



PREDIS

Milestone 27

Setting up characterisation process and operation of gamma spectroscopy

28/08/2024 version Final

Public

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This project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 945098.

Project acronym PREDIS	Project title PRE-DISposal management of radioactive waste	Grant agreement No. 945098
Milestone No. MS27	Milestone title Setting up characterisation process and operation of gamma spectroscopy	
WP No 4	Date version 28.08.2024 Final	Due date M48
Lead beneficiary NCSR		
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Abstract

PREDIS WP4 is focused on the decontamination and disposal of metallic radioactive waste. Task 4.5 aims minimization of metallic waste and increase of the amounts for recycling or declassification. This milestone focuses primarily on the non-destructive gamma spectrometry techniques developed in subtask T4.5.2 to help optimize the characterization and sorting of metallic radioactive waste from decommissioning of NPPs. Two semi-empirical techniques were investigated based on combination of gamma spectrometry measurements and MCNP Monte Carlo simulations. The first technique concerns the monitoring of metallic components or segments to distinguish activation from surface contamination. The second technique concerns the monitoring of metallic segments after dismantling and cutting for sorting in different management routes. The technique is aiming at reduction of the measurement uncertainties related to the density and activity distribution. For this purpose, a new non-destructive gamma spectrometry layout is proposed. Furthermore, in this subtask, the methodology for determination of scaling factors, which is a necessary procedure for the radiological characterization of radioactive waste, was discussed. T4.5.1 was dedicated for classification of the waste streams of the different types of reactors and to provide a scheme for classification of the reactor materials by using Nuclide Vector approach for characterization after dismantling. The need of non-destructive technique for metallic waste characterization was figured out to supplement the Nuclide Vector approach.

Keywords

Neutron calculations, scaling factors determination, nuclide vectors, gamma spectrometry, radiological characterization techniques, MCNP simulation, management routes for metallic waste, distinguish activation from contamination, sorting in different management routes

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Notification

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Acknowledgement

This project has received funding from the Euratom research and training programme 2019-2020 under grant agreement No 945098.

TABLE OF CONTENTS

1	MILESTONE DESCRIPTION.....	9
2	GENERAL INTRODUCTION TO TASK 4.5.....	9
3	STRUCTURE OF THIS MILESTONE.....	10
3.1	Specific introduction to T4.5. 1.....	10
3.2	Specific introduction to T4.5.2.....	11
4	MANAGEMENT ROUTES FOR METALLIC WASTE.....	11
4.1	Standard Routes.....	11
4.2	Controlled Reuse/Recycling.....	12
4.3	Specific Clearance for Disposal.....	14
4.4	Specific Clearance for Melting.....	14
4.5	Combined Strategies.....	15
4.6	Characterization for specific purpose.....	16
5	CLASSIFICATION OF THE WASTE STREAMS OF THE DIFFERENT TYPES OF REACTORS.....	17
6	RADIONUCLIDE GENERATION AND DISTRIBUTION IN MRW AT THE DIFFERENT NUCLEAR REACTORS.....	20
6.1	Composition of different nuclear reactors metallic components.....	21
6.2	Neutron spectrum, the neutron flux, runtime history of the reactors.....	24
6.3	Neutron activation products in different nuclear reactors metallic components.....	25
6.4	Neutron activation and corrosion as well as fission products released to reactor coolant in different nuclear reactors.....	27
7	SCALING FACTORS APPROACH.....	29
7.1	Key and Difficult-To-Measure (DTM) Radionuclides.....	29
7.2	Scaling factors in different metallic components of different nuclear reactor types and time evolution of scaling factors.....	30
7.3	Factors affecting scaling factors.....	33
7.4	Scaling Factor Determination methods.....	37
7.5	Characterization of waste streams of MRW.....	37
8	SAMPLING PROCEDURE.....	38
8.1	Estimation of sampling places.....	38
8.2	Sample preparation and registration.....	38
8.3	Estimation of representativeness and number of samples.....	39
8.4	A basic sampling optimization approach.....	41
9	MEASUREMENT TECHNIQUES FOR MRW CHARACTERIZATION.....	42
9.1	Destructive techniques.....	43
9.1.1	Samples preparation.....	44

9.1.2	Determination of ^{55}Fe and $^{59+63}\text{Ni}$	45
9.1.3	Determination of ^{90}Sr and ^{90}Y	45
9.1.4	Determination of ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu	46
9.1.5	Determination of ^{241}Am , ^{242}Cm and ^{244}Cm	46
9.1.6	Measurement alpha radionuclides.....	47
9.1.7	ICP-MS	47
9.2	Non-destructive techniques	47
9.2.1	Gamma spectrometry.....	47
9.2.2	Gamma-ray imaging	52
9.2.3	X-ray imaging.....	52
9.2.4	Neutron counting	53
9.2.5	Calorimetry	55
10	A NON-DESTRUCTIVE GAMMA SPECTROMETRY METHOD FOR CHARACTERIZATION OF RADIOACTIVE METALLIC COMPONENTS.....	55
10.1	APPROACH I.....	56
10.2	APPROACH II	56
11	PROPOSED SETUP FOR CHARACTERIZATION OF LOW ACTIVITY METALLIC SEGMENTS WITH LOW MEASUREMENT UNCERTAINTY.....	57
12	SUMMARY AND FUTURE PLANS.....	58
13	REFERENCES	60

ABBREVIATIONS

ABWR	Advanced Boiling Water Reactor
AGR	Advanced Gas Cooled Reactor
AK	Auxiliary Key
AMS	Accelerator Mass Spectrometry
AP	Activation Products
AR	Auxiliary Radionuclide
BWR	Boiling Water Reactor
CP	Corrosion Products
CPS	Control and Protection System
CZT	Cadmium Zinc Telluride
DA	Destructive Assay
DTM	Difficult to Measure
EC BSS	European Council Directive on Basic Safety Standards
ETM	Easy to measure
EW	Exempt Waste
FA	Fuel Assembly
FBR	Fast Breeder Reactor
FEPE	Full Energy Peak Efficiency
FP	Fission Products
GCR	Gas Cooled Reactor
GEB	Gaussian Energy Broadening
HLW	High Level Waste
HPGe	High Purity Germanium
IAEA	International Atomic Energy Agency
ICP-MS	Inductively Coupled Plasma Mass Spectrometry
ILW	Intermediate Level Waste
LA-MS	Laser Ablation Mass Spectroscopy
LILW	Low and Intermediate level Waste
LLW	Low Level Waste
LSC	Liquid Scintillation Counting
MCC	Main Circulation Circuit
MDA	Minimum Detectable Activity
MRW	Metallic Radioactive Waste
MS	Mass Spectrometry
NDA	Non Destructive Assay
NPP	Nuclear Power Plants
NV	Nuclide Vector
OSCAR	Outil de Simulation de la Contamination en Réacteur
PHWR	Pressurized Heavy Water Reactor
PMT	Photo Multiplier Tubes
PWR	Pressurized Water Reactor
	Pressurized Tube Water Reactor (in russian: Reaktor Bolschoi
RBMK	Motschnosti Kanalny)
RCS	Reactor Coolant System
RIMS	Resonance Ionization Mass Spectroscopy
RPV	Reactor Pressure Vessel
RSM	Radioactive Scrap Material
RW	Radioactive Waste

SF	Scaling Factor
SGS	Segmented Gamma Scanning
SNF	Spent nuclear Fuel
SNM	Special Nuclear Material
SS-MS	Spark Source Mass Spectroscopy
TGS	Tomographic Gamma Scanning
TIMS	Thermal Ionization Mass Spectrometry
TRL	Technological Readiness Level
VLLW	Very Low Level Waste
VVER	Water Water Reactor (in russian: Vodo-Vodyanoi Enyergeticheskiy Reactor VVER)
WAC	Waste Acceptance Criteria
WP	Work Package

