

Developments and experiences of the CHANCE, MICADO and PREDIS projects in radioactive waste characterization

Massimo Morichi^{1,*}, Erica Fanchini¹, Eric Breuil², Christophe Bruggeman³, Bertrand Perot⁴, Denise Ricard⁵, and Ernst Niederleithinger⁶

¹ MICADO Project Coordinator, CAEN s.p.a., Via Vetraia N.11, 55049 Viareggio, Italy

² Orano DS, Etablissement de Bagnols-sur-Cèze, RN 580 – ZA de l'Euze, 30200 Bagnols-sur-Cèze, France

³ SCK CEN, Belgian Nuclear Research Center, Boeretang 200, B-2400 Mol, Belgium

⁴ CEA CADARACHE, Bâtiment 224, F-13108 Saint-Paul-Lez-Durance, France

⁵ Andra, 1-7, Rue Jean-Monnet, 92298 Châtenay-Malabry Cedex, France

⁶ Bundesanstalt für Materialforschung und-prüfung (BAM), Unter den Eichen 87, 12205 Berlin, Germany

Received: 28 July 2022 / Received in final form: 26 October 2022 / Accepted: 10 November 2022

Abstract. Characterization is a very important step in dealing with materials and waste streams generated during the operational and decommissioning phases of nuclear installations, including nuclear power plants. Characterization allows differentiation between materials that can be released from regulatory control and those that require further treatment and conditioning to become a stable waste form suitable for future storage and final disposal according to its waste classification. Characterization is also needed in the pre-disposal stages of radioactive waste management to demonstrate compliance with the waste acceptance criteria of the facilities that will accept the different waste forms. This work will present the strategies developed and implemented by the three projects for in-depth and accurate waste characterization and investigation of the different radioactive waste packages considered. CHANCE, MICADO, and PREDIS will present their goals, the methods developed, the technologies used and the (preliminary) results contributing to the improvement of the safety and the data and information quality of the waste packages analyzed at the different stages of the waste management process. Special emphasis will also be given to complementary approaches highlighting the usability of the technologies, the accessibility of the data, and the problem-solving of the three projects within the European panorama.

1 Introduction

The characterization of radioactive waste is one of the key aspects of its management. Accurate knowledge of both the volume and the radiological and physicochemical content of nuclear waste generated in each national program will influence the strategies and technologies for its further management. Radioactive waste characterization is considered a key procedure required by national and international legislation, ensuring transparency of information for all stakeholders involved. It is also important with respect to safeguards to prevent nuclear proliferation, to ensure adequate radiation protection for workers in the relevant operation, and for the safety of the entire population and the environment. Detailed knowledge of the radioactive waste package (RWP) contents is also essential to establish the best operational procedures. Radiological and physicochemical assessments are important to determine the type of waste, the safety procedures for its handling, the type

of conditioning procedure to be adopted, and the resulting modalities of storage. This assessment in the early stages of waste production is very important to optimize subsequent waste management (treatment, conditioning, disposal conditions, and financial impact). It significantly reduces the risk associated with waste reconditioning.

Precise information on radiation levels is even more important for RWP with content that lies at the border between two categories. Precise information can be critical in determining whether to choose, for example, a surface or geologic repository, which can lead not only to a change in the final location, but also to a change in all activities down to the predisposal phase, and can result in significant costs.

Another important issue to be evaluated during the characterization phase is the identification of the chemical content, which is particularly relevant for contaminated sites and historical waste packages. Waste acceptance criteria at some current or future disposal facilities include chemical parameters that can limit the number of specific species.

* e-mail: m.morichi@caen.it

In this context, the CHANCE, MICADO, and PREDIS projects were launched under the Euratom calls. They focus on optimizing the handling, characterization, and monitoring of RWP, optimizing the data flow, and sharing the acquired knowledge within the community.

This paper describes the state of the art in characterization technologies and the innovations introduced by the three projects.

2 European nuclear waste and its sources

More than 60 000 tons of spent nuclear fuel are stored throughout Europe (excluding Russia and Slovakia), most of it in France. High-level waste, which also comes from the reprocessing of spent fuel, accounts for the largest share of radioactive waste. In Europe, more than 2.5 million m³ of low and intermediate-level waste has been generated, of which about 20% (0.5 million m³) has been stored and 80% (close to 2 million m³) has been disposed of.

The decommissioning of the European reactors may generate more than 1.5 million m³ of low and intermediate-level waste. During the entire lifetime of European nuclear reactors, about 6.6 million m³ of nuclear waste may be generated. Four countries account for more than 75% of this waste: France (30%), the UK (20%), Ukraine (18%), and Germany (8%) [1,2].

Excluding fuel chain facilities, the European power reactor fleet alone could generate at least another million cubic meters of low- and intermediate-level waste from decommissioning. This is a conservative estimate, as decommissioning experience is still growing.

There are more than 140 operating nuclear power plants in Europe. The ongoing generation of nuclear waste and the imminent decommissioning of nuclear facilities poses an increasing challenge as storage capacity in Europe will soon be exhausted. Moreover, in countries such as the United Kingdom and France, decommissioning waste is also strongly influenced by activities that are independent of the power reactors.

All countries publish information regularly but differ significantly in how they define, categorize nuclear waste, and report quantities of nuclear waste. The different national approaches reflect a lack of common standards and coherency in how countries manage some aspects of nuclear waste. To this end, initiatives have been taken, e.g., within the OECD-NEA, to achieve more consistent and standardized reporting [3,4].

