CHANCE project

(**Contract Number: 755371**)

TOPICAL DAY ON WASTE CHARACTERIZATION

DELIVERABLE (D6.4)
Work Package 6

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CHANCE

D6.4 Topical day on waste characterization

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Safe interim storage and final disposal of radioactive waste (RW) requires effective characterization and quality control of the waste. The CHANCE project therefore aims to address the specific and complex issue of the characterization of conditioned radioactive waste (CRW) by means of non-destructive analytical (NDA) techniques and methodologies. Characterization issues within CHANCE encompass both physico-chemical characterization and radiological characterization.

The CHANCE project will establish a comprehensive understanding of current characterization methods and quality control schemes for conditioned radioactive waste in Europe. Furthermore, CHANCE will develop, test and validate already-identified and novel techniques that will undoubtedly improve the characterization of CRW. These techniques include:

- Calorimetry as an innovative non-destructive technique to reduce uncertainties on the inventory of radionuclides;
- Muon Tomography to address the specific issue of non-destructive control of the content of large volume nuclear waste;
- Cavity Ring-Down Spectroscopy (CRDS) as an innovative technique to characterize outgassing of radioactive waste.

During the CHANCE Topical Day current needs, scientific evolutions and CHANCE highlights concerning conditioned waste characterization using non-destructive techniques were highlighted. The Topical Day took place on Thursday, March 21st 2019 at the Lamot Conference Center in Mechelen (Belgium) and was organised by the SCK•CEN Academy. The selected speakers have either operational experience with conditioned waste characterization, or are scientists working on new characterization techniques. This Topical Day therefore also aimed at bringing together these two groups of stakeholders. The book of abstracts of the Topical Day is presented in annex.

The Topical Day gathered a lot of interest from both research, industry, waste management organisations, waste facility operators and institutional/governmental bodies. A total of 102 persons participated to the Topical Day at the Lamot Conference Center. Apart from that, the Topical Day was also live-streamed. A total of 141 different devices logged onto the live-stream during the day, with a maximum peak audience of 42 different devices in the morning session and 33 devices in the afternoon session.

All the Topical Day participants afterwards received a link through email to the different transactions (encrypted pdf files). The video files from the live-streaming are also available upon request.

On Friday morning, March 22nd, a visit to the CILVA installation and AQ2 gammaspectrometry installation of Belgoprocess (Dessel, Belgium) was organised. Belgoprocess is the nuclear operator responsible for treating, converting into a stable end product, and storing the historical and current Belgian nuclear waste from numerous producers at its site in Dessel. The company has experience processing almost every type of low-, medium- and high-level radioactive waste at its sites, thereby covering a large segment of the back-end of the nuclear fuel cycle. Such processes lead inevitably to the generation of a number of different waste streams. Each of these streams is uniquely defined by its radiological and physicochemical characteristics. To select appropriate processing and conditioning techniques and disposal options, it is essential to characterize, sample and measure each of these waste streams in an economical manner while meeting all safety, operational and regulatory requirements.

During the technical tour, participants visited the following installations/

- CILVA-installation for overall treatment and conditioning of low-level waste, consisting of the following modules:
  - low-level waste incinerator;

- supercompactor;
- cementation unit;
- cutting and repackaging unit (inl. visual inspections, sampling, …)
- AQ²-installation (NDA segmented gammaspectrometry) for characterization of low-level waste (and free release).

Topical Day CHANCE | characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot conference centre, Mechelen, Belgium

Preliminary programme

08:30 – 09:00 h	Registration and welcome coffee

Opening

09:00 – 09:10 h	Welcome
	Hildegarde Vandenhove, Institute Manager Environment, Health & Safety, SCK•CEN, Belgium

09:10 – 09:30 h	Introduction to the CHANCE project
	Denise Ricard, ANDRA, France – CHANCE coordinator

Chair: Christophe BRUGGEMAN, SCK•CEN, Belgium

09:30 – 10:00 h	Radioactive waste characterization practices and gaps in Europe
	Crina Bucur, Raten, Romania

10:00 – 10:30 h	Waste characterisation in practice – lessons learned from operational experience
	Robin Tuerlinckx, Belgoprocess, Belgium & Tomas Vandoorne, NIRAS/ONDRAF, Belgium

10:30 – 11:00 h	Break

11:00 – 11:30 h	Waste acceptance and characterization in an operational waste disposal facility
	John Muller, ANDRA, France

11:30 – 12:00 h	Waste characterisation of conditioned waste in Sweden
	Claes Johansson, SKB, Sweden

12:00 – 12:30 h	Recent developments in calorimetry for nuclear wastes characterization
	Christophe Mathonat, KEP Technologies, France

12:30 – 13:45 h	Lunch + poster boards for CHANCE PhD students

Chair: Denise RICARD, ANDRA, France

13:45 – 14:15 h	Muon tomography | needs and challenges from a safeguards perspective
	Darius Ancius, EC Euratom and Paolo Checchia, INFN, Italy

14:15 – 14:45 h	Muon tomography | recent developments and CHANCE
	Jaap Velthuis, University of Bristol, UK

14:45 – 15:15 h	LINAC-based high energy X-ray imaging of radioactive waste packages at CEA, DEN, Cadarache
	Bertrand Pérot, CEA, France

15:15 – 15:45 h	Break

15:45 – 16:15 h	Novel optical methods for the characterisation of outgassing from radioactive waste
	Guillaume Genoud, VTT, Finland

16:15 – 16:45 h	Modelling of the CHANCE calorimeter and some general thoughts about NDA uncertainty quantification
	Wojciech Kubinski, WUT, Poland and Bart Rogiers, SCK•CEN, Belgium

16:45 – 17:00 h	Closing
	Denise Ricard, ANDRA, France

17:00 – 19:00 h	Reception
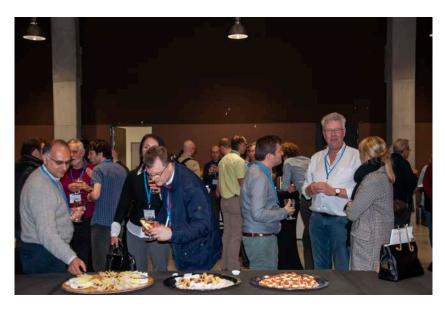
Book of abstracts

Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019

Book of abstracts

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Book of abstracts | BA-118 | SCK•CEN 32770025   Page 11 of 20
   1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Introduction to the CHANCE project

Denise Ricard
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Abstract

Context and objectives of the project
The CHANCE project aims to address the specific issue of the characterization of conditioned radioactive waste (CRW). The characterization of fully or partly conditioned radioactive waste is a specific issue because, unlike for raw waste, its characterization is more complex and needs specific non-destructive techniques and methodologies. There are different and varying reasons for this: 1) conditioned waste may no longer be in its initial form (e.g., due to incineration), 2) conditioned waste is typically embedded or surrounded by a matrix, 3) conditioned waste may contain wastes coming from different primary sources and therefore the radiological spectrum might become more complex.

CHANCE will focus on conditioned waste of the following activity levels:
- Very Low Level Waste (VLLW);
- Low Level Waste (LLW);
- Intermediate Level Waste (ILW);
- High Level Waste (HLW).

The first objective of CHANCE is to establish at the European level a comprehensive understanding of current conditioned radioactive waste characterization and quality control schemes across the variety of different national radioactive waste management programmes, based on inputs from end-users such as Waste Management Organizations and storage operators.
The second objective of CHANCE is to further develop, test and validate techniques already identified that will improve the characterization of conditioned radioactive waste, namely those that cannot easily be dealt with using conventional methods.

To address these specific issues, the CHANCE project proposes R&D actions to develop three innovative technologies for conditioned radioactive waste packages, namely:
- Calorimetry as an innovative non-destructive technique to reduce uncertainties on the inventory of radionuclides;
- Muon Tomography to address the specific issue of non-destructive control of the content of large volume nuclear waste;
- Cavity Ring-Down Spectroscopy (CRDS) as an innovative technique to characterize outgassing of radioactive waste.

1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

The activities performed and the results obtained within the project CHANCE are integrated, communicated and disseminated both between the project partners as well as with the broader European community involved in radioactive waste disposal. Dissemination of results and progress is an integral part of the CHANCE project. Generally stated, the goal of communication is to raise awareness concerning the CHANCE research topics and developments, and to inform the CHANCE stakeholders about the project's progress and achievements. The main communication tool to reach this goal is CHANCE website http://www.chance-h2020.eu/.

The final ambition of CHANCE is to improve the efficiency of the characterization of conditioned radioactive waste and therefore to improve the safety of the global radioactive waste management process throughout the full storage cycle, including transport, interim storage and final disposal operational and long term stability.

Project Overview
CHANCE is structured into six work packages:
- Management and coordination (WP1)
- Methodology for conditioned radioactive waste characterization: Problematic wastes and R&D proposal (WP2)
- Calorimetry associated with non-destructive assay techniques and uncertainties study (WP3)
- Muon imaging for innovative tomography of large volume and heterogeneous cemented waste packages (WP4)
- Innovative gas and outgassing analysis and monitoring (WP5)
- Dissemination activities (WP6)

WP1 is dedicated to the management and coordination of the project.

WP2 is dedicated to the establishment at the European level of a comprehensive understanding of current conditioned radioactive waste characterization and quality control schemes across the variety of different national radioactive waste management programmes, based on inputs from end-users such as Waste Management Organisations and storage operators.

WP3 objective is to complement existing, widely-used techniques for characterization of waste packages (gamma spectrometry and passive neutron measurement) with an exhaustive study of uncertainties related to such characterization methods in conjunction with calorimetry.

WP4 objective is to develop mobile muon tomography instrumentation to address the as-yet unsolved problem of the non-destructive assay (NDA) of large volume nuclear waste packages, such as large spent fuel casks and large concrete waste packages with heterogeneous waste.

WP5 will advance the use of Cavity Ring-Down Spectroscopy (CRDS) as an innovative technique to characterize outgassing of radioactive waste by developing new instrumentation for $H^{36}Cl$ and demonstrating an application of the technique to the monitoring of radiocarbon outgassing.

WP6 is dedicated to dissemination and training activities.

   1.   **Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments**

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Acknowledgment

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1.   **Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments**

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Radioactive waste characterisation: practices and gaps in Europe

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Abstract

The CHANCE project "Characterization of conditioned nuclear waste for its safe disposal in Europe" aims to address the specific issue of the characterization of conditioned radioactive waste in Europe. A questionnaire was elaborated by CHANCE Work Package 2 & 6 to obtain a broad overview on the end-users needs for the characterisation of conditioned radioactive waste and to appreciate how the members of End User Group (EUG) perceive the activity of waste characterization in relationship with the evolution of the national disposal programme, in terms of importance with respect to the whole back-end of the fuel cycle, or in terms of uncertainty management within each organization.

The answers collected from the CHANCE EUG members were used to identify:
- key parameters that need characterization;
- technologies/methods commonly used for characterization of conditioned waste;
- waste acceptance criteria applied and the possibilities of their harmonization in Europe
- specific problematic issues for the characterization of conditioned radioactive waste;
- R&D needs and potential on-going R&D programme on the topic of conditioned radioactive waste characterization;
- potential applications of R&D actions to be included in CHANCE;
- socio-technical and ethical issued associated with the waste characterization process

The methods used for characterisation of conditioned radioactive waste depend on the country approach. Spectroscopic techniques (alpha, beta and gamma) as well as neutron measurements and dose rate calculation are used in most European countries, but in some organizations, more advanced techniques such as muon tomography and Calorimetry are also applied in the characterisation process.

Despite the use of various conditioning technologies and matrices, there are common difficulties related to the characterization process of the conditioned waste packages. Characterization of the conditioned historical waste packages seems to represent a generalized problem. Radiologic characterisation of these waste packages represents a big challenge either with passive or active measurements because of the difficulty of the interrogators radiation to penetrate the waste matrix and of the strong attenuation of the measurable emissions (gamma or neutron) by the waste package itself.

1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Waste characterization in relation to the WAC: approach and challenges ahead

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Abstract

In Belgium, ONDRAF/NIRAS (National Agency for Radioactive Waste and Enriched Fissile Materials) is responsible for the long-term management of radioactive waste. Belgoprocess, as the industrial subsidiary of ONDRAF/NIRAS, is entrusted with the operational waste management. A very important aspect in this overall strategy are the waste acceptance criteria (WAC) and the characterization which is eventually performed in the assessment of these WAC.
Currently ONDRAF/NIRAS is in the final stages of obtaining the license for the near surface disposal site (cAt), which is expected to be operational in 2024. During the last years, a lot of effort has been made considering the final set of WAC. These final WAC have a great impact in the way characterization is to be performed, considering both radiological and chemical content. Some challenges were identified to tackle this need for detailed (extra) characterization information.
This presentation gives an overview of these criteria and the way that these needs can be fulfilled in the overall characterization program. Another important aspect is the control strategy, based on qualification and a control mechanism.

Book of abstracts | BA-118 | SCK•CEN 32770025   Page 16 of 20
1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Waste acceptance and characterization in an operational waste disposal facility

John Muller, Denise Ricard
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Abstract

The Centre de Stockage de l'Aube (CSA) is a basic nuclear installation designed to dispose Low and Intermediate Level Waste Short-Lived Waste (LILW-SL). The industrial operating of CSA started in 1992 and it is planned to operate this disposal facility for about 70 years (the operating period will be followed by a 300-year monitoring phase). The disposal capacity of the CSA is 1 000 000 m^3 which will correspond approximately to the filling of 400 disposal vaults (today about 140 vaults are filled).

Waste acceptance processes

The overall structure of the waste acceptance process includes the following phases:
- Approval (of a waste stream);
- Authorization (of a site, for the approved waste stream);
- Acceptance (of a waste production of an approved waste stream produced on an authorized site).

A document, called Compliance Matrix (MC) is gradually filled up during the process, and each stage of completion of the document is associated with a phase of the acceptance process.

Once the initial approval and acceptance delivered by Andra, the waste producer must define and perform a surveillance program to identify all the risks during the production of waste packages that could lead to produce and ship waste packages non-compliant with waste acceptance criteria of Andra. The surveillance process is defined and validated during the approval process.

Andra, as a part of the requirements of its license to operate the Low and Intermediate Level Waste Short-Lived Waste (LILW-SL) facility, verifies that the waste producers' capability to produce waste packages are compliant with the approval and authorizations delivered.

The surveillance program includes:
- Destructive control on waste packages at the waste producer's site and at the CSA after delivery;
- Non-destructive control on waste packages at the waste producer's site and at the CSA after delivery;
- Surveillance missions at the waste producer's site;
- Controls on the waste package information sent prior to delivery at the CSA;
- Controls performed upon delivery at the CSA (visual, radiological controls…).

Monitoring of waste package quality by Andra

Surveillance is a mission assigned by the terms of the disposal facility-operating license. Andra must periodically verify the actual and correct application by the producer of the technical and organizational conditions on which the authorization was notified. For this purpose and in addition to computerized verification checks and checks carried out at the reception of waste package at the CSA, Andra must carries out in its Annual Monitoring Program, which includes:

1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

- Destructive and non-destructive testing of packages carried out on delivery to the CSA. Destructive and non-destructive sampling checks are performed by Andra or contracted laboratories under the responsibility of Andra to ensure the compliance of the package (container, packaging, conditioning, chemical and physical nature of waste, evaluation of activity…).

  These controls are planned by Andra as part of the implementation of its Annual Monitoring Program. During these checks, Andra can detect non-compliances but also slightly abnormal situations (also called weak signals), which, although compliant, will trigger the attention of Andra.
- Surveillance missions at the production facilities. Andra carries out surveillance missions at the production sites to ensure the effective and correct application by the producer of the technical and organizational conditions on the basis of which approval and authorization have been notified. These missions can take the form of process audit, technical visits and participation in internal surveillance activities.

This monitoring is based on a 3 years planned quality-control inspections (destructive or non-destructive):
- To sample and control waste packages received at CSA integrating:
  - A minimal 3 years period for each package family;
  - A systematic control for new agreements and new delivery of suspended agreements;
  - Experience feedback from quality control.
- To check the consistency between results and declared properties for measurable parameters;
- To ensure of the compliance of corresponding parameters with specifications;
- To acquire knowledge.

These quality control inspections applies the same rules and methods as those defined in the specifications, technical tests and standards.

Focus on non-destructive controls

Non-destructive tests carried out on the packages delivered to the CSA aim at carrying out the verification of the characteristics of the waste packages declared by the producer by comparing them with the values measured by Andra and those contained in the acceptance folder.

In addition to a detailed review of the package file sent by the producer, non-destructive controls include visual and radiological controls (non-surface contamination and dose equivalent rate mapping), weighing and dimensional readings for all waste packages collected are performed. Gamma spectrometry measurements are also carry out for a large majority of waste packages (only cubic containers of 5 and 10 m^3 to be inject at the CSA are not measured in gamma spectrometry).