LIST OF FIGURES

Figure 1: Standard routes for radioactive waste management of metal scrap.	12
Figure 2: Extension of the standard management routes by controlled reuse/recycling	13
Figure 3: Different possibilities to use the clearance route allow a higher efficiency to clear metallic waste.	14
Figure 4: Optimized management route without disposal of radioactive waste	15
Figure 5 Waste classification scheme according to IAEA [8].	17
Figure 6 Neutron flux energy distribution for PWR, BWR, RBMK and CANDU type reactors (adapted according to L. Walters [41])	25
Figure 7 Separation of activation zones according to neutron flux in RBMK-1500 reactor by using MCNP6/SCALE6.2 modelling: highly activated zones (red line), zone of intermediate and low activation metal construction (green line) and non-activated materials (only surface contamination) above green line [40].	26
Figure 8 Cross-sectional cut of RBMK-1500 reactor vault [23], as in [21]. 1 - Central hall floor; 2 - Top metal structure with serpentinite; 3 - Concrete vault; 4 - Sand cylinder; 5 - Water tank; 6 - Graphite stack (core, top, bottom and side reflectors); 7- Reactor shell; 8 - Bottom metal structure; 9 - Reactor support structure; 10 - Shield and support steel blocks; 11 – Roller supports; 12 – Modeled segment [21].	27
Figure 9 Waste stream integration.	31
Figure 10 Chart of Nuclide vector determination and optimization.	32
Figure 11 Effect of fuel failure on scaling factor (BWR, total alpha/ ¹³⁷ Cs; homogeneous solidified waste). (a) Scatter diagram; (b) histogram of total alpha/key nuclide [52] [53].	34
Figure 12 ¹⁴ C/ ⁶⁰ Co scaling factor dependence on waste stream. (left) Scatter diagram of ¹⁴ C/ ⁶⁰ Co; (right) distribution diagram of ¹⁴ C/ ⁶⁰ Co [52] [53].	35
Figure 13 (a) Distribution and (b) scatter of ⁶³ Ni/ ⁶⁰ Co ratio for different radioactive waste streams generated at PWR plants from different countries [54].	36
Figure 14 Comparison of ⁶³ Ni/ ⁶⁰ Co ratios for a BWR and PWR. Scatter (a) and distribution (c) diagram for different reactor type of ⁶³ Ni/ ⁶⁰ Co; scatter (b) and distribution (d) diagram of ⁶³ Ni/ ⁶⁰ Co for PWRs in different countries [52] [53].	36
Figure 15 SF Accuracy & Uncertainty improvement by the manner of sampling	42
Figure 16 – Energy spectrum of a ¹⁵² Eu source acquired with different detectors: NaI (in blue), CZT (in red) and HPGe (in green). Credit to Prof. Zhong He.	48
Figure 17 – Example of peaks identification and area calculation in a complex region including 4 emissions (distinguishable by the different colour). The residual distribution (the plot at the bottom) laying in the 2-sigma region (red lines) testify the goodness of the fit.	50
Figure 18 – Example of non-destructive assay (NDA) gamma spectrometry setup: on the left the open-geometry, on the right the SGS (similar for TGS).	51
Figure 19 – Examples of TGS reconstruction: the matrix distribution on the left, the radioactivity distribution on the right.	51
Figure 20 – Examples of gamma ray imaging reconstruction. On the left, an ²⁴¹ Am source is imaged by means of the coded mask (visible as the red-contoured area). On the right, 2 ¹³³ Ba sources are reconstructed: one is in the forward direction respect to the detector, the other is in the backward direction.	52
Figure 21 – Application of x-ray imaging to make a digital radiography of a metallic box containing a glove box with other components.	53
Figure 22 – Neutron multiplicity distribution for ²⁴⁰ Pu spontaneous fission (adapted from ortec-online.com).	54
Figure 23 – FNDP (Fast Neutron Detector Package) Signals during an active neutron interrogation assay.	54

LIST OF TABLES

Table 1. The main differences for most common nuclear reactors in the world [17]	20
Table 2. Volume and activity of LILW generated annually by 1 GWe NPP (according to [18]).....	21
Table 3. Nuclide list of CP - radionuclides present in MRW generated at different nuclear reactors (the main radionuclides are presented with bold)	21
Table 4. Example of RBMK metallic structural elements	22
Table 5. <i>Chemical composition of metal materials of reactor RBMK-1500 and max impurity (wt%)</i>	22
Table 6. <i>Example of PWR metallic structural elements</i>	23
Table 7. <i>Example of PWR: Chemical composition of metal materials of PWR reactor, max impurity (wt%).</i>	23
Table 8. List of different reactors and maximum neutron fluxes achievable in various materials of core components in these reactors.	24
Table 9. List of radionuclides to be declared for MRW	30
Table 10: Dependence of the correlation coefficient on the number of samples.....	39

1 Milestone Description

Milestone 27, associated with Work package 4, Task 5 has been completed on 21.08.2024.

The justification for the readiness is described below and complies with the Grant Agreement Description of Action noting verification by M4.6 Deliverable 4.6 available.

The readiness of the milestone was reviewed and agreed upon by Abdessalam Abdelous (IMT) as WP4 leader.

2 General introduction to Task 4.5

In the framework of the European project PREDIS “Pre-disposal management of radioactive waste”, the work package WP4 “Innovations in metallic material treatment and conditioning”, the Task 4.5 “Optimization of metallic waste characterization and procedures for waste minimization and recycling” aims to optimize the metallic waste characterization and procedures for waste minimization and increase of the amounts of metals for recycling.

Adequate radiological characterization of a nuclear installation is important for final decommissioning planning and optimization of radioactive waste management. For decommissioning planning, the objective is to obtain a radiological understanding of the involved facility. The characterization at this stage could be carried out by means of: 1) neutron activation calculations, for components at the vicinity of the reactor core, based on the reactor design and neutron flux; 2) dose rate measurements; 3) in-situ gamma spectrometry. Especially for the contaminated parts, theoretical approaches based on mechanisms of transfer from the source of radionuclides by the coolant as well as on the chemical properties of the radionuclides compounds and the components materials can be applied. After approval of the final decommissioning plan, sampling for determination of the scaling factors in activated and/ or contaminated components is carried out.

The waste generated during decommissioning is collected and sorted according to the type, activity and scaling factors. The sorting of low activity waste (Low Level Waste (LLW), Very Low Level Waste (VLLW), Exempt Waste (EW)) is usually performed by using portable devices to measure dose rates or total counts and then are put into containers. The waste packages are monitored by non-destructive techniques for assessment of activity and determination of the management route: 1) to be processed as radioactive waste; 2) to be decontaminated for clearance or declassification, 3) to be cleared directly.

For essential decision-making process after dismantling and cutting of nuclear installation components, accurate and precise characterization of metallic waste is needed. The waste may be activated and/or surface contaminated with radionuclides which are products of neutron activation or come from the dispersion of the irradiated fuel and fission products due to fuel and fuel cladding defects. The ways of contamination with activation products are: 1) the structures in a reactor are activated by neutron flux and later on, the corrosion products of these structures are transferred by the coolant and contaminate the reactor components; 2) corrosion materials carried by the reactor coolant pass the reactor core and get activated, then are transferred by the coolant and contaminate the reactor core and MCC components. Although the major radionuclides in metallic waste from any Nuclear Power Plants (NPP) are more or less the same, the partitioning of the waste as well as the correlation between the radionuclides depend on the reactor type and operation history: the power, the operation time, presence of fuel cladding defects and leakage incidents, if exists. A radiological characterization should determine the activity of the waste and allow segregation into homogeneously activated, mixed (activated and surface contaminated) and only surface contaminated. The selection of decontamination techniques is based on determination of the radionuclides inside the material and/ or in the surface contamination. It is important to decide in which cases the decontamination will be efficient as well as to select the appropriate decontamination technique based on whether the waste is slightly activated or contaminated or both. The collective work in this task is contributing to:

- More effective selection of the management route for the metallic waste of the classes LLW, VLLW (i.e. management as radioactive waste, clearance after sand blasting, clearance and melt, melt and clearance etc.);
- Advanced characterization of MRW which allows selection of the more efficient decontamination and clearance procedures for the metallic waste streams;
- Minimization of the volumes of metallic radioactive waste and therefore save of resources by increasing the amounts of metals for recycling, reduction of the management cost [1] and saving disposal capacity.

3 Structure of this milestone

The work in Task 4.5 to achieve minimization of metallic waste and increase the amounts for recycling is carried out in the following three subtasks:

- T4.5.1 is dedicated to classification of the waste streams of the different types of power reactor;
- T4.5.2 is focused on characterization techniques and procedures for optimization of sorting of low activity metallic waste in different management routes as well as for selection of the most effective decontamination and clearance procedures;
- T4.5.3 deals with the development of robust and validated radiochemical procedures, which are lacking, for Difficult to Measure (DTM) radionuclides in metallic waste, significant for decontamination and clearance. (Results of this subtask are presented in separate deliverable and are not addressed in this).

This milestone, MS27, is divided into ten main chapters. The chapters from 4 to 8 focus on classification of the waste streams of the different types of reactors, i.e. T4.5.1 subtask. The next three chapters are devoted to the MRW characterization techniques and procedures for optimization of sorting, i. e., T4.5.2 subtask. The brief introduction of both subchapters is presented below. T4.5.3 is presented in a separate document (Deliverable 4.7 and MS28).

3.1 Specific introduction to T4.5. 1

T4.5.1 subtask is dedicated to the classification of waste streams from different types of reactors. Standard and extended routes for radioactive waste management of metal scrap have been analysed. Waste classification involves the quantification of specified radionuclides in the waste materials to comply with disposal requirements and disposal site performance objectives. The aim of classification of the Metallic Radioactive Waste (MRW) streams of different types of reactors is changing together with MRW activity: the higher activity MRW (High Level Waste (HLW) and Intermediate Level Waste (ILW)) needs classification related to radiation protection and best packaging concept, LLW and VLLW may needs decontamination and declassification afterwards. The largest volumes of waste from the dismantling of nuclear installations will mainly be VLLW and LLW.

The main aim of the 4.5.1 subtask is to provide a scheme for classification of the reactor materials regarding the level of specific activity: high (specific) activity, intermediate (specific) activity, low (specific) activity metallic constructions and non-activated materials in order to facilitate the procedure of decontamination and clearance or declassification. The methodology for characterization of the metallic waste is similar to all reactors and is based on Nuclide Vector (NV) determination. An essential part of characterization is the separate determination of activation activity and contamination activity to identify the best way of management.

There is no universal MRW characterization way for all installations, but the main steps include:

- Pre-dismantling classification of MRW using (MCNP6/SCALE6.2) modelling obtaining neutron activation map in the 3D of reactor core and peripheral hardware (experience in High-Power Channel-type Reactor (RBMK), Pressurized Water Reactor (PWR), Boiling Water Reactor (BWR) reactors).

- Detailed characterization of MRW during dismantling via experimental measurements applying available non-destructive and destructive measurement techniques (analysis of samples of surface contamination and activation).
- Optimized characterization: determination of NV for MRW stream. Together modelling and experimental measurements allows separating metallic waste streams to homogeneously activated, mixed (activated and surface contaminated) and surface contaminated MRW.