The CHANCE [5], MICADO [6], and PREDIS [7] projects contribute to improving the characterization of radioactive waste at different stages of the waste management process in order to reduce the impact of final disposal. The three projects focus on the largest part of the existing waste volumes, which are mainly composed of low and intermediate-level packages, legacy waste, and waste from European D&D activities. In the various European countries of origin, these are stored for final disposal or in geological repositories.

The future optimization of the waste management process towards the final repository is a combination of multiple activities that form the core of the three EU

projects described here: monitoring, handling, conditioning & packaging, and characterization with the digitization of key information.

Characterization aims at providing accurate knowledge of the radiological and physicochemical content of the waste package. To optimize the conditioning process, better manage the radiological inventory, digitally track the information, and support decisions to minimize volume and cost, several techniques are used simultaneously. This article addresses these radiological and physicochemical characterization techniques. Other important techniques, such as long-term waste monitoring assuring real-time knowledge of the nuclear waste status and enhancing global safety, pre-treatment and conditioning techniques, or knowledge transfer of lessons learned to future generations and countries not involved in this D&D phase, are not addressed in this article.

3 The importance of nuclear waste characterization

The RWP characterization is a complex analysis that involves not only the qualification and quantification of the radiological content but also the analysis of the physical (density, volume, shape, mechanics, cracking, diffusion, etc.), chemical (element composition, toxicity, presence of reactive substances, gas production, liquid, solid materials, etc.) and radiological (dose rate, spectroscopy, isotopic composition, spectrometry, calorimetry, etc.) parameters required to optimize the waste management. Destructive and non-destructive methods are used to measure and verify these parameters.

Destructive analysis (DA) offers the most accurate and unbiased activity determination, and the sampling provided should represent the whole package. This technique is mandatory since pure alpha and beta-emitting radionuclides or those emitting gamma or X-rays with a too-small intensity or energy are extremely difficult to measure even in already conditioned waste packages. Destructive analysis includes a sampling of the package, sample preparation, and chemical separation methods. Sampling is a critical step in the characterization process. Determination of representative samples and full compliance with sampling procedures ensure the reliability of results. Chemical and radiochemical treatment of the primary waste also enables measurements that assure traceability of the determined activity.

Non-destructive analysis (NDA) and testing methods are used to minimize the radiation dose to personnel, avoid secondary radioactive waste production, minimize costs, and provide a comprehensive characterization of waste packages in reasonable measurement times with respect to sampling. Several non-destructive methods for quality checking of radioactive waste packages have been developed and tested [8].

Despite the methods employed today, there is a need for the development of new non-destructive methods focused on conditioned waste.

The three projects CHANCE, MICADO, and PREDIS are mainly focused on non-destructive techniques of investigation. This section describes the state of the art of investigation approaches and methods used in Europe, focusing on radiological and physical characterization by using non-destructive methods and on the innovations introduced by the three projects.

4 Non-destructive measurement techniques

Non-destructive methods can be divided as follows: radiological methods measure the radiation generated by the content of the package. Non-radiological methods (visual, electromagnetic, acoustic, X-ray, or muon-based) are mainly used to retrieve information about the package's internal geometry, density distribution, or integrity. Active measurements use some kind of source (in X-ray, ultrasonic or electromagnetic methods and in radiological methods based on the emissions due to reactions to neutrons or photons from external sources) and the interaction of the source with the package and its content is recorded by some receivers/sensors. Visual and thermographic methods might also be considered active methods if they use external light or infrared sources, otherwise, they are passive. Passive methods use natural emissions (radiological methods based on spontaneous emissions), events (acoustic emission due to cracking), or particles (e.g., muon tomography) measured or tracked by detectors of various types.

The radiological measurements are used to determine the presence, quantity, and location of radioisotopes or the existence of toxic or reactive materials, neutron absorbers, or moderators in the RWP [9]. The main passive measurement technologies considered so far in waste characterization are:

- **Gamma dose rate measurements** are performed mainly for safety reasons: they are performed at contact and a distance of 1 meter from the RWP surface. They are used for the first radiological assessment and planning of the handling of the RWP by the operators. The dose rate can be converted into activity if the isotopic vector of the radioactive waste is known.
- **Gamma spectroscopy and spectrometry techniques:** mainly tomographic and segmented gamma scanning systems based on HPGe detectors are used to detect, identify, quantify, and localize gamma radioisotopes. They are suitable for scanning open, segmented, and angular geometries. Tomographic systems are also able to reconstruct radiological emissions and hot spots inside the RWP in 3D. They determine the presence of uranium and plutonium as well as the presence of radiotracers. Often, these systems are also equipped with a gamma radiation source to perform transmission measurements to obtain density information on the contents. Gamma results are also used in combination with neutron detection techniques.
- **Passive neutron measurement** is used to quantify actinides, mainly Plutonium. This type of measurement detects neutrons from spontaneous fissions, where several correlated neutrons are emitted per fission, or (α, n) reactions with only one emitted neutron per reaction. The measurements foresee both a total neutron count and a time correlation analysis. Looking at event coincidence allows distinguishing the signal due to fission neutrons from the accidental background due to (α, n) reactions and hence the type of isotopes in the RWP (e.g., ^{240}Pu vs. ^{241}Am). For medium size drums, this technique is less sensitive to high atomic number materials (metals) than to low atomic number materials (especially those containing hydrogen). Another limitation is the presence of curium because its very high specific activity of spontaneous fission can easily mask the passive neutron coincidence signal of plutonium in the RWP, as in the case of highly active metallic residues from spent fuel reprocessing.
- **Digital autoradiography** is a technique for determining alpha and beta-contaminated waste. It uses photographic-like screens that are applied to the waste. The radiological signature allows to extract graphical information on the presence of low-energy emitters such as ^3H , ^{14}C , and alpha particles.
- **Gas measurements:** chromatography or scintillation counting can be used to quantify the presence of radioactive or radiolysis gas released when the waste package is placed in a sealed enclosure. The gas can accumulate in this chamber and the quantity of gas emitted by the waste, such as ^{14}C , ^{36}Cl , and ^{226}Rn , can be measured to derive the release rate. In recent years, optical detection of radionuclides has been successfully demonstrated for radiocarbon. Laser spectroscopy can be used to provide selective and sensitive detection of molecules directly in the gas phase. Optical detection offers several advantages over conventional techniques for nuclear applications, such as liquid scintillation counting. One of the most sensitive laser spectroscopy techniques is cavity ring-down spectroscopy (CRDS).
- **Calorimetry** is an experimental technique for measuring the thermal power generated by nuclear decay in the radioactive matter, which is related to the specific power of the isotope to deduce the mass of the tested matter. Several prototypes and commercial instruments are currently in operation at different research centers around the world to determine the quantity of alpha emitters and the presence of tritiated waste.
- **Muon Tomography** is still considered to be in the R&D phase. Muons generated by cosmic rays can penetrate matter and be used to image the internals of structures. Since the early 2000s, several groups have extended the concept of muon radiography to tracking individual muons as they enter and exit a structure. Most current efforts have been aimed at demonstrating the potential of muon tomography for detecting the smuggling of special nuclear materials (SNMs) in cargo, the non-invasive characterization of legacy nuclear waste containers, and for nuclear material accountancy of spent fuel in dry storage containers.
- **Acoustic emission (AE):** this technique records acoustic events caused primarily by cracks in the cement fill of waste packages using receivers placed on the out-

side of the package. Localization and characterization of the events are possible. While AE is used as a standard tool in other applications, its use in waste characterization is so far limited. It can be considered very useful in the development and validation phases of treatment technologies involving cement or geopolymer, as well as in the design and testing of waste containers. This technique is also used to detect and monitor corrosion on the waste container walls.

The active measurement types are:

- **high energy photon** imaging performing radiography and/or tomography of the RWP is one of the initial measurements and provides information about the physical structure of the RWP such as density, cracks, presence of voids, position and shape of elements inside the volume, presence of liquids. This technique is usually used for legacy waste and serves as one of the key elements providing relevant information for further inspections.
- **Dual-energy X-ray imaging:** this technique is still in the R&D stage and is currently under investigation. Using tomographic acquisitions at different energies allows for enhancing the differences in the attenuation coefficients of the elements constituting the imaged materials and provide information about their effective atomic number in each pixel. This information is used in combination with the density already provided by X-ray imaging to improve the identification of materials with similar densities (e.g., aluminium and concrete in radioactive waste).
- **Active neutron interrogation technique** has been used industrially for decades, for instance at the spent fuel reprocessing plant of ORANO La Hague (France), where four measurement stations are currently in operation for the characterization of highly active metallic residues (hulls and end pieces) in the T1, R1 [10], and ACC facilities [11]. The measurement uses the die-away method to quantify the fissile material inside the package when plutonium gamma rays are masked by the intense emission of fission and activation products and when the passive neutron coincidence count signal is dominated by curium spontaneous fission neutrons or when the (α, n) accidental background is too intense. It uses the flux of a pulsed neutron generator (DD deuterium-deuterium or DT deuterium-tritium) inducing fissions in the presence of fissile material and counting the emitted prompt and delayed fission neutrons. The prompt neutron signal is detected a few hundred microseconds after the generator neutron pulse and is proportional to the fissile material mass. This is the key advantage of this technique for safety-criticality purposes, although it is more affected than passive neutron counting by the waste package matrix effects. The fission-delayed neutrons are also recorded between pulses from the neutron generator, but after the prompt neutron signal has disappeared. The delayed signal is sensitive to ^{238}U due to fast fissions, in addition to the fissile isotopes ^{235}U , ^{239}U , and ^{241}Pu (thermal fissions as in the prompt signal). Using both prompt and delayed neutron signals allows discrimination of U and Pu contributions in some instances.
- **Prompt gamma neutron activation analysis (PGNAA):** this technique is still in the R&D stage under study and is not yet widely used in the field. It requires a combination of gamma and neutron measurements to determine the presence of toxic elements (such as B, Hg, Cd, Gd, Cl, As), neutron moderators (such as H or C), or absorbers (such as B, Cl, Fe, Cr, Ni, Cd, Gd). It detects prompt gamma rays from radiative capture or inelastic scattering neutron reactions on the nuclei of interest.
- **Ultrasonic and electromagnetic (eddy current) techniques** can be applied to verify the thickness of steel containers. A reduction in thickness in some parts of the container may be caused by internal corrosion.
- **Visual techniques** using special sources (e.g., strip projection) and software, can be used to record and monitor deformations of waste packages caused by either improper handling or internal processes such as the cement-alkali-silica reaction.

