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New high-throughput measurement systems for radioactive wastes segregation and free release

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HIGHLIGHTS

- New-concept system for segregation and free release was developed.
- Patented material with low natural radioactivity is used as lead-free shielding.
- Accurate measurement is facilitated by new certified calibration/testing standards.
- Precise measurements reduce the quantity of waste sent to repositories.

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ABSTRACT

This paper addresses the measurement facilities for pre-selection of waste materials prior to measurement for repository acceptance or possible free release (segregation measurement system); and free release (free release measurement system), based on a single standardized concept characterized by unique, patented lead-free shielding.

The key objective is to improve the throughput, accuracy, reliability, modularity and mobility of segregation and free-release measurement. This will result in a more reliable decision-making with regard to the safe release and disposal of radioactive wastes into the environment and, resulting in positive economic outcomes.

The research was carried out within "Metrology for Decommissioning Nuclear Facilities" (MetroDecom) project.

1. Introduction

The world faces a major challenge of great urgency: the enormous costs of decommissioning many outdated nuclear facilities. Nuclear decommissioning covers all activities from shutdown to the environmental restoration of the site. More than 200 power reactors are presently being decommissioned or will be in some phase of the decommissioning process by the year 2025. Therefore, it is essential to achieve a significant reduction in the enormous decommissioning costs by means of the development and implementation of decommissioning methodologies and associated new measurement techniques.

Not only that the decommissioning process is very costly, but it is even being carried out in the context of low public confidence in both the comprehensive clearance of nuclear sites and the safe disposal of radioactive waste. Decommissioning needs support by improved metrology that will minimize the environmental burden by providing the means for improved handling and disposition of waste, thus building public trust in nuclear technologies.

Cost reduction requires the adoption of more precise and standardized methods and devices traceable to national standards. This will make it possible to discriminate precisely and rapidly between the various waste categories to reduce conservative approach based on disposal instead of free release or higher repository category selection then appropriate in case of high measurement uncertainty.

Commercially available systems that are currently in use have issues with:

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– low throughput,
– standardization of measurement method and design,
– spectrometric capability, while clearance levels are determined for individual radionuclides,
– modularity and transportability,
– expensive lead shielding unsuitable for construction of large facilities,
– insufficient sensitivity and uncertainty,
– fixed and unsuitable measurement geometry,
– versatilely usable measurement and transport containers,
– missing universally applicable certified calibration and testing standards.

One of the tasks in the "MetroDecom" (MetroDecom project websit) project is to overcome these issues, to develop a large-scale industrial prototype solid waste measurement systems with improved throughput, accuracy, reliability, modularity and mobility, to implement such a systems on decommissioning site and demonstrate the applicability of the same concept for segregation measurement (potential free release in the environment or storage in repository selection decision) and free release measurement.

2. Measurement system components

Novel measurement systems for the measurement of the activity of radionuclides with gamma and neutron emission present in solid waste go beyond the state of the art thanks to new components and features. New system components are described in this chapter, new system features in the following chapter.

2.1. Patented modular shielding

A special new patented (EPO patent family documents) modular shielding material from concrete having low natural radionuclide content, was implemented in unified, reusable ecological building blocks with unique features, thus facilitating easy and economical creation of large, site-optimized low-background facilities (chambers, tunnels).

Concrete segments consist of aggregate made of specific rock from very old Paleozoic formations containing low amounts of natural radionuclides and special cement based on very old sediments from the bottom of the sea. The activity concentrations of natural radionuclides in the building materials are A(Ra-226) ≈ 0.6 Bq/kg; A(Th-228) ≈ 0.3 Bq/kg; A(K-40) ≈ 6 Bq/kg. These activity concentration values are about ten times lower than it is usual, lowering significantly the background radiation inside the shielding.

The unified segments are interlocking, so it is very easy to build shielding facilities without wet processing. It is possible to build large facilities quickly, cost-effectively and optimized for an individual decommissioning site. It is easy to dismantle such facilities and reuse the segments at other sites. Plastic layer used inside the chambers enables easy decontamination. Detection array and electronics are autonomous within the shielding encompassing them. These costly parts can be easily removed and used in different systems with individual shielding chambers at different sites.

The new shielding overcomes the disadvantages of lead, e.g., high and increasing price, poor radionuclide purity and the inability to build large facilities; moreover, it is made of environmentally friendly material. For shielding factors comparison see Table 1.

