Methods for radiological characterisation

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Presentation outline

- Decommissioning
- Radiological characterisation
- Methods for radiological characterisation
- Measuring systems - examples
Introduction

At the present time, more than 560 commercial nuclear power plants are or have been in operation in the world.

About 120 plants have been permanently shut down and are at some stage of decommissioning.

About 10 % of all shutdown plants have been fully decommissioned (including 8 reactors with electric power more than 100 MWe).

A larger number of various types of fuel cycle and research facilities have been shut down and decommissioned (including facilities for extraction and enrichment of uranium, facilities for fuel fabrication and reprocessing, laboratories, isotope production facilities and particle accelerators.)
Decommissioning

The term „decommissioning“ is used to describe all the management and technical actions associated with ceasing operation of nuclear installation and its subsequent dismantling to facilitate its removal from regulatory control (delicensing).

These actions involve:

- Shut down
- Decommissioning plan
- Decontamination of components
- Dismantling of components
- Decontamination of structures
- Demolition of buildings
- Remediation of any contaminated ground
- Removal of the resulting waste
The final status of decommissioning

Some of the decommissioning projects have ended up in the brown field and the others in green field conditions.

In the course of the decommissioning of nuclear facilities is necessary to minimize the amount of waste (in terms of their chemical and radiological toxicity). It is therefore important to determine the content of radioactive substances in all the components that will be dismantled.
Brown field conditions

Brown field site / „brownfield“

The land previously used for industrial purposes or some commercial use.

The land (building) may be contaminated by low concentration of hazardous waste or pollution and has a potential to be reused once it is cleaned up.

In the nuclear facilities, the final liquidation of some contaminated objects is often postponed at a later time so as the radioactivity decreases by natural disintegration.

The reason is a significant radiation risk and associated high financial costs.
Green field conditions

The area that had been in industrial use is, in principle, restored to the conditions existing before the construction of the plant.

Example: NPP Yankee (USA – Connecticut, PWR, 600 MWe)
Radiological characterisation

What is the radiological characterisation?

The radiological characterisation is the knowledge of:

where is the activity?
   on the surface of component / structures?
     fixed?
     can be removed?
   inside the material?
     contamination (how deep, distribution ...) ?
     induced radioactivity (how deep, distribution ...) ?
     can be easy release by mechanical dividing (tritium)?

which nuclides?

dose rate in the vicinity of the device (radiation safety) ?
Radiological characterisation

Why we need the radiological characterisation?

Radiological characterisation is very important for each decommissioning project.

It is relevant for all phases of decommissioning and should be started as early as possible.

The radiological characterisation includes:

- collection and review of historical file
- performing calculation of radionuclide inventory from historical information
- in situ measurement
- sampling
- laboratory measurement
- review and evaluation of the data obtained
- comparison of the calculated result with measured value etc.
Radiological characterisation (relevance, timeline)

Relevance of the radiological characterisation:

1. For assembling the decommissioning plan
2. For radiological protection
3. For the selection of methods
   • of disassembling the equipment
   • of cutting objects (most suitable segmenting techniques)
4. For continuous refinement of the decommissioning plan
5. For estimation the quantity, form and activity of waste
6. For cost estimation

Timeline of radiological measurements:

1. Before decommissioning
2. During the decommissioning
3. After decommissioning
   area + radioactive waste measuring (for storing / releasing)
Several documents that describe the sampling and measurement methods, evaluation procedures, quality assurance and other relevant issues:

- **Multi Agency Radiation Survey and Site Investigation Manual (MARSSIM),**
- **Multi-Agency Radiation Survey and Assessment of Materials and Equipment Manual (MARSAME)**
- **Environmental Radiation Survey and Site Execution Manual (EURSSEM)**

**International Atomic Energy Agency (IAEA) Safety Standards for decommissioning:**

- The Joint Convention on the safety of radioactive waste management – 1 (2)
- Decommissioning of Nuclear Power Plants and Research Reactors – WS-G-2.1
- Decommissioning of Medical, Industrial and Research Facilities – WS-G-2.2
- Decommissioning of Nuclear Fuel Cycle Facilities – WS-G-2.4
- Safety Assessment for the Decommissioning of Facilities Using Radioactive Material – WS-G-5.2
- Decommissioning of Facilities Using NORM (planned)

........
Radiological data

Furthermore, the acquired data need to be stored, managed, retrieved and evaluated using versatile databases (decommissioning database).

In recent years, software for statistical evaluation of the data and for visualisation of the measurement results has been brought to perfection and is widely used.

