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REGULAR ARTICLE OPEN ACCESS

Non-destructive verification of materials in waste packages using QUANTOM®

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Abstract. The nuclear and non-nuclear industry has produced a considerable amount of low and intermediate-level radioactive wastes during the last decades. The material characterization of waste packages recently became more and more important in order to dispose of these waste packages in a final underground repository. Material characterization remains an indispensable criterion to prevent pollution of the groundwater with toxic materials and is usually required by the national licensing and supervisory authorities. Information on the nature of waste materials can be obtained based on existing documentation or, if the documentation is insufficient, on further destructive or non-destructive analysis. Non-destructive methods are to be preferred to minimize radiation exposures of operating personnel as well as costs. Existing non-destructive techniques (Gamma scanning, X-ray, active/passive neutron counting, muon tomography) do not allow the identification of non-radioactive hazardous substances. An innovative non-destructive measurement system called QUANTOM® (QUantitative ANalysis of TOxic and non-toxic Materials) has been developed. It is based on the prompt and delayed gamma neutron activation analysis (P&DGNAA). This technology is able to identify and quantify the elemental composition (Cd, Cu, B, Pb, Hg, Fe, Al, . . .) in radioactive packages such as 200-l radioactive drums. This information helps waste producers verify the content of their radioactive wastes, especially regarding the presence of hazardous substances. Different reference materials have been analysed by means of the same technology (P&DGNAA) at the research reactor of BUDAPEST. A comparison of those results for five reference materials is presented. The results show a very good agreement between QUANTOM® and standardized reference analyses.

1 Introduction

Disposal of nuclear waste is one of the major challenges facing the nuclear industry. Commercial nuclear power plants as well as the non-power industry have already produced a considerable amount of low-level (LLW) and intermediate-level (ILW) radioactive waste. Waste acceptance criteria have been developed according to different requirements for final repositories in several countries in order to assure safe and sustainable storage of the waste.

Due to the phasing-out of the German nuclear power production and dismantling activities, a considerable increase of LLW and ILW is expected in Germany. LLW and ILW are currently stored in intermediate storage. Such wastes are destined to be finally disposed of underground in the deep geological repository called Konrad, which is planned to go into operation in 2027. The German federal company for radioactive waste disposal called BGE (Bundesgesellschaft fuer Endlagerung) is responsible for the approval of conditioning procedures and the qualification of radioactive packages for final disposal in Konrad. Strict waste acceptance requirements [1] were defined based on the results of a site-specific safety assessment. They include requirements on waste forms, waste containers, activity limitations, as well as mass limitations of non-radioactive harmful substances. The latter is required in order to preserve the groundwater according to the Water Law [2]. Thus, the masses of several non-radioactive toxic substances (94 in total) are limited in the repository Konrad (e.g., mercury, cadmium, copper, arsenic, aluminium, antimony, lead, cyanide, etc.). The mass of these hazardous substances needs to be tracked and quantified in the repository inventory. This requires each waste producer to quantify and declare the amount of those materials if they exceed a specified value, usually 1% of the whole container mass (drum + material content), while for legacy waste the threshold is typically 5% [1]. Only qualified packages regarding the radiological inventory and

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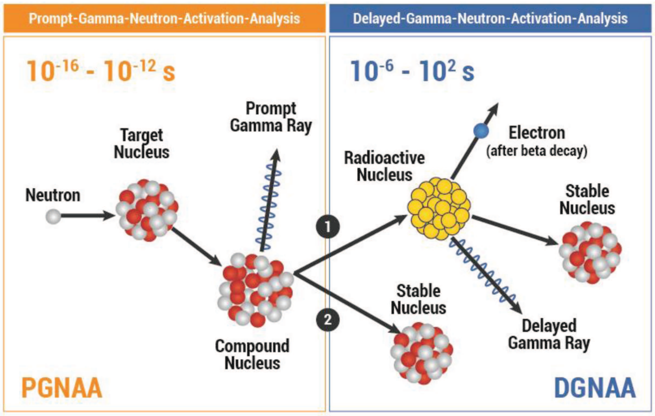

Fig. 1. Physical basics of Prompt and Delayed Gamma Neutron Activation Analysis (P&DGNAA).

the material composition can be received and subsequently disposed of in Konrad. Material characterization means that the materials in the drums need to be categorized into groups of substances (material vectors) and the mass fractions of these substances need to be quantified [1]. Waste producers declare their wastes based on these material vectors. Currently, more than 500 such material vectors have been declared and approved by BGE.

