New High-Throughput Measurement Systems for Radioactive Wastes Segregation and Free Release – 16166

Jiri Suran[*], Petr Kovar[*], Jana Smoldasova[*], Jaroslav Solc[*], Lukas Skala[**], Dirk Arnold[***], Simon Jerome[****], Pierino de Felice[*****], Bent Pedersen[******], Tatjana Bogucarska[******], Faidra Tzika [******], Raf van Ammel[******]

[*] Regional Branch Prague, Czech Metrology Institute, Radiova 1a, 10200 Prague, Czech Republic
[**] Envinet Nuvia Group, Modrinova 1094, 674 01 Trebic, Czech Republic
[***] Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany
[****] National Physical Laboratory, Hampton Road, TW11 0LW Teddington, United Kingdom
[*****] Agenzia Nazionale per le nuove tecnologie, l'energia e lo sviluppo economico sostenibile, Via Anguillarese 301 - S.M.Galeria, I-00123 Roma, Italy
[******] Joint Research Centre-European Commission, Via E. Fermi 2749, I-21027 Italy

ABSTRACT

The European Metrology Research Program (EMRP) is a coordinated research-and-development program that pursues the closer integration of national research programs. The EMRP Metrology for Decommissioning Nuclear Facilities “MetroDecom” Joint Research Project ENV54 (JRP ENV54) [1], a 36-month project started in September 2014, delivers research products addressing all aspects of the decommissioning process. This includes the characterization of waste materials present on the decommissioning site, pre-selection prior to free release or repository acceptance measurement, free-release measurement, the measurement of waste packages thermal power prior to the repository storage, and the monitoring of stored wastes and repositories in the very long term.

This paper describes the track record of two JRP ENV54 working packages, which address 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 more reliable decision-making with regard to the safe release and disposal of radioactive wastes into the environment and, resulting in positive economic outcomes.

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 based on the development and implementation of decommissioning methodologies and associated new measurement techniques.

The decommissioning process is not only very costly but is 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, to minimize the environmental burden by providing means for improved handling and disposition of waste, thus building public trust in nuclear technologies.

Cost reduction requires the adoption of standardized methods and devices traceable to national standards to make it possible to discriminate between the various waste categories precisely and rapidly. It allows in this way a safe release of waste into the environment and possible recycling, or its long-term storage in repositories.

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

- 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.
- 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” project is to overcome these issues, to develop a large-scale industrial prototype free release measurement system, to implement it 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).

DESCRIPTION OF MEASUREMENT SYSTEM COMPONENTS

Measurement systems 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.

Patented modular shielding

A special new patented [2] modular shielding material from concrete having low natural radionuclide content, as implemented in unified, reusable ecological building blocks with unique features, thus facilitating the easy, 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 Ra-226 ≈ 0.6 Bq/kg; Th-228 ≈ 0.3 Bq/kg; 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 them. 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 of 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 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°I below.

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

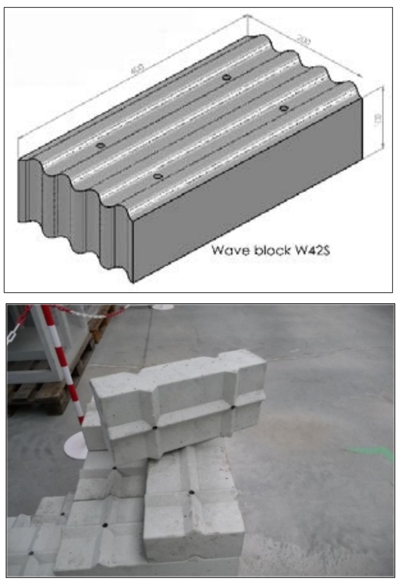

Fig. 1. Special bricks

Fig. 2. Method of constructing the shielding facility

The shielding capacity is high. Figure 3 shows a decrease in the dose rate from 101°nSv/h to 33 nSv/h during the construction of a shielding chamber of the experimental free-release measurement system. The position of the dose rate detector was identical prior and during construction. The chamber is prism-shaped, having floors being 60 cm thick, walls 40 cm, ceiling 50 cm and doors 20 cm. The ventilation is realized by blowing in by air free of radon and its decay products. Figure 4 shows a comparison of the background gamma-ray spectrum acquired by high purity germanium (HPGe) coaxial detector, with 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.