3.2 Specific introduction to T4.5.2

T4.5.2 is focused on characterization techniques and procedures for optimization of sorting of low activity metallic waste in different management routes (i.e. management as radioactive waste, clearance after sand blasting, clearance and melt, melt and clearance etc.).

Characterization of the metallic waste, which arisen from the dismantling and cutting of NPP components, involves the quantification of specified radionuclides to comply with WAC for disposal. Regarding the waste of low activity (which belongs to the classes EW, VLLW and possibly to LLW), the aim of characterization is to provide the basis for decision if decontamination will be efficient as well as to select the most effective decontamination techniques and clearance methodologies. It should be mentioned that the largest volumes of waste from the dismantling of nuclear reactors (NPPs or research reactors) are EW, VLLW and LLW and therefore the optimization of sorting of metallic waste of these classes will contribute crucially to metallic waste minimization and recycling. In the frame of T4.5.2, two semi-empirical techniques are proposed for further development. These techniques rely on a combination of gamma spectrometry measurements and MCNP Monte Carlo simulations. These techniques are focused on optimization of the radiological characterization of metallic waste to facilitate the procedures of decontamination and clearance or declassification.

The first technique concerns the monitoring of metallic segments after dismantling and cutting, aiming at reduction of the measurement uncertainties related to the density and activity distribution. For this purpose, a new non-destructive gamma spectrometry measurement setup is proposed. The technique is expected to be effective for determination of contamination of non-activated as well as of activated metallic waste which belongs to EW, VLLW and LLW. Therefore, it will allow better selection of the management route. Also, the technique will facilitate the selection of the appropriate decontamination techniques based on whether the waste is slightly activated or contaminated or both. The specific activities inside and in the surface contamination of metallic waste will be determined by using the measurement results in combination with the use of the scaling factors for activation and/ or contamination.

The second technique concerns the monitoring of metallic components or segments to differentiate between activation and contamination. Scaling Factors (SF) could be semi empirically determined in case of sampling and statistical analysis or totally theoretically determined in case of neutron calculations for activated components or of considering all the pathways, transfer mechanisms, chemical properties, etc. from the source to the contaminated component. This non-destructive gamma spectrometry technique is focused on the interpretation of the resulting gamma-ray spectra of the radionuclides in activated and/ or contaminated components.

4 Management Routes for Metallic Waste

4.1 Standard Routes

The question of what to do with the metallic waste generated during decommissioning of nuclear installations is of utmost importance. It is a very decisive question that must be discussed in advance the decommissioning process. Nevertheless, the answer to this question strongly depends on the characterization results of the metallic components to be decommissioned. In case of only two waste routes – clearance or disposal as radioactive waste – this answer is fairly easy to find (see Figure 1). The result of the characterization provides a basis for the decision, whether a clearance process is a successful option (possibly after a further decontamination step) or if the disposal as radioactive waste is unavoidable.

The requirements for characterization for clearance (complying with the general clearance values and their averaging parameters) are different from the characterization for disposal (complying with

the activity limit values given by WAC of the disposal facility). Therefore, the first characterization is only preliminary and has to be rerun according to the requirements of the chosen management route. This is a simple standard model, presenting an easy solution for sorting of radioactive waste (left or right sorting). In the following chapters, it will be shown that there are much more routes to get rid of the metallic scrap.

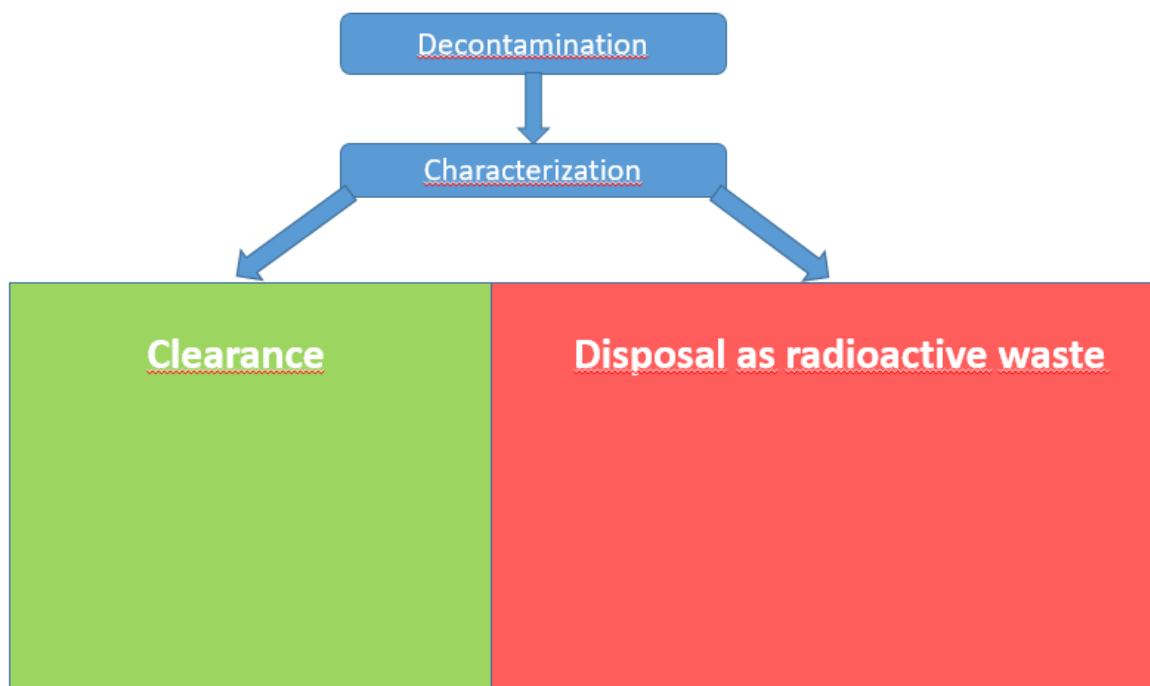


Figure 1: Standard routes for radioactive waste management of metal scrap.

4.2 Controlled Reuse/Recycling

Reuse in controlled areas

One of the easiest solutions for metallic waste reduction is the reuse of contaminated or activated metals in controlled areas, e.g. in the same or similar NPPs. Therefore, a reuse of metal components should be aspired and efforts to find suitable applications should be started prior the decommissioning process.

Controlled reuse offers several advantages:

- Limited effort for decontamination of the components.
- No effort and costs for managing disposal or clearance, the components remain under regulatory control.

If metal components need to be moved to another destination, the conditions for transport have to be considered. Nevertheless, in nearly all the cases the reuse of contaminated or activated metals in controlled areas is an easy and economical solution, in contrast to disposal or clearance.

Controlled Recycling

A second possibility is the recycling of contaminated or activated metals. A recycling of scrap is typically done by melting and there are several companies running melting facilities in controlled areas, thus performing controlled recycling. This process offers several advantages:

- A component has often hidden contamination not accessible for decontamination and impeding a reliable radiological characterization. The melting process is harmonizing the activity in the melt and by this enabling a reliable characterization.
- The majority of non-metallic contributions and heavy metals is separated and enriches in the slag and exhaust gas filters. Therefore, the melting enables an element-specific decontamination.
- The melting process will reduce the volume of the metallic component.

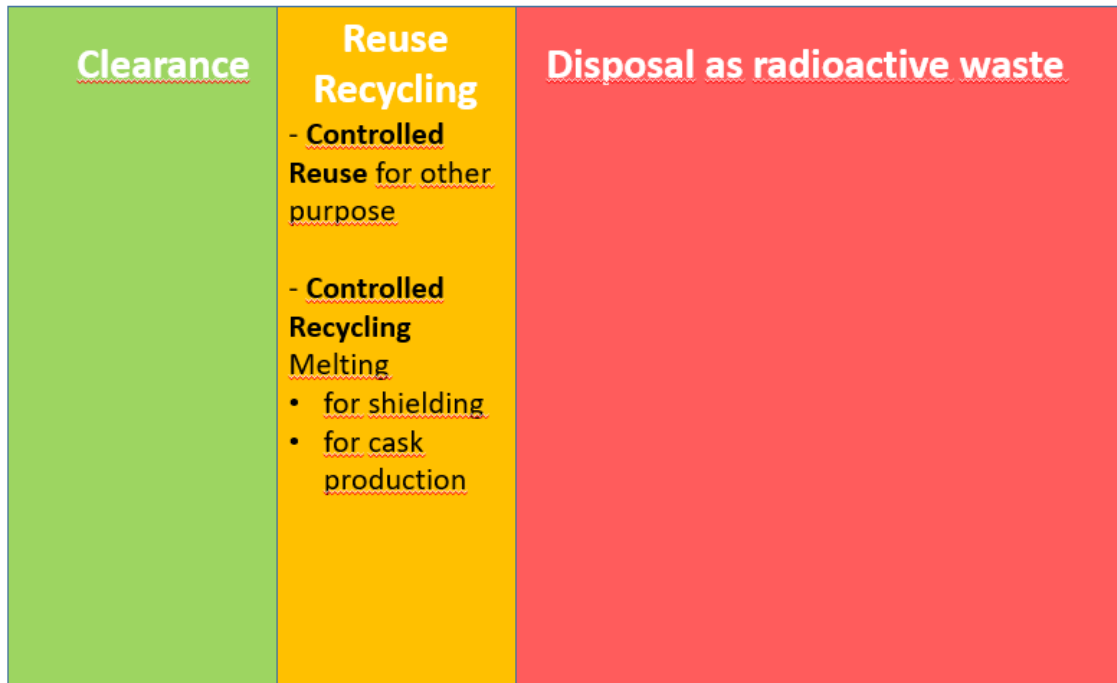


Figure 2: Extension of the standard management routes by controlled reuse/recycling

The melted metal is then available for future applications in controlled areas. Two examples for a controlled recycling and a future use of the product in a controlled area are:

- a) Melting for cask production

The first example of the use of the melting product is for the production of casks for radioactive waste (e.g. done by Siempelkamp, Germany [1]), if the metallic properties are appropriate. Nevertheless, the amount of radioactivity remaining in the metal of the cask has then to be included in the radionuclide content declaration of the radioactive waste package. The cask production is then client specific. Secondary radioactive waste resulting from the melting process, like slags and filters, will be sent back to the client.