For material characterization, legacy wastes are usually problematic: the existing documentation is poor and not sufficient to satisfy the requirements. Besides, the documented material vector may be too conservative or erroneous compared to the real material composition in the drum.

Until today, the material characterization of legacy wastes was performed based on documentation, if it exists sufficiently and, if not, by using destructive methods. Such methods are time-consuming, expensive and cause radiation exposure to the operating personnel. Additionally, they will lead to a repackaging of the waste, which will increase the waste's total volume. Furthermore, in Germany, repackaged wastes are subject to even more restrictive requirements.

To overcome the limitations of the current approaches for material characterization, a non-destructive technology has been developed: a fully automated mobile measurement device based on prompt and delayed gamma neutron activation analysis called QUANTOM®.

2 Non-destructive technology

2.1 State of the art of Non-Destructive Assay (NDA) techniques

Worldwide, segmented or integral gamma-scanning as well as active or passive neutron counting are used as the standard non-destructive measurement methods for the radiological characterization and quality assurance of radioactive waste packages [3]. These techniques determine the isotope-specific activities of radionuclides in waste packages, but they cannot detect non-radioactive hazardous substances such as cadmium, mercury, aluminium, etc. In addition to these methods, radiography or tomography of waste packages using a radioactive gamma source or X-ray is particularly useful to investigate the contents of heterogeneous waste drums. However, these imaging methods only show the attenuation of intense radiation and do not allow direct identification of substances. The existing imaging procedures only distinguish between metal, organic compounds and concrete by density categories, but they do not distinguish between different materials with similar densities such as cadmium and copper. Since the chemo-toxic potentials of these elements differ a lot and cannot be determined by the above-mentioned non-destructive technologies, an improvement in NDA techniques for waste characterization is needed.

2.2 Advanced methodology for scanning nuclear drums: P&DGNAA

In order to determine masses of non-radioactive substances in radioactive waste packages, especially drums, an innovative method based on Prompt and Delayed Gamma Neutron Activation Analysis (P&DGNAA) has been developed in Germany since 2007 [4–6]. P&DGNAA is a standard method at nuclear research reactors for the element mass analysis of small samples (mass range: mg - g). As described in Figure 1, PGNAA relies on the measurement of gamma radiation emitted promptly during the de-excitation of the compound nucleus after a neutron capture. These so-called prompt gamma rays are emitted within a time period of less than 10^{-12} s. Thus, the detection of these prompt gamma rays has to be carried out during neutron irradiation. Instead, DGNAA relies on the measurement of delayed gamma rays that are emitted later on from the activated radioactive products (see Fig. 1). The timing of this delayed emission is characterized by the half-life of the formed radioactive nucleus.

The main advantages of P&DGNAA for scanning waste packages are listed below:

- no need for conditioning or preparation of the waste matrix;
- the technology is applicable for any kind of waste form;
- the technology is non-destructive;
- high penetration capabilities of neutrons, which enable a full representative description of the entire waste package;
- a multi-element analysis with high sensitivity is achievable.

Current studies and analyses with QUANTOM® make use of PGNAA only, but DGNAA will be added as measurement mode in the future using the same equipment.