Fig. 1 shows one of the unified shielding segments, while Fig. 2 shows the method used to construct the shielding facility.

Fig. 3 shows a decrease in the dose rate from 101 nSv/h (external natural background) to 33 nSv/h (background inside the chamber) during the construction of a shielding chamber of the experimental free-release measurement system (Fig. 11). The position of the dose rate detector was identical prior and during construction. The chamber was

Table 1
Concrete blocks thickness equivalent to lead[a].

	662 keV (Cs-137)	1332 keV (Co-60)
5 cm lead	29 cm	22 cm
10 cm lead	58 cm	44 cm

[a] Greater lead-equivalent thickness of concrete for Cs-137 (662 keV) than for Co-60 (1332 keV) is caused by the atomic number (Z) of lead, which therefore attenuates lower-energy photons to a much larger degree than it attenuates higher-energy photons. In concrete with lower Z, this difference is relatively insignificant.

Fig. 1. Special bricks made of concrete with low content of natural radionuclides.

Fig. 2. Method of constructing the shielding facility.

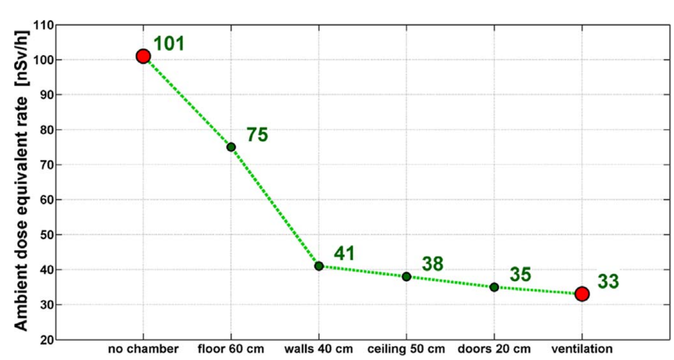

Fig. 3. Decrease of the photon ambient dose equivalent rate during the construction of the free release measuring system.

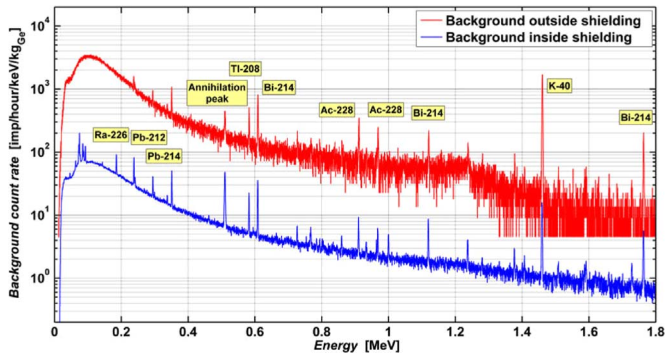

Fig. 4. Comparison of background gamma-ray spectrum inside and outside the shielded chamber.

prism-shaped, having floors being 60 cm thick, walls 40 cm, ceiling 50 cm and doors 20 cm. The chamber was ventilated by blowing in the air free of radon and its decay products using particulate and active carbon filters. Fig. 4 shows a comparison of the background gamma-ray spectrum acquired by high purity germanium (HPGe) coaxial detector with the resolution of FWHM = 1.8 keV and relative efficiency of 50% for Co-60 (1332 keV) inside the shielded chamber and outside the chamber (without shielding) in the same position.

In the regions of interest of two key radionuclides present in wastes, i.e. Cs-137 (661 keV) as a representative of fission products and Co-60 (1332 keV) as a representative of corrosion products, the background signal is reduced twenty-five times in the region of Cs-137 and fifty times in the region of Co-60. This leads to a significant decrease of minimum detectable activities to a few Bq/kg for low-density waste materials.

Table 1 specifies the thickness of the concrete shielding equivalent to the standard thickness of lead brick.

The geometry and parameters of each system are optimized on the basis of the Monte Carlo model (Solc et al., 2014, 2017). It takes into account the detectors and their collimators, the shielding blocks, the mechanical parts of the chamber, the hall parameters (for example content of radionuclides in the floor and walls), the natural background at the site, the natural radionuclide content in the air, the parameters of the measured material and the container in which it is placed. Fig. 5 shows an example of optimization of the shielding thickness below the conveyor belts for various lengths of this shielding.