- b) Melting for production of shielding blocks

A second example would be the production of shielding blocks, e.g. for future use in large acceleration experiments (e.g. done by Energy Solutions, USA). As the melting procedure for this use case is not client specific, the secondary radioactive waste, such as slags and filters, will be disposed by the melting facility.

Reuse of contaminated or activated metals in controlled areas and controlled recycling are two very interesting options, which should be included in the management routes for metallic waste (see Figure 2).

4.3 Specific Clearance for Disposal

The European Council Directive 2013/59/EURATOM [2] on basic safety standards (EC BSS) for protection against the dangers arising from exposure to ionizing radiation provides the legal framework for the release of radioactive material from regulatory control (clearance) and has already been implemented in the legislative of the European member states. The EC BSS provides threshold values of the specific activity of nuclides for general clearance, i.e. the release of the material from radiological control.

The EC BSS also offer a flexibility in cases, where the further process of the material is known and the dose constraint of some ten Microsievert (μSv) per year (de minimis dose concept) is kept. This is called specific clearance (see Figure 3). Some member states like Germany specified requirements for the clearance of materials if they are disposed on a conventional landfill. As a result, values for the specific clearance of materials on a landfill are implemented in the German Radiation Protection Ordinance, which are for some important nuclides like ^{60}Co and ^{137}Cs exceed the values for general clearance by a factor of 60 to 100. With this option the disposal route of clearance can be exploited more frequently, thus reducing the costs for decommissioning.

Nevertheless, in many member states it is a legal obligation to prove that recycling is economically not feasible. In cases of contamination with toxic substances like asbestos or if mixtures are not suitable for melting, the compliance with regulation is evident.

4.4 Specific Clearance for Melting

Secondly, a specific clearance for melting might be applied. It is specified in the European Commission Radiation Protection (EC RP) 89¹ and EC RP 117² and provides a guidance for the clearance of scrap, if the melting of a scrap guaranteed afterwards the clearance. In contrary to the controlled recycling, the melting process no longer has to be performed in a controlled area, in conventional facilities.

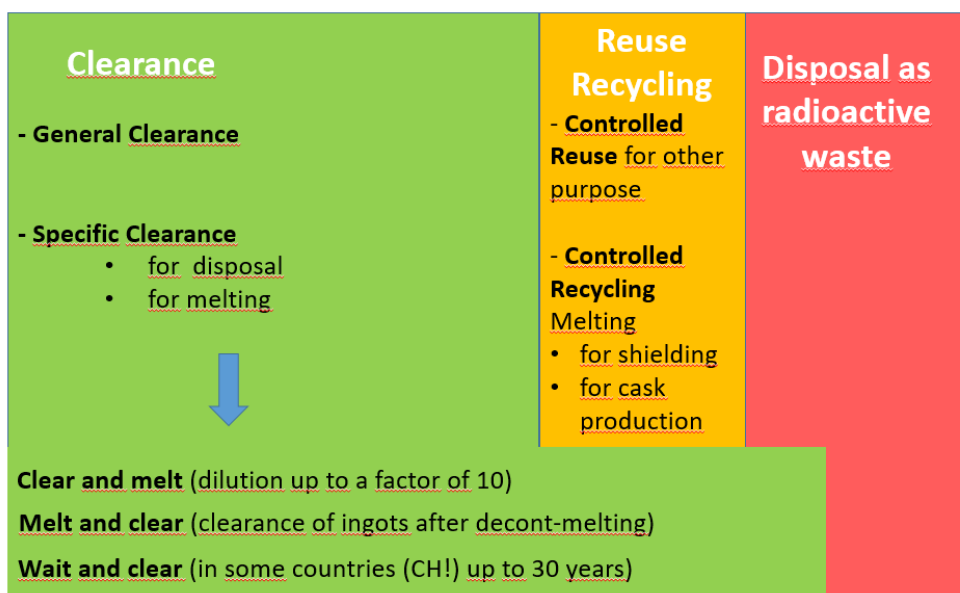


Figure 3: Different possibilities to use the clearance route allow a higher efficiency to clear metallic waste.

¹ EC RP 89 Recommended radiological protection criteria for the recycling of metals from the dismantling of nuclear installations, 1998

² EC RP 117 Methodology and models used to calculate individual and collective doses from recycling of metals from the dismantling of nuclear installations, 2000

In case of specific clearance for melting, the following conditions have to be adhered:

- ensured dilution of 1:10 as a minimum or
- an annual capacity of the melting facility of more than 40.000 tons per year.

If the requirements are met, specific clearance values of up to one order of magnitude higher than the values for general clearance (values published in EC RP 89) can be used. Regulatory control mechanisms might apply for the supervision of the dilution, but no radiation protection measures.

4.5 Combined Strategies

For the combined strategies presented in this subsection, it is assumed, that a decontamination of the component has been performed and further decontamination actions are not effective or economically feasible (see Figure 1).

Clear and Melt: The specific clearance for melting is a disposal strategy applying the specific clearance values for recycling of metallic waste, followed by a melting of the scrap in a conventional foundry. As the threshold values for specific clearance are approximately one order of magnitude higher than for general clearance, this route applies for much more material and the amount of metallic scrap classified as radioactive waste is getting lower.

Melt and Clear: But also, the reverse procedure makes sense. During the melting process of contaminated or activated metals in a controlled area, nuclide specific separation occurs as some specific nuclides are enriched in the slag and filters, thus less radionuclides will be found in the ingot. This procedure often enables a specific clearance of the ingot. Additionally, the resulting slags and filters might comply with the threshold values for a disposal on a conventional landfill. If both products of the melting process comply with the respective threshold values for release, an optimized melting in a conventional foundry might be possible depending on the local legislative (see Figure 4).

To prohibit accidental contamination of the melting facility and their products, most foundries have radiation detectors installed. Those detectors raise an alarm at a threshold of just above twice of background level, i.e. also material just below specific clearance levels will cause an alarm. Therefore, the willingness of the operators of the foundries as well as of the association of the operators of melting facilities and scrap dealers to accept cleared materials from nuclear installations for melting is very low.

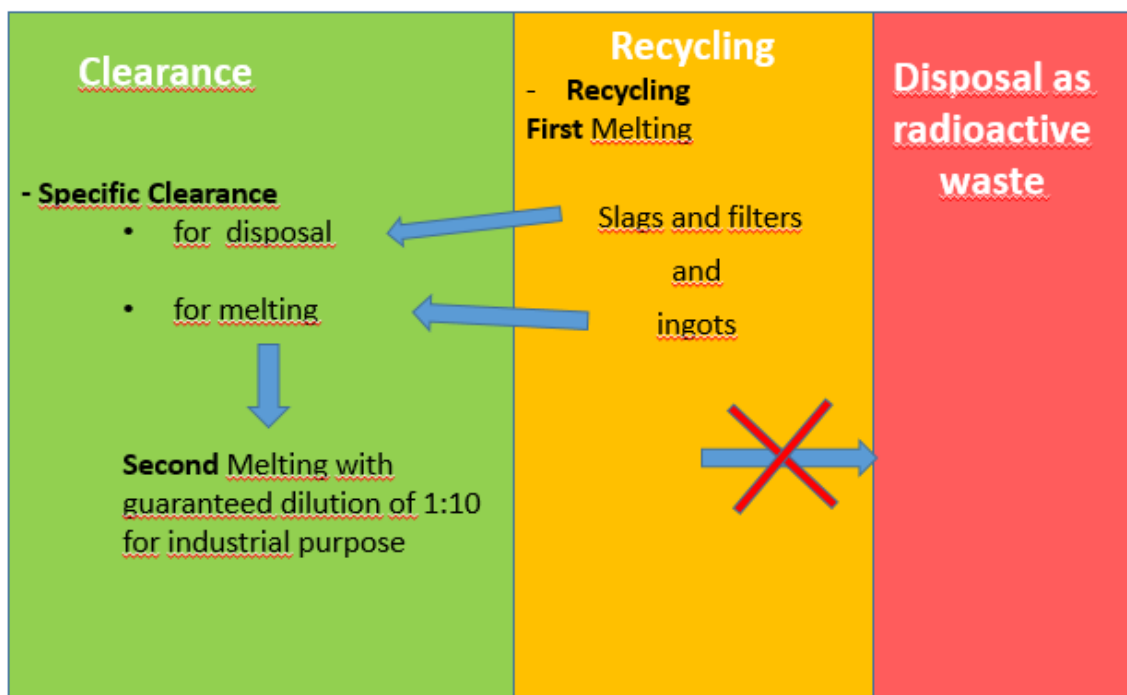


Figure 4: Optimized management route without disposal of radioactive waste

Wait and Clear: Especially for metals, hidden contamination of the component that are not accessible for decontamination or activation might impede the clearance process. For this reason, it is recommended to exploit the radioactive decay for a licensed limited time (e.g. used by Studsvik, Sweden; NPP Greifswald, Germany). In some countries, the operator is obliged to exploit the radioactive decay (the decay time in Switzerland is up to 30 years), if after this period of time a clearance might be successful. The exploitation of decay is a very effective instrument to reduce the amount of waste and to dispose of only those wastes that will still be radioactive during the operational lifetime of the disposal facility.