2.3 Measurement system

The QUANTOM® measuring system uses a deuterium-deuterium neutron generator as a neutron source, which emits neutrons isotropically with an energy of 2.5 MeV

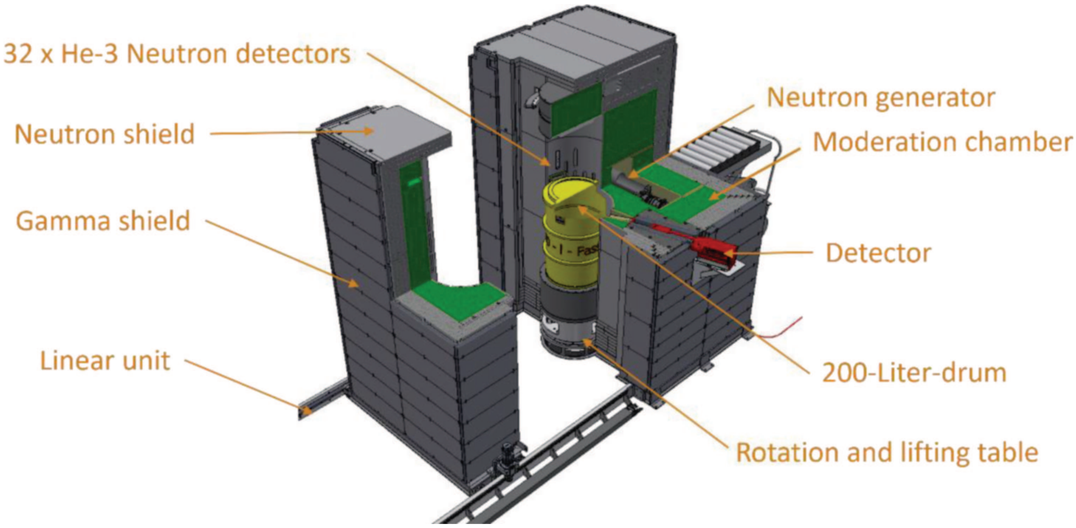

Fig. 2. Cut-view of the QUANTOM® measurement device. The measurement chamber can be opened by driving apart the left part of the facility using the linear unit.

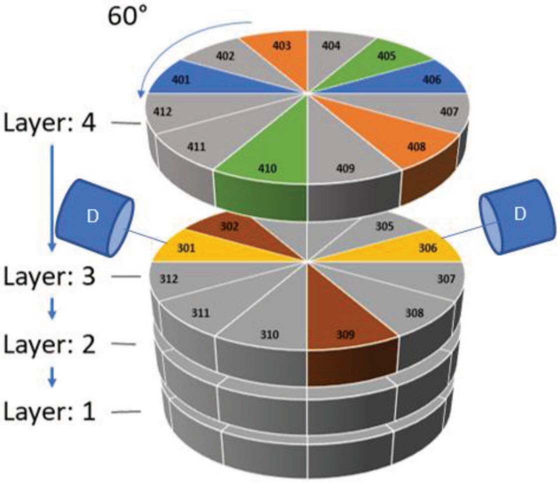

Fig. 3. Schematic representation of a segmented drum measurement.

Fig. 4. Overview of the measuring system QUANTOM®.

and yields a maximum source strength of 4×10^9 neutrons per second (model DD109.4 of Adelphi Technology Inc.). Inside the neutron generator, deuterium gas is ionized and accelerated towards a target by a high voltage of 130 kV. Fission reactions with a second deuterium nucleus take place inside the target, which emits free neutrons. The neutron generator can be operated either continuously or in pulsed mode. Figure 2 shows a cut-view of the facility and gives an overview of all installed components. For monitoring the source strength, a U-238 fission chamber is placed in the vicinity of the neutron generator. The fast neutrons are slowed down in a moderation chamber made of ultra-pure graphite and subsequently irradiate 200-L waste drum located inside the chamber. The graphite moderates and reflects the neutrons and thus maximizes the thermal neutron flux inside the waste drum. The neutron capture cross sections are high enough to induce a good signal-to-noise ratio only for thermal neutrons. The contents of the drum are activated, and the neutron-induced gamma radiation is measured by means of two N-type HPGe (high-purity Germanium) detectors with a relative photopeak efficiency of 60% each. The germanium detectors are electrically cooled which renders the handling of liquid nitrogen unnecessary. The two HPGe detectors are located on the sides of the moderation chamber shielded by collimators and thermal neutron shielding (see Fig. 2). Both detectors can be removed from the measuring position with little effort. The neutron flux surrounding the drum is monitored online by 32 ^{3}He proportional counters with a low partial gas pressure of 50 kPa

