*

[2]*AB Svafo, Nyköping, Sweden*

[3]*ELSE NUCLEAR S.r.l., Milano, Italy*

Abstract

The development of precise and efficient NDA techniques for nuclear materials evaluation is of fundamental importance for nuclear safeguards and security. In particular, the safeguarding of spent nuclear fuel and other types of long-lived radioactive waste potentially containing special nuclear materials (SNM) represents a major global challenge. SNM are difficult to detect, in particular in shielded containments and in the presence of strong radiation fields from other radioactive materials, due to their relatively weak radiation emissions. We report on the development of a scanning system for enhanced non-destructive assay (NDA) of radioactive waste using the novel technique Neutron-Gamma Emission Tomography (NGET). The technique relies on the detection of correlated fast neutrons and gamma rays emitted in spontaneous/induced fission or (a, ng) reactions using organic scintillation detectors. It enables sensitive detection and three-dimensional localisation of SNM without moving components. The scanning system under development is designed for the special category of radioactive waste called "legacy waste", which has special safeguards, security, and safety concerns due to its mixed, long-lived radioactive components. This application of the NGET technique was awarded the Euratom Innovation Prize 2022. The automatic scanning system, developed in collaboration between KTH, AB Svafo and ELSE Nuclear S.r.l., will serve as the first NDA characterization station for the Swedish legacy waste drum inventory at the Svafo interim storage facility in Studsvik, Sweden. Featuring additionally a collimated HPGe detector for high-resolution gamma-ray emission tomography and 3D gamma-ray densitometry, it will be able to load and perform vertical and azimuthal scanning of large numbers of waste drums with minimal manual intervention.

Introduction

The characterisation and localisation of special nuclear materials (SNMs) in radioactive waste is an important step in ensuring its safe handling and disposal. SNMs, such as uranium and plutonium, require a stringent disposal process and accurate characterisation helps determine the appropriate disposal method and ensures regulatory compliance. Proper management of SNMs is crucial, as they can pose significant risks to human health and the environment if not handled and disposed of properly, particularly if they fall into the wrong hands.

For large waste volumes, non-destructive techniques are preferable as they are non-invasive and do not require preparation, and later destruction, of samples, thus making these techniques faster. For waste management facilities with limited shielding capabilities, passive techniques are advantageous as they require no external neutron or photon source. Notwithstanding these advantages, it should be noted that most of these methods rely only on the detection of gamma or neutron emissions from radioisotopes and give a less accurate estimation of the activity than destructive techniques [1]. Thus, a combination of these techniques may be necessary to ensure a comprehensive and accurate measurement of SNMs in radioactive waste. Nevertheless, sampling the SNM in the waste package for destructive analyses requires that the material has been accurately localised.

Imaging techniques can provide valuable information on the location of radioactive sources within a waste package. External photon sources enable two- and three-dimensional radiographic imaging, but their limitation lies in capturing the structure of the waste matrix rather than identifying specific radionuclides [2]. On the other hand, collimated and segmented passive gamma emission spectroscopy, combined with waste package rotation, allows for the identification and tomographic imaging of gamma-emitting sources [3]. Combining active (transmission) and passive (emission) gamma tomography provides comprehensive information on source identity, position, and waste matrix composition. Attenuation-correction techniques, commonly used in nuclear medicine with single-photon emission computed tomography (SPECT) combined with computed tomography (CT), can further refine the accuracy of passive gamma spectroscopy by utilizing attenuation information from active photon transmission imaging [4].

However, gamma ray spectrometry has limitations in detecting weak emitters shielded by the waste matrix or with low counting statistics. Neutron imaging techniques, such as multi-layer detectors [5] and neutron scatter cameras [6], exist as alternatives. Even so, multi-layer detectors lack tomographic capability and neutron scatter cameras face trade-offs between detection efficiency and angular resolution due to collimation.