These controls, carried out on the CSA, can be supplemented by non-destructive tests carried out by external laboratories. These controls consist mainly of high-energy X-ray imaging (radiography and tomography), low-energy x-rays, but also passive and active neutron measurements coupled with gamma spectrometry measurements.

Depending on the nature of waste package, additional controls can be done to verify the internal characteristics of the packages (integrity, quality of the waste block, internal constitution of the packages) and also to reinforce the results of the activity measurements carried out at the CSA with others methods.

Note

In the near future, Andra will have a facility (called package control facility) on the CSA that will be able to perform on drums:
- Tritium and carbon 14 degassing measurements;

High energy X-ray imaging

1.   **Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments**

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Waste characterisation of conditioned waste in Sweden

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Abstract

Nuclear facilities and installations in Sweden consist of both nuclear power plants in operation (8) and plants (4) under decommissioning. In the next coming years two more NPP:s will be shut down.

SKB – Swedish Nuclear fuel and Waste Management was established of the owners of the NPP:s to take care of both the spent fuel after operation and the low and intermediate level waste from the NPP:s and also from the Studsvik nuclear site and some other producers in Sweden of radioactive waste.

Since 1988 SKB have a final repository SFR in operation at Forsmark that takes care of mainly short lived radioactive waste. Limitations due to the existing licence for receiving activity are 10E16 Bq in total. Most of the waste is transported by a special designed ship for radioactive waste from the nuclear facilities to SFR.

For the SFR facility there are existing waste acceptance criteria that must be fulfilled to be able to dispose of the waste. The waste is either examined before conditioning of the waste and/or after it is conditioned.

The normal waste characterisation methods for examining the conditioned waste consist of gamma dose rate measurements and gamma spectrometry measurements of the waste packages. The waste packages could be of different kind of geometries.

For old legacy waste that exist with very poor waste characterisation done, a special project has been performed at SKB:s sister company SVAFO at the Studsvik site. At this project (X-ray project) about 7000 drums was examined with x-ray examination in order to identify liquids, and other type of waste in this drums. At the same time also gamma specific measurements where performed as a quick scan for about 15 minutes each of the drums.

Book of abstracts | BA-118 | SCK•CEN 32770025   Page 19 of 20
1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Recent developments in calorimetry for nuclear wastes characterization

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Abstract

Calorimetry is a non-destructive method of quantitative measuring the heat emission to determine the masses or activities of nuclear material hidden inside nuclear waste compounds. Heat-flow calorimeters are in common use for measuring thermal powers over a wide range of 0.5mW (equivalent to 0.2g of low-burnup plutonium) up to 1kW and for samples of various sizes of about a few cm³ up to large waste drums of 60cm wide and 100cm long 0.

Calorimetry is measuring the overall thermal power due to the radioactive decay coming from the waste contained in a metallic drum or a different container. It has many advantages as it features a non-destructive method which remains independent of matrix effect or the chemical composition.

For measuring a multi-nuclide drum calorimetric assay is often combined with gamma ray and neutron measurements. Calorimetric determination of tritium and plutonium activities or masses has become common practice. However, the determination of beta-emitters (e.g. Sr/Y-90) or shielded sources inside a concrete filled waste compound is yet challenging for the comprehensive nuclide characterization of large heterogeneous waste drums.

In the context of the CHANCE work-package 3 0, 0, a benchmark of calorimeters and standard NDA methods for the characterization of large volume waste drums 0 have been undertaken and a study of the neutron and gamma ray signals that can escape various 200L waste drum matrices with different source configurations has been performed. A 200 liters calorimeter is under construction with an optimized detection limit and to reduce the methodological uncertainties of the inventory determination in a multi-nuclide matrix, the escaping radiation are quantized as much as the encapsulated gamma-rays and α-, β-, n- particles depositing radiological heat inside the drum. Monte Carlo simulations are utilized resembling the waste drum and the calorimeter in both, their geometries as well as the material compositions. For the analyses, the particle flux and energy deposition in each layer of the calorimeter are displayed. The results yield that a significant part of the emitted radiation leaves the system and does not contribute to the heat deposited inside the drum. Therefore additional layers of gamma and neutron absorber are contemplated and associated numerical studies are initiated.

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1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

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1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Muon tomography: needs and challenges from safeguards perspectives

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Abstract

European Union strategies related to Energy Union and Energy Security underline the importance of highest standards of safety, security, waste management and non-proliferation. It implies obligations for safe and responsible long-term spent fuel and waste management.

Raising number of spent fuel assembly loadings to Dry Storage Containers (DSC), the construction of new type facilities like Encapsulation plants and Geological repositories becomes serious challenge for EURATOM and IAEA. The proven traditional approaches based on maintenance of Continuity of Knowledge (CoK) of safeguarded materials are relying on continuous video surveillance and sealing. It requires considerable human resources. Moreover, given the big number of DSC (some thousands in EU in next decade) and interim storage for about 50 years, the cases with the loss of CoK are highly probable. Such cases would necessitate the verification that all spent fuel assemblies are present in DSC. However today no NDA technique can prove it and only way to confirm this is to unload the DSC in a water pool or a hot cell.

Cosmic muons penetrate metres of dense material and therefore can be used to image the contents of spent fuel held in DSC. Recent development of innovative imaging techniques using cosmic muons and muon trackers offers unique opportunities for safeguards. The National Institute of Nuclear Physics (INFN) in Italy and EURATOM tested the 8 x 8 drift-tube detector at proximity of CASTOR® container in Germany. The tests have convincingly proved that the detection of the muons that passed through the container is possible. The noise created by gamma and neutron radiation from spent fuel stored in this container represented only about 1% of useful signal. These experimental results justify other investigations related to the use of the muon trackers for spent fuel safeguards. Further tests with larger scale muon trackers are under consideration.

Keywords
Safeguards, muon detection, spent fuel, CASTOR.

1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Muon imaging for innovative tomography of large volume and heterogeneous cemented waste packages

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Abstract

A mobile muon tomography system is being commissioned in a non-laboratory environment to address the as-yet unsolved problem of the non-destructive assay of large volume nuclear waste packages, such as large spent fuel casks and large concrete waste packages with heterogeneous waste.

Muons are natural occurring particles with a flux at ground level of around 1 cm^{-2}min^{-1}; they are capable of passing through several hundred metres of rock. When they interact with matter, they scatter depending on the atomic number, Z, of the material. Hence, if muon trajectories are measured before and after traversing a volume, an image of the different materials in that volume can be produced. This mature and well-established technique is known as 'muon tomography' (MT). It has been successfully utilised in e.g. cargo screening for nuclear contraband. MT is fully passive and works for heavily-shielded volumes; it is also highly complementary to gamma and neutron tomography. The technique is particularly useful to detect heavy elements like lanthanides and actinides, but can also be applied to detect light elements and gas bubbles and density gradients or differences within a matrix.

The system is in an advanced stage of commissioning. Combining the knowledge and experience gained operating prototype systems and details and realistic Monte Carlo simulation program is well underway. A large Monte Carlo effort is well underway. The work includes comparison studies of the performance of different reconstruction algorithms to determine the contents of the waste drums focusing both of high-Z materials and gas bubbles. This has led to the development of a unique approach using figures of merit to enable performance comparisons between different algorithms and/or instrumentation techniques. For example, we have shown that we can resolve the presence of a uranium target object down to 4mm using our system in a concrete drum, can detect gas bubbles with a volume over 1L in concrete, discriminate between Pb, W and U, measure the size of objects with mm precision and study the minimum spacing between two objects to see them separately.

1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

LINAC-based high energy X-ray imaging of radioactive waste packages at CEA

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Abstract

In order to inspect the interior of a waste package without having to carry out a destructive action, high-energy photon imaging technique (radiography and tomography) can be used in the manner of a medical scanner. The package to be inspected is placed between the photon source and the detector. The image delivered by the detector reports the absorption of photons through the object. The thicker and denser the object, the greater the absorption. In order to image large and dense radioactive waste packages, such as 2 m3 concrete drums with a diameter of more than 100 cm, a high intensity and high energy X-ray source based on an electron linear accelerator (LINAC) has been implemented in CINPHONIE facility, at CEA DEN Cadarache. Our current "Mini-Linatron" LINAC produces 9 MeV electrons that collides a tungsten target, producing a Bremsstrahlung photon beam with a dose rate close to 20 Gy.min-1 at a distance of 1 m from the target, in the axis of the beam. Radiographic and tomographic images are produced with a millimeter resolution, allowing precise visualization of the interior of waste packages (pictures of the linear attenuation coefficient in each pixel, which can be converted to density through hypothesis on the nature of materials). A new "SATURNE" LINAC with an electron energy up to 20 MeV and a dose rate larger than 100 Gy.min-1 at 1m is currently being commissioned in CINPHONIE, which will allow imaging objects up to 150 cm of concrete or 40 cm of iron thick.

The use of such high-energy photons requires specific detectors and CINPHONIE is currently equipped with two acquisition systems. The first detector is a 2D wide-field screen of 80×60 cm^2 with a "Gadox" (Gd2O2S) scintillator coating for rapid imaging but with limited attenuation dynamics of about 3 decades, corresponding to 100 cm of concrete or 25 cm of steel. The second system is a fan array of 25 CdTe semiconductor bar detectors, with collimators oriented towards the focal spot of the photon beam, for quantitative computed tomography with attenuation dynamics of approximately 5 decades, i.e. 1.5 m of concrete or 40 cm of steel. New scintillator screens for large field of view detection are being tested in our laboratory, as well as large and continuous linear detectors (CdWO4 needles or flat CsI scintillators with on-edge X-ray beam) to avoid the horizontal sweeping scan of the package imposed by gaps between the collimated CdTe, and thus to reduce acquisition time. Recent tests of ModuPIX detector, which will equip the underwater X-ray imaging system of the future experimental Jules Horowitz Reactor, will also be reported. This detector based on TimePix chip (CERN) coupled with very thin cadmium telluride or silicon semiconductors with on-edge beam irradiation should allow reaching a very good spatial resolution (close to 100 μm).

1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Finally, we will report ongoing R&D studies aiming at determining the mean effective atomic number of materials in radioactive waste packages. The principle is based on performing acquisitions at different X-ray energies to take advantage of the differences in attenuation according to photon beam energy and atomic number of the elements. This method applied on computed tomography allows extracting both the density and the effective atomic number from the attenuation information, and thus refines the identification of materials. We will also mention the development of TOMIS, a transportable tomographic system to perform in situ imaging of large concrete waste packages, involving dual energy computed tomography as well.

1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Novel optical methods for the characterisation of outgassing from radioactive waste

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Abstract

Long-lived radionuclides are particularly challenging to detect in the context of nuclear facilities. They are the main source of radioactive gas emissions and must therefore be monitored. Radiocarbon is for example produced in all part of nuclear power plants and can then be released in the form of carbon dioxide or methane through the biodegration of radioactive waste. Long-lived radionuclide in general, such as radiocarbon and tritium, are problematic because they have a high residence time in the environment and can easily be assimilated into living matter. A proper management of nuclear wastes containing these nuclides is necessary due to their potential impact on human health and the environment. They are highly mobile in the environment [2], and must be monitored for long-time, because of their long half-life. In the context of conditioned nuclear waste, characterisation of the waste outgassing is necessary. Gaseous emissions from radioactive waste usually mostly consist of tritium and radiocarbon, mainly emitted in the form of water vapour (HTO), carbon dioxide ($^{14}CO_2$) and methane ($^{14}CH_4$). $H^{36}Cl$ is also of interest in the particular case of graphite waste.

Unfortunately, there are no method capable of rapid in-situ analysis of those radionuclides. Current methods require samples collection and off-line analysis in the laboratory by liquid scintillation counting (LSC). Scintillation peaks of those radionuclides overlaps in LSC limiting its sensitivity and often requiring time-consuming radiochemistry method to separate the different beta emitters. On the other hand, novel optical methods for detection of long-lived radionuclides from nuclear facilities have been developed in recent years. In particular, cavity ring-down spectroscopy (CRDS) has been shown to provide high sensitivity for radiocarbon and tritium detection in the form of carbon dioxide [3] and water vapour [4], respectively. CRDS is one of the most advanced and sensitive laser spectroscopy technique [5 - 6]. It is an isotopically selective laser spectroscopic technique and provides the highest sensitivity, allowing for the detection of extremely small amounts of radioactive gases, and thus detection of the smallest leaks.

In the CHANCE project, these methods are further developed to determine the outgassing rate of various radioactive molecules. The potential of CRDS for waste package monitoring is

    1.   **Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments**

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

explored, since this technique has not been used in this context before and has several advantages over conventional techniques. The CHANCE project focuses on the detection of radiocarbon and ^{36}Cl isotopes. CEA is developing a novel instrument for the detection of $H^{36}Cl$ based on CRDS. This is the first time that CRDS is used to detect this molecule, which is highly relevant in the case of outgassing from graphite waste. At VTT, an already-developed CRDS instrument for the detection of radiocarbon compounds will be used to study in detail their outgassing rate in various types of waste. The goal is to provide valuable information about the radiocarbon release kinetics and its speciation, including methane and carbon dioxide. Further work, in collaboration with ENEA, will consist of validating the technique and compare it to traditionally used techniques such as LSC. This work will therefore result in a major step forward in the development of the use of optical methods to detect radioactive gas emissions.

1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

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Book of abstracts | BA-118 | SCK•CEN 32770025   Page 28 of 20
1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Modelling of the CHANCE calorimeter and some general thoughts about NDA uncertainty quantification

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Abstract

Calorimetry is one method for the determination of nuclear material masses (or activities) by the quantitative measurement of heat flux, emerging from a sample. Calorimetry is used for non-destructive assay (NDA) which means that no sample needs be tempered with or destroyed by the analysis. Heat-flow calorimeters have been used successively for measuring a wide range of thermal powers i.e. from 0.5 mW (0.2g of low-burnup plutonium) up to 1kW for samples varying in size from about 2.5 cm up to 60 cm wide and 100 cm long [1,2].

The MCNP6-based numerical model of the Large Volume Calorimeter (LVC) designed by KEP Nuclear (France) was set up. Various scenarios were simulated for different particles types, energies and source locations to accompany and support the interpretation of calorimetric KEP-LVC measurements within the CHANCE project [3]. For the analyses, the particle flux and energy deposition in each layer of the calorimeter are plotted. The results yield that a significant part of the gamma and neutron radiation leaves the system and does not contribute to the heat deposited. Moreover, the high-energy part of the gamma and neutron radiation flux can reach the reference chamber of the calorimeter and deposit some energy there, compromising the calibration and causing double-bias.

A particle flux study of the 200L mock-up drum and the KEP-LVC calorimeter will be presented and discussed. A tally 4 with mesh option for both gamma and neutron of up to 5 MeV was used to simulate the mean flux in each cell and for various source location and source distribution patterns.

The simulations showed that, depending on source location and power, neutron radiation and high-energy gamma radiation leakage can influence the final heat measurement differently. In general, part of the realized simulations suggest that for some possible scenarios - with samples containing nuclei of high cross sections for (α,n) and other neutron-producing reactions, high activity beta-emitters (bremsstrahlung radiation) or gamma-emitters - the accuracy of calorimetric assay can be decreased due to radiation escape.

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Book of abstracts | BA-118 | SCK•CEN 32770025   Page 29 of 20
  1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium
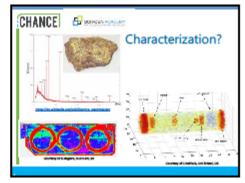
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1.   Topical Topical Day CHANCE | Characterisation of conditioned radioactive waste: status, challenges and new developments

March 21, 2019 | Lamot Conference Centre, Mechelen, Belgium

Slide 1

CHANCE

Introduction to the CHANCE project

Denise Ricard, H. Tietze-Jaensch, A. Rizzo, C. Mathonat, C. Bruggeman, J. Velthuis, L. Thompson, G. Genoud, C. Bucur, D. Kikola, G. Zakrzewska-Koltuniewicz, O. Guéton

Slide 2

CHANCE — Introduction

- CHANCE: "Characterization of conditioned nuclear waste for its safe disposal in Europe"
- Funded by Euratom research and training programme 2014-2018 under grant agreement N° 755371
- Within the NFRP 7-2016-2017 topic "Research and innovation on the overall management of radioactive waste other than geological disposal"
- Duration of 4 years; start date June 1, 2017; end date: 31 May, 2021
- Total CHANCE budget: 4,25 M€
- EC contribution: 3,98 M€
- Consortium: 12 partners from 8 European countries

Slide 3

CHANCE — Consortium

- Andra (FRA)
- CEA (FRA)
- ENEA (ITA)
- FZJ (GER)
- KEP Nuclear (FRA)
- SCK•CEN (BEL)
- University of Bristol (UK)
- University of Sheffield (UK)
- VTT (FIN)
- RATEN (ROM)
- WUT (POL)
- INCT (POL)

Slide 4

CHANCE — CHANCE objectives (1)

- To **establish at the European level a comprehensive understanding of current conditioned radioactive waste characterization and quality control schemes** across the variety of different national radioactive waste management programmes

  - Based on inputs from end-users such as Waste Management Organisations, regulators, waste producers and repository operators

Slide 5

CHANCE — CHANCE objectives

To **further develop, test and validate non-destructive techniques that will improve the characterization of conditioned radioactive waste (CRW)** and complement current methodology while particularly targeting large and heterogeneous waste compounds

  - **Calorimetry** as a non-destructive technique to reduce uncertainties on the inventory of radionuclides
  - **Muon Tomography** as a non-destructive technique to control the content of large volume nuclear waste
  - **Cavity Ring-Down Spectroscopy (CRDS)** to characterize outgassing of radioactive waste

Destructive technologies followed by chemical or radiological analyses are commonly used techniques for radioactive waste characterization but are not addressed in CHANCE, since these techniques are applied to the raw waste itself; this is not in the scope of this project.