Nevertheless, this strategy has the disadvantage, that all the material for decay needs additional storage capacity. Furthermore, there is a risk that legislation might change during the decay storage and a clearance can no longer be performed according to revised clearance regulation.

All the above discussed management routes for the reuse and recycling, as well as clearance procedures should be discussed before a decision for disposal as radioactive waste. It should be kept in mind that not only the way of reuse, recycling and clearance is much more economical than disposal, but also the capacity of the disposal facilities for radioactive waste is a very rare and valuable resource.

4.6 Characterization for specific purpose

Within the previous chapter, different routes for metallic waste were described. All of these routes have in common that a characterization of the material is necessary. Unfortunately, each route has its own requirements for the material characterization.

Clearance: The characterization must be appropriate to demonstrate that the specific activity is below the relevant clearance values. Further regulations, such as average parameters, have to be considered. Due to the “de minimis” concept of some ten μSv per year for the general public, clearance values are very low. Therefore, a characterization for clearance is often ambitious and sophisticated.

Controlled Reuse: In case of controlled reuse only transport regulations have to be taken into consideration, i.e. beside of dose rate and contamination measurements, an estimation of the activity is necessary.

Controlled Recycling: Next to the necessary characterization according transport regulations, the operator of the recycling facility has its own WAC, which need to be complied with. These WAC for recycling has its focus on the product (e.g. max. specific activity permitted) and the specific activity limits for the workers (e.g. permitted levels for dose rate and specific activity of aerosols in the working area).

Disposal as Radioactive Waste: Besides the necessary characterization according to transport regulations, a disposal facility for radioactive waste has defined WAC. If the disposal facility is still in the planning or construction phase, preliminary WAC should be available. These WAC include limit values and constraints for nuclide specific activities resulting from considerations about long-term safety, operational safety and accident safety. While limit values define specific numerical thresholds for radionuclide activity levels in the waste, constraints encompass broader requirements or conditions related to waste characteristics, disposal facility operations, and safety considerations. It should be mentioned that the requirements for long-term safety include nuclides with a very long half-life and less or no gamma emission. Characterization in case of disposal can also be very ambitious, but is very different from the characterization for clearance.

All these considerations demonstrate that each waste route has its own kind of characterization. There is no one-fits-it-all characterization. However, before starting treatment and characterization according to the waste route chosen for the component, there must be a pre-characterization to decide, which of these routes is appropriate for the component.

5 Classification of the waste streams of the different types of reactors

The radioactive waste management and disposal strategy are the major issues, which are important for each state having operating NPP or NPP under decommissioning.

According to IAEA [3] at the end of 2020 the total number of NPPs at various stages of their life cycle was 686 worldwide. Among them, 172 are permanently closed and 442 are operational. The age profile of operational NPPs indicates that most of them will proceed to permanent shutdown and decommissioning within the next few decades. This is obvious by the fact that Germany, Japan & USA are planning to reduce their capacity by 131.5 GW until 2047 [4]

Regarding the cost of decommissioning a huge amount of money is needed. Indicatively the total cost of decommissioning for an NPP ranges from 0.5 billion to 1 billion USD. Especially for the waste management, the amount is around 1-2 hundred million USD [5]. It is important to mention that the US decommissioning market is valued at about USD 70 billion [6]

MRW in different reactors usually has a wide list of radionuclides of different concentrations. General estimations suggest that the majority (two-thirds) of the NPP waste resulting from decommissioning and dismantling is radiologically unrestricted material (buildings, auxiliary equipment which do not have contact with direct contamination or only contamination with very low activity material etc.) i.e. exempt waste [7]. The remaining one-third of all generated waste is radioactive waste with different activity levels, where the dismantled metallic parts forms about 12%, other are concrete (5%) and remaining structures (83% having contact with radioactive contamination/activation) [7]. There is a variety of alternatives for processing waste and for short term or long-term storage prior to disposal. Likewise, there are various alternatives for the safe disposal of waste, ranging from near surface to geological disposal. Various schemes have evolved for classifying radioactive waste according to the physical, chemical and radiological properties that are of relevance to particular facilities or circumstances in which radioactive waste is managed [8]. International Atomic Energy Agency (IAEA) classification system puts an emphasis on the aspect of disposal safety, while there may be some differences in the national waste classification systems. Regulation practices are based on guidelines from the International Commission on Radiological Protection [9]. This is valid also for metallic waste: it has to be collected, segregated, characterized, treated, stored and finally disposed of in special disposal facilities.

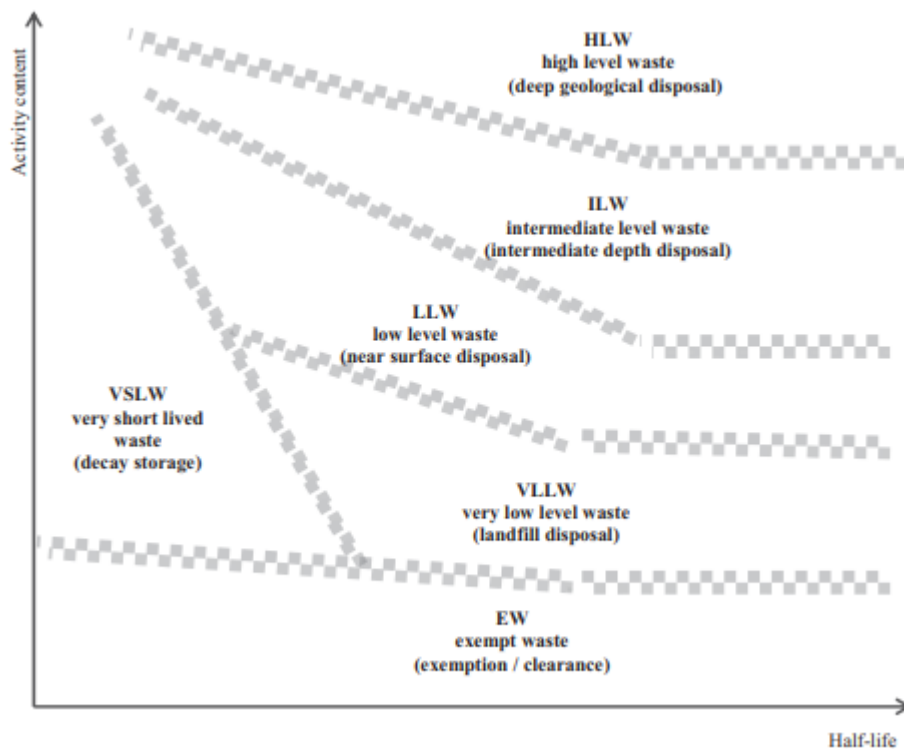


Figure 5 Waste classification scheme according to IAEA [8].

According to EURAD project WP 9 ROUTES analysis on categorization/classification of radioactive wastes (RWs) in member states there is no completely unified approach for RW classification [10]. Various countries apply different types of RW classification, and sometimes several types of classification are used simultaneously in the same country. The IAEA General Safety Guides-1 (GSG-1) approach to classification [8] is applied in the majority of participating countries, but the classes “low level waste” (future disposal in near-surface disposal facilities) and “intermediate level waste” (disposal facilities at intermediate depth) do not always have the same meaning as in IAEA GSG-1. In many participating in ROUTES countries, low and intermediate level waste (LILW) are combined as one class, which, in turn, is often subdivided into short-lived and long-lived RW. Generally, short-lived LILW could be associated with LLW within the meaning of IAEA GSG-1, whereas long-lived LILW could be associated with ILW within the meaning of IAEA GSG-1. However, such an interpretation is correct only in case that division of LILW into short-lived and long-lived is explicitly linked to the disposal route of waste.

As it can be seen from *Figure 5* the radioactive waste classes are: HLW, ILW, LLW, VLLW, EW and the separate category – Very short lived waste (VSLW) which is stored waiting for decay.

VLLW: Waste that is above clearance levels and does not need a high level of containment and isolation and is suitable for disposal in near surface landfill type facilities with limited regulatory control. After certain decontamination procedure it could possibly meet exempt waste criteria for clearance (effective doses to individuals should be of the order of 10 μ Sv or less in a year). As concerning metallic waste it mainly includes decommissioning process equipment, construction materials, tools, etc. For this waste:

- Characterization methods like gamma spectroscopy and dose rate measurements, waste inventory determination by utilizing NV determination procedure, surface contamination estimation is needed.

Decontamination methods should be applied considering the list of radionuclides and assessment of the possibility for clearance. Clearing as much as possible of materials from decommissioning is beneficial, not only that it saves potentially valuable raw material, but it also saves the repository capacity and saves financial funds that would otherwise be spent on treatment and disposal. In cases of a surface contamination, decontamination methods can be used like steel grit blasting, water blasting, chemical flushing, mechanical scrubbing or brushing, high-pressure washing, electrochemical decontamination [11].

- Classification optimization should be applied according to waste activity level after decontamination and dedicated disposal/clearance methods. Scrap metals is one of the possible methods for management of VLLW after decontamination, as after decontamination not only the removable but also the fix contamination can be removed (e.g., by abrasive blasting), after clearance the scrap is considered non-radioactive. The annual average release of surface contaminated metallic waste, after decontamination, from reactors above 100 MWe worldwide is approximately 110 kt [11].