Fig. 5. Universal drum adapter surrounding a drum to be measured.

of ^{3}He and 100 kPa of Argon. The number and the positions of the neutron detectors have been optimized based on Monte Carlo simulations [7].

As part of the analysis process, collimated sectoral measurements are carried out. Therefore, the drum to be measured is virtually divided into 48 sectors (4 horizontal segments (layer) with 12 sectors each), as shown in Figure 3.

The drum is virtually divided into four horizontal segments (layers), which are further partitioned into 12 angular sectors. The gamma spectra are recorded individually for each sector at discrete positions. The measurement of two sectors is performed simultaneously in front of the collimated field of view of two HPGe detectors, avoiding double measurements of the same sector. The use of two detectors reduces the measuring time by a factor of two and increases the overall sensitivity of the measuring system. After two sectors with the same colour in Figure 3 were measured in parallel, the drum is automatically rotated (60°) by a rotating table and the next two sectors are measured. After the 12 sectors of a layer have been measured, the drum is lifted up and the next layer can be measured. The entire drum can be scanned within 24 single "sector"-measurement positions which yields a complete non-overlapping surface scan. The moderation chamber is surrounded by a neutron and a gamma shield. These shields consist of borated polyethylene plates and lead-steel composite plates. The QUANTOM® measuring system has been successfully set up and is currently being operated in Stolberg, Germany, in the AiNT Technical Center. A picture of the measuring system is shown in Figure 4.

3 Status of QUANTOM®

3.1 Building and commissioning

The measuring system shown in Figure 4 has been fully built and put into operation in 2020 after having been licensed according to §12 Para. 1 No. 1 of the German Radiation Protection Act (StrlSchG). In addition, this license was extended in accordance with §12 Para. 1 No. 3 (StrlSchG) for handling unsealed radioactive materials up to 3×10^9 times the exemption limit and for handling sealed radioactive materials up to 10^6 times the exemption limit. The commissioning of the measuring system was authorized by a technical expert in 2020. Non-destructive measurements of radioactive waste drums using the current installation site have already been performed.

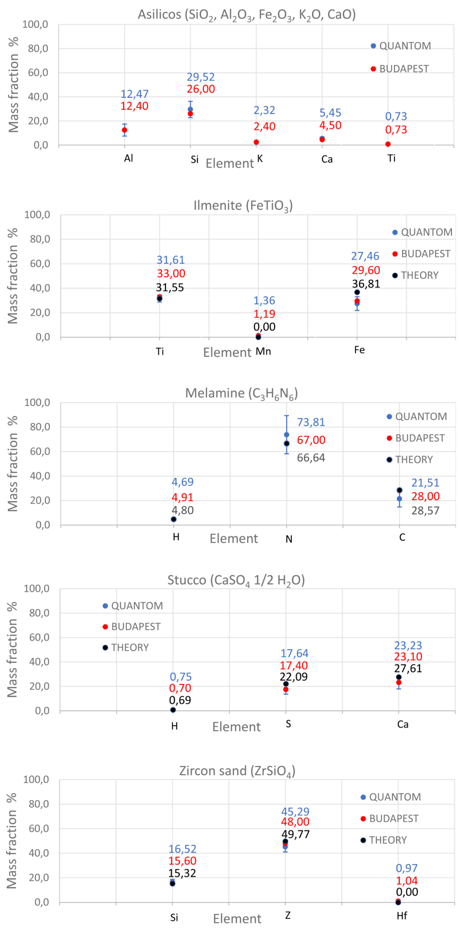

Fig. 6. Reconstructed mass fraction for five homogeneous drums filled with reference materials (QUANTOM®) and compared with the reference analysis made at the research reactor of BUDAPEST. The additional black points called "THEORY" show the theoretical mass fractions calculated by using the known molar masses and the stoichiometry.