While new waste is typically characterised and sorted before packaging and disposal, countries with a nuclear research and power history face challenges with legacy waste [7]. Despite meeting contemporary waste acceptance criteria (WAC) for disposal during production, this waste must now be reconditioned due to changing WAC or lack of documentation [8]. In Sweden, AB Svafo, a nuclear decommissioning and waste management company, is tasked with characterising Swedish legacy waste (SLW) [9]. A significant portion of this waste consists of drums filled with concrete after packaging, originating from nuclear facilities, research, and medicine, often lacking complete documentation. Consequently, some drums may contain one or multiple SNM sources, for instance low enriched uranium and actinides in liquid form. Given the complexity of the SLW and the concrete filling, characterising, opening, sorting, and repackaging the contents according to current WAC's radioisotope and material requirements pose significant challenges. Therefore, accurate localisation and identification of radionuclides are necessary for effective remediation of the SLW.

This paper presents a prototype design of an automated scanning system for non-destructive waste characterisation. Developed in collaboration with AB Svafo and KTH Royal Institute of Technology, and currently under production by ELSE Nuclear S.r.l., the system integrates the advanced techniques NGET, active and passive gamma tomography. The implementation of the NGET technique for waste characterisation received the Euratom Innovation Prize in 2022 [10]. The primary focus of the scanning system is on accurate localisation and identification of radioactive sources in waste materials through imaging, aiding in radiation protection during waste sorting and management. The system's automation and efficiency are crucial for managing large waste volumes effectively. While detailed results have not yet been acquired as the system is still in the manufacturing process, the system exhibits substantial potential for facilitating comprehensive waste analysis. This work sets the stage for future evaluations and advancements in waste imaging technology.

Instrumentation and methods

NGET detector array

The NGET technique relies on the detection and differentiation of correlated fast neutrons and photons originating from spontaneous fission events and (α, nγ) reactions. To accomplish this, detectors must possess the ability to discriminate between these particles and have a high temporal resolution in the nanosecond range to distinguish individual events. Given that the events considered are coincident gamma-rays and neutrons, originating from spontaneous fission in waste packages of sizes in the order of one meter, the time window is between a few and 100 ns.

Organic scintillators exhibit the desired characteristics and can effectively separate neutrons from photons using pulse shape discrimination (PSD) techniques. Fast-neutron interactions in organic scintillators are primarily governed by elastic scattering on hydrogen nuclei, whereas when gamma rays interact with the scintillator material, they deposit the energy directly; this gives the different pulse shapes. The organic scintillator EJ-309 has demonstrated excellent neutron discrimination capabilities even in the presence of high gamma-ray backgrounds [11]. Additionally, EJ-309 possesses other advantageous qualities that make it well-suited for challenging environments, distinguishing it from other PSD scintillator materials available [12].

The present detector setup consists of a total of 20 cylindrical detectors filled with EJ-309 scintillator material, each connected to a photomultiplier tube (PMT). Among these detectors, eight have dimensions of 127 mm in diameter and 127 mm in length, while the remaining 12 detectors have dimensions of 76 mm in diameter and 76 mm in length. The suitability of the former was initially tested for radiation portal monitors (RPMs) by Petrović et al. at KTH Royal Institute of Technology, who provide a more detailed description of the NGET setup, data analyses, and image reconstruction in their article [13], as well as in the subsequent performance evaluation of the system [14].

The PMTs are powered by a set of five high-voltage power supplies. The anode pulses from the PMTs are read out by three eight-channel digitizer boards equipped with PSD capabilities, allowing for the distinction between neutron and gamma-ray interactions in the scintillators. To distinguish gamma-ray interactions from neutron interactions, the PSD algorithm utilised relies on the charge comparison method [15]. Furthermore, a digital constant fraction discrimination algorithm and a moving window deconvolution algorithm are applied to the digital signal to extract the timestamp and energy information respectively.

The image is then reconstructed from the signal information. During the initial RPM development, two image reconstruction approaches have been used. What significantly distinguishes NGET from many other imaging techniques is that collimation is unnecessary due to the inherent characteristics of these algorithms. Furthermore, they enable the measurement of SNM outside of the confines of the scanning system, although limitations can be set to define the anticipated volume, such as the waste drum being scanned. The first is a deconvolution algorithm based on Bayesian inference and is an iterative reconstruction approach [16], which can be applied to cumulative event distributions [13]. An event is defined as the neutron and gamma-ray coincidence. On an event-by-event basis, the algorithm computes a sphere-shaped probability distribution for the point of origin of the event by calculating the neutron time-of-flight using the time difference between the gamma and fast neutron and the positions of the detector elements that detected the event to then calculate the kinetic energy of the neutron. The probability of emission of a neutron with that kinetic energy is derived from evaluated spectral data of prompt fission neutrons [17]. Notably, only a single neutron is required per event.