In particular, robust statistical analysis of the data enables the reduction of the number of samples and measurements without losing the significance of the results.

**Motto:**
The more accurate the preliminary survey is performed, the less additional control / decontamination cycles are needed to reach clearance levels.
Methods for radiological characterisation

- Historical data (very carefully, the old data ....)
- Historical investigation (what happened?)
- Data quality objectives
- In-situ investigation
  - measurements (Non Destructive Analysis - NDA)
  - sampling (Destructive Analysis - DA)
- Laboratory measurements (DA)
- Implementing of calculation methods (activation ....)
- Statistical methods
- Calculations of scaling factors
In-situ measurements
(Non destructive analysis – NDA)
Methods for in-situ measurements (NDA)

1. Dose rate measurements
2. Surface contamination measurements
   - gross alpha activity
   - gross beta / gamma activity
3. Gamma scanning
4. Gamma imaging camera (distribution activity with picture)
5. Gamma spectrometric measurement (nuclide specific measurement)

For example, a simple radiation dose rate measurement will give an indication of the total quantity of gamma emitting radionuclides, but will not identify the individual radionuclides or their concentrations.

Gamma spectroscopy will identify the individual radionuclides and, when properly calibrated, their quantities as well.
Dose rate measurement

Especially for the radiation protection of the workers, localization of hot-spots

Hand held equipments

Detectors: Geiger Mueller Tube (GMT)  
Proportional counter tube  
Scintillation detector  
    NaI/Tl, NaI/Cs, plastic scintillator  
      Ionisation chamber  

Measuring range: from background to 1 Sv/h (10 Sv/h)

Teleprobe (up to 4 meters distance)  
    for difficult accessible places  
    for sources with high activity (with high dose rate)
Dose rate measurement

Frequently used monitors: Dose Rate Meter 6150AD, FH 40 G .......

6150D

6150D with external probe

FH 40 G

IF 104
Surface contamination measurements

Measurement of surface contamination is an important aspect of the decommissioning. From the measured data, it is possible to decide on the protection of workers and to prevent the unacceptable discharges to the environment.

Hand held instruments are mainly used for direct measurement of:

- gross alpha surface activity
- gross beta / gamma surface activity

Detectors:
- Proportional counter tube
  - Gas flow counting tube - butane, counting gas P10 (Ar + methane)
  - Xenon counter tube - permanent filling
  - Scintillation detectors – thin layer plastic scintillator with ZnS/Ag-coating

Important parameters:
- counting sensitivity for alpha and beta
- detection area (100 – 170 cm²)
- distinguish between alpha and beta particles (gamma radiation)
- very low sensitivity to gamma radiation
- small mass
- detector resistance against damage (tearing of foil detector)
**Surface contamination measurements - monitors**

**Frequently used monitors:**
- LB 124 SCINT
- FHT 111M
- CoMo 170
- Electra with DP6 probe

**LB 124 SCINT**
- (scintillation detector)
- (gas flow detector)
- (scintillation detector)

**Additional protective grid**

**LB 124 SCINT in floor trolley**
Surface contamination measurements - monitors

- FHT 111M – Contamat
- CoMo 170 – trolley for floor measurement
- Electra – survey meter
- DP6 Alpha-beta probe
- Large area alpha / beta detector + GPS
Surface contamination measurements - monitors

**Large-area gamma detector PL 525**

- Detector surface: 525 cm²
- Basic version detector lift
- Maximum high approx. 5 m
Gamma scanning

Gamma scanning provides relevant information to the contamination of the object.

A scintillation detector is usually used.

Its sensitivity (the size of a crystal in the detector) is chosen so as the statistical error by counting of impulses will be sufficiently small.

When performing gamma scanning:

a) the detector moves in a specified height by a fixed speed above the surface
b) the detector is gradually placed into individual points of measurement

For reducing the impact of surrounding radioactive sources the detector is inserted into shielding (most commonly lead collimator).
Gamma scanning – example

Detector: 3 x 3 inch NaI(Tl)
Collimator: 5 cm lead
Measuring time: 10 s
Measuring unit: Integral count rate [cps]

Report – example:
The gamma imaging camera:

- Provides indication of shape and positions of main radioactive sources
- Enables to locate radioactive hot spots

Principle:

Main characteristic of a gamma camera:

1. Optical part
   - collimator, pinhole, coded mask
2. Detector
   - NaI/Tl scintillator, pixellated detector
3. Acquisition SW
Gamma imaging – RadScan 800

RadScan 800

Detection head, pan / tilt unit, tripod

Look inside the detection head
Shielded scintillation detector of RadScan 800
angle 2 degrees
for high angular resolution