2.2. Integrated modular detection array

Detector array consists of Stirling-cycle cooled gamma spectrometric compact modules, plastic scintillators total gamma modules and neutron modules. Detector arrays can be optimized for specific uses by integrating the individual detectors separately in the array or removing them if needed.

Stirling-cycle cooled gamma spectrometric compact modules, such as e.g., Ortec IDM-200V, and neutron modules based on proportional counters filled with He-3 or BF3 are used for free-release measurement. The use of spectrometric modules for free-release measurement is essential because clearance levels are specified for each released radionuclide considering its respective toxicity (Council directive, 2013; IAEA, 2014). Only spectrometric detectors can be used to direct activity measurement of gamma emitting radionuclides. The activities of the selected easily measurable key radionuclides can be correlated with those of difficult-to-measure nuclides using scaling-factor method (IAEA, 2009; ISO 21238:2007). During the decommissioning of nuclear facilities, it is not possible to employ the radionuclide vector method using non-spectrometric detection, because that method is only applicable for exactly known and stable compositions of wastes (ratios of the activities of various radionuclides).

For segregation purposes, non-spectrometric plastic scintillator detectors employed as high, narrow prisms to allow the optimization of their number and positions are usually sufficient.

Four detectors of the same type are usually used to ensure optimal detection geometry. In this way, it is possible to scan the containers in order to check the homogeneity of their radionuclide content and the presence of hot spots and at the same time to attain a high measurement sensitivity.

Fig. 6 shows the design of the detection array as a component of the measurement system, with three types of detectors (four HPGe detectors, three neutron slab counters each consisting of five He-3 tubes and four plastic scintillation detectors) mounted around the rollway inside the shielding tunnel.

During decommissioning, low-density materials (e.g., filters, wood, paper) and high-density materials (e.g. steel tubes, concrete) are measured. The measurement systems currently available have fixed detector positions, so they cannot be optimized to measure lower amounts of high-density materials or higher amounts of low-density materials. That is why upper detectors are supported by a movable platform. Threaded bars and laser sensors ensure the vertical movement of the upper detector positions, so it is possible to measure two containers

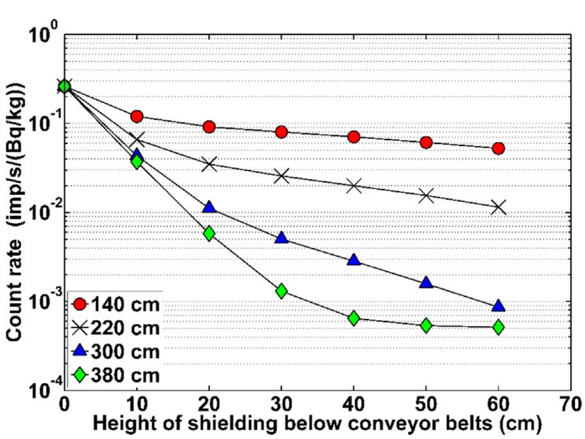

Fig. 5. An example of Monte Carlo shielding optimization. The dependence of HPGe detector count rates in Co-60 peak of 1332 keV photons on thickness of shielding below all conveyor belts, for various lengths of the shielding.

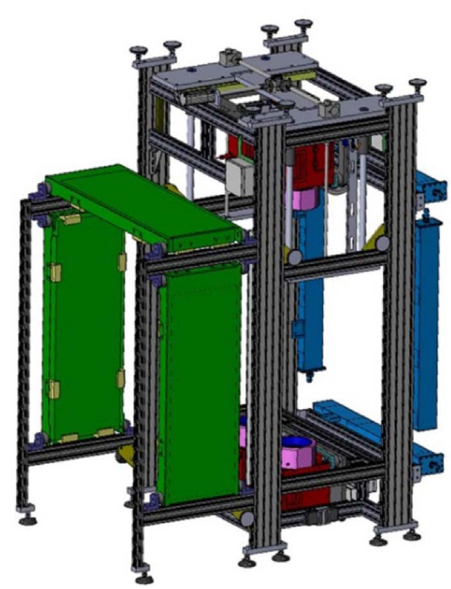

Fig. 6. Design of detectors array with three types of detectors: four HPGe detectors (red), three neutron He-3 counters (green), and four plastic scintillators (blue). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

with a low-density material at the same time or one container with a high-density material in close measurement geometry.