5 Destructive measurement techniques

When handling samples, non-destructive analysis is preferred and, usually, is the first type of measurement. However, under certain conditions, the second type, characterization of the waste by destructive measurement techniques, becomes mandatory. Legacy or alpha and beta-contaminated waste are examples in which destructive measurements of the content are necessary since the radiological emission from outside the package is not detectable. In these cases, there is a necessity to open the packages and perform precise measurements on-site. The decision on the type of measurement and the number and locations of the samples to be extracted and analyzed is made by the results of the non-destructive techniques and is applicable only to conditioned waste.

The main measurement techniques available are:

- **coring and cutting:** this involves the extraction of a coring element to perform multiple tests and controls, beginning with a visual examination, evaluation of composition components, and ending with the extraction of waste samples for analysis.
- **Chemical, radiochemical, and physical analyses** are performed on the extracted disks coming from the coring. Tests to check containment, detect and measure the presence of toxic materials, identify long-lived radioactive elements, measure pure alpha and beta emitters, and determine isotopic composition will be conducted.

6 The three projects and their innovations

The previous sections refer to the main characterization and conditioning technologies used nowadays in the nuclear waste management sector. The list is not exhaustive but gives an overview of the measurement techniques used. The three projects fit into this framework and are focused on these measurement techniques.

This section describes the aims of the projects and the technologies developed, examining the innovations in light

of the panorama described earlier. The innovations behind these projects not only refer to the investigation technologies under development. Describing the single technologies falls short, as the waste management procedures can improve the waste package characterization itself and can be considered an element of innovation. These processes under study combine information from the different detection techniques to improve the knowledge of the package contents.

Figure 1 reports all the technologies of the CHANCE, MICADO, and PREDIS projects, as well as the leading institutes or partners of the projects during their lifetime. The timing of the projects varied: CHANCE ended in March 2022, MICADO will end in February 2023, and PREDIS will end in 2024, so it is not possible to report results for all elements in the table.

The three projects are in the field of decommissioning and/or general waste treatment. The CHANCE project (Characterization of conditioned nuclear waste for its safe disposal in Europe) aimed to further develop, test, and validate techniques that will improve the characterization of conditioned radioactive waste, in particular for radiological and physicochemical characterization. Besides the technological developments, another objective of the project was to establish a comprehensive understanding at the European level of the current conditioned radioactive waste characterization and quality control schemes across the different national radioactive waste management programs and the remaining needs in this field. In order to collect information on characterization, particularly on the links and overlaps between these requirements and the waste specifications for the different national disposal facilities, a questionnaire was produced and distributed to the operators of radioactive waste disposal facilities in Europe through the CHANCE project end user group (EUG). A Synthesis of the questionnaire answers is provided in the deliverable [12]. Another report has been prepared to describe the state of the art regarding techniques and methodologies on waste characterization in the R&D stage to meet the needs identified by end users [13]. An important document is the CHANCE report D2.3 [13] enlightens important points on the expected challenging R&D techniques in the field. The most relevant topics are:

- high energy and dual-energy X-ray imaging to characterize the physicochemical content and structure of large and dense waste packages
- photofission: active photon interrogation with high-energy X-rays to measure and identify nuclear materials in large, dense, concrete matrixes or containers
- passive neutron measurements: neutron coincidence counting techniques based on cheaper alternatives to ^3He detectors
- active neutron interrogation techniques to measure the fissile mass for high-level radioactive waste
- neutron activation analysis to characterize chemical elements and long-lived isotopes difficult to measure by gamma-ray spectroscopy
- radiation attenuation corrections depending on the waste matrix and the localization of radioactive products in the package based on machine learning tech-

niques trained by intensive calculations (e.g., Monte Carlo simulations of large measurement data sets covering a wide range of waste matrices and activity distributions)

- bayesian approaches interactively adjust the prior distributions of activity, radionuclide location, matrix density, composition, etc. according to all available measured data
- mobile measurement systems are to be reused at different sites and avoid the transport of the radioactive waste packages to the measurement system, which is generally very difficult for legacy waste.

The list does not include technologies already available under the CHANCE project. The remaining 6 of 8 points are already covered by the MICADO and PREDIS projects.

The techniques studied in the CHANCE project have seen major improvements in development, data acquisition, and modeling. However, as future developments remain:

- for muon tomography:
 - development of algorithms for the monitoring of gas bubbles in bituminized waste to study their formation and behavior and assess the associated safety risks
 - improvements in the algorithms for material identification of objects inside the waste drums to limit the data acquisition time and identify smaller objects
 - imaging of the contents of CASTOR drums; not only to verify the presence of fuel assemblies but also their structural integrity and to image the contents of waste silos.
- For calorimetry:
 - developments to improve the insulation of the calorimeter to measure the smaller effect and be able to work in facilities with significant room temperature fluctuations
 - developments to decrease measurement time and increase accuracy in quantifying radioelements emitting high gamma radiation.
- For cavity ring-down spectroscopy:
 - validation of the method under real industrial conditions
 - characterization of the method in different gas matrices to ensure the functionality of the CRDS in all situations
 - implementation of simultaneous detection of different radionuclides of interest (e.g., ^{14}C , tritium, ^{36}Cl) to have a single system capable of detecting all relevant radioactive elements present in outgassing.

Most of these topics are covered by the three projects with their different collaborations and backgrounds.