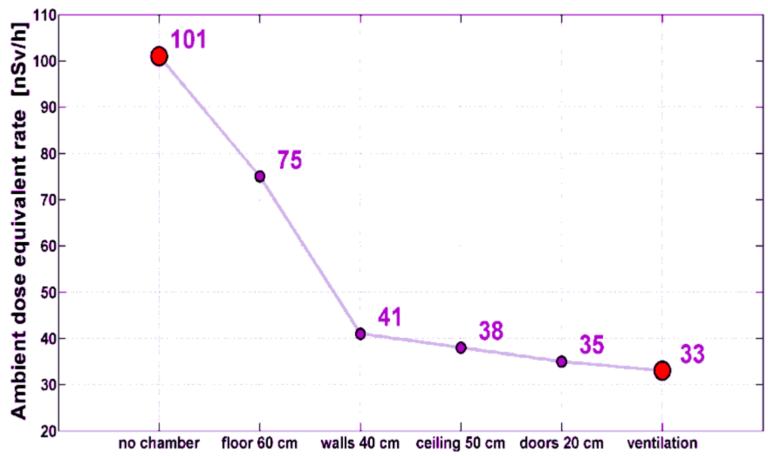

Fig. 3. Decrease of the photon ambient dose equivalent rate during the construction of the Free Release Measuring System

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 in the region of Cs-137 is reduced twenty-five times and in that of Co-60 fifty times. This leads to a significant decrease of minimum detectable activities to a few Bq/kg for light waste materials.

TABLE I 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 [3], [4], [5]. 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. Figure 5 shows an example of optimization of the shielding thickness below the conveyor belts for various widths of shielding.

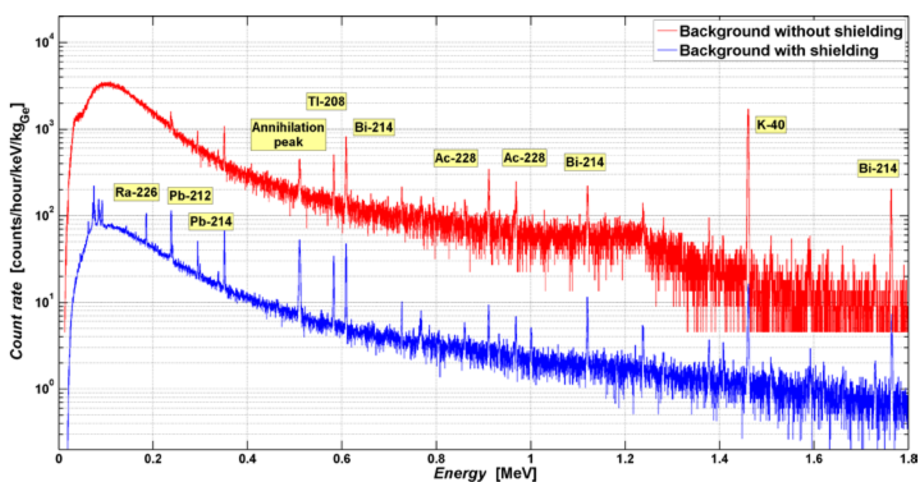

Fig. 4 Comparison of background spectrum with and without shielding

TABLE I. Concrete blocks thickness equivalent to lead[1]