Slide 6
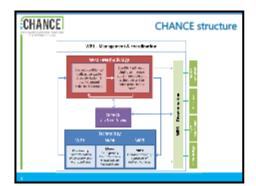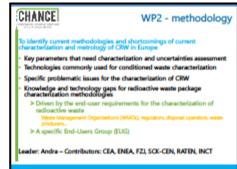
CHANCE — CHANCE objectives (3)

- CHANCE will focus on the following waste forms (IAEA classification):

  - Very Low Level Waste (VLLW)
  - Low Level Waste (LLW)
  - Intermediate Level Waste (ILW)
  - High Level Waste (HLW)

- Very Short Lived Waste and Exempt Waste are beyond the scope of CHANCE as these kinds of waste are not destined for a specific radioactive waste disposal

Slide 1: CHANCE structure

CHANCE — CHANCE structure

WP1 - Management & coordination

WP2 - methodology

WP6 - Dissemination

Technology
WP3 · WP4 · WP5

Slide 2: WP2 - methodology

CHANCE — WP2 - methodology

To identify current methodologies and shortcomings of current characterization and metrology of CRW in Europe

- Key parameters that need characterization and uncertainties assessment
- Technologies commonly used for conditioned waste characterization
- Specific problematic issues for the characterization of CRW
- Knowledge and technology gaps for radioactive waste package characterization methodologies
  - Driven by the end-user requirements for the characterization of radioactive waste
    - Waste Management Organizations (WMOs), regulators, disposal operators, waste producers...
  - A specific End-Users Group (EUG)

Leader: Andra – Contributors: CEA, ENEA, FZJ, SCK-CEN, RATEN, INCT

Slide 3: WP2 - methodology

CHANCE — WP2 - methodology

A questionnaire concerning the characterization of conditioned radioactive was produced and send to EUG members

- Synthesis of questionnaire answers will be presented by Crina Bucur (RATEN)
  - Radioactive waste characterization practices and gaps in Europe

Slide 4: WP3 - calorimetry

CHANCE — WP3 - calorimetry

Objectives:
- To test and evaluate the performance of calorimetry for inventory of radionuclides (measure Beta or alpha radiation heat source)
- To identify how calorimetry can complement existing, widely-used techniques (gamma spectrometry and neutron passive measurement)
- To carry out an exhaustive study of uncertainties assessment related to calorimetry and its coupling to other non-destructive techniques

A dedicated calorimeter (200 litres) will be designed and built by KEP Nuclear

A dedicated experimental program will be carried out with mock-up drums (CEA and SCK-CEN)

Leader: KEP Nuclear – Contributors: CEA, FZJ, SCK-CEN, WUT

Slide 5: WP3 - calorimetry

CHANCE — WP3 - calorimetry

- Talk of Christophe Mathonat (KEP Technologies)
  Recent developments in calorimetry for nuclear wastes characterization

- Talk of Wojciech Kubinski (WUT) & Bart Rogiers (SCK-CEN)
  Modelling of the CHANCE calorimeter and some general thoughts about NDA uncertainty quantification

Slide 6: WP4 – muon imaging

CHANCE — WP4 – muon imaging

To develop mobile muon tomography instrumentation to address imaging of large volume and heterogeneous nuclear waste packages

Objectives:
- To build a suitable mobile muon detection system
- To demonstrate real waste drum muon tomography
- To evaluate performances of the technique

WP4 Leader: University of Bristol – Contributors: FZJ, SCK-CEN, University of Sheffield, WUT

- Talk of Jaap Velthuis (University of Bristol)
  Muon imaging for innovative tomography of large volume and heterogeneous cemented waste packages

CHANCE — WP5 – outgassing analysis and monitoring

WP5 Leader: VTT – Contributors: CEA, ENEA, FZJ

Objective:
- Advance the use of Cavity Ring-Down Spectroscopy (CRDS)
  - Develop new instrumentation for H^{36}Cl
  - Demonstrate an application of the technique to the monitoring of ^{14}C outgassing

Cavity ring-down spectroscopy

- Talk of Guillaume Genoud (VTT)
  Novel optical methods for the characterization of outgassing from radioactive waste

CHANCE — WP6 – dissemination and training

WP6 Leader: SCK•CEN – Contributors: Andra, ENEA, FZJ, INCT

To integrate, communicate and disseminate CHANCE results within the European community involved in radioactive waste management
- Communication to broader European community involved in radioactive waste disposal
- Study on the socio-technical concerns and uncertainties associated with the principles and characterization methodology of radioactive waste
- Training and education of young professionals
  - CHANCE Mobility-fund in the field of characterization of conditioned radioactive waste by means of non-destructive analytical techniques and methodologies
    - Open to Master students, PhD students and junior professionals
      Internships, thesis preparations or scientific visits hosted by a partner of the CHANCE project
      Participation in conferences and workshops
  Further information available on www.chance-h2020.eu
- Synthesis report integrating all CHANCE results

CHANCE — WP6 – dissemination and training

Communication tools
- Public website : www.chance-h2020.eu
  - News about the project, events
  - Public Deliverables
  - Publications

- Participation to national and international events (conferences, workshops,...)
- Specific communication through IGD-TP (website, newsletter,...)
- Organization of this **Topical day**
- Training course (will be organized towards the end of CHANCE project)

CHANCE — CHANCE End-User Group

CHANCE EUG gather experts from European (WMOs, regulators, repository operators including waste producers, TSOs...)

EUG role in the project
- Participate in EUG meetings organized by the CHANCE consortium
- Answer a specific questionnaire related to WP2 & WP6, and review related deliverables
- Provide feedback and insights with respect to practical concerns that might benefit the project progress

CHANCE — CHANCE End-User Group

Current members

Organisation	Country	Role
Andra	France	WMO
ANDR	Roumanie	WMO
Areva	France	Nuclear Facility Operator
CEA	France	Nuclear Facility Operator
EMT	Allemagne	IAEA Technical Expert Consulting Group
EDF	France	Nuclear Facility Operator
Enresa	Espagne	WMO
IRSN	France	TSO
NCBJ Demolition	Grèce	Nuclear Facility Operator
NRG	Pays-Bas	Nuclear Facility Operator
Nucleco	Italie	WMO
RWM	Royaume-Uni	WMO
SCK-CEN	Belgique	Nuclear Facility Operator
SEB	Suède	WMO
SOGIN	Italie	Nuclear Facility Operator
ZUOP	Pologne	WMO

- EUG will remain open for new participants during the project life

CHANCE — CHANCE expected impact

- To develop new solutions for the characterization of conditioned radioactive waste to solve specific issues
- To improve radioactive waste classification
- To improve the global safety of the whole chain of radioactive waste management
- To contribute to an European homogenized approach for the characterization of conditioned radioactive waste
- To improve common practice in terms of conditioned radioactive waste characterization
- To maintain and enhance knowledge and competences
- To improve public confidence

Slide 1

CHANCE
Characterization of Conditioned Nuclear Waste for its Safe Disposal in Europe

Radioactive waste characterization: practices and gaps in Europe

Crina BUCUR

Institute for Nuclear Research Pitesti (RATEN ICN), Romania

Slide 2 — Outlines

CHANCE

- ❑ **General considerations on radioactive waste characterization**
  - ○ Why characterisation of RW is important ?
  - ○ On what is based the characterisation process?
  - ○ Responsibilities in a waste characterization programme
  - ○ Techniques/methodologies used in RW characterization
    - ✓ NDA methods
    - ✓ Destructive analysis (DA)
- ❑ **CHANCE outcomes on RW characterisation practices**
  - ○ CHANCE WP2 objectives
  - ○ Content of the CHANCE questionnaire
  - ○ Characterisation techniques used by CHANCE EUG members
  - ○ Difficulties in RW characterisation
  - ○ How these problems could be solved?
  - ○ EUG interest in applying techniques addressed by CHANCE

Slide 3

CHANCE

General considerations on radioactive waste characterisation

Slide 4 — Why characterisation of RW is important ?

For producing a **"quality end product"** for all disposed RW

➢ it could be achievable by obtaining a thorough and accurate assessment of the **physical, chemical and radiological characteristics** of the waste

According to IAEA recommendations [*IAEA-TECDOC-1537*], RW characterisation must be:

- accomplished or verified with a **clear demonstration** of the compliance with WAC for interim storage, transport and final disposal
- accomplished in a **systematic manner** using **proven** methodologies, technologies and techniques with emphasis on **quality assurance (QA) and quality control (QC)**.

*Proper control of safety relevant parameters of RW within the entire waste management life cycle, and careful **testing of the quality of final waste forms and waste packages**, are principal components in any waste management strategy*

Slide 5 — On what is based the characterisation process? (1/2)

The **type and extent of the characterization efforts** depend on many factors, such as:
- the type of waste or waste form;
- the disposal concept;
- the regulatory regime;
- the amount of process knowledge that is available;
- the kind of characteristics to be measured.

➢ *It is impossible to define or recommend a single characterization procedure for even similar waste forms or packages.*
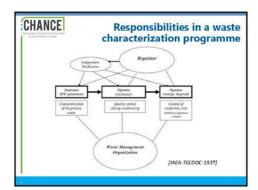
Slide 6 — On what is based the characterisation process? (2/2)

According to the IAEA recommendations [*IAEA-TECDOC-1537*], the **strategy for the RW characterization** should be based on understanding and assessment of the:

- the **waste acceptance criteria** and the rationale behind it;
- the **safety assessment** for the disposal concept: assumptions made, information needed ...
- **process control and quality assurance** requirements, transportation requirements, and worker safety requirements.

*Planning for waste characterization should take place in advance of waste generation to ensure **that all stages in the waste management are accomplished in a controlled manner.***

Responsibilities in a waste characterization programme

[IAEA-TECDOC-1537]

Techniques/methodologies used in RW characterisation (1/2)

- **Process knowledge**: knowledge and documentation of the processes that generate RW is an effective way of assessing the RW characteristics
- **Non-destructive examination and assay** (NDE and NDA):
  - NDE - involves checking the **physical and mechanical properties** of the waste package
  - NDA - involves analysis of the **content of one or more RN** in waste package without affecting the physical or chemical form of the material. Selection of a specific NDA techniques depend on the **radionuclides** (γ-X ray emitters or fissile material) and the **relevant parameters of the waste package** (type of matrix, radioactivity/ sources and density distribution, shape and dimensions of the container and inner/outer shielding)
- **Destructive analysis** (DA) - involves **sampling**, sample preparation, chemical separation methods (radiochemistry) and **radiometric determination**

Techniques/methodologies used in RW characterisation (2/2)

The measured RNs are categorized in:
- **easy to measure (ETM)** - can be determined by direct measurements (i.e. γ-emitters);
- **difficult to measure (DTM)** - cannot be determined by direct measurement in the waste form (i.e. pure-β emitters, low-energy X-ray emitters, α emitters);
- **impossible to measure (ITM)** – measurement of their activity would require unjustified costs and effort

The **nuclide vector (NV)** methodology: qualitative identification of all RNs on specific waste stream

The **scaling factor (SF)** methodology: determines the activity of DTM and ITM radionuclides using correlations between them and key nuclides (KN) chosen among the ETM. SFs are **waste stream and case specific**

A combination of different techniques (e.g. alpha spectrometry, mass spectrometry and gamma spectrometry) are used together with modeling activities to determine the NVs and SFs. **The sample and location for sampling are very important and have to be representative for the waste stream considered.**

Using a NDA system, waste packages are measured to determine KN concentrations and by using the SFs and NVs the associated DTM, ITM and ETM nuclides are then **calculated and declared for each package.**

NDA methods

Use standards calibration or modelling and could have high uncertainty associated to the activity determined.

- **Gamma methods** - rely on capturing gamma photons emitted from the waste package as result of radioactive decay of the radionuclides present in the waste
- **Neutron methods** - typically rely on capture of neutrons emitted either through spontaneous fission or (α,n) reactions and are primarily used to identify and quantify fissile or fertile isotopes in the waste
- **Calorimetry** relies on measuring the specific heat generated from radioactive decay to quantify the amount of activity present

Destructive analysis (DA)

Provides the **most accurate and unbiased** activity determination
- Involves **sampling**, **sample preparation** (dissolution/mineralization), **chemical separation methods** and **radiometric measurements**
- **Sampling - a critical step** in the characterization process. Designing the sampling procedures and checking the **homogeneity and representativeness** of the samples assures the reliability of the final results
- **Radiometric determination** is performed by instrumental analysis such as:
  - LSC - allow α-β spectrometry
  - α spectrometry
  - high resolution γ spectrometry - high and low energy γ emitters
  - mass spectrometry

CHANCE outcomes on RW characterisation practices

CHANCE WP2 objectives

- Collect **end-users requirements** in terms of **characterization**, especially links and overlaps between these requirements and waste specifications for different national disposals;

- Identify **critical parameters to be measured** for each waste type and their final disposal solutions and the **commonly used evaluation techniques** and **problematic cases** for radioactive waste package characterization;

- Identify **R&D needs** to develop and evaluate innovative technologies complementing current characterization techniques and providing supplementary

---

How to accomplish these objectives?

Based on a **survey** of **characterization methodologies currently used** by European waste generators, disposal operators and waste management organisations

A questionnaire was sent to CHANCE EUG members to identify:
- **key parameters** that need characterization;
- **technologies/methods** commonly used for characterization of conditioned RW;
- **WAC** applied and the possibilities of **their harmonization** in Europe
- **specific problematic issues** for the characterization of conditioned RW;
- **R&D needs** and potential **on-going R&D programme** on the topic of conditioned radioactive waste characterization;
- **potential applications of R&D actions** included in CHANCE;
- **socio-technical and ethical issues** associated with the waste characterization process.

Content of the questionnaire (1/2)

18 questions related to the:
- RW, including SNF, managed by the EUG members
- The storage / disposal options
- The WAC for the storage / disposal facilities and the possibility of WAC harmonization across Europe
- Impact of the evolution of the WAC and of the final disposal concepts
- Methods applied in characterisation of conditioned RW and the associated uncertainties (technical, conceptual, social, political, ethical)
- Waste categories and/or waste forms that do not have a dedicated option for storage / disposal
- The major difficulties characterisation of conditioned radioactive waste
- The R&D needs and on-going R&D programmes on characterisation of conditioned RW
- Potential application of characterisation techniques improved in CHANCE: **Calorimetry, Muon Tomography and Cavity Ring-Down Spectroscopy**

Content of the questionnaire (2/2)

- Characterisation and control processes: responsible organizations, structure and ways to followed up these processes
- The role of host communities in the characterisation and control processes
- Disciplines / fields of expertise / actors involved in the characterization of conditioned RW
- The importance of RW waste characterization
- Lessons learnt

Answers received

Nr.crt.	Country	Organisation	Type
1	BE	ONDRAF/ NIRAS	WMO
2	ES	ENRESA	WMO
3	FR	ANDRA	WMO
4	IT	Nucleco S.p.A.	WMO
5	PL	Radioactive Waste Management Plant	WMO
6	RO	ANDR	WMO
7	SE	SKB	WMO
8	UK	RWM	WMO
9	BE	SCK-CEN	RE
10	DE	JULICH	RE
11	FR	CEA	RE
12	PL	INCT	RE
13	RO	RATEN ICN	RE

Characterisation techniques used by CHANCE EUG members

- The methods used for RW characterisation depend on the country approach. The majority of the respondents to CHANCE questionnaire apply **spectroscopic techniques** (alpha, beta and gamma) as well as **neutron measurements** and **dose rate calculation**. Only few of them more advanced technique such as calorimetry.