MICADO (Measurement and Instrumentation for Cleaning And Decommissioning Operations) started as the second project in 2019 and will run until the beginning of 2023 [6]. MICADO aims at changing the manual procedures used for the non-destructive assay (NDA) characterization techniques of the waste packages. To better qualify the waste package to be analyzed, a waste type-dependent analysis procedure is applied and information from different relocatable detection stations is combined. The project

Method/Technology/Sensor		Lead Institute	Purpose	Project
Calorimetry		KEP	Quantification of the alpha emitters or low energy emissions	CHANCE
Cavity ring-down spectroscopy (CRDS)		VTT	Characterization of radioactive waste outgassing	CHANCE
Gamma station	Gamma camera	ORANO ENEA	Hot spot search and identification	MICADO
	Gamma dose rate and spectroscopy & RFID	CAEN	Gamma dose at contact and fast open geometry measurement for survey application	MICADO
	Gamma spectrometry	ENEA	Quantification of gamma emitters (open geometry, angular scanning, tomography)	MICADO
Neutron station	Neutron active measurements	CEA	Quantification of nuclear materials (Pu, U) in not cemented packages	MICADO
	Neutron passive measurement	CEA		MICADO
Photofission station	Photofission measurements	CEA	Quantification of fissile materials in cemented packages	MICADO
Long term monitoring	SciFi technology	INFN	Gamma dose rate monitoring for storage or repository sites	PREDIS MICADO
	SiLiF technology	INFN	Neutron monitoring for storage or repository sites	PREDIS MICADO
	Timepix network	CTU	Gamma and neutron monitoring system	MICADO
Data assessment	Uncertainties analysis	SCK-CEN	Monte Carlo error propagation, including pre- and post-processing of measurement data and results, tailored to radiological characterization	MICADO
Muon tomography		UNIBRIS INFN	Internal structure of drum	CHANCE PREDIS
Ultrasonic echo (high frequency, metal)		BAM,>NNL	Container Wall: thickness, internal, corrosion, cracks	PREDIS
Ultrasonic echo (low frequency, concrete)		BAM	Container fill: structure, objects, cracks	PREDIS
RFID Monitoring inside of the package, p, T, H		BAM	Tracking cement hardening, monitoring fill evolution	PREDIS
Acoustic emission analysis (AE)		MAGICS	Changes in concrete fill, crack development, stiffness	PREDIS
RFID solid state gamma and neutron sensors for long term monitoring		UNIPIISA	Gamma and neutron monitoring for waste packages	PREDIS

Fig. 1. Technologies investigated in the CHANCE, MICADO, and PREDIS projects.

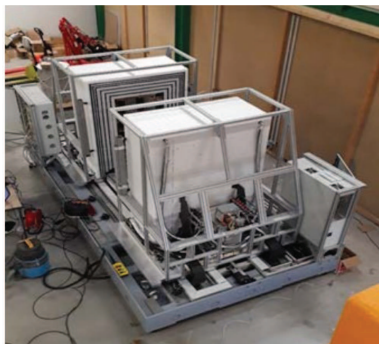


Fig. 2. The CHANCE calorimeter at the SCK–CEN being tested with real waste packages.

established a characterization procedure, data analysis, and information storage suitable for different types of waste activities (VLLW, LLW, ILW, legacy waste), matrix types (metallic, organic, and concrete fillings), and drum dimensions, including unconditioned waste but excluding high activity level waste (HLW) packages.

The infrastructure for data collection and visualization of the data provided by each technology represents another innovative approach in addition to the newly developed technologies. The RCMS software infrastructure can collect, store, display all data and implement advanced algorithms (such as Monte Carlo calculations, machine learning, or Bayesian inference) for error propagation analysis. This software allows a new approach to data management and more secure data processing. These aspects and the mobility of the detection devices are included in the list of topics prepared by CHANCE in Deliverable D2.3. These topics demonstrate the importance and complementarity of the projects, which provide continuity in the nuclear waste characterization field.

The third project PREDIS (Pre-disposal management of radioactive waste) started in 2020 and will run for 4 years. The PREDIS project aims to develop and increase the technological readiness level (TRL) of treatment and conditioning methodologies for wastes for which no adequate or industrially mature solutions are currently available. PREDIS project is also developing innovations in cemented waste handling and pre-disposal storage by testing and evaluating them. PREDIS will develop decision-making tools regarding the added value of the developed technologies and their impact on the design, safety, and economics of waste management and disposal. PREDIS encompasses the wider European Community allowing cross-fertilization and interaction between different national programs. The main objectives of the project are to develop solutions (methods, processes, technologies, and demonstrators) for future treatment and conditioning of waste, to improve existing solutions with safer, cheaper, or more effective alternative processes where they bring measurable benefits, and to analyze criteria, parameters, and specifications for materials and packages with associated waste acceptance criteria (WAC) for pre-disposal and disposal activities to support homogenization of waste management processes.

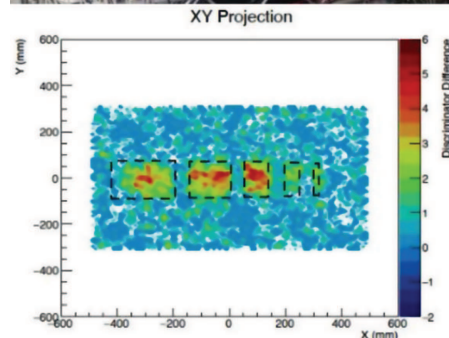
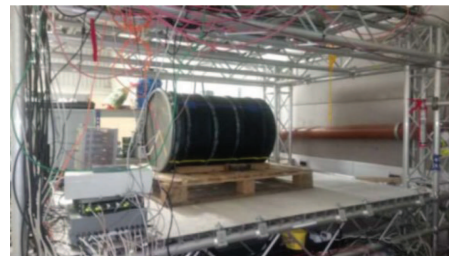


Fig. 3. Mock-up drum characterization for the CHANCE project and reconstructed data.