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

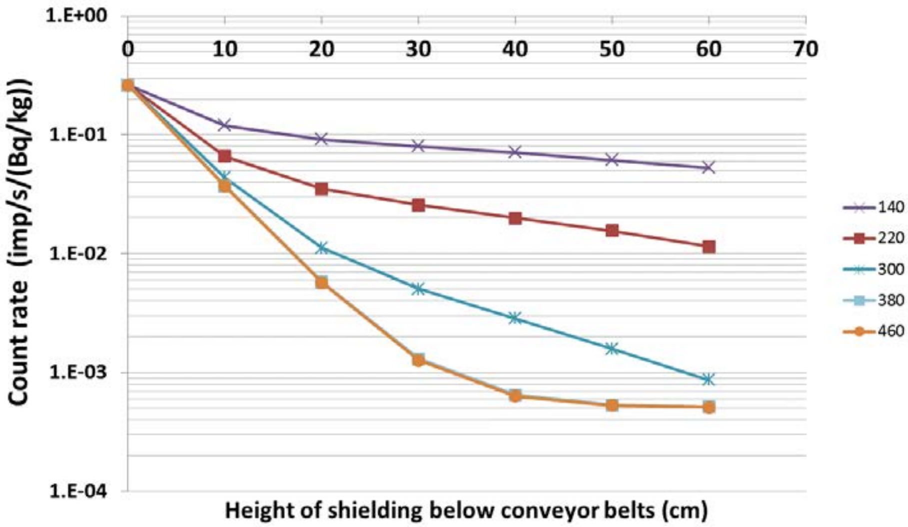

Fig. 5. Dependence of count rates on thickness of shielding below all conveyor belts, for various widths of this shielding.

Integrated modular detection arrays

Detector arrays, as other components of measurement systems, consist of Stirling-cycle cooled gamma spectrometric compact modules, plastic scintillators total gamma modules and neutron modules collectively facilitate site-optimized detection solutions, high sensitivity, scanning of homogeneity and transportability.

Detector arrays use compact and modular elements, and therefore they can be optimized for specific use by integrating the individual detectors separately in the system and, if needed, remove them. 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 [6], [7]. Only spectrometric detectors can be used to direct

[1] The 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 greater degree than it attenuates higher-energy photons. In concrete with lower Z, this difference is relatively insignificant.

activity measurement of gamma emitting radionuclides. The activities of selected easily measurable key radionuclides can be correlated with that of difficult-to-measure nuclides using scaling-factor method [8], [9].

During the decommissioning of nuclear facilities it is not possible to employ the radionuclide vector method using non-spectrometric detection, because that method of counting rate/activity conversion is only usable if the composition of wastes (ratio of the activities of different radionuclides) is exactly known and stable.

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 selected to ensure optimal detection geometry. It makes possible to scan containers in order to check the homogeneity of their radionuclide content and the presence of hot spots as well as to attain high measurement sensitivity.

Figure 6 shows the design of the detection array, as component of the measurement system, with three types of detectors (four HPGe detectors, three neutron slab counters each consisting of 5 He-3 tubes and four plastic scintillation detectors), mounted around the travel route in the shielding tunnel.

During decommissioning, low-density (e.g., filters, wood, paper) and high-density (e.g. steel tubes, concrete) materials are measured. The measurement systems currently available have fixed detector positions, so they cannot be optimized to measure smaller amounts of high-density materials or greater amounts of low-density materials. The structure permits the vertical movement of the upper detector positions so it is possible to measure two containers with a low-density material at the same time or one container with a high-density material in close measurement geometry.

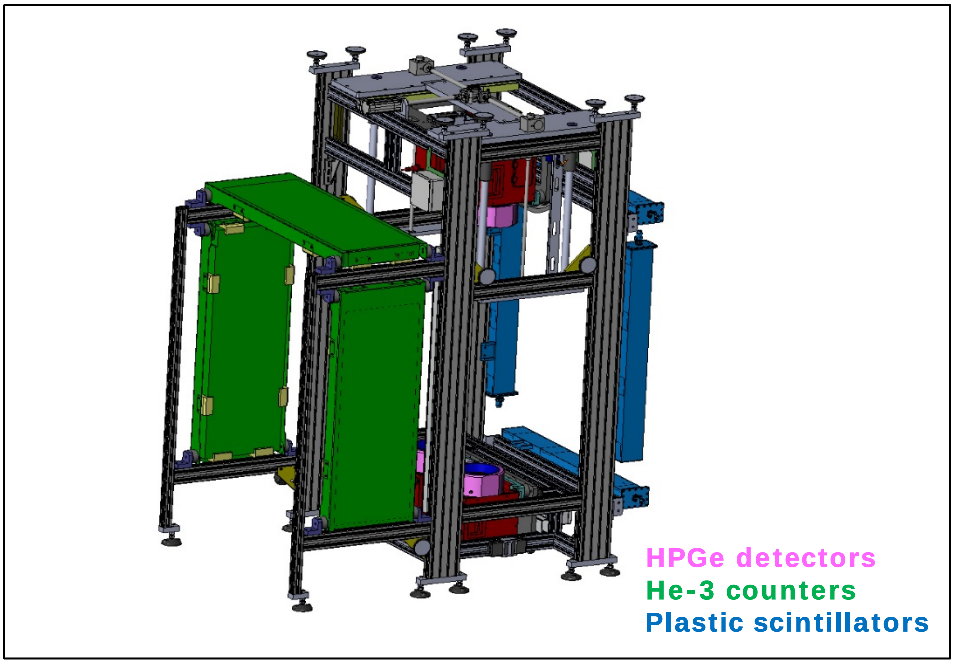

Fig. 6. Design of detectors array with three types of detectors
(four HPGe, three neutron He-3 counters and four plastic scintillators)