- **Nuclide vector and Scaling Factor** methods are used by a large number of European countries, such as Belgium, France, Germany, Spain, Sweden, the UK.

CHANCE — RW characterisation in Belgium

The characterisation process is based on the following techniques:

- **gamma-spectrometry** (for KN determination) combined with an **approved nuclide vector** (base on models, calculations, and sometime on destructive measurements);
- **dose rate** (measured on non-conditioned waste packages) **conversion with approved NVs** (nuclide inventor is calculated through specific conversion factors - mSv/h to MBq - determined using software such as MicroShield);
- **destructive analysis** and **approved NV**;
- **neutron measurements** on conditioned waste;
- **mass spectrometry, gamma spectrometry, alpha spectrometry**...on samples of non-conditioned waste.

CHANCE — RW characterisation in France

The characterisation process is split into 2 phases:

- **Determination of NVs and SFs** for waste streams;
- **Measure the waste itself**: when a practical spectrum is known, it is needed to gain access to the waste activity. In this case, the aim is to measure one or more tracer(s) that can lead, with the knowledge of spectra, to the knowledge of every significant radionuclide by means of SFs and NVs. The measurement of this / those tracers is aimed to be easy and not hard-to-analyse.

Depending on the waste type, different methods are used :

- γ spectrometry on waste package;
- X-ray inspection;
- active neutron measurements for prompt or delayed neutrons;
- passive neutron measurements;
- DA: samples of waste package may also be collected (with destruction of the waste package), dissolved, then gamma spectrometry or radiochemistry are made;
- measurement of **tritium degassing** with LSC;
- calorimetry

CHANCE — RW characterisation in Germany

For operational waste:

- **pre-characterization**: source/origin of RW, radiological and material pre-sorting (mainly material stream separation and gamma activity / dose rate survey);
- **conditioning input characterization**: sampling & radio-chemical analysis (RCA), QA and ID/origin checks, **temperature-range stability, outgassing control, temperature survey, dose rate** at surface and 1m distance;
- **measurement portfolio**: dose rate/gamma spectroscopy, calorimetry;
- determination of **heat, density, volume, mass, homogeneity, centre-of-mass**
- **sampling-statistics**, random sampling for RCA;
- **applied methods**: AAS, ICP-MS, OCP-MS, gamma, neutron spectroscopy, QA-process control parameters, uncertainty assessment, LS/CRDS

For decommissioning waste:

- **pre-characterization**: source/origin of RW, radiological and material pre-sorting (mainly material stream separation and gamma activity / dose rate survey);

CHANCE — RW characterisation in Italy

An integrated approach combining different methods, such as **physical scanning, gamma spectrometry** and **neutron counting** measurements.

- For the **physical scanning** an X-ray tube is used to analyse **the matrix and its density distribution**.
- The **gamma spectrometry** is performed by using HPGe detector and keeping the item (usually a drum with variable volume) in rotation to reduce the radial matrix effect.
  - Open-geometry: one measure per waste package
  - Segmented scanning: the waste package is be "cut" in segments (reconstructing the vertical distribution of both matrix and contamination)
  - Tomography: a 3-dimensional reconstruction of matrix and contamination distributions
- For the **neutron counting** measurements, a dedicated equipment able to detect neutrons coming from both spontaneous fissions (**passive mode**) and induced fissions (**active mode**) is used – able to detect both fertile and fissile materials

CHANCE — RW characterisation in Poland

To characterise the RW intended for storage, **radiation activity measurements** and eventually **radiation spectroscopic measurements** are made.

Usually, radioisotope composition of waste is determined on the basis of **the end users' declaration**.

Characterisation of the RW is simple, limited to the **gamma dose rate** in a single container sent for the treatment and disposal.

Each type of the waste is generally described: the volume, form of the waste, radionuclides present (according to the knowledge of the user) and time of generation.

CHANCE — RW characterisation in Romania

- For institutional waste sent for final disposal to National Repository Baita Bihor:
  - **gamma dose rate** measurement
  - **gamma spectrometry**
  - the activity of α and β emitters is based on **the measurements carried out on row waste** before conditioning or on the **generators declaration**.
- Cernavoda NPP: no conditioned RW packages were produced up to date
  - **C-14, H-3 and gamma emitters** are measured **on raw waste** that are regularly sent for incineration at an European operator
  - For the LLW, the process for developing scaling factors is on-going. The waste **streams were identified** and gamma measurements for **ETD radionuclides** were carried out. Also, the **DTM radionuclides are measured** on the identified waste streams by using adequate radiochemistry methods for their separation and purification and **LSC and alpha spectrometry** for quantification

RW characterisation in Spain

The ETM radionuclides are determined:
- by **gamma dose rate** measurement for waste with known gamma isotopic composition;
- **segmented radiography** for heterogeneous waste or for better improvement;
- by **samples taken before the conditioning process** - for homogeneous waste, and further extrapolation as a function of the mass introduced; a final dose rate factor correction is applied if necessary;
- **gamma spectrometry** to the whole package/item;
- **segmented /collimated gamma spectrometry** for heterogeneous waste or for better improvement.

The DTM radionuclides are assessed by using **Scaling factors**

RW characterisation in Sweden

The radiometric measurements are split in two categories:
- *nuclide specific measurements of waste packages*
- *non-package-specific data*

The **waste suppliers** are requested to perform **gamma spectrometry for each waste package disposed** of in SFR (the final disposal for LILW-SL).

The activation product **Co-60** and the fission product **Cs-137** are considered **"key nuclides"** and are used in most of the calculation methods to determine the activity or distribute other DTM nuclides in the different waste vaults in SFR (final disposal facility for LILW-SL at Forsmark).

RW characterisation in the UK

Characterisation activities are undertaken by the **waste producers, on raw waste**, prior to any conditioning process.

Routine characterisation of row RW consists of radiological characterisation, routine sampling and analysis to confirm satisfactory operational processes, effluent characterisation, condition, monitoring and inspection (CMI) on inside existing storage facilities –based on British Standard inspection requirements.

Condition, monitoring and inspection techniques:
- **Laser measurement** of waste package geometry - to detect waste container expansion
- **Muon tomography** – was used in the past to support an understanding of the evolution of specific waste packages
- **air sampling and discharge monitoring** in storage facilities to confirm satisfactory operation
- Gas sample collection for selected packages

Difficulties in RW characterisation (1/3)

- The proper characterization of the **conditioned legacy/ historical waste packages** represents a general problem, especially for countries with long-term utilization of nuclear technology (Be, De, Es, Fr, It, the UK).
  - radiologic characterisation of these waste packages, especially in the case of **large volume packages**, represents a **major challenge to either passive or active measurements** because of the difficulty of the interrogators radiation to penetrate the waste matrix through strong attenuation before (gamma or neutron) emissions arrive at detectors placed around the waste package.
  - the type of different materials in historical waste is often difficult to identify
- **Accelerator based waste** for which the radionuclide vectors and material vectors were not defined yet and SNF for direct disposal are also considered categories of **difficult to be characterized** RW.

Difficulties in RW characterisation (2/3)

- Determination of a **viable source term** in already **conditioned RW** - a common problem mainly because the α and β emitting radionuclides are DTM due to the **signal attenuation** caused by the waste package itself and the backfill. These effects, in case of low or very low radioactivity or low energy emissions (such as the 59.54 keV gamma line for Am-241), strongly compromises the measurement efficiency, leading to significant uncertainty of the measured activity.
- **Detection of declarable DTM isotopes** (such as Sr-90) and **sealed radioactive sources**.

Difficulties in RW characterisation (3/3)

- Other identified problems in characterisation of conditioned RW consist of:
  - **little traceability of the chemical content** of waste packages, when the conditioning is done by the waste producer;
  - **accessibility of the waste for sampling** - due to the limited access at the waste packages;
  - **difficulties in monitoring and periodical control** of the waste drums packed deeply in a storage facility;
  - **characterization and reconditioning of the waste already stored** in a repository
  - the **lack of standardized processes for the characterization and repackaging** (or reconditioning) of spoilt drums/containers.

How these problems could be solved?

- By developing *non-destructive methods* capable to detect the **radiological (including α and β emitters) and fissile mass** content with an acceptable uncertainty, compatible with the WAC for storage, transportation and final disposal.

  These methods should be **able to be applied** for:
  - **homogeneous and heterogeneous waste**
  - waste packages of **different sizes**, including **SNF** casks.

EUG interest in applying techniques addressed by CHANCE

Calorimetry is seen as **a useful technique in:**

- **diminution the uncertainty and/or detection limit** for conditioned RW such as waste containing H-3 and Pu (in conjunction with other nuclear measurement techniques)
- **to identify sealed spent sources or alpha/beta emitters** with considerable heat load hidden in packages to be disposed of.
- Combined with gamma spectrometry, **Calorimetry** is considered to **bring improvements to the currently used non-destructive techniques** for DTM isotopes in conditioned waste packages.

CHANCE attempts to make the calorimetry applicable for large volume, heterogeneous and multi-nuclide waste packages

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EUG interest in applying techniques addressed by CHANCE

Muon Tomography is regarded as a technique that:

- **could replace the X-ray** high energy for package with high biological protection
- stand-alone or combined with gamma imaging, **Muon Tomography** is considered useful to identify the:
  - **location and amount of high-Z**
  - **potentially fissile material** in a waste package
  - **position and integrity of SF-assemblies** in very large storage casks
  - unwanted **density depletion from voids or bubble formation** in cemented drums
- Possibly useful for **monitoring interim storage halls**, maybe even filled and sealed repository galleries.

EUG interest in applying techniques addressed by CHANCE

Cavity Ring-Down Spectroscopy is considered very useful tool for:

- the **extremely low limit outgassing monitoring** of waste packages containing **graphite waste** exuding very small amounts of H-3 or C-14 or for the detection of Cl-36 outgassing.
- **measuring in real time the emission of radioisotopes** (in water vapour, CO_2,...) **in repositories** and even in the **reactor caissons**, in case of graphite moderated reactors.

Conclusion

- The **requirements for characterisation are driven by the WAC**, either for storage or disposal facilities
- RW characterization is a very complex process that requires the use of **in-situ measurements (NDA), sampling and analyses (DA)** complemented with **modeling and theoretical calculations.**
- **Process knowledge** represents an important component of the characterization process
- Despite of various techniques already used in the RW characterisation process, there is need of developing new *NDA methods* capable to detect the radiological (including α and β emitters) and fissile mass content with an acceptable uncertainty, compatible with the WAC for storage, transportation and final disposal

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Questions?

NIRAS **BELGO PROCESS**

Waste characterization in relation to the WAC for surface disposal

Approach and challenges ahead

Robin Tuerlinckx (Belgoprocess)
Tomas Vandoorne (ONDRAF/NIRAS)

Content

1. **Introduction**
2. Waste acceptance criteria for surface disposal
3. Upstream characterization
4. Control strategy
5. Challenges ahead

---

1. Introduction

- Near-surface disposal site (cAt), for low-level waste in Belgium is expected to be operational in 2024
- Safety report of near-surface disposal site is in final stage of approval by safety authority
- Recent developments have lead to a re-evaluation of the waste acceptance criteria (WAC)
  - Impact on characterization
  - Emphasis on control strategy

---

1. Introduction

- A total production of about 26 000 disposal packages is foreseen
- Waste inventory overview
  - » NPP filters conditioned in cement matrix
  - » NPP concentrates and resins homogeneously conditioned in cement matrix
  - » (Supercompacted) waste, heterogeneously cemented → from NPP's, fuel fabrication, research, industry, medicine,...
  - » Incineration ashes, supercompacted and cemented
  - » Bituminized sludges
  - » Dismantling wastes: construction materials, metals,...

Content

1. Introduction
2. **Waste acceptance criteria for surface disposal**
3. Upstream characterization
4. Control strategy
5. Challenges ahead

---

2. Waste acceptance criteria

- Radiological content limitations on primary waste packages
  - 28 RNs are essential in characterization:
    C-14, Cl-36, Ca-41, Ni-59, Ni-63, Se-79, Sr-90, Zr-93, Nb-94, Mo-93, Tc-99, Ag-108m, Sn-126, I-129, Cs-135, Cs-137, U-234, U-235, U-236, U-238, Np-237, Pu-238, Pu-239, Pu-240, Pu-241, Am-241, Am-243, Cm-244
  - Ra-226/Th-232 excluded
  - Fissile material (remark: RN-limit value is more stringent)

	Limit
U-235 + Pu-239	≤ 50 g
U-235 (in case of HEU > 20%)	≤ 15 g

2. Waste acceptance criteria

- Physical content limitations on primary waste packages
  - No free liquids
  - Concrete insensible to ASR (alkali-silica-reaction)
  - Concrete insensible to DEF (delayed-ettringite-formation)
  - For concrete rubble (dismantling waste):
    - Based on known properties of concrete
    - Or based on test on representative samples

---

2. Waste acceptance criteria

- Chemical content limitations on primary waste packages
  - Water soluble chlorides

    Chlorides can increase corrosion of rebars, thus disturbing the concrete barriers.
    - mass of chloride in waste ≤ 0,4 m% of cement mass

  - Sulphates

    Released sulphates can attack concrete barriers of repository.
    - maximum 12 g sulphates per kg of waste

---

2. Waste acceptance criteria

- Chemical content limitations on primary waste packages
  - Cellulose

    Degradation of cellulosic materials leads to the formation of complexing agents (e.g. ISA), which may lead to an increase of radionuclide release.
    - maximum 100g of cellulose per waste package

  - Water soluble chlorides

    Chloride can act as ligands, with a possible increase of radionuclide release.
    - mass of chloride in waste ≤ 0,4 m% of cement mass

Content

1. Introduction
2. Waste acceptance criteria for surface disposal
3. **Upstream characterization**
4. Control strategy
5. Challenges ahead

---

3. Upstream characterization

- Characterization prior to and during production!
  - Decrease of risk for production non-compliant to WAC
  - Less information → Analysis effort ↑ → Increased complexity

- Characterization of non-conditioned waste includes:
  - Radiological content
  - Chemical (and physical) content

- Characterization of conditioned waste is based on characterization of its content (non-conditioned waste)

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3. Upstream characterization

Characterization of conditioned waste is based on characterization of its content (non-conditioned waste) : **importance of tracking system!**

3. Upstream characterization

- **Physico-chemical characterization**
  - Driven by clear sorting procedures and training!!
    - → material specific waste packages
  - Organic compounds will be directed preferentially to the combustible waste (e.g. cellulose, complexing agents)
  - Material identification based on:
    - material inventory
    - visual inspection
    - sampling & analysis

3. Upstream characterization

- **Chemical characterization:** tools to support training

3. Upstream characterization

- **Radiological characterization**
  - Based on a combination of:
    - A. Measurement of ETM radionuclides
    - B. Scaling factors for DTM radionuclides (nuclide vector)

  - Approach is developed case-by-case for every relevant waste stream: mostly dependant on waste origin

3. Upstream characterization

- A. Measurement of ETM radionuclides / key nuclides
  - Most frequently used
    - Dose rate ("dose-to-Bq")
    - Segmented gamma spectrometry
    - Sampling and analysis

3. Upstream characterization

- A. Measurement of ETM radionuclides / key nuclides
  - Assess appropriate measurement technique for individual waste stream depending on
    - Desired detection limit
    - Nuclide vector (and "gamma ray signature"): e.g. variable ratio $^{60}Co/^{137}Cs$
    - Heterogeneity: e.g. assumption of homogeneity

3. Upstream characterization

- B. Scaling factors for DTM radionuclides (nuclide vector)
  - Starting point: - process knowledge (type nuclear activity, ...)
    - historical data (records, accidents, ...)
  - Level of documentation ↓ → Complexity ↑
  - Modeling: depletion / activation code [ORIGEN, FISPIN, SCALE, MCNPX]
    → Assumptions based on available data
  - Evaluation by sampling and analysis
    - Adjustment / fine-tuning of nuclide vector
    - Need for supplementary data?
    - Necessity of segregation/zoning in decommissioning

Slide 19

3. Upstream characterization

- B. Scaling factors for DTM radionuclides (nuclide vector)

- Scaling factors
  >> evaluation of scaling factors
  (case-by-case)

- WAC evaluation
  >> make sure all safety-relevant radionuclides
  are sufficiently covered for the waste stream

Graph legend: NPP's
other waste producer (fuel cycle, research)

Slide 20

Content

1. Introduction
2. Waste acceptance criteria for surface disposal
3. Upstream characterization
4. **Control strategy**
5. Challenges ahead

Slide 21

4. Control strategy

1. Administrative control (e.g. qualification)
   - During all stages of waste life cycle
   - Visual inspection (audit) prior to qualification/acceptation

2. Inspection prior to conditioning (non-conditioned waste)
   - Visual inspection and sampling: physico-chemical content
   - Segmented gamma spectrometry: radiological content
   - 10% of incoming waste

3. Control prior to disposal (conditioned waste)
   - Non-destructive test (NDT) / Destructive test (DT)
     - Frequency to be defined for individual waste stream / relevant WAC
   - Approach is currently subject of a study
     - Discussion and development ongoing

Slide 22

4. Control strategy

2. Inspection prior to conditioning (non-conditioned)

In case of specific findings or non-conformities:
→ Continuous improvement of WAC
→ Communication with waste producer
   - improvement of sorting/characterization procedure
   - supplementary cost in case of operational issues

Slide 23

4. Control strategy

3. Control prior to disposal (conditioned): *in study*
   - **Non-destructive tests (NDT) if possible** - high frequency
     - Hard gamma-ray radionuclides: (segmented) gamma spectrometry
     - Cement matrix and "voids": high energy X-ray tomography
     - Cellulose, ... : high energy X-ray - nuclear resonance fluorescence C/O

   - **Destructive test (DT) if needed**
     - Cl$^-$, SO$_4^{2-}$: sampling , leaching and analysis
     - Phenomena ASR/DEF: sampling and petrographic analysis
     - DTM radionuclides: sampling and radiochemical analysis

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Slide 24

Content

1. Introduction
2. Waste acceptance criteria for surface disposal
3. Upstream characterization
4. Control strategy
5. **Challenges ahead**

5. Challenges ahead

- Approach for waste produced before disposal criteria: pre-disposal control by random checks
  - Study goes on
  - Emphasis added on physical and chemical content!
- Necessity of an approach for historical waste with poor characterization: case by case
- Management of uncertainties in characterization
- Reconditioning of non-compliant waste?
  - E.g. thermal treatment will destroy all organic compounds
  - Characterization remains necessary!