2.3. Scanning gamma irradiator

Collimated gamma irradiator component of the measurement system is used for improved scanning of wastes with heterogeneous composition and density.

In most countries, the clearance levels in the activity concentrations are usually determined together with the lowest mass unit for which the clearance level must be met. These masses range in different countries between 1 kg and 100 kg (Council directive, 2013; IAEA, 2014; Maringer et al., 2011). The purpose of controlling the potential averaging is to prevent the release of small objects (hot spots) with an activity concentration above the clearance level. The method of multiple detectors and measurement positions is used for segmenting the measured load and cross-comparison of the segments is used to check the homogeneity of the radionuclide content in the scanned container. If the homogeneity criterion is met, it is possible to divide the activities of radionuclides by the net load mass of the waste and to compare the activity concentrations to the clearance levels. However this test of radioactivity distribution cannot reveal the presence of a radioactive source that is shielded by a high density waste material.

Consequently, the scanning gamma irradiator module contains a collimated Co-60 source and a detector to measure the material density profile along several axes of the passing container during the system's travel route. This scanning reveals high-absorption spots and improves the accuracy of the data on the distribution of density entering the software algorithm based on Monte Carlo model. The data on density distribution in each individual container can be used to improve the accuracy and reliability when measuring radionuclide content and to test the homogeneity of radioactivity distribution.

Fig. 7 shows the design of the collimated scanning gamma-ray irradiator module.

2.4. Universal transport-and-measurement containers

Universal transport-and-measurement IP1 and IP2 containers are used. They are optimized for safe transport and low absorption during measurement, thus eliminating waste reloading.

The specification of ideal containers for the transport and measurement of wastes is a complex issue because many properties must be considered. Particularly important are the standardized transport dimensions, optimum volume in terms of measurement capacity and acceptable radiation absorption, and the qualifications for safe transport (IAEA, 2012). The goal is to limit, as much as possible, the need to reload and handle the material. While design using thin wall material due to low radiation absorption is suitable for measurement, a mechanically stronger container design offering increased absorption is required for transport.

Fig. 8. "MetroDecom" container with IP-1 certification.

The "MetroDecom" project developed optimized containers shown in Fig. 8 (IP-1) and Fig. 9 (IP-2).

These containers have the following features:

– Standardized dimensions corresponding to the dimensions of "Europallet" ($100 \times 120 \times 60$ cm), with a volume of approx. 0.5 m^3, which is optimal in terms of handling of the container and the limited absorption layer of the material inside as well as a load capacity up to 500 kg.

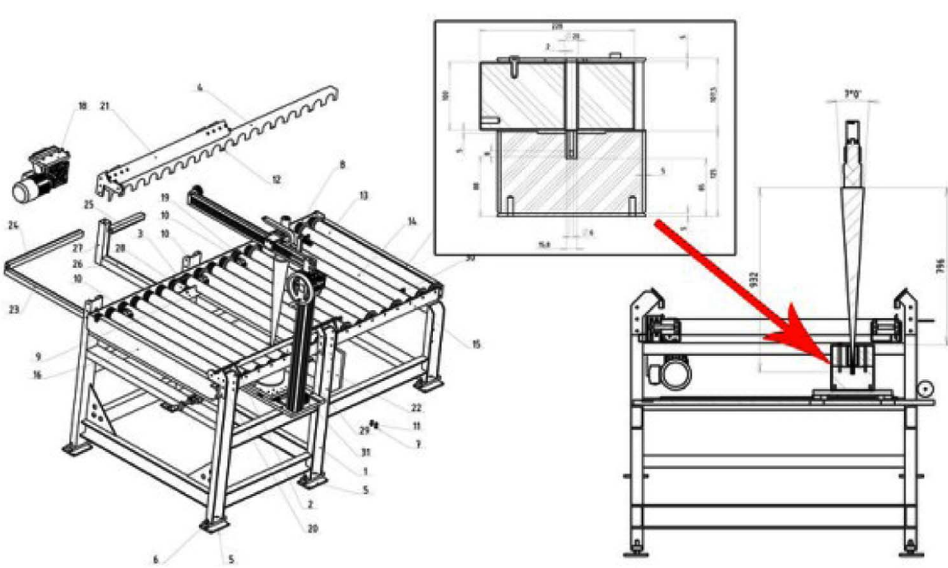

Fig. 7. Design of collimated scanning gamma-ray irradiator module. The cut shows a detail on the collimator and the shielding of the radionuclide source. The cone represents a collimated beam.