3.2 Validation

To test and validate the measurement technology, reference drums were filled with various reference materials (e.g., zircon sand, melamine, stucco plaster, asilicos, etc.) and measured with QUANTOM®. The reference materials were selected as a function of their neutron affinity and gamma absorption properties. Diverse drum and cap types were used to simulate the real diversity of the waste drums used in the past. A universal adapter has been developed for handling all 200 l drum types with different drum caps (see Fig. 5). This adapter has been successfully tested and used so that all drum and cap types can be safely handled.

Samples of the reference materials used for the validation campaign were analysed at the research reactor in

BUDAPEST (neutron source intensity = 1 × 10^{15} n/s) using the same technique (PGNAA) and standardized processes. In this paper, we present the results for five reference drums filled with different homogeneous materials (asilicos, ilmenite, melamine, stucco, and zircon sand). The integral measurement of a drum takes about 2–4 h. The data analysis process is explained in Section 4. The achieved sensitivity (detection limits) depends on the element to be analysed. For metals such as Al, Cr, Fe, Cu, Ni, Mn, Mo, etc., a detection limit of approx. 100 ppm can be achieved. These sensitivities are based on simulations studies with MCNP modelling a homogeneous concrete matrix (density = 2 g/cm^3) with a measurement time of 4 h for the entire drum. For other toxic elements such as Cd or Hg, an even lower detection limit of approx. 10 ppm can be achieved for a measurement time of 4 h. These low detection limits can be further reduced if, for example, the measurement time is increased.

The following Figure 6 shows the results of the analysis of the five reference materials. Overall element masses were calculated by summing over the spatially distributed masses. Note that with QUANTOM® the entire matrix is analysed (i.e. 200 L). At the research reactor of BUDAPEST, the sample volume analysed was about a few grams (i.e. ∼1 cL). Due to this large difference in scanned volume, some discrepancies are expected since the materials contain impurities and are not perfectly homogeneous. With QUANTOM® major, minor and trace components can be detected. However, we plotted only the major components in Figure 6. In order to compare the results between QUANTOM and BUDAPEST, the mass fractions have been plotted in Figure 6. The uncertainties for the BUDAPEST values are very small (about a few %). In some cases, (ilmenite, melamine, stucco, and zircon sand) it is also possible to calculate the theoretical mass fractions by simply using the stochiometric ratio and the corresponding molar masses of the elements. This approach is called "THEORY" in Figure 6.

The evaluation of drum measurements includes a data-driven approach for the calibration of the fission chamber to determine the neutron generator's source strength. Measurement uncertainties were calculated using Monte-Carlo sampling with respect to all input quantities of the reconstruction method. The sources of uncertainties considered include, among others, calibration uncertainties, model uncertainties in the simulations and uncertainties regarding the positioning of the drum inside the measurement chamber. In addition, some gamma lines have to be corrected for a background signal in the active underground of the measurement (e.g., H, Al). This results in a significantly increased measurement uncertainty for the corresponding element mass.

All measurement results show a very good agreement with the known reference masses from BUDAPEST within the stated measurement uncertainties. The "THEORY" values show small discrepancies. This was expected and can easily be explained: the theoretical calculation does not consider any impurities (e.g. Mn in ilmenite), which, of course, does not correspond to reality. In the future, measurement uncertainties can be reduced by further studies and additional measurement campaigns which will yield more detailed information about the variability of measurement parameters.

3.3 Mobile drum inspection system

After the successful validation in 2021, the QUANTOM® measuring system will be integrated into a 25-Foot container. This mobile unit (see Fig. 7) can be brought directly to the site where the drums are stored or to the waste conditioner. In this case, the operation of the system only requires a notification notice according to §17 of the German Radiation Protection Act (StrlSchG), since the local dose rate at a distance of 0.1 m from the surface of the container is below 10 µSv/h. This significantly reduces the licensing efforts for commissioning the measuring system.