Scanning gamma irradiator

Collimated gamma irradiator component of 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 [6], [7]. The purpose of controlling the potential averaging is to prevent the release of small objects (i.e., hot spots) with an activity concentration above the clearance level. The described method of segmenting the measured load (multiple detectors and multiple container positions during measurement) and cross-comparison of the segments is used as 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. Although the average density of materials in an individual container also enters the homogeneity test based on the Monte Carlo model, this test 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 measured radionuclide content and to test the homogeneity of radioactivity distribution.

Figure 7 shows the design of the collimated scanning gamma irradiator module.

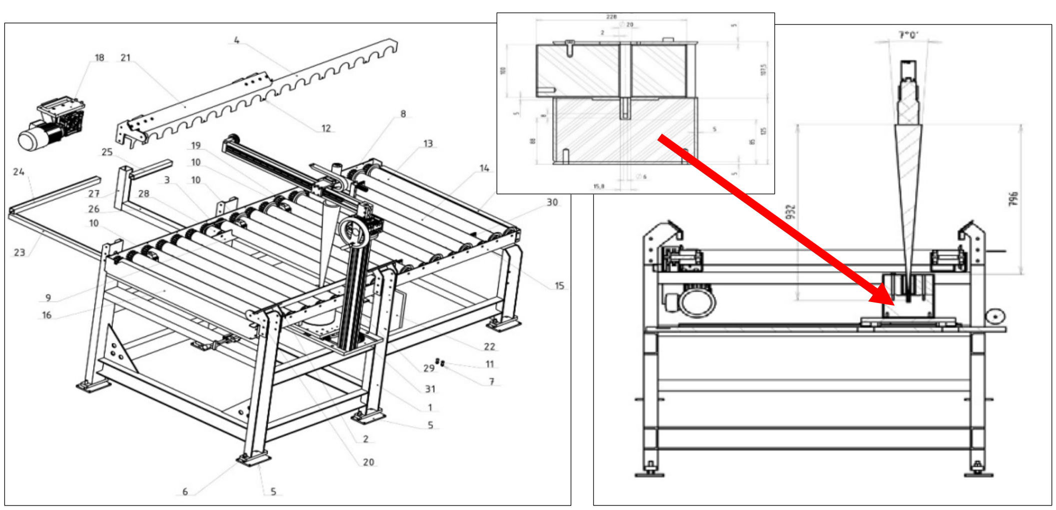

Fig. 7. Design of collimated scanning gamma irradiator module

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 the standardized transport dimensions, optimum volume in terms of measurement capacity and radiation absorption, and the qualifications for safe transport are important [10]. The goal is to limit, as much as possible, the need to reload and handle the material. While containers with low radiation absorption due to the wall material are suitable for measurement, a mechanically stronger container design offering increased absorption is required for transport.

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

Fig. 8. "MetroDecom" container IP-1

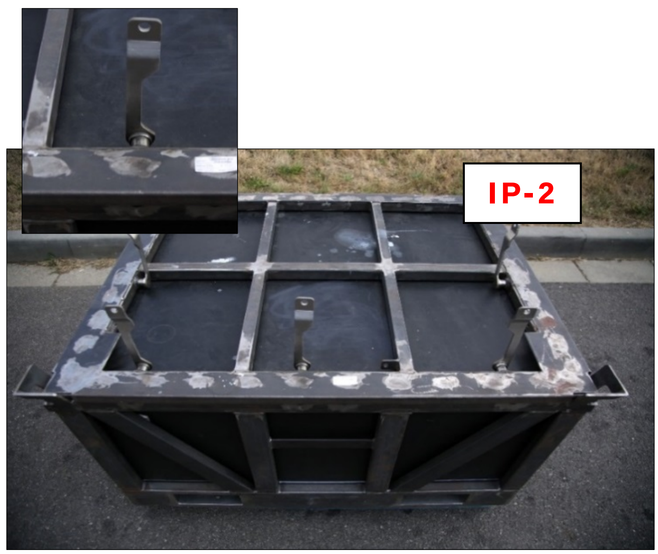

Fig. 9. "MetroDecom" container IP-2

These containers have the following features:

- Standardized dimensions corresponding to the dimensions of "Europallet" (100x120x60 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 is up to 500 kg;
- A design combining a rigid frame and a plastic inner tray, which is an optimal compromise between the requirements for transport, 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.

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 using special holders. Figure 10 shows examples of such holders.

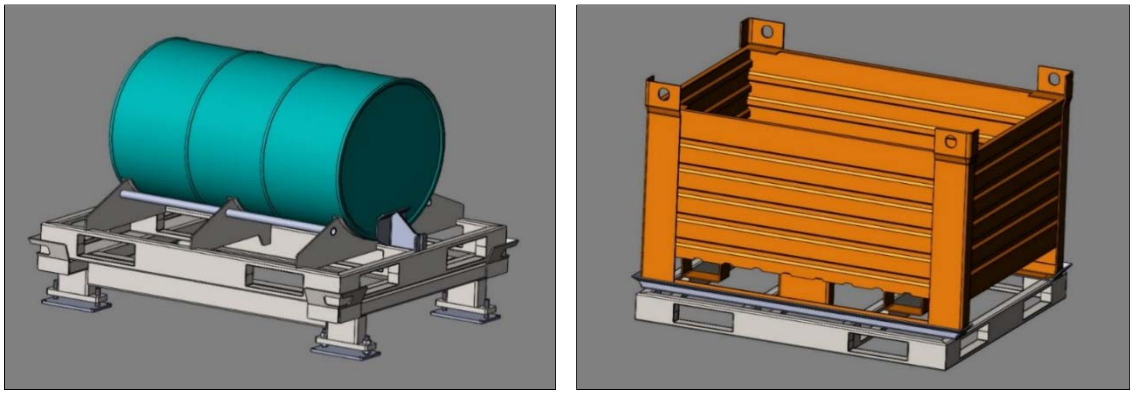

Fig. 10. Special holders for different types of containers

Filtration and ventilation equipment

Equipment for ventilation and filtration is used to filter radon and its decay products out of the blown in air which 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. Movable doors are not used because their limitations regarding high throughput, safety and affordability.

FEATURES OF THE MEASUREMENT SYSTEMS

Experimental and industrial systems

As part of the “MetroRWM” project [11], which preceded the “MetroDecom” project, an experimental free-release system was built as shown in the Figures 11 and 12. The testing of this system confirmed the properties of 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 3x60 s (three container positions) for a spot source in the middle of the empty container.
MDAs for homogeneously contaminated materials filling up the 0.4 m^3 container are shown in TABLE II.

Fig. 11. Experimental Free Release Measurement System

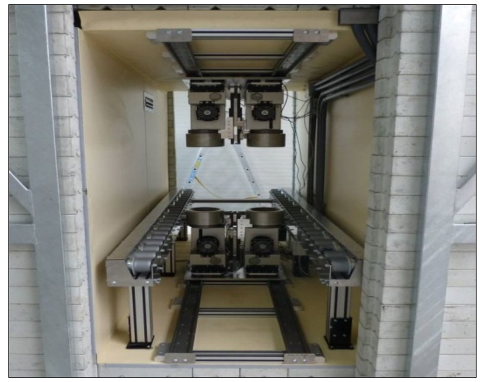

Fig. 12. HPGe detector array (4pcs) in experimental Free Release Measurement System

TABLE II. 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

Experimental system experiences resulted in a fully modular concept for an industrial system shown in Figure 13 permitting optimization based on local needs.