7 The investigated technologies

Calorimetry [14] technique is used as a comprising non-destructive technique to reduce uncertainties in the inventory of radioactive waste containing shielded and hidden material difficult to measure by other means. Calorimetry is a method for the determination of nuclear material masses (or activities) by the quantitative measurement of the heat flux emerging from a sample. Calorimetry is used for NDA, which means that no sample needs to be tempered or destroyed by the analysis. In combination with gamma spectroscopy and neutron measurements, calorimetry is well suited for multi-nuclide assay of conditioned waste drums with large volumes and possibly heterogeneous contents and for reducing uncertainty assessment. Figure 2 shows the CHANCE calorimeter prototype being tested with real waste packages. It is designed to measure 200-l drums emitting heat in the range of 10–3000 mW and has already been tested with mockup drums containing Pu pellets in the center of a concrete matrix. A value of 99.7 ± 16.4 mW was measured and compared to the expected value of 110 ± 6 mW from simulations. Increasing the power emission showed better accuracy than expected.

Muon tomography is an NDA technique with a long exposure time (days/weeks) that can produce a full 3D image of a volume of interest. It allows viewing individual objects inside the drum and obtaining information on their atomic number Z and density. The CHANCE system (Fig. 3) is a device tested in a real environment that consists of two detection technologies: drift chambers and resistive plate chambers (RPCs). Data in combination with multivariate analysis (MVA) classifiers and clustering algorithms allow us to approximately identify the locations and shapes of objects stored in a concrete-filled waste drum. Once the categorized data is trained

in terms of different materials (lead, iron, etc.), machine learning techniques are applied to the reconstructed data to identify the unknown material inside the package [13]. The possibility of identifying bubbles of 5 cm in 4 weeks of measurement time was also tested. Finally, Monte Carlo simulations were used to evaluate the possibility of using this technique for large CASTOR drums with weeks-long measurements to determine the presence of materials or the absence of materials due to unauthorized material removal. The same type of measurement is performed with the prototype developed by INFN for the PREDIS collaboration. Within this project, the prototype is made of 2 layers of drift chambers (Fig. 4). The main aim is to validate the properties of concrete waste packaging including density analysis. At this stage of the project, an initial test was conducted to detect iron and other objects inside a concrete block.

Cavity ring-down spectroscopy (CRDS) is a transportable instrument for in situ airborne radiocarbon detection. It operates in the mid-infrared range. Within the CHANCE project, the system demonstrated the feasibility of the method to measure the H^{36}Cl and the application of the technique to monitor the outgassing of ^{14}C waste, showing good repeatability and accuracy. Figure 5 shows a sample line developed to be connected to the CRDS prototype for the measurement of ^{14}C outgassing.

The waste management procedure was devised to characterize the different waste packages in order to minimize the measurement time of each step and be able to select the required detection technology based on previous measurement results. This procedure avoids multiple identical measurements and optimizes the process. An example of a predefined procedure is represented by the block scheme in Figure 6. This is the decision-making process developed for the MICADO gamma station, in which the data provided by two detection systems drives the choice of the spectrometric system.

The gamma station procedure is used for gamma characterization as one of the first and mandatory analyses for all waste packages. The MICADO station is not a single detection technology, but a procedural integration of three technologies. It is the first measurement station to pass through the waste package. The RadhHAND system [15] is used for contact, dosimetry, and spectroscopic measurements in open geometry. The Nanopix [16] is a gamma imaging system for the localization and identification of hot spots in open geometry. The SEA radioactive waste gamma analyzer (SRWGA) [17] is a tomographic segmented gamma scanner performing spectrometry and tomographic measurements of the package.

Although gamma spectrometry and tomography are considered traditional techniques, the main innovation is the procedural integration of the three techniques. The procedure consists of four steps (see Fig. 6): identification, screening, preliminary, and characterization phases. The identification phase starts with a UHF-RFID tag used to tag the waste package, providing a unique identifier, and associating its EPC number to the entry in the database identifying the package and its characteristics. During the inspection phase, an initial dose rate at contact is taken for safety reasons and radiation protection activ-

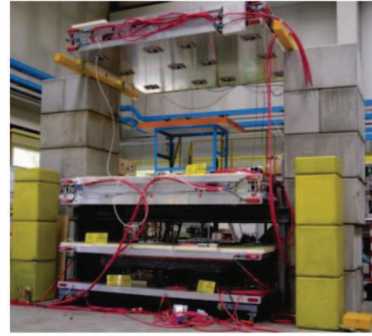


Fig. 4. Muon tomography system of the PREDIS project.



Fig. 5. New sampling line for use with ^{14}C CRDS prototype.

ities. The preliminary characterization phase consists of measurements of radiological homogeneity/inhomogeneity of the contents, identification of higher intensity radionuclides, acquisition of package picture recordings using the Nanopix and RadHAND systems, and identification of the tag on the package. During characterization, SRWGA will complete the gamma characterization phase. The SRWGA measurement type will be decided based on the results of the preliminary phase helping to select the best measurement technique for a complete package characterization. The result provided by the gamma station is used to determine the second step of the characterization process and select the measurement to be performed (neutron or photofission measurements).