As a complement to analytical approaches such as the Bayesian one, the other reconstruction algorithm investigated is based on machine learning. It operates on time-difference distributions recorded within the prompt time window of the detector array. This imaging method offers a simplified, yet efficient, approach to event-by-event image reconstruction for localizing the neutron-gamma events as this technique does not require gamma-neutron discrimination capabilities or energy measurements. In a system comprising N detector elements, there are N(N-1) unique time difference distributions, considering which detector element detected the first particle in a time-correlated pair emitted from a fission event. The cumulative neutron-gamma event timing technique utilises this comprehensive set of time difference distributions. By employing an artificial neural network, the technique determines the three-dimensional location of SNM. Throughout the measurement process, the time difference distributions are continually updated and analysed, progressively improving the accuracy of source position determination.

With the current implementation of the NGET system, SNM can be localised within centimetres of their position as the spatial resolution in the image is on that scale [14].

Gamma-ray transmission tomography

Alongside the implementation of NGET, the system will incorporate gamma-ray transmission tomography to generate 3-dimensional images of the structural composition of the waste matrix. Given that the drums are filled with concrete, the utilisation of high-energy x-rays or gamma-rays becomes necessary to penetrate the material and produce tomographic images. This can be achieved using either a linear accelerator (linac) [2] or a source containing one or multiple gamma-emitting radionuclides [18]. The benefit of using radionuclides over linacs is the accessibility not only in terms of cost-effectiveness, but also in power supply and shielding capabilities. Depending on the chosen radionuclides, a wider range of gamma-ray energies may also be available. Using multiple energies to enhance contrast is increasingly common in medical CT [19], known as dual-energy or spectral CT. In single-energy CT, contrast arises from the varying photon attenuation in different patient tissues. Nevertheless, certain tissues and materials can exhibit similar attenuation at a specific photon energy despite having distinct elemental compositions. Varying the attenuation caused by the constituent materials by using multiple photon energies hence results in diverse contrasts. To reconstruct a tomographic image, the gamma-ray projections need to be made segmentally both vertically and horizontally, as well as angularly. One scanning technique to achieve that, often referred to as the step-and-shoot scan, is using a pencil beam and a single detector which undergo a simultaneous translational motion, rotate and repeat the translation until a full circle has been completed [20].

A Co-60 source will be used initially to develop the prototype drum scanning system, due to its well-defined high-energy gamma-ray emissions. The first implementation will be the so-called step-and-shoot scan with a shielded and collimated source on one side of the waste drum and a collimated sodium-iodide gamma spectrometer on the other. In its early stages the transmission scanning system can essentially be regarded as single-energy due to the low resolution of sodium-iodide scintillators and similarities in energy of the Co-60 gamma emissions.

High-resolution gamma-ray emission tomography

In a manner similar to transmission tomography, the scanning system will encompass gamma-ray emission tomography of the waste drum contents. The measurements will be performed with a shielded and collimated high-purity germanium (HPGe) detector to facilitate both tomography and radionuclide identification. Again, the first implementation will be the so-called step-and-shoot scan, wherein the detector moves laterally, and the drum travels vertically and rotates.

Scanner assembly

The drum scanning system will integrate the previously described imaging techniques into a singular automated apparatus. This machine will be equipped with multiple safety systems, including guard rails to prevent the drum from falling and interlock systems that halt the machine in the event of unexpected motion, thereby averting crush injuries. Currently, the machine possesses dimensions of approximately 414 cm in length, 244 cm in width, and 280 cm in height. Figure 1 presents two perspectives of the scanning system, figure 2 accentuates the NGET-detector pillars, while figure 3 depicts a schematic representation illustrating the movements of the various components of the machine.

The machine will feature a loading platform onto which the drum is placed. Subsequently, via conveyor rollers, the drum is automatically conveyed to the scanning platform, which can move vertically. Furthermore, integrated into this platform is an additional cross-shaped platform for the rotation of the drum.