The entire system is fully automated. The final design of the mobile measurement device is shown in Figure 7 including an automatic drum conveyor and the container. The system can be loaded with at least up to 10 drums so that approximately only one loading per day is necessary. The measurement time for each drum is in the range of 2–4 h. The system automatically and autonomously transports one drum after the other into the final drop-off position for measurement, where it will be weighed by a high-precision scale (±0.03%). The drum is then automatically taken by a rotating crane and transported into the irradiation chamber on the lifting turntable.

4 Methods and software development

For the evaluation of the gamma spectra, a new gamma-spectroscopy software called PEAK® has been developed. It is especially suited for PGNAA and enables an automatic fingerprinting of the elements (see Fig. 8, which shows a typical PGNAA spectrum) using special algorithms coupled to current and verified nuclear physics databases of prompt and delayed gamma lines of the IAEA [8]. The net peak area is determined by fitting a physical model [9] to the data.

The gamma spectra are analysed individually for each sector measurement (48 in total) to calculate net peak areas. The mass reconstruction is then performed by jointly evaluating all measurements based on simulations which take into account signal contributions of all sectors within one measurement. This enables a spatial resolution for the element quantification in a discrete model. Thus, inhomogeneities can be taken into account within the drum.

The mathematical algorithm for spatial-dependent mass quantification is based on the virtual partitioning of the drum into four axial segments (layers) and twelve radial sectors per axial segment (see Fig. 3). In the data analysis, the sectors are additionally subdivided into seven radial partitions. The mass reconstruction algorithm takes into account the attenuation of the gamma radiation and absorption of the neutron flux within the waste matrix