The neutron station presented in Figure 7 combines passive neutron coincidence counting and active neutron interrogation techniques for waste packages containing nuclear material. Packages with fissile or fertile material, but also large metallic waste packages, which are difficult to characterize by gamma spectroscopy, come to this station. The structure is mobile and can process packages of different sizes up to 220l. Moreover, this technique is combined with data analysis based on matrix characteristics (material, density, filling) using artificial intelligence with trained data (based on many MCNP simulations) to apply signal attenuation corrections (also called “matrix effect” corrections) based on internal monitors and neutron transmission measurements to characterize the matrix. Monte Carlo simulations [18] were performed to optimize the design. In order to achieve a good compromise between the performances in passive mode and active

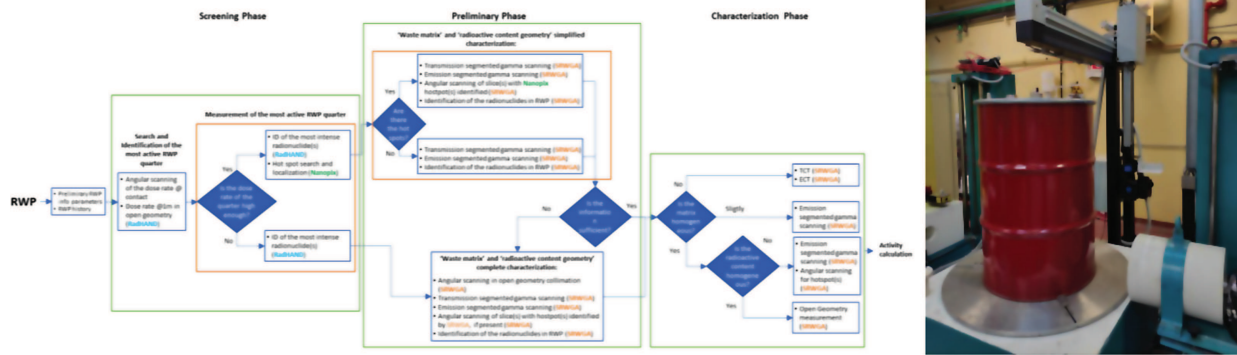


Fig. 6. Gamma station procedure on the left and the integration of the RadHAND sensor with the SRWGA on the right.

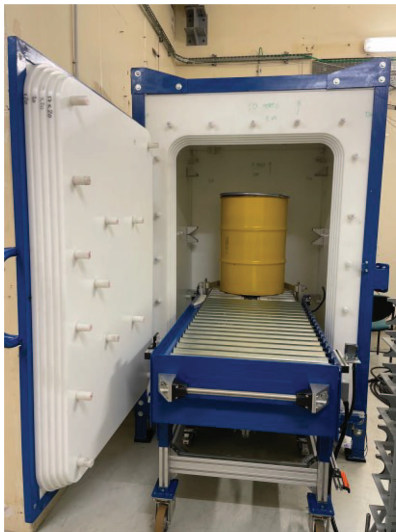


Fig. 7. Neutron detection system installed at CEA Cadarache.

interrogation mode, the differential die-away technique was used. Since the latter strongly depends on the characteristics of the nuclear waste drums to be measured, the matrix effect correction is mandatory to limit the measurement uncertainty.

The photofission station is used to characterize large volumes of packages with concrete or polyethylene matrices, which are considered difficult to characterize with both passive and active neutron measurements. Within MICADO, the possibility of using a mobile photofission system based on “low energy” X-rays (6 MeV) has been studied to minimize the hazard and safety shielding usually required for this technique. To provide this information, simulations and laboratory tests are performed to test two-photon beam energies, 7 MeV and 9 MeV. It is common practice to use 9 MeV. However, the ability to perform a package characterization using a 7 MeV beam as well will facilitate the use of a mobile system. This will require a more flexible shielding structure that is transportable or can be easily prepared on-site to ensure safety procedures.

Pipeline data assessment: the data analysis pipeline (DAP) is not a detection technology for char-

acterizing a waste package, but rather a software infrastructure capable of propagating the uncertainties related to the individual techniques and combining the results of the individual techniques to reduce the global uncertainty of the final inventory. One of the main goals is to better determine the levels of radioactivity to optimize the waste package classification. This concerns in particular radioactive waste packages whose values approach the limits for surface storage or storage in a geological repository. This software infrastructure uses Monte Carlo particle transport (MCPT) simulations [19] to evaluate the sources of uncertainty in all detection technologies involved in the MICADO project. An analysis of the feedback mechanisms between different measurements enables the results integration and provides the overall uncertainty based on a full Bayesian uncertainty quantification using a probabilistic programming language (PPL) for Markov chain Monte Carlo (MCMC) sampling. The combined data analysis fuses different measurement results to extract information that is not available in the individual systems and to reduce individual uncertainties. This reduction is a solution to the problem of properly categorizing complex waste packages, for release or more accurate disposal.

Data digitalization: all MICADO technologies, software, and hardware provide data that is saved in a common software framework. This framework stores, processes, and displays the information required during and after the characterization of an RWP. A key element is the RCMS database, from which the organized collection of stored data can be accessed by multiple computers and operators. Using blockchain technology this data is securely stored to ensure the reliability of the data content.

Small and affordable gamma and neutron sensors are developed under MICADO and upgraded under PREDIS with a focus on electronics. Unlike the other projects, the main aim is long-term monitoring of waste packages (e.g., in intermediate storage facilities). There are two types of sensors, scintillating fibers for gamma detection (SciFi) [20] and a solid-state detector with lithium (SiLiF) for the detection of neutrons [21]. The sensors are coupled either directly to the package or the surrounding framework and provide continuous trends of neutron and gamma count

rates to monitor the status of the stored package in real-time. Wireless communication with a prototype data collection platform is studied in PREDIS.