Fig. 9. "MetroDecom" container with IP-2 certification.

- A design combining a rigid frame and a plastic inner tray, which is an optimal compromise between the requirements for transport, requirements for measurement and corrosion resistance.
- A design that meets the requirements of qualification IP-1 or IP-2 for transport, minimizing the need to reload the material.
- A tight-fitting lid to prevent the spread of contamination during transport and free-release measurement.

While the design IP-1 is sufficient for the free-release measurement of materials that have passed through segregation, a container with the IP-2 design may be necessary in order to measure materials for segregation. The goal of these containers optimized for safe transport and low absorption during measurement is mainly waste reloading elimination.

2.5. Special holders

Special holders enable the use of various site-specific containers and/or drums. Decommissioning sites frequently use their own drums or containers, which can vary in dimensions. The new measurement system is able to measure such non-standard containers provided with special holders. Fig. 10 shows examples of such holders.

2.6. Filtration and ventilation equipment

Equipment for ventilation and filtration is used to filter radon and its decay products out of the blown-in air that would increase the background. Overpressure is created within the shielding to prevent the leakage of contaminated dust. In the flow-through tunnel structures overpressure is created using automated aluminum shutters on the tunnel openings for industrial system (Fig. 16) or movable doors for experimental system (Fig. 11).

Fig. 11. Experimental free release measurement system.

3. Measurement systems features

3.1. Experimental systems

As a part of the "MetroRWM" project (MetroRWM project website), which preceded the "MetroDecom" project, an experimental free-release system was built as shown in Figs. 11 and 12. The testing of this system confirmed the properties of the components specified above and the functionality of the corresponding control and measurement software.

For this experimental equipment with four HPGe detectors IDM-1 (Ortec), the minimum detectable activity (MDA) of 10–30 Bq was achieved (depending on the radionuclide) with a measurement time of 3×60 s (three container positions) for point reference sources in the middle of the empty container.

MDAs for homogeneously contaminated materials filling up the 0.4 m^3 container are shown in Table 2.

3.2. Industrial systems

Experimental system experience resulted in a fully modular concept for an industrial system shown in Fig. 13 permitting optimization based on local needs of individual decommissioning sites.

The number of tunnels and detection arrays can be selected according to the needs. Sliding doors were replaced with a shielding labyrinth, which is a cheaper solution increasing the throughput and safety. Currently, a system with a single tunnel is being prepared. Its shielding is shown in Fig. 14, and its internal structure is depicted in Fig. 15.

The current stage of construction is shown in Fig. 16. A prototype of the industrial system will be tested and operated at the Ispra site (Italy) of the European Commission's Joint Research Centre.

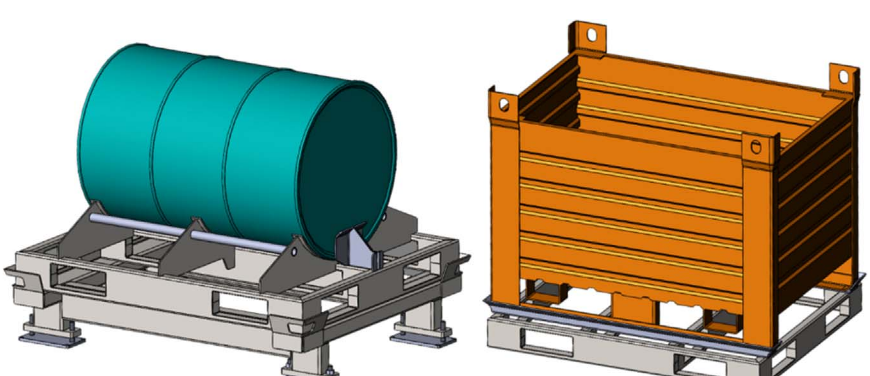

Fig. 10. Special holders for different types of containers.