The NGET system will be strategically positioned on the sides of the machine, comprising two opposing pillars with scintillation detectors that cover the entire side of the drum simultaneously. While the NGET technique does not necessitate drum rotation for image generation, the option for rotation remains available as it is required for the other imaging techniques. On the rear side of the machine, a nitrogen-cooled HPGe detector, along with its dewar, will be situated on a shelf capable of lateral movement. This arrangement facilitates step-and-shoot tomography as the drum rotates and travels vertically.

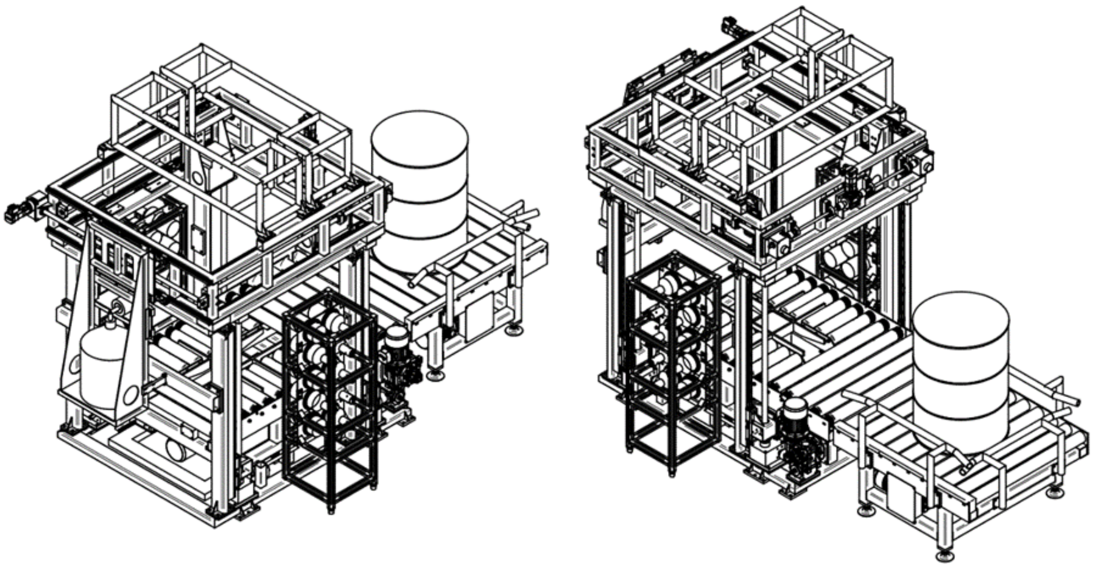

Figure 1. Two schematic views of the drum scanning system with a drum in the loading position. To the left, the drum scanning system seen from behind, facing the HPGe detectors and one of the NGET detector pillars. To the right, the system shown from the drum loading platform from which it is transported via rollers into the scanning position.