WASTE ACCEPTANCE AND CHARACTERIZATION IN AN OPERATIONAL WASTE DISPOSAL FACILITY

SUMMARY

1. GENERAL OVERVIEW OF CSA : LOW AND INTERMEDIATE LEVEL WASTE - SHORT LIVED WASTE (LILW-SL)

2. ACCEPTANCE PROCESSES

3. MONITORING OF WASTE PACKAGE QUALITY BY ANDRA (NON DESTRUCTIVE CONTROLS)

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1. GENERAL OVERVIEW OF CSA

Basic Nuclear Installation
95 ha including
30 ha reserved for disposal

Date of commissioning : 1992

3 local implantation in Aube :
Brienne-Dépôt, Ville-sur-Aube, Epothémont

In 2018

10 812
9 540
8 166
11 367
343

Disposal capacity
1 000 000 m³

33,5 %

392 748

143

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1. GENERAL OVERVIEW OF CSA

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1. GENERAL OVERVIEW OF CSA

Barrier n°1 : waste package
Inside which a containment material coats the waste

Barrier n°2 : vault
Completed by the network of control galleries and the final cover

Barrier n°3 (natural) :
The geological environment

Vaults filled with concrete

Vaults filled with gravel

---

1. GENERAL OVERVIEW OF CSA

Number of waste packages
Volume in m3

Slide 1 — 1. GENERAL OVERVIEW OF CSA

Slide 2 — 2. WASTE ACCEPTANCE PROCESSES

The overall structure of the waste acceptance process includes the following phases :
- Authorization (of a site, for the approved waste stream)
  - The authorization is delivered after the determination and the share of operating/technical parameters that will be used in package production
    - Producer gives reference used to ensure the compliance of waste package with Andra's requirements at the end of production
- Acceptance (of a waste production of an approved waste stream produced on an authorized site)
  - Acceptance of delivery imposed, on the basis of quality-control requirements, that :
    - Find user gives confidence in its ability to produce packages according to operational / technical parameters
    - Operational / technical parameters are auditable (defining who does what, when, how) and sustainable

A document, called Compliance Matrix (MC) is gradually filled up during the process, and each stage of completion of the document is associated with a phase of the acceptance process. This document :
- Contains an exhaustive review of Andra specifications requirements,
- Contains an identification of derogations,
- Constitutes a quality recording related to the agreement and acceptance.

Slide 3 — 2. WASTE ACCEPTANCE PROCESSES

Filling stages of the compliance matrix

			MC0 stage (Andra)	MC1 stage (Producer)	MC2 stage (Producer)	MC3 stage (Producer)	MC4 stage (Producer)

Slide 4 — 2. WASTE ACCEPTANCE PROCESSES

Authorization instruction

Goal
- Limit the risk of non-compliant package production
- Ensure the detection of non-compliant package before delivery to the CSA
- Ensure "stability" of production over time

Stage 1 (MC1) → Stage 2 (MC2) → Stage 3 (MC3) → Authorization notification

Slide 5 — 2. WASTE ACCEPTANCE PROCESSES

Instruction of the acceptance

Goal :
- Decline in the production site applicable documentation:
  - The technical provisions (column 5 of the MC)
  - Andra organizational specifications (for example: those related to the provisions to be applied by the production site in case of producing a non-compliant package, or those related to reporting procedures and shipments)

Stage 4 (MC4) → Acceptance notification → Delivery

Slide 6 — 2. WASTE ACCEPTANCE PROCESSES

Once the initial approval and acceptance delivered by Andra, waste producer and Andra must define and perform a surveillance program to identify all the causes that could lead to the production or the shipment to the disposal site of a non-compliant waste package.

The surveillance program includes :
- Destructive control on waste packages at the waste producer's site and at the CSA after delivery
- Non destructive control on waste packages at the waste producer's site and at the CSA after delivery
- Surveillance missions at the waste producer's site
- Controls on the waste file transmitted before waste package delivery at the CSA
- Controls performed upon delivery at the CSA

Slide 1:

3. MONITORING OF WASTE PACKAGE QUALITY BY ANDRA (NON DESTRUCTIVE CONTROLS)

1. CONTROLS AND CONTROLS OBJECTIVES

Andra, as operator of the radioactive waste disposal facility, is responsible for protecting the interests covered by the environmental code. This responsibility of operator of the waste disposal facility includes monitoring actions, defined as Important Activities for the Protection of Interests (IAP), helping to control the quality of packages accept in its facilities.

As part of the monitoring arrangements established, Andra carries out checks on packages to verify that the packages received and disposed comply with the acceptance criteria of the facility and to identify non-compliance requirements or possible malfunctions.

The technical specifications (Waste acceptance criteria) for the management of waste packages constitute the reference for controls.

Controls can be performed on CSA or on external laboratories.

Slide 2:

3. MONITORING OF WASTE PACKAGE QUALITY BY ANDRA (NON DESTRUCTIVE CONTROLS)

⇨ **Visual and radiological controls (about 200 waste packages by year)**

Control objective

- Check the mass, the dimensions, the physical state of the package, in particular the respect of the confining nature of its envelope (cracks, depression, perforation, ...)
- Check the dose rate of the package and its level of surface contamination

Checks carried out on all waste packages taken for checks

Slide 3:

3. MONITORING OF WASTE PACKAGE QUALITY BY ANDRA (NON DESTRUCTIVE CONTROLS)

⇨ **Activity measures**

Control objective

- Check the activities declared by the producers
- Ensure the respect of the thresholds (Maximum Activity Limit, Ratios, Fissile material ...)
- Radiological capacity and long term safety of the CSA

Controls performed on all cylindrical geometries and parallelepipedal geometries of non-immobilized waste for gamma spectrometry, and only on 200 L drums for neutron measurements

Gamma spectrometry
About 200 controls by years

Passive / active neutron measurements
Punctually realized

Slide 4:

3. MONITORING OF WASTE PACKAGE QUALITY BY ANDRA (NON DESTRUCTIVE CONTROLS)

⇨ **Imagery : X radiography Low energy (about 60 waste package by year)**

Control objective

- Check the nature of declared waste, the absence of prohibited or restricted waste (massive pieces, liquids, Waste Electrical and electronic equipment, ...)
- Provide input data for activity measurements and their exploitation

Controls carried out on 200L drums to be compacted

Slide 5:

3. MONITORING OF WASTE PACKAGE QUALITY BY ANDRA (NON DESTRUCTIVE CONTROLS)

⇨ **Imagery : X radiography (High Energy) and Tomography (punctually realized)**

Control objective

- Check the nature of declared waste and the absence of prohibited waste
- Check the constituent elements and materials of the waste package (internal and external envelopes, internal configurations, natures and dimensions)
- Provide input data for activity measurements and their exploitation

Checks on immobilized waste packages of cylindrical geometries

Slide 6:

3. MONITORING OF WASTE PACKAGE QUALITY BY ANDRA (NON DESTRUCTIVE CONTROLS)

⇨ **Measurements of the release rate of ³H and/or ¹⁴C by the waste package (punctually realized)**

Control objective

- Check the release rate of ^3H (respect of the specified value of <2.0E⁴ Bq.t⁻¹.d⁻¹ for the ^3H) and/or ^{14}C of the waste package

Controls carried out on 200 L drums, concrete shells <2 m³ and non-injected metal boxes

Slide 1:

THANK YOU FOR YOUR ATTENTION

Slide 2:

DESTRUCTIVE CONTROLS

⇨ Inventory

Control objective

- Check the types of waste declared, the possible presence of restricted waste and the absence of prohibited waste.
- Provide input data for carrying out activity measurements and their exploitation.
- Eventually carry out samples for analyses or reconditionings of waste in geometry allowing the measurement of activity (all or part of waste conditioned in metallic boxes/are placed in a drum)

Controls carried out on the 200 L drums to be compacted and on metallic box of 5 m³ to be injected.

Slide 3:

DESTRUCTIVE CONTROLS

⇨ Coredrilling

Control objective

- Check the waste block (nature, homogeneity, quality of materials)
- Collect waste samples for radiochemical analyses and part of waste or materials from the packages to perform technical tests

Controls carried out on homogeneous waste packages (sludge, concentrates, incineration residues, etc.) and water circuit filters, immobilized.

Slide 4:

RADIOCHEMICAL ANALYSIS ON SAMPLES

⇨ Radiochemical analysis objective

- On homogeneous or assimilated waste
- Check the reported activities on a larger range of radionuclides than SPγ measurable on packages:
  - Not measurable by SPγ: α and β pure radionuclides
  - Measurable by SPγ: Radionuclides γ difficult to measure without prior chemical separation (interference, background compton, etc)
- Several measures to have access to a representative average and to be able to approach the global activity of the package
- Make sure the thresholds are respected
- Acquisition of data on the activities of long-lived RN or difficult to measure to allow to retire the ratios used by the producers during their declaration → control of the producer ratios (radiological capacity and long-term safety of the CSA)

Slide 5:

RADIOCHEMICAL ANALYSIS ON SAMPLE

⇨ Analytic methods

- Pre-treatment
  - Dissolution of the sample
  - Eventual chemical separation of the dissolved sample
    - Suppression of interferents
    - Sample concentration
- Several analytic methods are used
  - SPγ on liquid sample
  - SPγ from liquide sample
  - SPα on liquid sample
  - Liquid scintillation
  - Chemical measurements of RN by ICP-MS (Mass spectrometry Inductively coupled plasma) or SMA (accelerator mass spectrometry)
  - COT (Total organic carbon) on solid

Slide 6:

CHEMICAL ANALYSIS ON SAMPLE

⇨ Objectives of chemical toxic analysis

- Mainly on homogeneous or assimilated waste
- Chemical species inventory - comparison with producer declaration
- Reagent content with hydraulic binder - verification of abrasive of impact (package, radionuclide transfer, etc.)
- Presence of chelating elements

⇨ Analytical methods

- Elementary analysis - or ICP-MS
- Ionic species : anionic and cationic chromatography
- Sample isolation

Characterisation of conditioned Waste in Sweden
Topical Day Chance End User Meeting
21 march. 2019

Nuclear Sweden on the Map

- 8 (12) reactor units at 3 sites
- 40 % of total electricity supply is nuclear
- 12,000 tonnes of spent fuel

- **SFR** – Final Repository for Short-lived Radioactive Waste
- **Clab** – Centralized Interim Storage for Spent Nuclear Fuel

Radioactive Waste Management System

SFR – Final Repository for LILW

SFR today

Planned extension

Waste Acceptance Criteria – SFR

General
- Construction
- Weight
- Marking

Radiological
- Radionuclides, activity content
- Surface dosrate
- Surface contamination
- Radiation impact
- Homogenity

Waste Acceptance Criteria – SFR

Chemical and physical
- Composition and structure
- Homogenity
- Hydraulic quality
- Temperature
- Free liquids
- Gas formation
- Fire resistance
- Chemical reactivity (Complexing agents)
- Leaching
- Environmentally hazardous materials

Mechanical
- Strengt
- Inner mechanical stability
- Corrosion resistance

Waste Packages – SFR

Concrete tank
Steel Container
Concrete mould
Steel mould
Steel Drums

Example from Forsmark NPP
Characterisation of Conditioned Waste

HPGe
- Free release detector 30 - 50 % efficiency, long measurement time
- For active waste < 15 % efficiency

- Detectors has a Canberra **factory characterization**

- Software from Canberra: **APEX** counting system

- Efficiency calibration is performed by using **ISOCS**

- Distance between detector and waste package normally 3 meters, up to 10 meters for ISO containers

---

Surface contamination

Free release levels of surface contamination as an average over maximum of 0,03 m²
- 40 kBq/m² for other beta and gamma emitting nuclides
- 4 kBq/m² for alpha emitting nuclides

Radiation Protection staff are responsible for surface contamination checks by:
Smears/Joint Survey and
Gamma radiation monitor box

SVAFO – A Legacy Waste Challenge

- Ownership of all waste at the Studsvik site 1991 transferred to NPPs
- Waste handled and characterised according to old standards
- Limited additional efforts were foreseen
- A handfull of drums deviating from the standard types corrected
  Odd dimensions / high dose rate / suspected liquid content
  ➢ Severe Pu/Am contamination
  ➢ Documentation not trustable
  ➢ Waste handling halted

Were there more drums with surprising content in our inventory?

Further NDT needed!

SVAFO – A Legacy Waste Challenge

To verify that the 20 drums remaining were the only ones containing liquids a RTR method was selected

➢ Labour intensive
➢ 450 kV/10 mA X-ray tube – limited to ~30 cm concrete
➢ 4 axis scanning
➢ Limited throughput
➢ Great (or bad) results

7 303 drums handled in 2009-2011

SVAFO – A Legacy Waste Challenge

As the drums were RTR'd the following parameters were also verified and measured

➢ Weight
➢ ID number
➢ Gamma nuclides (ISOCS)
➢ Dose rate

The findings in the RTR-project has changed the view on SVAFO's legacy waste

- All SVAFO's waste is subject to further studies, handling and characterisation – a huge undertaking.
- Earlier time schedules and cost calculations will be re-evaluated

Thank you for your attention !

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Slide 1:

CHANCE

Characterization of Conditioned Nuclear Waste
for its Safe Disposal in Europe

Topical Day CHANCE
Recent Developments in Calorimetry for Nuclear Waste
Characterization

Christophe Mathonat (KEP Nuclear), Holger Tietze-Jaensch (FZ Juelich), Bart Rogiers (SCK•CEN), Wojciech Kubinski (WUT), Cédric Carasco (CEA)

This project has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 755371

Slide 2:

CHANCE — Outline

WP3 Leader: KEP Nuclear – Contributors: CEA, FZJ, SCK•CEN, WUT

Outline / synopsis:
- Calorimetry, methods and merits
- LVC CHANCE development
- Experimental plan
- Calorimetry and NDA methods
- Numeric modelling support
- Conclusion and outlook

Slide 3:

CHANCE — Calorimetry

- Calorimetry is a technique for measuring the **thermal power** produced by the nuclear decay of any radioactive material
- It is a **non destructive assay** used mainly for **tritium, plutonium and americium** quantification based on the knowledge of the isotopy.
- The item to be studied is placed in the calorimeter measurement chamber and the heat flow at equilibrium, that is, the thermal power, from the item is determined by temperature sensors and associated electronic equipment
- The thermal power emitted by a test item
  is directly related to the quantity
  of the radioactive material in it

Slide 4:

CHANCE — Calorimetry

- Heat flow calorimetry is best suited for steady-state measurements, when the samples has a constant heating rate, like nuclear samples. A heat-flow calorimeter configuration is most commonly used for nuclear material characterisation

  Object heating

  Object at 34°C put in the calorimeter field at 35°C

  Heat released by the sample
  ↓
  Heat-flow Φ through the heat sensors
  ↓
  Temperature difference between the heat sensors/°C
  ↓
  Potential / µV
  ↓
  Heat / mW

Slide 5:

CHANCE — Twin-cell design

- In order to reduce the experimental errors from thermal noise in the heat sink, the twin design was introduced in calorimetry based on thermopiles

  Heat flow of the measurement cell
  Heat flow of the reference cell
  Differential calculation

  Measuring cell   Reference cell

- In calorimeters designed for the characterisation of large samples (> 100 L), the implementation of a ghost reference cell placed below the measurement sample and with the same thermal capacity is favourable

Slide 6:

CHANCE — Energy deposit

Pure Beta emitters:
^{3}H, ^{14}C, ^{36}Cl ...