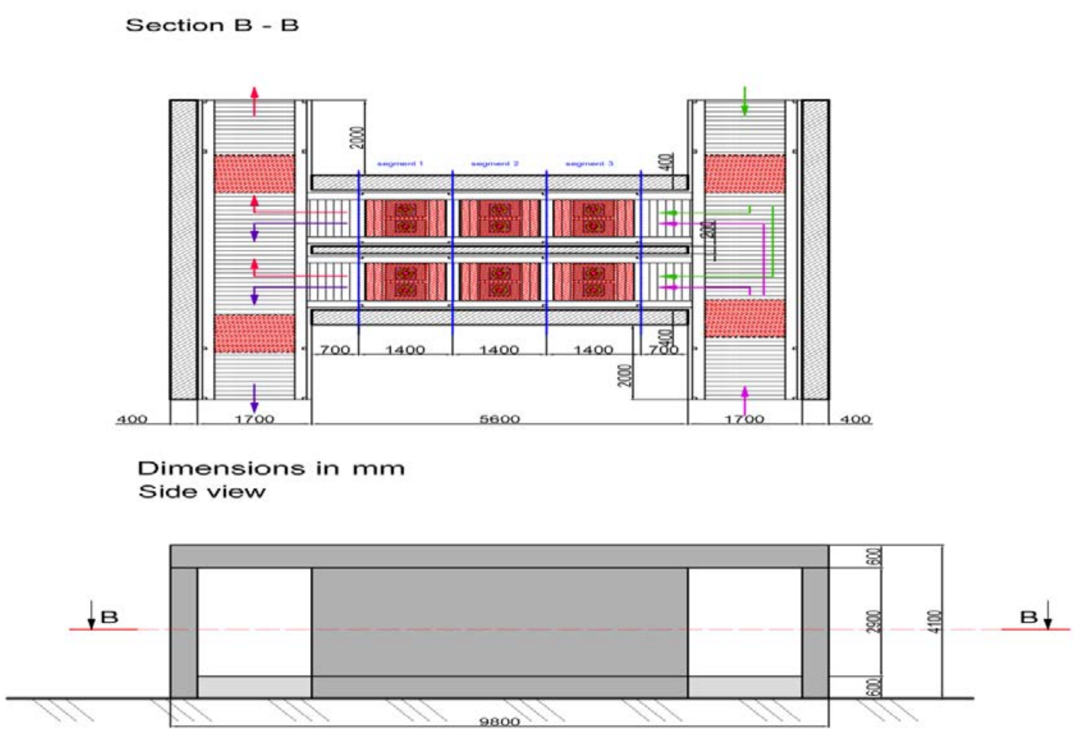

Fig. 13. Concept of industrial Free Release Measurement System

The number of tunnels and detection arrays can be selected according to 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 Figure 14, and its internal structure is depicted in Figure 15.