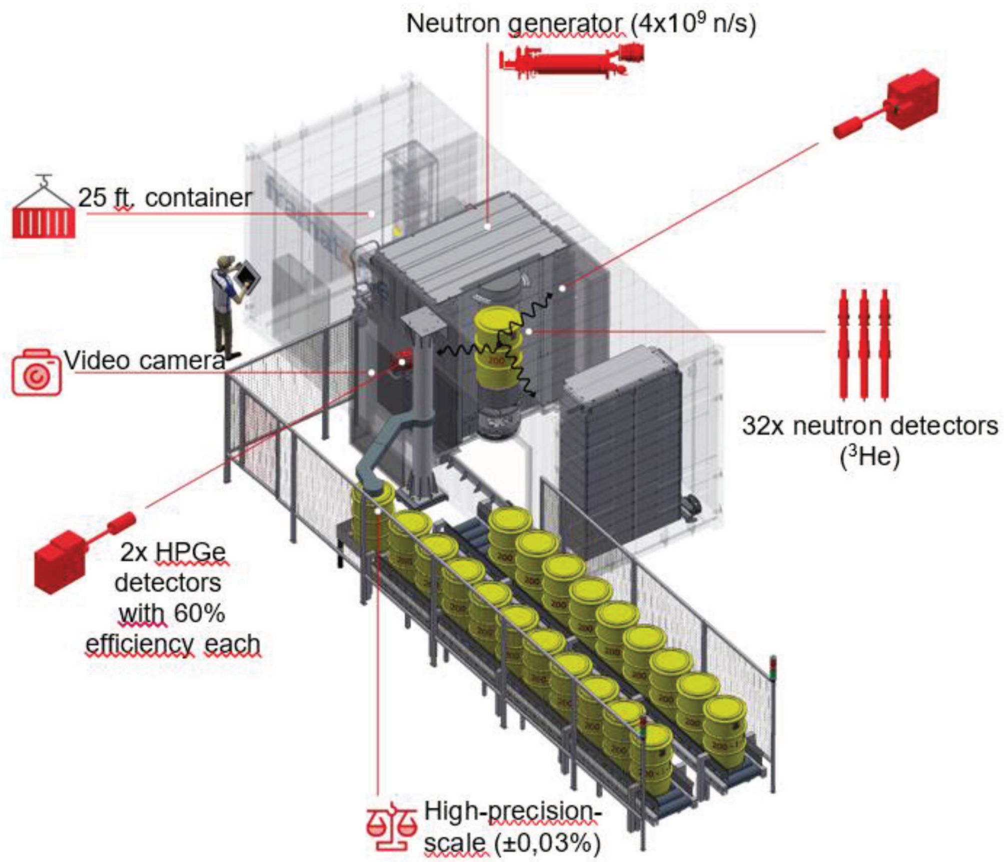

Fig. 7. Design of the mobile QUANTOM® measurement device integrated into a specially designed transportable container as well as the automatic drum loading system.

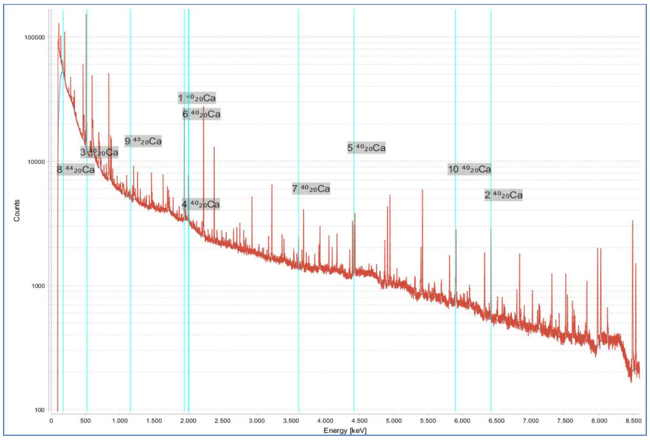

Fig. 8. Fingerprint of Calcium in a stucco sample, where the blue lines show the 10 most intense prompt gamma peaks from Calcium.

and the drum wall. The data analysis must be carried out iteratively because the measurement parameters depend on the material composition itself. Iterations with regard to the elemental composition of the drum are carried out until the computed composition of the partitions stabilizes within a range smaller than a predefined threshold value. The neutron flux within a respective partition and resulting partial cross-sections can either be calculated deterministically based on a diffusion approximation of the space and energy-dependent linear Boltzmann equation [10,11] or by using MCNP simulations [12]. This is done for each individual iteration step by considering the physical boundary conditions. The design of the measuring system and the data analysis algorithm have been successfully patented [13].

5 Conclusion

The qualitative and quantitative determination of elements (and in some cases the ruling out of some materials) in waste drums is possible with PGNAA. A full-automated drum measuring system called QUANTOM® has been developed and validated. It enables non-destructive verification of the plausibility of material descriptions of (radioactive) waste packages. QUANTOM® is a first-of-a-kind commercial system and will be ready as a mobile unit to be used directly where (legacy) wastes are stored or conditioned. The main benefits of QUANTOM® are summarized below:

- non-destructive multi-element analysis of the entire matrix;
- fast measurement process (2–4 h per waste drum) with high measurement precision;

- no repackaging and no increase in waste volume;
- reduction of costs (min. 50% per waste drum) compared to destructive analysis processes;
- minimizing the transportation of radioactive waste drums and radiation exposure of the operation staff.

Conflict of interests

The authors declare that they have no competing interests to report.

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Data availability statement

Analyzed Data associated with this article cannot be disclosed due to legal reasons.

Author contribution statement

Laurent Coquard: technical lead and coordination of project, writing. Julian Hummel: mechanical design. Günter Nordhardt and Max Georgi: automation. Andreas Havenith: conception, technical lead. Kai Krycki: codes and methods development. Bo Fu: measurement and data analysis. Christopher Helmes: software development. Marcel Heidner: simulation & software development. Frederic Simons: software development. Theo Köble, Olaf Schumann: conception, neutronic measurement.

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