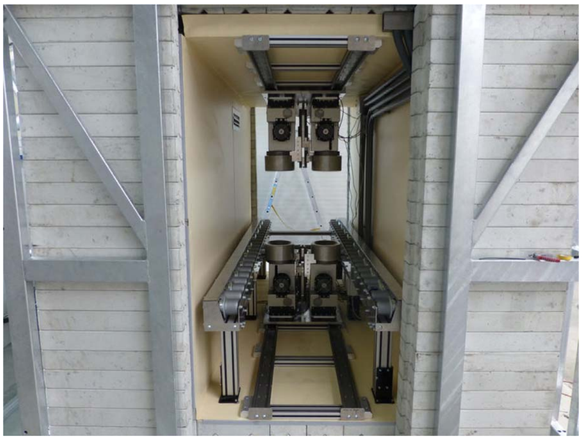

Fig. 12. Detector array made of 4 pieces of HPGe detectors in experimental free release measurement system.

Table 2
Minimum detectable activities for different materials.

Radionuclide	MDA plastic		MDA gravel		MDA iron tubes	
	Bq/kg	Bq	Bq/kg	Bq	Bq/kg	Bq
Cs – 137	0.3	70	0.6	100	0.8	150
Co – 60	0.2	100	0.4	150	0.4	180

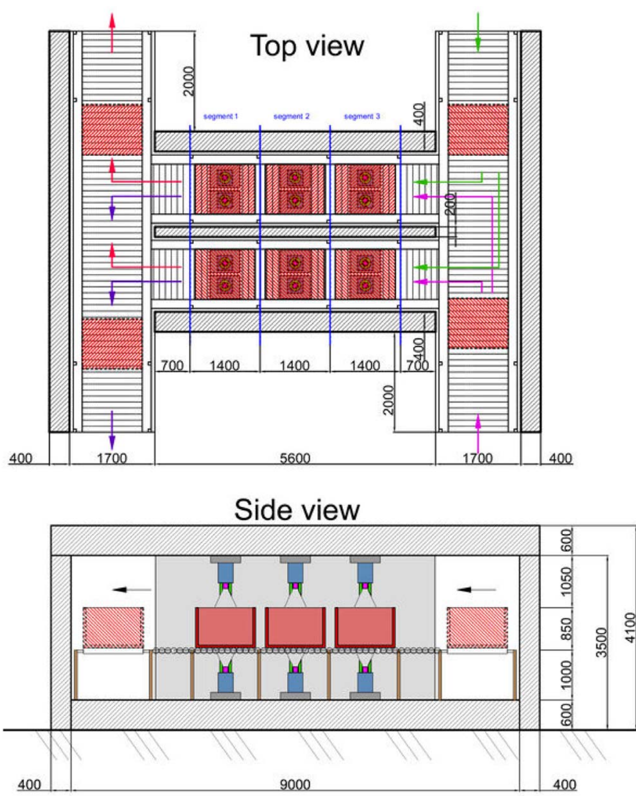

Fig. 13. Concept of industrial free release measurement system.

3.3. New and improved features of the systems

The measurement systems, incorporating all the aforementioned components, will have the following new or improved features:

– standardized modular, transportable, reusable, site-optimization-enabling concept,
– high throughput (up to 10 t/h, 20 m^3/h),
– high sensitivity and wide detection range, individual multi-detector modular arrays,
– adaptation for segregation or free-release measurement purposes,
– adjustable measurement geometry (optimization for low- and high-density materials),
– flow through, without movable doors configuration (high throughput, safe, affordable),
– universal use for any type of container,
– simplified handling of materials using a new optimized type of container,
– radon decay products background and internal contamination reduction.

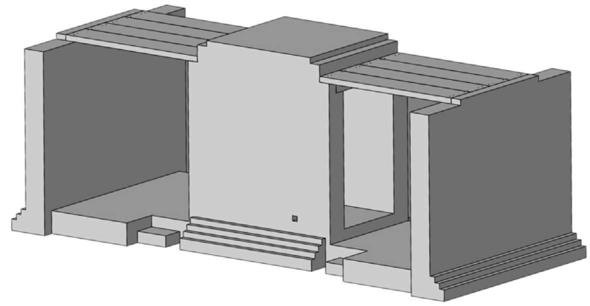

Fig. 14. Industrial free release measurement system shielding.