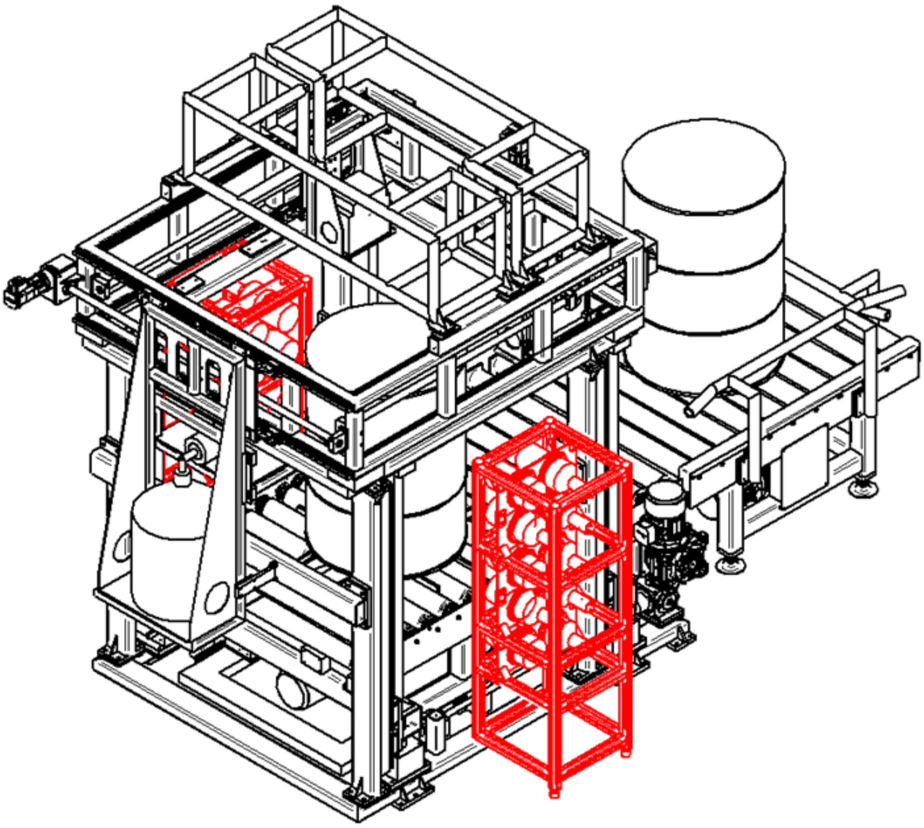

Figure 2. The NGET-system, comprising two pillars with ten scintillation detectors each, highlighted.

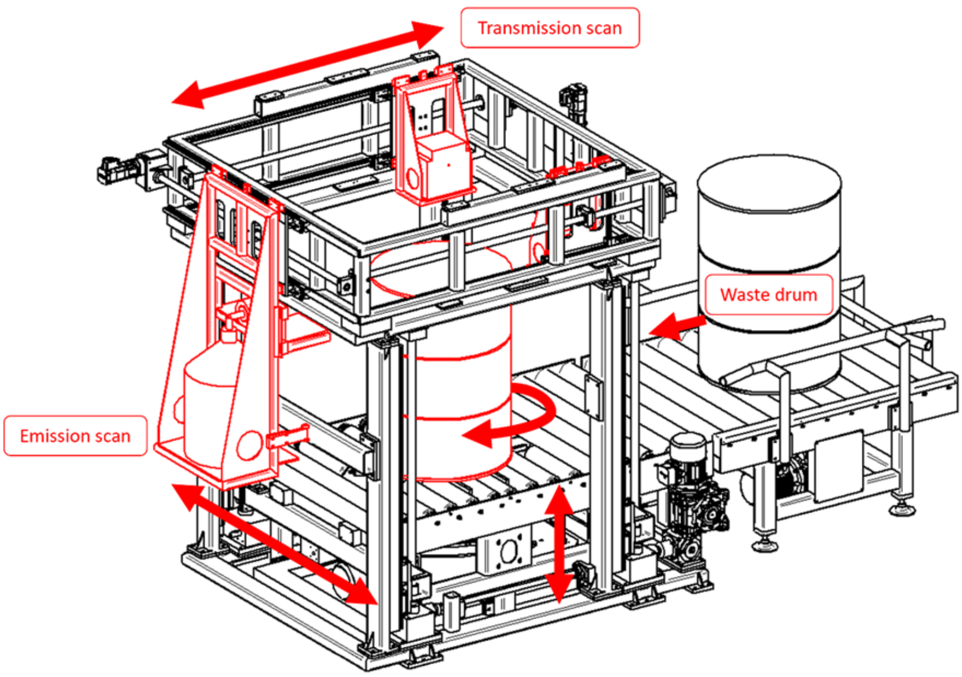

Figure 3. A schematic drawing of the drum scanning system, excluding the NGET detector pillars and the upper guard railing. The arrows indicate the different motions involved in the scanning process, such as rotation of the drum, vertical movement of the platform on which the drum is positioned during the scanning and the horizontal motion of the gamma-ray transmission tomography setup and the high-resolution gamma-ray emission tomography setup.

Positioned above the scintillation detector pillars, the gamma-ray transmission scanner will be placed. This scanner comprises two mutually facing shelves that execute simultaneous lateral movement. One shelf houses the collimated sodium-iodide detector, while the other the shielded and collimated source. The collimation of the source narrows the Co-60 radiation field into a pencil beam, directed towards the drum.

To prevent interference between the Co-60 source and the passive scanning techniques, NGET and emission tomography image acquisitions will be performed separately from transmission tomography. However, it should be noted that the drum will remain within the machine until all acquisitions have been successfully completed.