LLW: Waste that is above the limit of VLLW with limited amounts of long-lived radionuclides. It requires isolation and containment for periods of up to a few hundred years and is suitable for disposal in engineered near surface facilities. Usually, it does not require shielding or needs little shielding during normal handling and transport, dose rate at the surface of a drum containing LLW under 2 mSv/h, disposal down up to 30 m, specific activity limit of 400 Bq/g on average (and up to 4000 Bq/g for individual packages) for long lived alpha emitting radionuclides. For long lived beta and/or gamma emitting radionuclides, such as ^{14}C , ^{36}Cl , ^{63}Ni , ^{93}Zr , ^{94}Nb , ^{99}Tc and ^{129}I , the allowable average activity concentrations may be considerably higher up to tens of kBq/g. To LLW belongs metallic waste contaminated with fission products and activation products during operation of nuclear power plants of different types. Waste includes contaminated primary coolant tubes and the off-gas system contaminated equipment. For this waste:

- Application of nuclear spectroscopy characterization methods, NV determination procedure, surface contamination and neutron activation estimation are needed. Consistent scaling factors for a wide variety of LLW materials can be determined for ^{63}Ni as well as for ^{93}Mo and $^{93\text{m}}\text{Nb}$ relative to ^{60}Co over a concentration range of four orders of magnitude [12].

- Decontamination methods according to NV should be applied. Melting is usually considered as a suitable method for metal recycling and decontamination of separate parts [13], when elements and their radioisotopes are redistributed from metal to slag and dust/fumes. In case of contamination of the cooling circuit of a NPP, it is usually necessary to decontaminate the whole circuit or at least its disconnected parts on site at the decontamination centre. The on-site treatment is mostly performed by chemical methods (decontaminating solution), either under static (at specified time interval at a specified temperature) or, more often, dynamic (solution is circulated in a separate closed loop) conditions [14].
- Classification optimization and dedicated disposal could be set according to waste level after decontamination.

ILW: Waste containing long lived radionuclides, requires a greater degree of containment and isolation in disposal at greater depths, of the order of tens of meters to a few hundred meters. It is waste generated from the replacement and decommissioning of slightly activated components outside core. For this waste:

- For estimation of the amount of ILW by MRW characterization, determination of neutron activation zones by the neutron flux at the reactor is needed. Modeling validation according to non-destructive or indirect measurements of important nuclides should be performed.
- Nuclear spectroscopy methods, NV determination, surface contamination and neutron activation estimation are needed for estimation of declassification possibility.
- Usually ILW is not suitable for decontamination, but waste-treatment and storage (segmentation, treatment, conditioning-packaging and storage) optimization and selection of most suitable disposal facility and packing (having in mind the best radioprotection) is needed.
- Disposal strategy (cut and dispose, pack and go, pack and wait, pack and dispose) considering radiation safety of workers and other factors should be considered.

HLW: Activity concentration of this waste is high enough to generate significant quantities of heat by the radioactive decay process or waste with large amounts of long-lived radionuclides that need to be considered in the design of disposal in deep, stable geological formations usually several hundred meters or more below the surface. The levels of activity concentration of waste are of 10^4 TBq/m³. National authorities determine that certain waste constitutes ILW or HLW depending on safety cases.

HLW are solid waste such as fuel rod cladding hulls, hardware. Waste contains activation products, as well as some undissolved fission products, uranium and minor actinides. HLW is generally stored in tanks prior to its eventual vitrification. For this waste (similar as for ILW):

- Neutron activation estimation according to modeling should be performed: determination of neutron activation zones by the neutron flux in the reactor obtaining the amount of High-Level metallic waste. Modeling validation according to non-destructive or indirect measurements of important nuclides is needed.
- Both nuclear spectroscopy and modeling methods are applied for NV and MRW nuclide composition (in terms of WAC and long-term radiotoxicity) determination.
- HLW is not suitable for decontamination, but estimation of declassification possibility for the selection of most suitable disposal facility and packing (having in mind the best radioprotection) is needed for waste-treatment and storage optimization.
- Disposal strategy (cut and dispose, pack and go, pack and wait, pack and dispose) considering radiation safety of workers and other factors should be considered.

Classification of MRW should be performed following an iterative process driven by weighting of a set of different aspects [15]: protection of workers, risk assessment, protection of the public and environment, know-how on the nuclear facility (specific of reactor: no accidents or there was fuel leakage); management options for radioactive wastes, including predisposal and disposal solutions that are available or selected:

- Waste inventory determination;
- Decommissioning strategies and end state;
- Applicability/type of the techniques for decommissioning including dismantling capacity;

- Secondary radioactive waste generation;
- Auxiliary facilities for dismantling;
- Infrastructure/workspace needed;
- Maintenance and dismantling time;
- Suitability of predisposal radioactive waste management options (meeting WAC);
- Decontamination processes;
- Clearance;
- Transport requirements (national WAC issue, distance of disposal territory, etc.);
- Cost estimation.

All recommendations for decision-making must consider environmental and radiation protection aspects, as well as the overall financial aspect of the selected decommissioning strategy. The selection and application of suitable dismantling and decontamination techniques become crucial elements of the successful conduct of decommissioning projects [16].

6 Radionuclide generation and distribution in MRW at the different nuclear reactors

If considering radionuclide generation and distribution in MRW at the different nuclear reactors there are certain similarities and differences, which depends on particular reactor type, fuel and moderator material applied, operation history etc. (see detail information in the Table 1). Due to the structural design of the different facilities (PWR, BWR, RBMK (Reaktor Bolschoi Moschtschnosti Kanalny), VVER (Vodo-Vodyanoi Enyergeticheskiy Reactor)) the radiation control area for MRW is different. In PWR technology, only the reactor building and the reactor auxiliary building are in radiation control area, while in BWR also the turbines and the generator are located in radiation control area [11]. The volume and activity of LILW generated annually by 1 GWe in different type NPP is given in Table 2. One can notice that highest volumes and activities belong to Gas-Cooled Reactor (GCR) and RBMK type reactors, while the lowest to PWR and Pressurized Heavy-Water Reactor (PHWR) reactors.

Table 1. The main differences for most common nuclear reactors in the world [17]

Reactor type	Countries	Number of the reactors	GW _e	Fuel	Coolant	Moderator
Pressurized Water Reactor (PWR) (VVER - Russian version)	USA, Russia, France, Germany, Japan	252	235	enriched UO ₂ , Mixed Oxide Fuel (MOX)	H ₂ O	H ₂ O
Boiling Water Reactor (BWR)	USA, Japan, Sweden	92	83	enriched UO ₂	H ₂ O	H ₂ O
Gas-cooled Reactor (Magnox & AGR)	Great Britain	34	13	natural U (metal), enriched UO ₂	CO ₂	graphite
Pressurized Heavy Water Reactor " CANDU " (PHWR)	Canada, China, India	33	18	natural UO ₂	D ₂ O	D ₂ O
Light Water Graphite Reactor (LWGR) (RBMK)	Russia, Ukraine Lithuania	14	14.6	enriched UO ₂	H ₂ O	graphite

Similarities of radionuclide production and distribution in MRW generated at different nuclear reactors are usually as follows:

- Reactor operation generates radioactive fission, activation and corrosion products;
- Methodology for characterization of the metallic waste is quite similar;
- Key and DTM radionuclides usually are the same;
- Scaling factors determination and NV methodology is quite similar.

Factors determining different radionuclide production and distribution in MRW are:

- Composition and construction geometry of different nuclear reactors metallic components;
- Neutron energy distribution and fluence in the reactor core;
- Reactor operation specific (normal/ incidents/ accident), time after reactor shutdown;
- Volume and activity of long-lived neutron activation products;
- Volume and activity of neutron activated fission and corrosion products released to reactor coolant.

Table 2. Volume and activity of LILW generated annually by 1 GWe NPP (according to [18])

Reactor type	Volume (m ³)	Activity (TBq)
Advanced Boiling Water Reactor (ABWR)	500	500
AGR	650	600
BWR	500	500
Fast Breeder Reactor (FBR)	500	500
GCR	5000	1000
LWGR (RBMK)	1500	1000
PHWR	200	100
PWR	250	100
VVER	600	600

6.1 Composition of different nuclear reactors metallic components

The main reactor materials differ depending on the reactor type: carbon steel and/or stainless steel are dominant in BWRs and Inconel or Incoloy in PWRs. As the element composition of these reactor materials differs, it is considered reasonable for Corrosion Products (CP) nuclides to be differentiated according to the reactor type. For BWR's the main metal activation products are ⁶⁰Co, ⁵⁹Ni, ⁶³Ni, ⁹⁴Nb, ¹⁴C [16]. See Table 3, for the details.