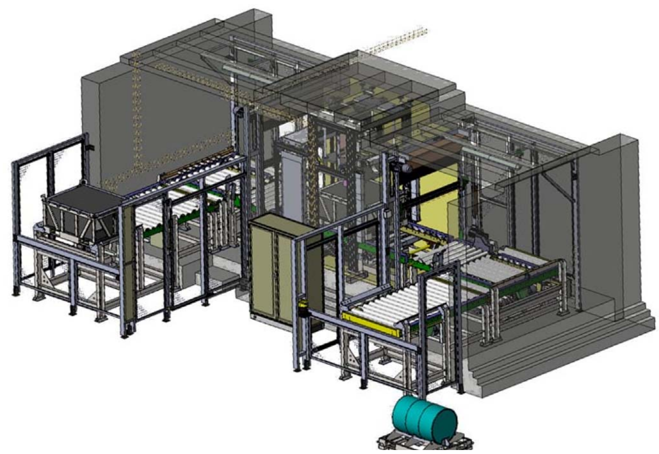

Fig. 15. Internal structure of industrial free release measurement system (e.g., rollway, detector array in the center of the tunnel).

Fig. 16. Current phase of industrial free release measurement system construction.

Fig. 17. Phantom filled with low radionuclide content gravel.

4. Metrological traceability management

4.1. Calibration and testing standards

New, universally applicable certified calibration and testing standard sources and reference materials are used to facilitate accurate, traceable measurement.

Different standards and reference materials were prepared for the purposes of adjustment, calibration, stability checking and, especially, verification of the Monte Carlo models:

– Phantoms consisting of standardized containers filled with non-active materials having different densities, with embedded tubes for use in the insertion of standard sources of radiation; Fig. 17 shows a phantom filled with aggregate gravel having a low content of natural radionuclides.
– Reference materials, such as ordinary gravel, containing naturally occurring radionuclides; special metallurgically produced tubes with known activity of Co-60, Ag-110m (Fig. 18) and contaminated light material.
– Special ball standard sources from metal and plastic in which standard cylindrical sources containing different radionuclides can be introduced (Figs. 19 and 20). These balls simply fill any large-volume container, and a combination of balls with sources (or without them) can be used to simulate hot spots and verify the performance of homogeneity tests. The metal balls (ordinary petanque balls) are hollow having a wall thickness approx. 6 mm. These balls simulate metal tubes very well.

Fig. 18. Metallurgically produced tubes (reference material, Co-60, Ag-110m).).

Fig. 19. Ball standard sources.

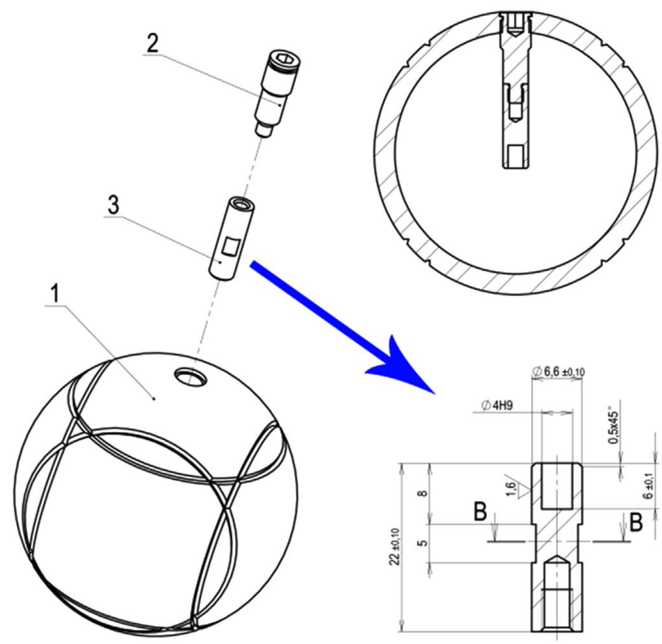

Fig. 20. Diagram of the ball standard source. Active material inside is covered by steel forced on plug (2 mm thickness).

5. Conclusions

The new-concept measurement system for the segregation and free release of radioactive waste using new shielding building blocks makes it possible to significantly improve the throughput, accuracy, reliability, modularity and mobility of the measurement.

The results will allow decommissioning measurements to be performed using standard methods with traceability to national standards of radionuclide activity. In this way the requirements of regulators and industrial stakeholders are met, guaranteeing the integrity and cost-effectiveness of the clearance and disposal processes with improved safety and accuracy.

New high throughput, rapid and precise segregation and free release measurement technologies will enable rapid, more reliable and less conservative discrimination of various waste categories. It will reduce the quantity of very low level waste and low level waste incorrectly sent to repositories (including recyclable) thus reducing the high disposal costs.

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