The system will be controlled by a computer to ensure simultaneous registration of motions and data acquisition. In future development, this will enable real-time imaging capabilities during the scanning process.

Discussion

The development of an automated scanning system for non-destructive waste characterisation addresses the crucial need for efficient and accurate localisation and identification of radioactive sources in waste materials. The system presented in this paper integrates the advanced techniques NGET, and active and passive gamma tomography. By employing imaging techniques, the system enables the precise detection and localisation of radioactive sources within waste materials, thus enhancing radiation protection measures during waste sorting and management processes. This capability is of utmost importance for ensuring the safety of personnel involved in waste management operations and to fulfil WAC.

Efficiency and automation are key considerations in the design of this system, primarily due to the management of large waste volumes. The automated nature of the scanning process enables swift and accurate characterisation, contributing to the overall effectiveness of waste management practices. By streamlining the identification process, time and resources can be saved, leading to improved operational efficiency in the continued management of the SLW.

As further advancements in the NGET technique are pursued, several areas warrant investigation. One avenue for future development of NGET involves the exploration and implementation of new image reconstruction algorithms. In addition to algorithmic advancements, the testing and integration of new detectors are another possibility for the evolution of NGET. To optimise image quality and performance, the exploration of different detector arrangements is another area for future development. By strategically arranging the detectors in the system, potential limitations can be mitigated and improve overall image quality. This could involve exploring different geometric configurations, optimizing the spacing and positioning of detectors. Exploring alternative algorithms, detector technologies and detector arrangements could offer improved sensitivity, temporal and spatial resolution, and energy discrimination capabilities. These refinements can lead to enhanced imaging capabilities, enabling the detection and localisation of radioactive sources with higher precision and accuracy.

The inclusion of gamma-ray emission tomography within the scanning system presents a valuable extension to the capabilities of the equipment. By incorporating a shielded and collimated HPGe detector, the system can facilitate both tomography and radionuclide identification of potential gamma-emitting sources in the waste package. This is particularly important for accurate assessment and categorisation of radioactive materials and an improvement of the current standard methods for waste handling, which often involve gamma-ray spectrometry of the complete waste package without localisation. Future advancements may involve refining the scanning technique by considering alternative motion patterns and beam configurations or exploring dynamic acquisition schemes to further improve image quality, reduce scanning time, and enhance overall efficiency.

While this prototype scanner will implement a Co-60 source for the transmission tomography, using a linac could also provide benefits such as controllability of the beam and consequently safety as it can be turned off. The energy of the beam can also be adjusted with filters, making a spectral CT. Due to the WAC of the final repositories for radiological waste requiring both radiological and material separation, a spectral CT is desirable, and is a natural progression in the development of this equipment, presumably with a different radionuclide and a detector with a higher resolution to begin with. Inherently, using multiple energies to find differing photon attenuation in the waste matrix materials will provide a better attenuation-correction for the entire acquired gamma spectrum in the other imaging techniques.

While the distinctive feature of the waste drum scanning system lies in its multi-modal imaging capabilities, the system remains functional even if one imaging modality fails. In the absence of SNM, the remaining imaging techniques aim to offer valuable insights into the composition of the waste drum contents, encompassing both radionuclide identification and waste matrix material analysis. Essentially, each imaging technique can also be utilised independently to provide meaningful information.

Conclusion

A complete waste drum scanning system is under development in collaboration with KTH Royal Institute of Technology and AB Svafo. Although detailed results from the system's implementation are still to be obtained, the system holds promise for delivering accurate and reliable waste characterisation. As ongoing research and development efforts continue, the prototype system's performance can be further refined and optimised. Future studies will focus on acquiring more detailed results to assess the system's effectiveness and reliability in real-world waste management scenarios.

In conclusion, the prototype design of an automated scanning system presented in this paper represents a significant advancement in non-destructive waste characterisation. By combining advanced imaging techniques and prioritising source localisation, automation and efficiency, this system offers the potential for improved waste sorting, management, and radiation protection. Although the strength of the complete scanning system lies not only in the novel NGET technique, but also in its multi-modality, each imaging technique can be used individually. Further research and evaluation will be necessary to fully exploit the capabilities of this system and drive advancements in waste imaging technology.

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