- Steel collimator
  - 2 degrees
- Tungsten shielding
  - $\rho_0 = 19.3$ g/cm³
- Detector NaI (TI)
  - 10 x 16 mm
- Photomultiplier

Kolimátor - 2 stupně - EN.vsd
The gamma imaging system collects data on a predefined array of measurement points. The detection head is moved to appropriate pan and tilt position, gamma spectrum is acquired for the dwell time defined by user.
Gamma imaging – RadScan 800 – gamma snaps

Measurement at EC JEC Ispra – building: 20a  pipe line: L0008 8001 BS0020  above the tank VB0020  
range: 4,5 m    collimator: 3 degree    scan time =  200 s    count rate for Cs-137:  max. 1,9 cps, 
contact dose rate: 1 010 µSv/h
Gamma imaging camera CARTOGAM uses the results of the development of the gamma camera in nuclear medicine.

CARTOGAM – detection head
Gamma imaging – Cartogam

CARTOGAM – hot spots imaging

gamma
Gamma imaging – GAMPIX

New generation – model GAMPIX:
- lower weight
- higher sensitivity
- lower price
In-situ gamma spectrometric method takes advantage of the fact that many radionuclides during its conversion emit gamma photons. These gamma photons have energy typical for each radionuclide.

It is possible to determine the activity of gamma radionuclides

- in solid and liquid substances
- on a surface of the objects (from simple to relatively complex shapes)
- inside materials
- on large surfaces (on the floor, on the walls, in the open field ....)

**Stage of measurement:**

- 1st stage: uncollimated detector
- 2nd stage: collimated detector
In-situ gamma spectroscopy system consists of:

- detector (scintillator, semiconductor)
- shielding (collimator)
- electronic unit (high voltage power supply, preamplifier ....)
- analog to digital converter (ADC) / digital signal processing (DSP)
- multichannel analyzer
- evaluation unit (PC, NB, laptop, tablet) with gamma spectrometry SW
Gamma spectrometric measurement ranks among the so-called relative measurements. For each geometry of the measurement, a calibration should be performed.

The calibration is performed using:
- radioactive standards
- calculations (SW: MicroShield, ISOCS, Monte Carlo – MCNP)
Low resolution gamma spectrometry

As the name says, by low resolution gamma spectrometry, detectors with relatively low energy resolution are used. That brings problems with the interpretation of the measured data, but on the other hand, it is a cheap and commercially available device.

For the detection of gamma photons, so called scintillation detectors are used. The most widespread is the detector with NaI/Tl (CsI/Tl).

Recently, new scintillation materials (lanthan bromide - LaBr$_3$/Ce, cadmium zinc tellurid - CdZnTe, ...) begin to be used. They have a better energy resolution, but the disadvantage is their high price.
Low resolution gamma spectrometric measurement - example

Detector: 3 x 3 inch NaI(Tl)
Collimator: 5 cm lead
Multichannel analyzer: 1024 channels
Measuring time: min. 600 s
High resolution gamma spectrometry

Using in-situ high resolution gamma spectrometry, almost all the radionuclides emitting gamma photons can be detected.

For the measurement a semiconductor detector is used (today almost exclusively from high purity germanium - HPGe).

Comparison of HPGe detector with scintillation detectors:

- Better energy resolution
- Needs cooling to the temperature of liquid nitrogen (-196 degree of C)
- Less sensitive (small volume of the crystal)
- More expensive

Equipment cost:
HPGe detector (high efficiency, small Dewar, liquid nitrogen cooling), multichannel analyzer, SW cca 60 k€

Cooling:
For cooling of HPGe detector, liquid nitrogen is mainly used. Recently, detectors with electrically cooled HPGe crystal are more and more used (the most commonly by Stirling´s cooler).
High resolution gamma spectrometry - example

In-Situ Object Counting System (ISOCS):
High resolution gamma spectrometry – Micro-Detective HX

Portable Hand-Held Radioisotope Identifier
(HPGe detector, battery powered, dose rate meter, neutron detector, GPS)

Weight: less than 7 kg
Radiological laboratory analysis
(Destructive analysis – DA)
Radiological laboratory analysis

There are several reasons why it is necessary to make some measurements in the laboratory:

- high radiation background in the place where you want to determine the concentration of radionuclides
- bad geometry for measurement
- inaccessible place for in-situ measurement
- in-situ measuring systems do not measure some radionuclides

Some examples:

In the vicinity of the measured object is another object with significantly higher activity (it cannot be removed, shielded).

We want to determine the distribution of activity inside the material.