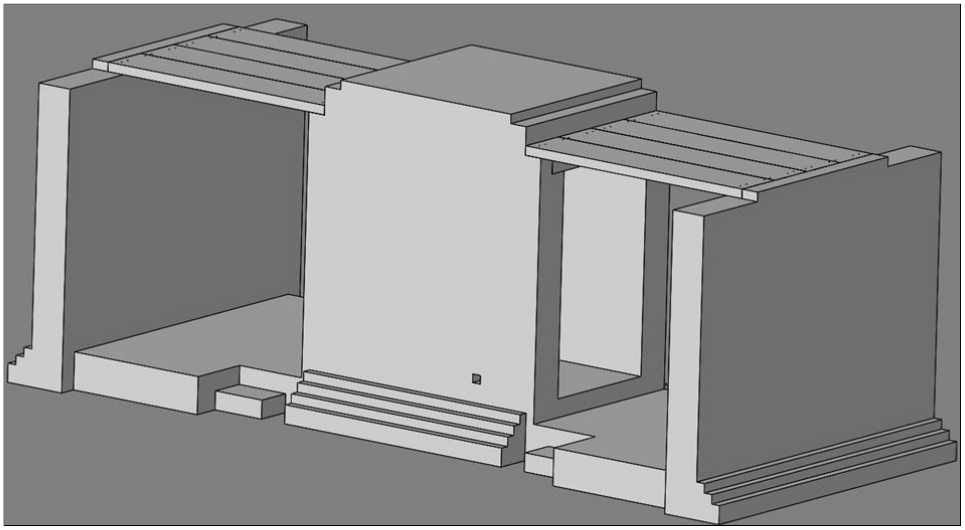

Fig. 14. Industrial Free Release Measurement System shielding

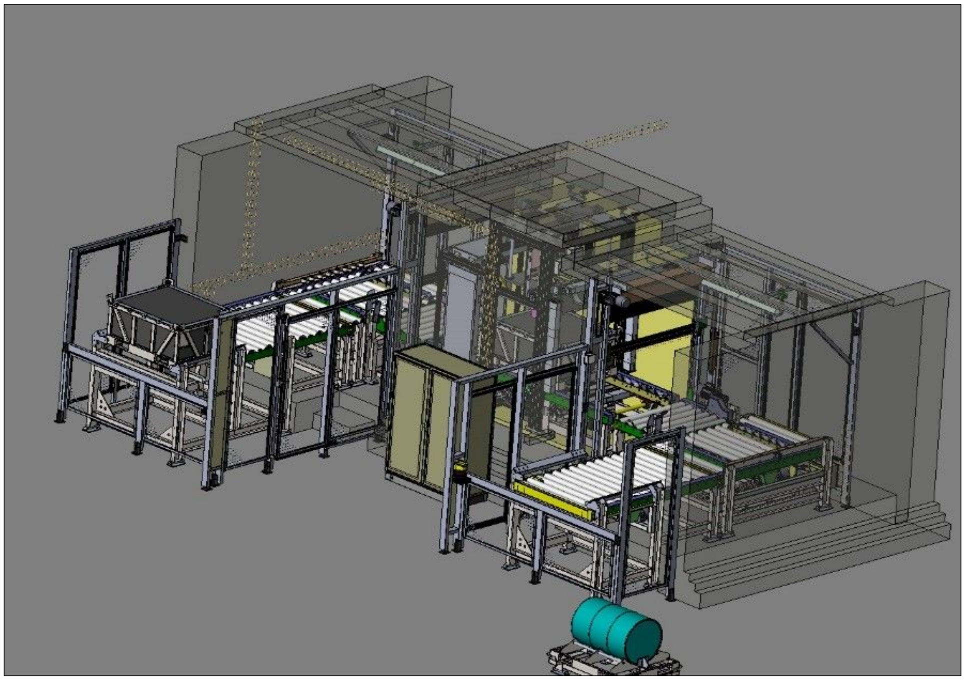

Fig. 15. Internal structure of industrial Free Release Measurement System (e.g., rollway, detector array in the center of tunnel)

The current stage of construction is shown in Figure 16. A model of the equipment (with the front wall removed) is shown in Figure 17. 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. 16. Current phase of industrial Free Release Measurement System construction

Fig. 17. Industrial Free Release Measurement System model (front wall removed)

New and improved features of the systems

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

- A standardized modular, transportable, reusable, site-optimization-enabling concept.
- High throughput (up to 10 t/hr, 20 m3/hr).
- 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 container.
- Simplified handling of materials using a new type of container.
- Radon decay products background and internal contamination reduction.

METROLOGICAL TRACEABILITY MANAGEMENT

Calibration and testing standards

New, universally applicable certified calibration/testing standards 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; Figure 18 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 (Figure 19) and contaminated light material;
- Special ball standard sources from metal and plastic in which standard cylindrical sources containing different radionuclides can be introduced (Figure 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. Phantom filled of low radionuclide content gravel

Fig. 19. Metallurgically produced tubes (reference material, Co-60, Ag-110m)