Pure beta emitter

$n \rightarrow p + e^-$

β particle emission

Alpha emitters
^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am ...

Heavy nucleus

Main type of decay

Spontaneous fission

α particle emission

- Calorimetry is sensitive to all heat sources, thus RN inside a waste drum, as all radiation emitted turns into heat
- Charged particles deposit all their energy inside the surrounding material (unless they escape the calorimeter)
- The energy deposit induce heating
- Linear relationship between thermal power and alpha (or beta) activity

Slide 1 — Applications

CHANCE — Applications

- Alpha emitters: trans-uranium: Pu and isotopes, ^{241}Am : calorimetry + gamma spectrometry
- Pure Beta emitters : ^{3}H, ^{14}C, ^{36}Cl, ^{63}Ni, ^{90}Sr: calorimetry is the unique non destructive method
- In the frame of CHANCE, the experimental task of calorimetry NDA is:
  - to establish or rule out the existence of RN that are not easily detectable by either gamma or neutron spectrometry
  - namely ^{3}H, ^{90}Sr/^{90}Y, ^{106}Ru/^{106}Rh, ^{238}Pu, ^{243}Am, ^{244}Cm)
  - hidden/shielded gamma or neutron source such as shielded ^{60}Co or ^{99}Mo/^{99}Tc or even ^{252}Cf (and hidden alpha sources, of course)
- CHANCE progress aim:
  - large and heterogeneous waste drums
  - multi-radio-nuclide content
  - quantitative assay with integrated numerical modelling support

Slide 2 — Tritium example [1]

CHANCE — Tritium example [1]

- The mass (m) of tritium is calculated from the measured thermal power of an item (W_{item}) :

$$m = \frac{W_{item}}{P_i}$$

- P_i for tritium = 324 mW / g
- Test of a sample containing a certified amount of tritium
  - 35.9858 TBq at the test date
  - Shielded metallic container
- KEP LVC580 calorimeter used for the test
  - Vessel volume equal to 25L
- Result
  - Measured value in mW equivalent to 35.9644 TBq.
  - Relative error 0.06%

[1] K. M. Song et al., Fusion Engineering (2009)

Slide 3 — Plutonium and Americium example [2]

CHANCE — Plutonium and Americium example [2]

- The mass (m) of Pu and ^{241}Am is calculated from the measured thermal power of an item (W_{item}) using the following relationship:

$$m = \frac{W_{item}}{\sum P_i \times R_i}$$

- P_i is the specific power of each isotope (Plutonium and Americium isotopes)
- R_i is the ratio of each isotope determined by gamma spectrometry
- The mass m from the ith isotope is $m_i = m \times R_i$

Slide 4 — Plutonium and Americium example [2]

CHANCE — Plutonium and Americium example [2]

Sample: Plutonium
We assumed the following composition to calculate the power emitted by 1 g of Calex working reference :

Isotope	Fraction [wt. %]
^{238}Pu	0.01
^{239}Pu	93.73
^{240}Pu	5.85
^{241}Pu	0.3712
^{242}Pu	0.03
^{241}Am	0.061

Isotope	Mass (g)	Specific power (mW/g)	Power (mW)
238 Pu	0.0001	567.57	0.0567
239 Pu	0.9373	1.9288	1.8078
240 Pu	0.0585	7.0824	0.4143
241 Pu	0.003712	3.412	0.0126
242 Pu	0.0003	0.1159	~0
241 Am	0.00061	114.2	0.6966
TOTAL	1 g		2.99 mW

0.6 mW corresponds to 200 mg of Pu

ASTM C1458 Standard Tests Methods for Nondestructive Assay of Plutonium, Tritium and 241Am by Calorimetric Assay

B. Srinivasan K. J. Mathew U. I. Narayanan W. F. Guthrie T. E. Sampson, J Radioanal Nucl Chem (2009) 282:963–970

Slide 5 — LVC CHANCE

CHANCE — LVC CHANCE

- Development of a new 200 liters calorimeter with optimized lower detection limit

- Opening in two half shell
- Ghost reference cell underneath the drum to be measured

Slide 6 — LVC CHANCE

CHANCE — LVC CHANCE

1,2 m max.
1,1 m

CHANCE — handling completed

CHANCE — LVC CHANCE

- Development of a new insertion system and a new reference chamber makes it possible to insert a reference with thermal properties similar to the sample to be studied

  Insertion system
  Reference chamber

- Insertion of the drum in the calorimeter by means of grippers or clamping system

CHANCE — Calorimeter under construction

CHANCE — Transport of the calorimeter

CHANCE — Experimental investigation

- Experimental investigation with LVC CHANCE will measure plutonium and other possibly hidden RN in realistic cases with 200L mock-up waste drums and real unconditioned waste drum at CEA Cadarache and SCK.CEN

CHANCE — Experimental investigation

- Use of a mock-up drums with a plutonium source in a -concrete matrix

  Mock-up with concrete matrix from CEA
  522 kg
  92 cm high
  58 cm diameter

- Measurements in Cadarache of a 200 L drum with plutonium sources
  - Concrete drum ready: neutron & γ ⇒ 2018 (CEA), calorimetry ⇒2019 (KEP)
  - Experimental hall DANAIDES cleared to welcome KEP calorimeter (CEA)
  - Calorimeter implementation in DANAIDES studied by CEA with KEP

Slide 1: Experimental investigation

At SCK•CEN, the experimental program will consist of maximum six configurations with well-defined matrix and radiological content (PuO_2 pins).

In order of priority:
- A real unconditioned waste drum
- Effect of matrix:
  "Homogeneous" source distribution inside
  - A new mortar mock-up
  - An existing mock-up with ethafoam
  - An existing mock-up with ethafoam, PVC and steel
- Effect of heterogeneity:
  The new mortar mock-up with
  - The largest-activity source in the center
  - The largest-activity source at the border

Slide 2: Effect of source distribution

γ vs. n emission

- The gamma ray attenuation is mainly determined by the matrix density, but is poorly affected by matrix type
- Neutron attenuation is very sensitive to the amount of hydrogen in the matrix. Thus passive measurement cannot be envisaged for matrices with high hydrogen concentration (intense incoherent neutron scattering)
- For other types of waste, passive neutron counting is a valuable option as long as density stays below 1g.cm^{-3}
- Most considered matrices are challenging for HRGS at their true density
- For PNCC, bitumen and polyethylene are more challenging because of their hydrogen content

Slide 3: Effect of source distribution

- For assessing the effects of the source distribution within a drum, a geometry with a minimum distance of 1 cm between the sources and the inner surface of the drum, and a drum thickness of 1 mm
- 7 radial and 3 axial locations for Plutonium sources

3 sources configurations

Most conservative   Least conservative   homogeneous

Slide 4: Effect of source distribution

- The comparison between the different geometries reveals that the uncertainty related to the exact position of sources in a drum can be large
- For bitumen matrix, the ratio of gamma vs. neutron emission rates is more or less constant, and the effect of source position is comparable to that of the matrix density
- For the other matrices, if the density is more or less known, the ratio of gamma versus neutron emission rates can provide information on the sources position. For bitumen it cannot

Slide 5: Calorimeter MC Modelling

- Compile generic data of calorimeter geometry and material properties of generic mock-up drum & set-up numerical tests and computational environment
- Estimate escape radiation vs. encapsulated and heat depositing radiation, thus reduce lower detection limit
- Establish and improve uncertainties from either part
- Estimate effect of additional absorber material to trap heat depositing radiation
- Listen to Wojciech Kubinski's talk for further interesting details on the numerical modelling
- Some results summarized here:

Slide 6: results γ

Example of a ^{60}Co source: 1.173 MeV and 1.333 MeV

γ Most conservative γ	Energy [%]	Rel. Error [%]	Homogeneous γ	Energy [%]	Rel. Error [%]
Deposited	96	< 1	Deposited	87	< 1
Detected	87	< 1	Detected	73	< 1

« Most conservative »   homogeneous

Slide 1

CHANCE — Results n, α

Example of a 5.2 MeV mono-energetic neutron source

Most conservative n	Energy [%]	Rel. Error [%]	Homogeneous n	Energy [%]	Rel. Error [%]
Deposited	98	< 0.5	Deposited	95	< 0.5
Detected	94	< 0.5	Detected	78	< 0.5

« Most conservative »

homogeneous

Slide 2

CHANCE — Simplified MCNP model **gamma flux distribution**

a) Flux along z-axis   b) Flux along x-axis

For details see talk presented by Wojciech Kubinski

- Flux – number of particles per source particle per cm2
- Source – 0.1, 1 and 5 MeV homogenously sampled particles

Slide 3

CHANCE — Real case Pu sample simulation

Alpha simulations – real case Pu sample
most conservative source

composition:

Isotope	Fraction [%]	Normalised [%]	Specific power [mW/g]	Specific power contribution [mW/g]
^{238}Pu	1.35	1.30	568.00	7.372
^{239}Pu	86.54	83.88	1.93	1.232
^{240}Pu	23.22	22.28	7.10	1.582
^{241}Pu	4.66	4.28	3.30	0.141
^{242}Pu	4.43	4.25	0.10	0.006
^{241}Am	4.2 % of Pu	4.03	115.00	4.635
			Total	15.0

- Calculated power: (14.4 ± 0.2) mW/g
- 99.99% of the radiation is deposited inside the drum (so 99.99% is potentially detected)
- As a first approximation, ^{241}Pu being mainly a beta emitter (98%) it was omitted in this simulation (it'll be taken into account in the next simulations).
- A part of the gap between the two values is due the ^{241}Pu contribution omission (about 0.14 mW/g).

Slide 4

CHANCE — Modelling conclusion

Particle	source	Deposited energy [%]	Relative error [%]	Detected energy [%]	Relative error [%]
gamma	Most conservative	90.4	0.6	78.9	0.6
	Homogenous	77.2	0.7	47.6	0.7
neutron	Most conservative	97.7	0.4	94.4	0.4
	Homogenous	95.0	0.1	78.3	0.1
Alpha	Most conservative	100.00	0.00003	100.00	0.00001
	homogenous	100.00	0.00003	100.00	0.00003

- In case of gamma and neutron radiation:
  - from 2% up to about 25% of the energy escaped the system
  - from 50% up to 95% was detected
  - A part of the flux goes through the ghost cell and the aluminum block which cause in an additional bias in mass measurements
- For alpha radiation 99.98% of the radiation is deposited inside the drum (all the heat is potentially detected)

Slide 5

CHANCE — Conclusion and Outlook

- Existing calorimeters :
  - Measurement of drums from 3.3 to 380 liters
  - Easy to use
  - Power range : from 100 µW to 100 W (from few mg to tens grams of tritium and from grams to kilograms of Pu)
- Calorimetry measures the radioactive heat flux
  - Calorimetry measures all heat emitted from the drum and deposited inside the calorimeter
  - Can't miss any heat emission / deposition (above detection limit and except escaping particles)
- New target for calorimetry is to focus on multi-radionuclide compounds for instance in bituminous and cemented waste drums with quite hydrogenous matrix material.
- Numerical modelling makes possible to properly analyze the data and therefore reduces the detection limits and better quantifies the uncertainties.

Slide 6

CHANCE — Conclusion and Outlook

- Calorimetry is a power tool complementary and supplementary to gamma- and neutron spectrometry
- Calorimetry measures all heat deposited inside the drum whatever the source distribution
- Calorimetry is independent from type of matrix and density
- Calorimetry cannot discriminate / locate the heat source(s)
- Calorimetry is simple to set up and to use

- Task 3.2 Experimental investigation
  - Calorimetric measurement in CEA starting in June 2019
  - Calorimetric measurement in SCK•CEN starting 3rd trimester of 2019

Slide 1

Topical Day CHANCE | characterization of conditioned radioactive waste: status, challenges and new developments

Muon tomography | needs and challenges from a safeguards perspective

Darius Ancius, EC DG ENER and Paolo Checchia, INFN, Italy

Lorenzo Barcellan, Massimo Benettoni, Franco Gonella, Fabio Montecassiano, Gianni Zumerle (Istituto Nazionale di Fisica Nucleare (INFN)),
Sara Vanini, Andrea Paccagnella (Università di Padova);
Nicole Erdmann, Konrad Shoop, Peter Schwalbach (EC DG ENER);
Katharina Aymanns (Forschungszentrum Jülich);
Detlef Jung, Thomas Lamprecht, Bernard Lukas (Kernkraftwerk Neckarwestheim).

March 21, 2019 Lamot conference centre, Mechelen, Belgium

Slide 2

Contents

- *Spent nuclear fuel in EU Member States (MS)*
- *Existing Safeguards of spent fuel*
- *Safeguards of Dry Storage Casks: challenges*
- *Muon detection techniques*
- *Field test with INFN muon tracker*
- *Conclusions*

European Commission

Slide 3

Spent nuclear fuel in EU

Status	Power reactor	Research reactor	Geological repository
In operation	127	36	-
In construction or being licensed	4	2	2
Planned or under discussion	21	?	12
Shutdown or decommissioned	93	151	-
Total:	245	189	14

Sources:
15.5.2017 COM(2017) 236),
IAEA power and research reactor DB,
www.world-nuclear.org

European Commission

Slide 4

Spent fuel in EU: Quantities

66 400 tHM* of Spent Nuclear Fuel in EU MS (end of 2018)

=

~ 750 t Pu
~ 620 t ^{235}U

Significant quantity (SQ) – the approximate amount of nuclear material for which the possibility of manufacturing a nuclear explosive device cannot be excluded.

e.g. 1SQ = 8 kg of Pu, or
= 75 kg of high enriched, ^{235}U ≥ 20%

(52 kg bare sphere of pure ^{235}U makes the critical mass)

DRY STORAGE
10 – 12 SQ in 1 CASK

WET STORAGE & Reprocessing Plants
Many SQ !

tHM* – tons of Heavy Metal

European Commission

Slide 5

Dry Storage: Safeguards

FORK detector

Fuel assembly loading into cask verification by EURATOM

Based on burn up calculations and prediction of neutron and gamma count rates

European Commission

Slide 6

Dry Storage: Safeguards

Electronic seal

Containment and surveillance

Video Surveillance 24h/24H

Ultra-sonic seal bolt

"Cobra" seal

NEW!
3D laser seal

European Commission

Safeguards challenges: Loss of continuity of knowledge, resources

- Continuity of Knowledge (CoK) – basic principle of Safeguards
- Number of dry storage casks in EU MS **today:1547**
- Storage time – up to 100 years!

Possible loss of CoK, Significant EURATOM's human resources, Doses to EURATOM Inspectors.

- *SOLUTION:* timely detection of possible diversion – NDA verification of Nuclear Material in heavy shielded casks

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NDA of nuclear material in Dry Storage Casks: possible novel techniques

- *Muon tomography (muography)*
- *Fast neutron camera*
- *Laser induced pulsed neutron source and induced fission*
- *Gamma tomography*

Muography

Field test: INFN muon tracker

- A small prototype: 8 layers of 8 Al tubes: 50 mm diameter, 1.5 mm thickness, central 100 μm anode wire (+3000 V)
- FE electronics : signal amplification and shaping
- RO chain: time digitization, trigger and remote transmission

Calibration with CMS chambers:

Testing:
- **Performance of drift tubes in radiation environment**
- **Proof that the muons that crossed CASTOR could be tracked**

Field test: Neckarwestheim site

Field test: measurement positions

The detector in data-taking positions (Left) far from and (Right) near the CASTOR®.

Field test: measurement background

Relativistic muons cross the detector in 1.3 nS

The drift time in tube is 1µs

Some photons from SF may hit the tube and create noise within 1.5µs data acquisition window

14 µSv/h

neutrons 25 µSv/h

European Commission

Field test: recorded muon tracks

Neckarwestheim, 2 – 6 July 2018

The number of additional hits due to γ interaction in coincidence with a cosmic muon does not disturb the track reconstruction

They represent <1% of total hits

CASTOR

Field test: Results

- Muon tracks can be successfully recorded at proximity of a loaded CASTOR

- Asymmetrical azimuthal and elevation distribution of muon tracks prove that muons that have crossed CASTOR were recorded

- Larger scale muon trackers to measure in and out muons are feasible

Azimuthal Angle

Elevation Angle

European Commission

Field test: Future plans

- Testing 1/3 full-scale trackers

- Validation of Monte Carlo simulations

- Production of tomographic images

- Prove that the absence of 1 SQ (1 PWR or 4 BWR assemblies) can be detected

Geant4 simulation 360° coverage

1/3 full-scale tracker

European Commission

Conclusions

- Presently, Safeguards inspectorates face challenges due to increased amounts of spent fuel in EU MS. No feasible traditional NDA technique could be used for verification of NM in dry storage casks.