A number of radionuclides emit during its conversion such a radiation that cannot be measured directly, and it is necessary to perform the radiochemical separation. These radionuclides are called "hard to detect" radionuclides – HTD or “difficult to measure” - DTM.
Radiological laboratory analysis – important radionuclides

Laboratory destructive analyses for common radionuclides

<table>
<thead>
<tr>
<th>Laboratory method</th>
<th>Radioisotopes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Alpha spectrometry</td>
<td>Np-237, U-233, U-234, U-235, U-238, Pu-238, Pu-239, Pu-240, Am-241, Cm-242, Cm-243, Cm-244</td>
</tr>
<tr>
<td>Beta counting</td>
<td>Sr-90, Tc-99</td>
</tr>
<tr>
<td>Liquid Scintillation</td>
<td>H-3, C-14, Ni-63, Kr-85, Sr-90, Sm-151, Ra-226, Pu-241 (gross alpha)</td>
</tr>
<tr>
<td>ICP/MS</td>
<td>Tc-99, I-129, Pd-107, U, Th, Np-237</td>
</tr>
<tr>
<td>Mass spectrometry</td>
<td>U-233, U-234, U-236, U-238, Pu-238, Pu-239, Pu-241, Pu-242</td>
</tr>
</tbody>
</table>

Typical preparation time before measurement: from hours to days
Typical counting time: from hours to days
Price to detect one radionuclide: between 100 and 300 €
Radiological laboratory analysis – alpha emitters

The most commonly used instruments to detect alpha radiation is an alpha spectrometer:

- silicon detector
- evacuated counting chamber
- power supply
- amplifier
- analog to digital converter
- multichannel analyzer
- computer
- SW

Measurement often requires:
- complex chemical separation
- special sample preparation before alpha counting
Radiological laboratory analysis – beta emitters

The most commonly used instruments to detect beta radiation is a liquid scintillation spectrometer.

Usually, it is necessary to carry out the chemical separation of the radionuclide.
Some radionuclides emit gamma photons with very low energy during their conversion.

For the detection of these photons it is necessary to use a special semiconductor detector (entrance window is made from a material with a small absorption – carbon fiber, ion-implantated contact on the HPGe crystal).
Radiological laboratory analysis – gamma emitters

**Gamma spectroscopy:**
Gamma spectrometry in laboratory is used to define isotopic composition of materials.

The sample together with the detector is inside the high-quality shielding.

The advantage of this method is that a very low detection limit can be reached.
**Inductively Coupled Plasma – Mass Spectrometry (ICP-MS)**

**Principle:**
The solution with the isotopes is sprayed into flowing inert gas (argon) and passed into a torch which is inductively heated to approximately 10 000 °C.

At this temperature, the gas and almost all components are atomized and ionized, forming a plasma which provides a rich source of both excited and atomized atoms.

Positive ions in the plasma are focused down a quadrupole mass spectrometer.

Data can be obtained for almost the entire periodic table just in minutes with detection limit 0.1 µg/L (0.1 ppb).

It is possible to measure radioactive and stable elements.

The measurement of some elements is complicated by interference.

For some elements, calibration standards are not available.
Scaling factor - SF

Laboratory methods are much time consuming and more expensive than direct methods. Therefore, it is an effort to reduce the amount of laboratory analyses.

The SF method was developed from operating experiences at nuclear power plants for low and intermediate level radioactive waste.

The SF method is a technique for evaluating the concentration of difficult to measure (DTM) nuclides which are typically represented by beta and alpha emitting nuclides, such as C-14, Ni-63, Pu-240 ......

The scaling factor is the ratio of the concentration between DTM nuclide and easy to measure (ETM) nuclide (key nuclides: Co-60, Cs-137)

Other information in the document: IAEA No. NW-T-1.18 “Determination and Use of Scaling Factors for Waste Characterization in Nuclear Power Plants”.
The use of scaling factor:

1. the determination of the scaling factor (correlation factor)
2. determine the concentration of easy to measure nuclide
3. calculate the concentration of radionuclide difficult to measure

\[ C_{DTM} = SF_{DTM} \times C_{ETM} \]

Scaling factor – Ni-63 to Co-60 – NPP Dukovany
Measurement of waste packages
Measurement of waste package

The reason for the measurement:
Prevent the release of material with significantly higher activity than the other material.

Gamma Excavation Monitor (GEM) – NUKEM UK

Gamma Excavation Monitor (GEM) is a gross gamma system capable of real-time assay of excavated materials in support of clearance initiatives. The bulk monitoring system has gained Environment Agency approval for effective material segregation of volumes up to 1m³.
Total gamma measuring

On the market are available measuring systems with 24 plastic scintillations detectors.