Internal monitoring RFID devices to be integrated into new waste packages are developed in PREDIS to wirelessly deliver data on temperature, humidity, pressure, and other parameters.

Ultrasonic techniques [22,23] are used and developed in several variants in PREDIS to verify the physical state of the packages. The high-frequency ultrasonic echo technique is used to measure the thickness of the outer metallic container wall and to detect and monitor corrosion and cracks on the metallic container wall. A low-frequency variant can be used to inspect and monitor the concrete fill and to detect voids, objects, and cracks in the cemented matrix and potentially in the wall of concrete containers.

Acoustic emission: in this technique, sensors are placed on the container wall to detect events that generate elastic waves (“sounds”), e.g., cracking, crack extension, or internal friction. TAE can be used to monitor concrete hardening or any change in the cemented matrix of RWP due to degradation processes.

Machine learning algorithms are used in PREDIS to provide a fast and accurate prediction of RWP geochemical evolution and integrity, continuously updated by monitoring data (digital twins).

8 Treatment and conditioning of the waste

The only project that focuses on technologies for the physical treatment and conditioning of waste is PREDIS. The project investigates technologies for the treatment and conditioning of radioactive wastes, such as metallic and radioactive organic wastes, whether liquid (RLOW) or solid (RSOW). The focus is on steel and Ni-alloys radioactive metallic wastes, which are major components in nuclear installations. These metallic wastes are often surface contaminated, in the form of corrosion layers of a few tens of micrometers retaining radionuclides, including but not limited to activation corrosion products (^{60}Co , ^{63}Ni , ^{55}Fe) and fission products such as ^{137}Cs . Treatment of surface-contaminated metallic wastes includes chemical decontamination using chemical solutions for complex geometries (pipes) and gels for simple geometries such as flat surfaces and walls. Optimization of chemical formulations is being conducted and includes several parameters such as the reactants concentrations, treatment cycles, and temperature. Optimization of the gel-based treatment process will investigate the adherence capacity and the dissolution of the contaminated metal surface.

The PREDIS project also develops innovative solutions for the direct conditioning of RLOW as well as reactive metallic waste such as Al and Be. Hence, three geopolymers (based on blast furnace slags, metakaolin, and mixtures) have been selected to study the encapsulation of different types of RLOW, such as oils, organic solvents, scintillation cocktails, and decontamination liquids. Due to the nature of the RSOW (e.g., ion exchange resins, cemented and polymerized/bituminized wastes) and their

current incompatibility with long-term waste management solutions, the physicochemical characteristics need to be modified. Within PREDIS, several treatments are investigated, promoting thermal processes. Subsequently, the treated wastes are embedded using similar types of geopolymers, as well as cementitious materials or glass coatings, but also densification processes (pelletization or hot isostatic press). Regardless of the type of waste studied, synthesis procedures are currently being finalized. Testing of these materials will be conducted by studying the matrix’s mechanical performance, durability under various conditions (acid and alkaline waters, endogenous conditions, and air), chemical durability, and resistance to radiation. Magnesium phosphate cements (MPC) are studied as the best conditioning solution for the encapsulation of reactive metallic wastes. They allow high protection of metallic waste from oxidation leading to hydrogen production. MPC optimization mainly includes raw material selection and cost reduction. The tested parameters include mechanical and chemical durability as well as resistance to ionizing radiation.

9 Conclusions

This paper describes the innovations coming from the three Euratom projects CHANCE, MICADO, and PREDIS for the characterization of RWP. The technologies developed or under development are described, but also the procedures and statistical approaches used to improve RWP qualification and its content. Better determination of the radiological content and physicochemical status of RWP are the key points for better handling and storage of the packages. Improving the current characterization results will not only help stakeholders and end users to reduce costs and operational work but will also help future generations and countries that will approach the process in the near future with more confident and reliable procedures.

Conflict of interests

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

Acknowledgements

The authors appreciate the opportunity to share and improve our ideas, made possible by grants from the European Commission. We would also like to thank our colleagues from the CHANCE, MICADO, and PREDIS projects for their support in the respective projects and in writing this paper.

Funding

The CHANCE project falls within the NFRP 7-2016–2017 topic “Research and innovation on the overall management of radioactive waste other than geological disposal” of the H2020 Euratom Work Programme 2016–2017. The MICADO project has received funding from the European Union’s Horizon 2020 research and innovation program under grant agreement No. 847641. The PREDIS project

has received funding from the Euratom Research and Training Programme 2019–2020 under grant agreement No. 945098. The work presented in this paper reflects only the author’s views. The Commission is not liable for any use that may be made of the information contained therein.

Data availability statement

No data were generated and/or analyzed for this article that cannot be disclosed for legal, ethical, or other reasons.

Author contribution statement

All the authors of this text are partners in, at least, one of the three projects described. Morichi, Fanchini, Breuil, Perot, and Bruggeman are the main contributors to the MICADO project description, for CHANCE we have Ricard and Perot and finally Niederleithinger, Bruggeman and Breuil have written the PREDIS part.

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Cite this article as: Massimo Morichi, Erica Fanchini, Eric Breuil, Christophe Bruggeman, Bertrand Perot, Denise Ricard, and Ernst Niederleithinger. Developments and experiences of the CHANCE, MICADO and PREDIS projects in radioactive waste characterization, EPJ Nuclear Sci. Technol. **9**, 12 (2023)