- Some novel technologies can be potentially used for verification purposes.

- Muography is a source-free technique, which is well advanced.

- Simulations and field tests show high potential of muography for dry storage cask verifications

- The content and configuration of SF in dry cask is supposed to be well known initially – thus one looks for any deviations from known configurations

European Commission

3

LINAC-BASED HIGH ENERGY X-RAY IMAGING OF RADIOACTIVE WASTE

Bertrand PEROT

CEA DEN Cadarache, Nuclear Measurement Laboratory

On behalf of Nicolas ESTRE, Léonie TAMAGNO, David TISSEUR, Eric SIMON, Bernard CORNU, Marc KISTLER, Emmanuel PAYAN, Daniel ECK

www.cea.fr

CHANCE TOPICAL DAY | MECHELEN, BELGIUM | 21 MARCH 2019

OUTLOOK

❑ The Nuclear Measurement Laboratory

❑ From gamma to high-energy X-ray imaging

❑ Imaging of radioactive waste packages

❑ Ongoing developments and R&D
  ➢ New LINACs
  ➢ High-energy photon detectors
  ➢ In situ imaging system
  ➢ Dual energy imaging

❑ Conclusion and prospects

| PAGE 2

CEA CADARACHE

Largest European center involved in low carbon energy research

1600 hectares (900 enclosed)

CEA sites

Cadarache

Agencies in Cadarache

	cea	orano	IRSN	iter
people →	2400	810	330	5000 soon

| PAGE 3

CEA CADARACHE MAJOR STRATEGIC MISSIONS

❑ *Preparing tomorrow's reactors*

❑ *Providing support for the current French nuclear infrastructure*

❑ *Promoting alternative energies to replace fossil-energies*

❑ *Exploring fusion energy for the near future*

| PAGE 4

THE NUCLEAR MEASUREMENT LABORATORY

❑ 30 permanent staff and 5 to 10 non-permanent staff (PhD...)
❑ 50 different projects and collaborations

| PAGE 5

NUCLEAR NON-DESTRUCTIVE ASSAY @ LMN

❑ **Radiological characterization**
  ✓ Gamma-ray spectroscopy
  ✓ Passive and active neutron measurements
  ✓ Photofission

❑ **Physical characterization**
  ✓ Photon radiography and tomography
  ✓ Neutron radiography

❑ **Elemental characterization**
  ✓ Thermal neutron activation analysis (PGNAA)
  ✓ Fast neutron activation analysis (APT)

| PAGE 6

1

APPLICATIONS OF NUCLEAR NDA @ LMN

- **Nuclear fuel cycle**
  - ✓ Uranium mining, enrichment, fuel fabrication & reprocessing
  - ✓ Radioactive waste characterization
  - ✓ Dismantling and decommissioning

- **Reactors**
  - ✓ Contamination transfers in circuits and components
  - ✓ Non-destructive exams for experimental reactors like JHR
  - ✓ Nuclear accident studies and monitoring

- **Homeland Security and Safeguards**
  - ✓ CBRNE threats detection
  - ✓ Nuclear material controls

| PAGE 7

THE NUCLEAR MEASUREMENT LABORATORY

GAMMA AND HIGH-ENERGY X-RAY IMAGING

NEW LINACS AND DETECTORS

DUAL ENERGY IMAGING

CONCLUSION AND PROSPECTS

PHOTON TRANSMISSION RADIOGRAPHY

Detection system

Photon source

| PAGE 9

PHOTON TRANSMISSION DENSITOMETRY

Source Φ_0 Φ Detector

L

$$\Phi = \Phi_0 \times \exp[-\mu(E) \times L] = \Phi_0 \times \exp\left[-\frac{\mu(E)}{\rho} \times \rho \times L\right]$$

$$\Rightarrow \boxed{\rho = \frac{\ln(\Phi_0/\Phi)}{\frac{\mu(E)}{\rho} \times L}}$$

ρ density (g.cm^{-3})

$\mu(E)/\rho$ mass attenuation coefficient (cm^2 g^{-1}) relatively independent on materials for 1.17 + 1.33 MeV γ in ^{60}Co imaging

LINAC

^{60}Co source $\overline{E}\gamma = 1,25$ MeV

| PAGE 10

PHOTON TRANSMISSION TOMOGRAPHY

Projections >1000 on 360°

Reconstructed 2D cross section

| PAGE 11

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^{60}CO IMAGING: PRECISE DENSITY BUT VERY LONG

TRANSEC, legacy ^{60}Co imaging (CHICADE facility, Cadarache)

50 mm   120 mm   190 mm   260 mm
330 mm   400 mm   470 mm   540 mm
610 mm   680 mm   750 mm   820 mm

Density
0   4

| PAGE 12

LINAC-BASED HIGH ENERGY X-RAY IMAGING

CINPHONIE, high energy X-ray imaging (CHICADE, 2013)

CINPHONIE Vs. TRANSEC:
- Radiography: 240 x faster (40 h → 10 min)
- Tomography: 36 x faster (15 h → 25 min)

Mirror

Bremsstrahlung X-ray beam

LINAC   Lead collimator   Object (waste drum...)

Scintillation plate   CCD camera

9 MeV LINAC, 15 Gy/min @ 1m   Up to 5 tons platform   Scintillation plate 800x600 mm² Gadox Gd₂O₂S ; 1.5 mm

N. Estre, D. Eck, J-L. Pettier, E. Payan, C. Roure, E. Simon, High-Energy X-Ray Imaging Applied to Non Destructive Characterization of Large Nuclear Waste Drums, IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 62, NO. 6, DECEMBER 2015 | PAGE 13

HIGH ENERGY IMAGING OF LARGE CONCRETE DRUMS

Concrete waste (density > 2)
Less than 1 s per single radiography

- Radiography
- Tomography

1,2 tonne
H=1,2m
⌀=0,84m

0°

N. Estre, D. Eck, J-L. Pettier, E. Payan, C. Roure, E. Simon, High-Energy X-Ray Imaging Applied to Non Destructive Characterization of Large Nuclear Waste Drums, IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 62, NO. 6, DECEMBER 2015 | PAGE 14

DETAILED EXPERTISE OF A LARGE CONCRETE DRUM

500 l waste blocked in a concrete matrix
(external volume 1.2 m³)

Radiographies (duration 10 min)   2D/3D Tomographies (duration 30 min)

3D waste checking

2.7 t
H=150 cm
⌀120 cm

| PAGE 15

COMBINED USE WITH OTHER NDA TECHNIQUES

X-ray of a 220 L drum filled with technological waste → MCNP model for gamma-ray spectroscopy and neutron measurements interpretation

homogeneous matrix of light waste up to 58 cm filling height

Dense metallic objects at the bottom of the drum

B. Pérot, C. Jallu, C. Passard, O. Gueton, P.G. Allinei, L. Loubet, N. Estre, E. Simon, C. Carasco, C. Roure, L. Boucher, H. Lamotte, J. Coste, M. Bertaux, P. Fichet, F. Carrel, A. Lyoussi, The characterization of radioactive waste: a critical review of techniques implemented or under development at CEA, France, EPJ Nuclear Sci. Technol. 4, 3 (2018) 1-24 | PAGE 16

OTHER EXAMPLE OF IMAGING + GAMMA SPECTROSCOPY

1.2 m³ drum with 3 filters blocked in a concrete matrix

Eq. Dose Rate (µSv/h)

Average dose rates across from filters (purple dots surface proportional to dose rate)

Numerical model and Monte Carlo simulation

internal filter checking

⇒ Measured activity of gamma emitters consistent with expectations (expertise)

| PAGE 17

THE NUCLEAR MEASUREMENT LABORATORY

GAMMA AND HIGH-ENERGY X-RAY IMAGING

→ NEW LINACS AND DETECTORS

DUAL ENERGY IMAGING

CONCLUSION AND PROSPECTS

SATURNE: A NEW MULTIPURPOSE HIGH ENERGY LINAC

- Replaces the legacy 9 MeV MINILINATRON in CINPHONIE (see slide 13)
- SATURNE LINAC ⇒ 150 Gy/min @ 1m, E_{min} = 9 MeV up to E_{max} = 20 MeV
- Commissioning underway with Thales
- ⇨ Imaging of large concrete waste packages
- ⇨ "Super-controls": radioactive waste package expertise for ANDRA
- ⇨ R&D on dual-energy imaging: 9 MeV / 20 MeV
- ⇨ R&D on photofission: 15 MeV

| PAGE 19

COMPARATIVE STUDY OF SCINTILLATOR SCREENS FOR LARGE FIELD OF VIEW DETECTOR

Scintillator	Density (g/cm³)	Thickness	Configuration
Medex, GOS	7.32	750 µm	On 2 mm brass plate
Lanex, GOS	7.32	290 µm	See [11]
CsI	4.51	2 mm	On 2 mm Cu plate
BGO	7.13	3 mm	
LYSO	7.25	25 mm	Blackened surfaces other than exit surface
GLO	9.10	1.58 mm	

Scintillator name	Surface density in mg/cm²	Light yield (photons/MeV)	Optical index
Medex	347	65000	
Lanex Fast Back	134	65000	
CsI	902	54000	
BGO	899	8000-10000	2.15
LYSO	13500	30000	1.82
GLO	1438	55000	1.89

Setup (see slide 13)

Spatial resolution vs. sensitivity

⇒ **2 mm CsI is a good tradeoff**

D. Tisseur, H. Estre, L. Taragno, C. Biron, D. Eck, E. Payan, H. Chrespy, Performance evaluation of several well-known and new scintillators for MeV X-ray imaging, NSS 2016, 10-17 November 2016, Sydney, Australia | PAGE 20

HIGH ENERGY RADIOSCOPY

- **Dynamic radiography**
  - Originally one camera with 50 pictures / sec and 230 × 240 pixels
  - Now 4 cameras ⇒ up to 200 pictures / sec and 2560 × 2160 pixels
- **Application to corium / water interaction**
  - ⇨ MC3D code validation (nucl. accident studies)

Water surface

In yellow: 9 MeV LINAC
In black: imaging plate

H. Estre, E. Payan, L. Berge, Fast Megavoltage X-Rays Radioscopy, Nuclear Science Symposium, 29 Oct. – 4 Nov. 2016, Strasbourg, France. | PAGE 21

LINEAR DETECTORS: LEGACY SYSTEM AT LMN

Segmented and collimated CdTe array

- 25 CdTe detectors (25 mm length)
- Attenuation up to 5 decades (40 cm steel)
- Radiography and 2D plane tomography
- 3D helical tomography
- 0.5 to 2 mm spatial resolution
- 1 h (2D) to several days (3D) acquisitions

Collimated CdTe detectors

Simulations of a 5 m³ concrete waste package

9 MeV LINAC

15 MeV LINAC

H. Estre, D. Eck, J.L. Rother, E. Payan, C. Roure, E. Simon, High-Energy X-Ray Imaging Applied to Non Destructive Characterization of Large Nuclear Waste Doses, IEEE TRANSACTIONS ON NUCLEAR SCIENCE, VOL. 42, NO. 6, DECEMBER 2015. | PAGE 22

COMPARISON OF LINEAR DETECTION SYSTEMS

- CdWO₄ needle scintillators with optical fibers and photodiodes

- Flat CsI scintillators with X-ray beam reading on the edge, coupled to visible cameras

CsI sample

X-ray beam

Relative signal of the linear detectors

H. Estre, D. Eck, C. Biron, M. Kistler, E. Payan, L. Taragno, D. Tisseur, Design of a very efficient detector for High Energy Tomography, NSS 2016, 10-17 November 2016, Sydney, Australia | PAGE 23

MODUPIX FLAT DETECTOR FOR ON-EDGE IRRADIATION

- TIMEPIX chip (CERN)
- 256 × 256 pixels (55 µm x 55 µm)
- 14 bits (stops at 11810 counts)
- Acquisition frequency up to 850 fps
- No dark current
- Photon counting mode (spectroscopy) or charge integration mode
- Coupled to Si or CdTe substrates

Tests in view of JRH imaging system (see next slide: 100 µm resolution objective on the object)

9 MeV beam

W collimator with a 0.4 mm

MODUPIX

- ~ 150 µm measured detector resolution (Gaussian FWHM) with 1 mm CdTe
- 75 µm ultimate detector resolution objective ⇒ ~ 0.1 mm Si substrate needed

| PAGE 24

Slide 1: JHR UNDERWATER HIGH ENERGY X-RAY IMAGING

- **High resolution mechanics** to move the JHR test devices in 3D
- **Highly collimated X-ray camera** WidePIX semi-conductor sensor (~ 50 µm x 50 µm pixels)

"ORIATRON" 6 MeV LINAC ~ 500 µm focal spot

⇒ Very high resolution objective ~ 100 µm to observe cracks, corrosion...

C. Roure, B. Cornic, E. Simon, H. Estre, et al...Non-Destructive Examination Development for the JHR Material Testing Reactor, ANIMMA 2013, doi:10.1109/ANIMMA.2013.6728002, 23-27 June 2013, Marseille, France.

| PAGE 25

Slide 2: TOMIS : A TRANSPORTABLE TOMOGRAPH IN DEVELOPMENT

- "TOMIS" French Project with CEA and THALES (PIA ANDRA framework) https://www.andra.fr/sites/default/files/2018-06/Fiche%20projet%20TOMIS%20VF.pdf
- Self-shielded LINAC under development with Thales
- 30 Gy/min @ 1 m (100 mA, 200 Hz)
- 6, 9 and 15 MeV
- ⇒ *In situ* imaging of large concrete waste packages
- ⇒ *In situ* Dual Energy Tomography

Legacy waste, clean-up and dismantling activities

Slide 3

THE NUCLEAR MEASUREMENT LABORATORY

GAMMA AND HIGH-ENERGY X-RAY IMAGING

NEW LINACS AND DETECTORS

➡ **DUAL ENERGY IMAGING**

CONCLUSION AND PROSPECTS

Slide 4: DUAL ENERGY IMAGING

- **Single-energy tomography** → density ρ of a material

Low Energy Tomography
Linear absorption coefficient μ_{BE} (cm⁻¹)

- **Dual-energy tomography** → both ρ and Z

Z_{eff}

High energy Tomography
Linear absorption coefficient μ_{HE} (cm⁻¹)

Mass attenuation coefficient µ/ρ

LE HE

➡ **Better material identification in nuclear waste packages**
Powders, compacted materials, materials having similar densities but different Z_{eff} (e.g. aluminum vs. concrete, organic vs. low apparent density metallic pieces, etc.)

Likonia Tassapino, Cyrille Eleon, Nicolas Estre, Stéphane Plumeri, David Tisseur, Albe in nuclear reconstruction using multi-MeV X-ray DECT in concrete drums, NSS 2019, 10-17 November 2019, Sydney, Australia

| PAGE 29

Slide 5: TESTING THE METHOD BY NUMERICAL SIMULATION

- Simulation of 200 L (Ø 60 cm) concrete drums, containing cylinders filled with various materials
- 10 inserted materials simulated
- High-energy X-ray source : LINAC 9 MeV (low-energy) and 20 MeV (high-energy)
- Source description (flux, angular and energy distribution)
  Object description (dimensions, materials)
  Detector description
  Geometry of acquisition

MODHERATO code Developed at CEA, DEN, Cadarache, Nuclear Measurement Lab

Simulated tomography images

Likonia Tassapino, Cyrille Eleon, Nicolas Estre, Stéphane Plumeri, David Tisseur, Albe in nuclear reconstruction using multi-MeV X-ray DECT in concrete drums, NSS 2019, 10-17 November 2019, Sydney, Australia

| PAGE 29

Slide 6: NUMERICAL SIMULATION RESULTS

LINAC emitting at 9 MeV and 20 MeV ⇒ tomography of large concrete drums

	Theoretical Z_{eff}	Extracted Z_{eff}	Reconstructions error
Water	6.60	5.72	-0.88
Solvent 1	4.60	3.61	-0.99
Solvent 2	5.37	4.68	-0.69
Solvent 3	6.06	5.36	-0.70
Graphite	6	5.39	-0.61
PVC	11.38	11.35	-0.03
Plaster	12.95	12.99	+0.04
ZrO₂	30.86	30.52	-0.34
PbO	65.8	61.30	-4.50
Pb	82		

Table 1 Z_{eff} reconstruction of the 10 test materials inside a 200 L drum filled with concrete. (*) Density artificially reduced to 1.