Disadvantage of this system:
We need a good knowledge of nuclide composition (different gamma photons emissions for individual nuclides, different energy of photons).
Measurement of waste package – nuclide specific measurement

**High Resolution Assay Monitor - HiRAM (producer NUKEM UK)**

This trailer-mounted system is designed for the on-site assay of waste materials, is easily transportable and can remain on site during the measurement campaign.
# Measurement of waste package – nuclide specific measurement

HiRAM – result of measurement, using of scale factor for difficult to measure nuclides

## HiRAM Results

Using CP7 Project Fingerprint

<table>
<thead>
<tr>
<th>Reference</th>
<th>1904</th>
</tr>
</thead>
<tbody>
<tr>
<td>Net Weight (kg)</td>
<td>902</td>
</tr>
<tr>
<td>Date of Measurement</td>
<td>04/11/2012</td>
</tr>
<tr>
<td>Waste Material</td>
<td>Soil</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Measured Activity (Bq g(^{-1}))</th>
<th>Fingerprint Activity Assessment (Bq g(^{-1}))</th>
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<tbody>
<tr>
<td></td>
<td>Result</td>
<td>Uncertainty</td>
</tr>
<tr>
<td>Cs137</td>
<td>1.39E+00</td>
<td>2.81E-01</td>
</tr>
<tr>
<td>*Am241</td>
<td>Not Detected</td>
<td>n/a</td>
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<tr>
<td></td>
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</table>

This colour denotes data entry fields.

* This value is for information only. The fingerprint derived value is the only value applied in.

Waste Category: VLLW
Measurement of waste package – nuclide specific measurement

Drum measuring
One HPGe detector – scanning system
Measuring before sending the drums to disposal site at NPP Dukovany
Measurement of waste package – nuclide specific measurement

Drum measuring
3 HPGe detectors
Measuring before sending to Radioactive waste depository at NPP Dukovany
Characterization of Waste at NPP Dukovany before sending to incinerator (at Studsvik, Sweden)
Measurement of waste package – free release measurement

The reason for the measurement:

Prevent the release of the material with an activity that exceeds the free release level.
3 HPGe detectors – 10 cm lead shielding - measuring before sending to landfill disposal.
Measurement of waste package – nuclide specific measurement
SUE SIA „Radon“, Russia

RADIOACTIVE WASTE CHARACTERIZATION SYSTEM
- Stationary system with HPGe detector and a conveyer for 25 drums for automatic RW measurement
- 2 mobile systems with HPGe detector and a turntable ISO-CART + MCA DigiDart device
- SW GamControl, RAOS, GamaVision, Isotopic 32
- Generating reports for the State control and accounting system of RW&RM

Conveyer for 25 drums
Drum rotating unit
MUM – advanced free release measurement facility

Prototype of unified Europe wide FRM methodology system.

- Low-background measuring tunnel
- 4 HPGe Interchangeable Detector Modules with lead collimators
- Conveyor for moving the measuring container
- Air-conditioning and filtration unit for measuring tunnel
- A conservative approach to the calculation of the radioactivity
MUM – advanced free release measurement facility

Low – background concrete like shielding material.
High density material – bulk density in the interval 2 300 – 3 250 kg/m³

Key advantages:
- Easy, dry and clean construction.
- Modular construction system.
- Self-supporting construction.
- Cheap production.

Internal content of radionuclides:
- K-40 10.6 Bq/kg
- Ra-226 1 Bq/kg
- Th-228 0.7 Bq/kg
Metrology for decommissioning nuclear facilities

European Metrology Research Program SRT-v19 project

Start in the half of 2014.

- Radionuclide characterization of materials present on decommissioning sites, automated radiochemical analysis
- Preselection of waste into streams (repository or potential free release)
- Implementation of ‘free release measurement facility’ on decommissioning site, scanning of heterogeneous wastes
- Monitoring in radioactive waste repositories and decommissioning sites
- Calibration reference materials and standard sources

It can be measured at the same time 6 boxes with dimension 120 x 80 x 40 cm (384 L).
Each box shall be measured with two detectors from top and two detectors from bottom - always in 3 positions for the assessment of homogeneity of distribution activities.
24 pcs. HPGe detectors, electrically cooled
High throughput of material (6 * 384 kg = 1,2 t / 30 min).
Capacity: More than 2 tons per hour.
The device is easy to move to another site.
Methods for radiological characterisation

Thanks a lot for your attention.

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