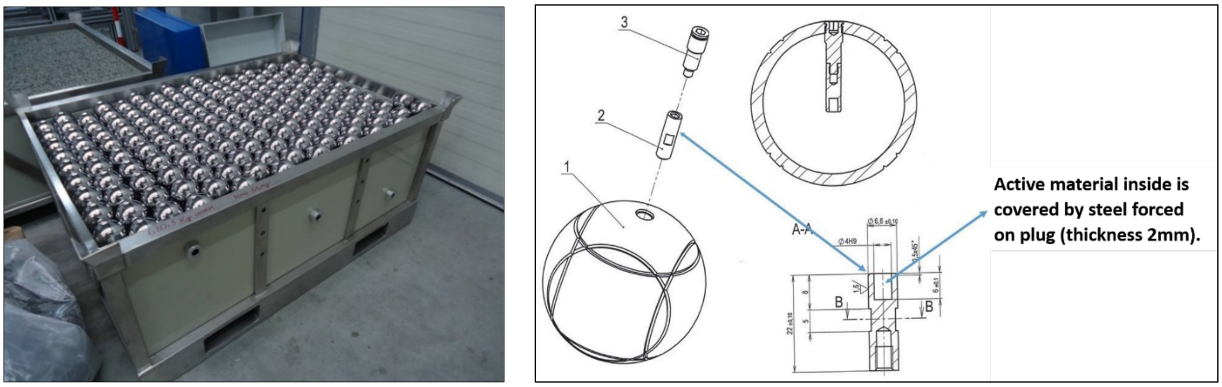

Fig. 20. Ball standard sources

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 and mobility of the measurement leading to economic savings.

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 reduce the quantity of solid waste materials incorrectly sent to repositories (including recyclable) thus reduce the disposal costs by about 10%. For the decommissioning of a single nuclear power plant it would lead to millions of Euros of savings. Typical cost of one cubic meter of very low level waste (VLLW) and low level waste (LLW) repository space is about EUR 5,000 to 30,000. For example decommissioning Germany's power plant at Greifswald produced more than half a million tons of VLLW and LLW.

The underpinning of harmonized methods and productive infrastructure will allow thorough implementation of the legislation contained in a series of EC Directives and IAEA Safety Standards.

Testing of the prototype of the industrial system at the European Joint Research Centre in Ispra (Italy) will be accessible to professionals in the area of decommissioning and waste management by participation in workshops. This creates an opportunity for stakeholders and end users to launch and implement a new generation of radioactive waste monitoring systems with special shielding.

The details of the physical design and algorithms of the operational and measurement software will be subject of future articles.

REFERENCES

1. http://www.decommissioning-emrp.eu/

   https://www.euramet.org/research-innovation/search-research-projects/details/?eurametCtcp_project_show%5Bproject%5D=1221&eurametCtcp_project%5Bback%5D=473&cHash=0ecb77224cc4401c34162aa1106a415b
2. EPO patent family documents: EP 2684855 A2 20140115; EP 2684855 A3 20140903; CA 2821148 A1 20140112; CZ 20120478 A3 20140910; CZ 305447 B6 20150923; US 2014027676 A1 20140130
3. JRP EMRP ENV09: MetroRWM, Data set D1.3.3, "Optimization of FRMF Measurement Geometry", (2013)
4. JRP EMRP ENV54: MetroDecom, Data set D2.1.2, "MC Calculations to Derive Optimum Parameters for Segregation Facility", (2015)
5. J. Solc, P. Kovar, J. Suran, V. Peyres and E. García-Toraño, "Optimization of a measurement facility for radioactive waste free release by Monte Carlo simulation", *Applied Radiation and Isotopes*", 87, pp. 348-352 (2014)
6. COUNCIL DIRECTIVE 2013/59/EURATOM of 5 December 2013 laying down basic safety standards for protection against the dangers arising from exposure to ionising radiation, and repealing Directives 89/618/Euratom, 90/641/Euratom, 96/29/Euratom, 97/43/Euratom and 2003/122/Euratom, Annex VII, Table A
7. IAEA Safety Standards Series No.GSR Part 3, Radiation Protection and Safety of Radiation Sources: International Basic Safety Standards, General Safety Requirements, Schedule I, Table I.1, I.2; International Atomic Energy Agency, Vienna, 2014
8. ISO 21238:2007, "Nuclear energy — Nuclear fuel technology — Scaling factor method to determine the radioactivity of low- and intermediate-level radioactive waste packages generated at nuclear power plants".
9. IAEA Nuclear Energy Series MW-T-1.18 "Determination and Use of Scaling Factors for Waste Characterization in NPPs".
10. IAEA Specific Safety Requirements, Series No. SSR-6, "Regulations for the Safe Transport of Radioactive Material - 2012 Edition".
11. http://www.radwaste-emrp.eu/wp-content/uploads/downloads/2013/03/T1.1-Free-Release-and-Repository-Acceptance-Legislation.pdf

    https://www.euramet.org/research-innovation/search-research-projects/details/?eurametCtcp_project_show%5Bproject%5D=1126&eurametCtcp_project%5Bback%5D=470&cHash=4753be2ba34fe4f840994e959cd9b571

The EMRP is jointly funded by the EMRP participating countries within EURAMET and the European Union.