- Error reconstruction < 1 on Z_{eff} for materials up to Z_{eff} ~ 30 (ZrO₂)
- Metallic materials recognized as "heavy" even if they have a low apparent density
- Work ongoing to improve the precision for Z_{eff} > 40 materials

Likonia Tassapino, Cyrille Eleon, Nicolas Estre, Stéphane Plumeri, David Tisseur, Albe in nuclear reconstruction using multi-MeV X-ray DECT in concrete drums, NSS 2019, 10-17 November 2019, Sydney, Australia

| PAGE 30

CONCLUSION AND PROSPECTS

■ **Developments of new LINACS: SATURNE, TOMIS, ORIATRON**
  - High energy X-ray imaging of large concrete packages
  - Dual-energy imaging
  - Very high resolution imaging for JHR
  - Photofission of large concrete packages ⇨ nuclear material characterization

■ **Developments in high-energy photon detectors**
  - Large 2D panels for fast imaging & radioscopy: BGO, CsI GOS, LYSO, GLO...
  - Linear detectors for large dynamic range: CdTe, CdWO₄, CsI
  - Thin high resolution detectors with photon beam on the edge: Si + WidePIX

■ **Developments in simulation, image reconstruction & processing**
  - MODHERATO simulation code ⇨ acquisition, image reconstruction and processing
  - Helical, cone beam, partial filed-of-view acquisitions ⇨ 3D tomography
  - 3D viewing, object segmentation, dimensions and volume calculations...

PHOTON INTERROGATION: PHOTOFISSION (CEA DEN)

❑ **U and Pu** measurement in **large concrete waste packages**
❑ **High energy X rays** produced by an electron LINAC (photofission threshold ~ 6 MeV)
❑ **Delayed neutron counting** between the pulses
❑ **Delayed gamma-ray spectroscopy** after stopping irradiation

Thank you
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iCHANCE

Characterization of Conditioned Nuclear Waste
for its Safe Disposal in Europe

Novel optical methods for the characterisation of outgassing from radioactive waste

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JÜLICH — Holger Tietze-Jaensch

Optical methods extensively used in emission and environmental monitoring

---

Can the same methods be applied to the monitoring of radioactive gaseous emissions ?

Conventional radiation detectors

ALPHA PARTICLE

ATOMIC NUCLEUS

GAMMA RAY

BETA PARTICLE

Liquid scintillation counting

- laboratory technique
- not ideal for in-situ analysis
- in some cases: complex sample preparation
- produces additional waste
- cannot analyze directly gas samples

Overlapping peaks in the LSC spectrum

Scintillation peaks of commons radionuclides

Principles of laser spectroscopy for molecular detection

Molecular vibration modes

Water

Methane

Mid-infrared spectroscopy for trace gas detection

Absorption spectrum of CO_2 isotopes

Basics of laser spectroscopy

Beer-Lambert law:
$$I_{out} = I_{in}e^{-\sigma LN}$$

$$\frac{I_{out}(\lambda)}{I_{in}(\lambda)} \rightarrow$$

Cavity ring-down spectroscopy

Compact instrumentation

Compact instrumentation

New alternative for in-situ measurements

- Compact, low cost
- Real time measurement
- Gas samples
- No complex sample preparation
- Small sample volume of a few ml
- Only sensitive to one type of molecule
- No additional radioactive waste

Example: C-14 detection

Experimental results

CO_2 vs $^{14}CO_2$ spectra

$^{14}C/C = 1$ ppb

C-14 detection using cavity ring-down spectroscopy

C-14 analysis from atmospheric samples

On-site C-14 stack monitoring

On-site C-14 stack monitoring

Tritiated water detection with CRDS

Key features of CRDS

4

CRDS for waste outgassing monitoring

CRDS in CHANCE

CRDS in CHANCE

Find suitable absorption lines

Energy of a rotation vibration level of a diatomic molecule

Line intensities of different HCl isotopes

H^{36}Cl detection: next steps

cea

- Experimental validation of the transition with ^{35}Cl standards
- Develop a chemical method of the transformation of Na^{36}Cl into H^{36}Cl
- Experimental measurements using the developed CRDS equipment on graphite samples

ap
Le

CRDS in CHANCE

$^{14}CO_2$
$^{14}CH_4$

C-14
- Apply the CRDS technique for C-14 outgasing studies

Study C-14 from irradiated graphite

VTT

Study of CO$_2$/CO outgassing

VTT

CO$_2$ outgassing

CO outgassing

Isotopic fractionation in graphite combustion

VTT

Blue: ^{13}O
Red: °C

CRDS for C-14: next steps

VTT

- Study of the release behaviour of C-14 from irradiated graphite waste
- Release behaviour of C-14 from another type of wastes such as organic waste
- Comparison with traditional techniques

Outlook

New alternative for in-situ measurements

- Compact, low cost
- Real time measurement
- Gas samples
- No complex sample preparation
- Small sample volume of a few ml
- Only sensitive to one type of molecule
- No additional radioactive waste

Characterisation of waste outgassing

Health and occupational safety during decomissioning

Monitoring of radioactive emissions from power plants stacks

In-situ characterisation of nuclear waste

:CHANCE

Characterization of Conditioned Nuclear Waste
for its Safe Disposal in Europe

Modelling of the CHANCE calorimeter and
some general thoughts about NDA uncertainty quantification

Wojciech R. Kubinski, Christophe Mathonat, Holger Tietze-Jaensch,
Cedric Carasco, Daniel Kikola, Bart Rogiers, Dariusz Tefelski

CHANCE Topical Day, Mechelen, 21ᵗ March 2019

This project has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 715311

:CHANCE

Outline

1. On the power of calorimetry
2. LVC calorimeter and MC model
3. Escaping gamma and neutron fluxes
4. Possible use of shielding
5. Conclusion

Wojciech Kubinski et al, Modelling of the CHANCE calorimeter, CHANCE Topical Day, Mechelen, 21ˢᵗ March 2019

:CHANCE

CHANCE project
WP3 objectives

WP3 Leader: KEP Nuclear
Contributors: CEA, FZJ, SCK•CEN, WUT

Objectives:

To test and evaluate the performance of calorimetry as a
non-destructive technique for inventory of radionuclides

To identify how calorimetry can complement existing,
widely-used techniques (gamma spectrometry and
neutron passive measurement for instance)

To carry out an exhaustive study of uncertainties
assessment related to calorimetry and its coupling to
other non-destructive techniques

To construct the new LVC calorimeter (200L, 10-3000mW
range) with optimized lower detection limit (1.5mW)

Opening in two half shells.
Ghost reference cell underneath the drum to be measured

Wojciech Kubinski et al, Modelling of the CHANCE calorimeter, CHANCE Topical Day, Mechelen, 21ˢᵗ March 2019

:CHANCE

Calorimetry

The measurement of the heat from nuclear radiation
using calorimetry combined with the determination of
the nuclear isotopic mass ratios of the samples by
another Non Destructive Assay (NDA) technique (e.g.
High Resolution Gamma Spectrometry) provides a
convenient and accurate measure of the total
radioactive mass of the sample.

Fission + γ-rays and
neutrons emissions

Alpha particle emission

The daughter nucleus is in
its ground state (for β-
emitter) or in an excited
state and γ-rays are emitted

A neutron is converted into a proton by the
emission of an electron

Wojciech Kubinski et al, Modelling of the CHANCE calorimeter, CHANCE Topical Day, Mechelen, 21ˢᵗ March 2019

:CHANCE

Calorimetry

- Calorimetry is sensitive to all RN inside a waste drum, as all radiation emitted
  turns into heat
  - Calorimetry measures all heat deposited inside the drum
  - Can't miss any heat emission / deposition (above detection limit)
- Calorimetry is well adapted for alpha and beta emitters (e.g. Pu, Am, ^{3}H) – all
  the heat emitted by radioactive decay can be measured (less 1 % of
  uncertainty)
- Calorimetry can't discriminate / locate the heat source(s)
- The experimental task of calorimetry NDA is
  - Measure beta or alpha radiation heat source
  - to establish or rule out the existence of RN that are not easily detectable by either
    gamma or neutron spectrometry
  - namely ^{3}H, ^{90}Sr/^{90}Y, ^{106}Ru/^{106}Rh, ^{238}Pu, ^{241}Am, ^{244}Cm)
  - hidden/shielded gamma or neutron sources such as shielded ^{60}Co or ^{99}Mo/^{99}Tc or
    even ^{252}Cf
- Calorimetry is complementary and supplementary, mainly to gamma- and
  neutron spectrometry

Wojciech Kubinski et al, Modelling of the CHANCE calorimeter, CHANCE Topical Day, Mechelen, 21ˢᵗ March 2019

CHANCE

Summary and conclusions
possible improvements and further analyzes

- As the energy distribution caused by the radionuclides is already calculated, and after converting it into power, the heat transfer equations can be solved (using i.e. COMSOL tool) and show the **temperature distribution, the time required to achieve thermal equilibrium and time extension due to the additional mass of shielding**

- The model can also be used to improve the existing LVC construction by finding the **optimum shielding thickness and geometry which would minimize the radiation leakage at the optimized time of measurement**

- **The model is implemented and it can be used as a tool for the uncertainties assessment support.** It can also be an indicator for the anticipated measurements

Wojciech Kubinski et al, Modeling of the CHANCE calorimeter, CHANCE Tranical Day, Mechelen, 21st March 2019

CHANCE
Characterization of Conditioned Nuclear Waste
for its Safe Disposal in Europe

This project has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 755371

Thank you for your attention!

CHANCE
Characterization of Conditioned Nuclear Waste
for its Safe Disposal in Europe

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4

Slide 1

SCK·CEN — SCK·CEN ACADEMY
FOR NUCLEAR SCIENCE AND TECHNOLOGY

Topical Day
CHANCE
Characterisation of conditioned radioactive waste
Status, challenges and new developments

Modelling of the CHANCE calorimeter and
some general thoughts about NDA uncertainty quantification

Bart Rogiers
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Slide 2

Slide 3

Outline

- Basic concepts
- Error propagation
- Application to radiological characterization
  - Example from literature
  - SCK·CEN example
    - Isotopic vector
    - Measurement techniques
    - Total gamma example
  - Link with CHANCE WP3

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Slide 4

Slide 5

Basic concepts

$$5 \pm 2$$

- Can this variable be equal to 2?
- What is the probability that it is between 5 and 7?
- Is the probability that it equals 3 or 7 the same?
- What is the probability that it is larger than 8?

You cannot really know without more details!

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Slide 6

Basic concepts

The GUM [1] provides important guidelines. A summary:
- Provide all details of a measurement including how the uncertainty was evaluated
- The uncertainty can be communicated in two ways
  - As a (combined) standard uncertainty (± standard deviation)
  - As an expanded uncertainty (≈ confidence interval) with coverage factor k (± k * standard deviation)
    - Normal distribution:
      k = 1 for 68% confidence interval
      k = 2 for 95% confidence interval
      k = 3 for 99.7% confidence interval
- State clearly what type of uncertainty is provided!

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1

So how can we quantify the uncertainty of the final result?
- List all parameters used for calculating the final result
  - We should now this, right?
- List all uncertainties associated with these parameters
  - This can be problematic in some cases, but I suppose we can often come up with a good guess?
- Do error propagation
  - This might seem to be the most difficult step, but it really is not!

- Basic concepts
- Error propagation
- Application to radiological characterization
  - Example from literature
  - SCK•CEN example
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- First order Taylor expansion
  $y = f(\mathbf{x})$ output mean   output variance
  → variance → covariance → sensitivity coefficient (partial first derivative)

- Second order Taylor expansion
  $E[y] = f(\mathbf{x}_0) + \frac{1}{2} tr(\mathbf{H}_{xx}\mathbf{C}_x)$ output mean   output variance
  → trace → covariance matrix → Hessian (partial second derivatives) → gradient matrix (partial first derivatives)

- Monte Carlo simulation
  $y = \frac{\sum_{i=1}^{m} f(\mathbf{x}_i)}{m}$ output mean   output variance   number of samples $\mathbf{x}_k = (x_{k,1}, x_{k,2}, \dots, x_{k,n})$ random sample

- Other methods
  - Simulation-based (cf. Monte Carlo) e.g. importance sampling
  - Local expansion-based (cf. Taylor expansion) e.g. perturbation method
  - Functional expansion-based e.g. Neumann expansion
  - Most probable point-based e.g. first-order reliability method
  - Numerical integration-based e.g. full factorial numerical integration

---

- First order Taylor expansion
  - (without correlation) results in the well-known rules for
    addition/substraction and **multiplication/division**

    w = relative standard uncertainty

  - is the most commonly used approach (e.g. ISO 11929 [2], GUM [1])
  - but has its limitations
    - normal distribution approximations
      - can easily lead to non-physical results (e.g. ISO 11929 [2])
      - might deviate a lot from the real distributions
    - assumes linearity, so has limited accuracy for nonlinear expressions

---

- Second order Taylor expansion
  - corrects for bias in non-linear expressions
- Monte Carlo simulation
  - provides the full probability distribution (not just mean and variance)
  - poses no problems with non-linearity
  - works with all kinds of distributions (no simplifications needed)
  - is more computationally intensive (but simple to implement)
  - is more and more being incorporated in guidelines and norms (e.g. GUM supplement [3], new version of ISO 11929 in preparation)

Slide 1 (Outline)

Outline

- Basic concepts
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Slide 2

Application to radiological characterization
Example from literature

Kirkpatrick *et al.* (2013) [4]
- "Minimum detectable activity, systematic uncertainties, and the ISO 11929 standard."

Conclusions

The results presented here should serve as a caution that the method of calculating MDA prescribed by the ISO 11929 standard may be poorly suited for adoption in some applications. In particular, *in vivo*, *in situ*, and waste assay measurements routinely see relative systematic uncertainties in the range of several tens of percents, and often higher. Based on the results of our statistical modeling approach, we conclude that the ISO 11929 method may significantly overestimate the MDA under these conditions or fail to yield any physically meaningful result at all.

Slide 3

Application to radiological characterization
Example from literature

Slide 4 (Outline)

Outline

- Basic concepts
- Error propagation
- Application to radiological characterization
  - Example from literature
  - SCK·CEN example
    - Isotopic vector
    - Measurement techniques
    - Total gamma example
  - Link with CHANCE WP3

Slide 5 (Outline)

Outline

- Basic concepts
- Error propagation
- Application to radiological characterization
  - Example from literature
  - SCK·CEN example
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  - Link with CHANCE WP3

Slide 6

Application to radiological characterization
SCK·CEN example: Isotopic vector

Example presented by Rogiers *et al.* (2017) [5]:
BR3 pilot PWR reactor
- Reactor shutdown: 1987-06-30
- Reference date isotopic vector: 1998-07-01, about 11 years after reactor shutdown
- Key nuclides (Co60 and Cs137) and scaling factors (SFs)
- Monte Carlo error propagation flow chart:

Application to radiological characterization
SCK•CEN example: Isotopic vector

Outline

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- Basic concepts
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Application to radiological characterization
SCK•CEN example: Measurement techniques

Application to radiological characterization
SCK•CEN example: Measurement techniques

Application to radiological characterization
SCK•CEN example: Measurement techniques

5

Outline
- Basic concepts
- Error propagation
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Outline
- Basic concepts
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Application to radiological characterization
SCK•CEN example: Total gamma example

Example presented by
Rogiers et al. (2017) [5]

Material to be measured:
- HDPE tubes
- Outer diameter: 75 mm
- Length: 500 mm
- Thickness: 5 mm
- Density: 0.97 g/cm³
- Cut in half

Application to radiological characterization
SCK•CEN example: Total gamma example

Total gamma parameters:
- Measurement times (considered to be constant)
  - Background: 120 seconds
  - Gross: 20 seconds
- Count rates
  - Background: 1500 CPS
  - Gross: 2300 CPS
- Self absorption: Correction factor with uniform distribution between two extreme cases.

Application to radiological characterization
SCK•CEN example: Total gamma example

Application to radiological characterization
SCK•CEN example: Total gamma example

Outline

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Application to radiological characterization
Link with CHANCE WP3

- After the experimental program, the results will be processed and an exhaustive study on uncertainties will be performed
- This could be done with the previously discussed approaches, but everything is still open at this point
- Monte Carlo particle transport simulations like those discussed earlier today by Christophe Mathonat and Wojciech Kubinski allow us to estimate systematic uncertainties (*i.e.* those on the efficiency or calibration factor) accounting for potential heterogeneity in matrix and source composition/distribution

---

Take home

Take home

- Basic concepts
  - Clearly communicate the type of uncertainty and how it was obtained
- Error propagation
  - Taylor expansion-based error propagation is not fit for all purposes
- Application to radiological characterization
  - Count statistics can be relevant, but systematic uncertainties are often more important
- CHANCE WP3
  - We still have lots of interesting data analyses ahead!

Questions?

References

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