Table 3. Nuclide list of CP - radionuclides present in MRW generated at different nuclear reactors (the main radionuclides are presented with bold)

Nuclide	PWR	BWR	VVER	RBMK
¹⁴C	+	+	+	+
⁵⁴ Mn				+
⁵⁵ Fe	+	+	+	+
⁶⁰Co	+	+	+	+
⁵⁹ Ni	+	+		+
⁶³ Ni	+	+	+	+
⁹⁴ Nb		+	+	+
¹³⁷ Cs (FP)	+	+	+	+

Radionuclide composition directly influences the choice of dismantling and waste management technologies, which allows the minimization of volumes of radioactive waste and the cost of its management [19]. Metal structures are made from various grades of steel and metal alloys during the construction of a nuclear reactor. The choice of a specific type of material depends on the conditions in which the reactor will operate. The most commonly used are stainless steel, steel and various metal alloys such as zirconium for fuel rods and assemblies as resistant material to high temperatures and neutron activation. The materials used in the construction of the reactor become

radioactive during neutron activation, and also can be contaminated with radionuclides due to leakages during normal reactor operation or incident events. All these materials have impurities, the amount of which is regulated during production, and their permissible values are usually indicated in their passports. However, maximum permissible concentrations are often indicated only for the main composing chemical elements. As an example in Table 4, for structural elements of RBMK-type reactors, the following metal materials are used: steel 25 (support blocks and shielding plates), steel 10HN1M (support structures, upper and lower parts of biological shield), steel 08X18N10T (parts of pipes of technological channels, channel paths), zirconium-niobium alloy E125 (parts of pipes of technological channels located in the reactor core), etc. [20] [21].

Table 4. Example of RBMK metallic structural elements

Part of the reactor	Construction elements	Structure material
Fuel channels	Pipes in the stack	Zr-Nb alloy E125
	Pipes above and below the core	Steel 08X18N10T
Shielding plates	Steel plates	Steel 25
Support blocks	Top/ Bottom steel plates	Steel 25
Metal of serpentine construction	Steel plates	Steel 10HN1M

Table 5. Chemical composition of metal materials of reactor RBMK-1500 and max impurity (wt%)

Element	ZrNb alloy	Element	Steel		
			10HN1M	08X18N10T	25
Al	$7.5 \cdot 10^{-3}$ [22]	C	0.1 [23]	0.08 [24]	0.26 [25]
B	$8.0 \cdot 10^{-5}$ [20]	Si	0.27 [23]	0.8 [24]	0.27 [25]
Be	$2.0 \cdot 10^{-3}$ [20]	Mn	0.45 [23]	2.0 [24]	0.65 [25]
C	$2.7 \cdot 10^{-2}$ [20]	Cr	0.85 [23]	18.0 [24]	0.25 [25]
Ca	$3.0 \cdot 10^{-2}$ [20]	Ni	1.25 [23]	10.0 [24]	0.3 [25]
Cd	$5.0 \cdot 10^{-5}$ [22]	Ti	-	0.7 [24]	-
Co	$2.5 \cdot 10^{-2}$ [20]	Mo	0.5 [23]	-	-
Cu	$5.0 \cdot 10^{-3}$ [20]	S	0.03 [23]	0.020 [24]	0.04 [25]
Fe	$1.8 \cdot 10^{-2}$ [20]	P	0.03 [23]	0.035 [24]	0.035 [25]
H	$2.5 \cdot 10^{-3}$ [22]	Cu	0.3 [23]	-	0.3 [25]
Hf	$4.0 \cdot 10^{-2}$ [20]	As	-	-	0.08 [25]
Mg	$2.0 \cdot 10^{-3}$ [22]	Co	0.01 [20]	0.04 [20]	0.01 [20]
Mn	$5.0 \cdot 10^{-3}$ [22]	Fe	96.21	68.33	97.8
Mo	$5.0 \cdot 10^{-3}$ [22]				
N	$8.0 \cdot 10^{-3}$ [20]				
Nb	$2.5 \cdot 10^0$ [20]				
Ni	$1.1 \cdot 10^{-2}$ [20]				
O	$1.3 \cdot 10^{-1}$ [22]				
Pb	$5.0 \cdot 10^{-3}$ [20]				
Si	$2.2 \cdot 10^{-2}$ [22]				
Ti	$6.0 \cdot 10^{-3}$ [20]				
Zr	$9.72 \cdot 10^1$ [22]				

If there are several sources of information on the concentration of the same chemical element, a higher concentration value is used in the calculations, i.e. the conservative option is chosen (see Table 5). If concentration limits are specified, the average value is taken, if only the upper limit is specified, the upper limit is taken.

The concentration of all necessary impurities is often not indicated in the design documentation of the NPP; therefore, scientific literature is used, which publishes the real results of measurements of these concentrations of impurities. Usually, the assessment of the neutron activation of metal structures of the RBMK-1500 reactor is made considering either maximal concentrations of

impurities according to metal production standards and scientific literature data or corrected concentrations taking into account experimental (validation) measurements [19].

Table 6. Example of PWR metallic structural elements

Part of the reactor	Construction elements	Structure material
Fuel Assembly (FA) structure	Springs/Bolts/Spacers	Inconel X 750 Inconel 718
Fixed core components	Large components Springs/Bolts/FA centring pins	Steel 1.4550 / 1.4541 Inconel X 750
Reactor Pressure Vessel (RPV)	Vessel, Lid	Steel 1.6310
Activated structures outside RPV	Primary circuit components Water-wetted structures	Steel 1.6310 / 1.6751 Steel 1.4550 / 1.4541

Table 7. Example of PWR: Chemical composition of metal materials of PWR reactor, max impurity (wt%).

Element	Steel				Inconel 718	Inconel X 750
	1.4541	1.4550	1.6310	1.6751	2.4668	2.4669
Al			0.010-0.040 [26]	0.05 [26]	0.50 [26]	0.40-1.00 [26]
As			0.025 [26]			
B					0.050 [26]	
C	0.08 [26]	0.08 [26]	0.17-0.23 [26]	0.17-0.25 [26]	0.040 [26]	0.08 [26]
Cr	17.0-19.0 [26]	17.0-19.0 [26]	0.20 [26]	0.30-0.50 [26]	19.0 [26]	14.0-17.0 [26]
Co	1.5 [27]	0.221 [28]	0.030 [29]		1 [30] [30]	1.00 [31]
Cu		0.123 [28]	0.12 [26]	0.18 [26]	0.3 [30]	0.50 [26]
Fe					18.5 [26]	5.00-9.00 [26]
Mn	2.00 [26]	2.00 [26]	1.20-1.50 [26]	0.50-1.00 [26]	0.20 [26]	1.00 [26]
Mo		0.306 [28]	0.40-0.55 [26]	0.50-0.80 [26]	3.3 [30]	
N		0.035 [28]	0.013 [26]			
Ni	9.00-12.0 [26]	9.00-12.0 [26]	0.50-0.80 [26]	0.60-1.20 [26]	52.5 [26]	≥70.0 [26]
Nb		10x%C≤1.00 [26]				0.70-1.2 [31]
Nb/Ta					5.30 [26]	
P	0.045 [26]	0.045 [26]	0.012 [26]	0.020 [26]	0.15 [30]	0.02 [31]
S	0.030 [26]	0.015 [26]	0.008 [26]	0.020 [26]	0.15 [30]	0.010 [26]
Si	1.00 [26]	1.00 [26]	0.15-0.30 [26]	0.35 [26]	0.30 [26]	0.50 [26]
Sn			0.011 [26]			
Ta			0.030 [29]			
Ti	5x%C≤0.70 [26]				0.90 [26]	2.00-2.75 [26]
U						
V		0.057 [28]	0.02 [26]	0.03 [26]		

Table 6 summarises the most used materials in structural elements of PWRs. For FA structures and fixed core components these materials are Inconel X 750 and Inconel 718, used e.g. for springs,

bolts or spacers, or steel 1.4550 and steel 1.4541 for larger components. Steel 1.6310 is used for the reactor pressure vessel and its lid, as well as components of the primary circuit. Additionally, steel 1.6751, 1.4550 and 1.4541 are used. Table 7 lists the main elements of these six materials, as well as often found impurities.

6.2 Neutron spectrum, the neutron flux, runtime history of the reactors

A numerical modelling is indispensable tool for estimation of neutron flux properties and radionuclide activation in different metallic construction of the reactor during nuclear power plant operation and during decommissioning process. The different complexity 1D-2D-3D models of the reactor are created for assessment of activation of metallic constructions by the neutron flux in the different NPP. Various numerical simulation codes with different modelling approaches are used for modelling of nuclear processes and calculation of main performance parameters/activation of radionuclides in the reactors. The modelling methods and code packages are chosen depending on the research needs and the goal of the particular investigation. The deterministic methods, i.e., code packages like HELIOS, SCALE, and WIMS are usually used for numerical modelling of the critical reactor [32] [33] [34], while Monte Carlo is used for modelling both critical and sub-critical systems [35]. The irradiation history and reactor design information are necessary to perform a precise modelling of spent nuclear fuel (SNF) composition and generated neutron flux properties in the reactor. Additionally, the modelling should be validated by the actual experimental data, to ensure that the considered model is an accurate representation of the real reactor construction. The validated calculation models for evaluation of reactor performance parameters and nuclear fuel composition serve as a guiding tool during the change in operation parameters or fuel enrichment/design without costly and complicated experiments [36] [37].

The most commonly used codes are various versions of SCALE and MCNP, which were used for SNF composition and neutron activation, criticality safety, radioprotection calculations both for interim storage and as source term for disposal. One should note that the USA produced codes (e.g., SCALE package) are optimized for the PWR and BWR type reactors [33], but they have been successfully adopted also for other types of reactors [38]. As it was mentioned above the needed information for neutron activation calculation (MCNP6/SCALE6.2) is as follows:

- reactor core constructions geometry and materials (with impurities),
- fuel type, enrichment and geometry,
- power history,
- time after reactor shutdown, etc.

Reactor components subjected to a neutron flux are activated and also undergoing radiation induced changes that impact their ability to function as designed [39]. The neutron flux properties in different reactors metal constructions are presented in Table 8 and Figure 6.

Table 8. List of different reactors and maximum neutron fluxes achievable in various materials of core components in these reactors.

Reactor	Acronym	Reactor type coolant	Fast flux (E > 1 MeV) $n \cdot cm^{-2} \cdot s^{-1}$	Thermal flux (E < 0.5 eV) $n \cdot cm^{-2} \cdot s^{-1}$	Total flux (E > 0 eV) $n \cdot cm^{-2} \cdot s^{-1}$
Boiling Water Reactor	BWR	Pressure vessel Light water	0.06×10^{15a}	0.04×10^{15a}	0.3×10^{15a}
Pressurized Water Reactor	PWR	Pressure vessel Light water	0.07×10^{15a}	0.06×10^{15a}	0.4×10^{15a}
Channel Reactor of High Power	RBMK	Pressure tube Light water	0.02×10^{15a}	$0.07 \times 10^{15a*}$	0.2×10^{15a}
Canada Deuterium Uranium	CANDU	Pressure tube Heavy water	0.02×10^{15b}	0.1×10^{15b}	0.3×10^{15b}

^aMid-core flux, ^bMean pressure tube flux, RBMK case corrected according to [40]

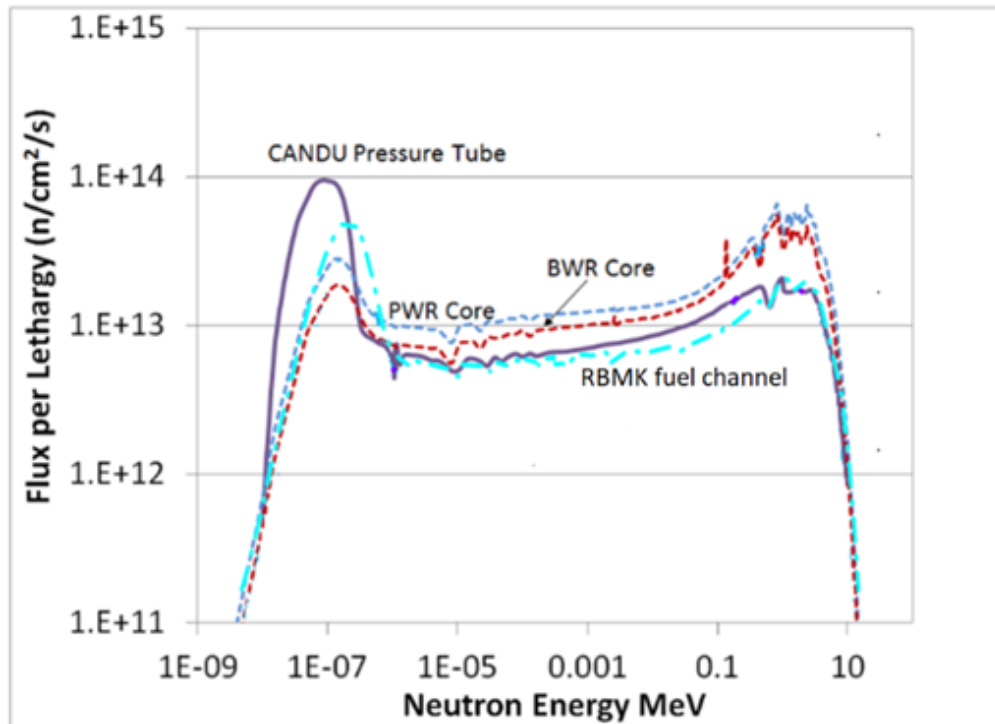


Figure 6 Neutron flux energy distribution for PWR, BWR, RBMK and CANDU type reactors (adapted according to L. Walters [41])

6.3 Neutron activation products in different nuclear reactors metallic components

Pre-dismantling classification of the metallic waste using modeling approach allows sorting of reactor materials regarding the level of activation: highly, intermediate, low activated metal constructions and non-activated metals.

Simulation models of the reactors can give the neutron flux distribution in a 3D reactor core and evaluation of activation of the reactor materials can be performed. Neutron activation in the nuclear reactor metallic construction materials (reactor pressure vessel, pipelines, fuel channels, reactor internal constructions, Control and Protection System (CPS) rods, equipment of low or medium activation) can be determined for different reactors. After validation of the models it becomes possible to classify the reactor materials regarding the level of activation (see Figure 7 as an example for RBMK case). Furthermore, the nuclide composition in the reactor materials in the HLW and ILW can be preliminary determined. Similar approach has been used for German BWR Philippsburg 1 (KKP1) and the PWR Neckarwestheim 1 (GKN I) characterization and neutron induced activation calculations for the structural components of the reactors. This technique helps classification of higher activity RW (HLW, ILW) concerning dismantling radiation protection and best packaging concept selection and allows to predict future costs for conditioning and packaging [42].

As concerning RBMK case separation of activation zones according to neutron flux in RBMK-1500 reactor by using MCNP6/SCALE6.2 modelling was done [40]. The neutron flux in the entire reactor using full scale 3D (MCNP6) model was calculated. The SCALE6.2 COUPLE sequence was used to evaluate the macroscopic cross-section of neutrons in the materials depending on the calculated neutron fluxes. Further, multigroup (252) neutron cross-section libraries were employed in the ORIGEN-S (of SCALE 6.2) for nuclide concentration evaluation in RBMK-1500 reactor construction materials including distant structures with strongly reduced neutron flux. The details are presented in Figure 7: highly activated zones are indicated with red line, zone of intermediate and low activation metal construction with green line and non-activated materials, for which only surface contamination is present - zone above green line [40].

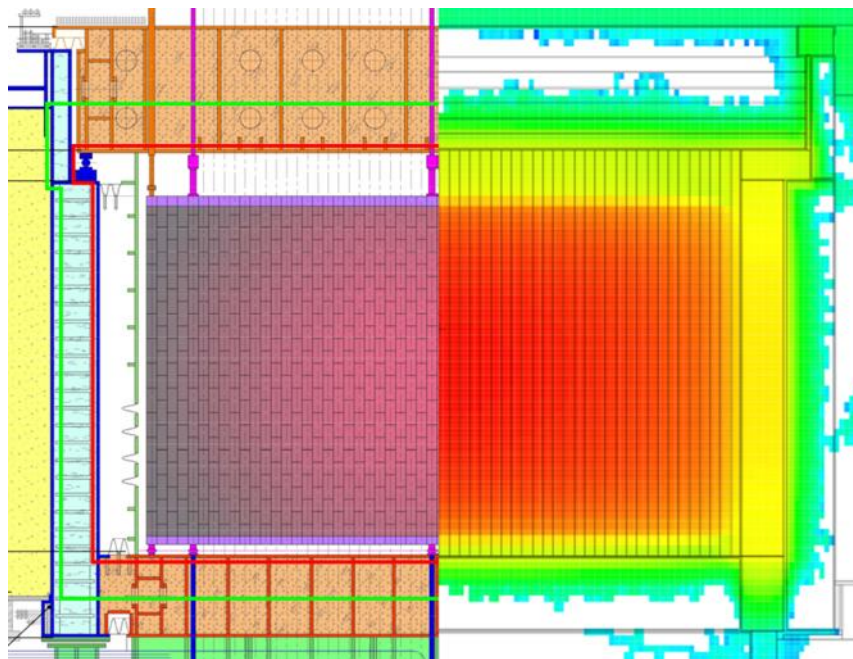


Figure 7 Separation of activation zones according to neutron flux in RBMK-1500 reactor by using MCNP6/SCALE6.2 modelling: highly activated zones (red line), zone of intermediate and low activation metal construction (green line) and non-activated materials (only surface contamination) above green line [40].

Similar approach but with simplified geometry (segment 12 indicated in Figure 8) was used in [21], where the activation process was modelled by using SCALE 5 software ORIGEN-S. The results of the neutron activation modeling of the reactor components show that the maximum total specific activity immediately after the final shutdown of the RBMK-1500 reactor in the central parts of the fuel channels reaches up to $\sim 1,9 \cdot 10^{10}$ Bq/g, while during 150 years it decreases to $6,7 \cdot 10^6$ Bq/g. During ~ 40 years the specific activity of the fuel channels is practically determined by the specific activity of ^{60}Co (including ^{95}Nb and ^{95}Zr during the $\sim 0,5$ years), while during $\sim 50 - 150$ years the main radionuclide becomes ^{94}Nb . For the activity of the fuel channels are also important ^{63}Ni and ^{14}C radionuclides.

The main radionuclides that determine the activity of the top and bottom of fuel channels are ^{55}Fe , ^{60}Co , ^{63}Ni , ^{59}Ni and ^{14}C . The estimated maximum total specific activities are in the parts of these structures closest to the core, where they are exposed to the most intense neutron fluxes. Based on the results of the assessment, the maximum total specific activities of the upper and lower parts of the fuel channel during reactor shutdown are, respectively, $\sim 1,8 \cdot 10^8$ and $2,3 \cdot 10^8$ Bq/g, while after 150 years they decrease to $\sim 1,3 \cdot 10^6$ and $\sim 1,7 \cdot 10^6$ Bq/g.

The specific activity in protective and support steel plates immediately after the final shutdown of the RBMK-1500 reactor is found at about $3,4 \cdot 10^8$ and $5,6 \cdot 10^8$ Bq/g, respectively. For the first 30 years after the final shutdown of the reactor the ^{55}Fe and ^{60}Co have the highest specific activities. After 150 years, the specific activity in protective and support steel plates decrease to $\sim 1,3 \cdot 10^5$ and $\sim 2,2 \cdot 10^5$ Bq/g, respectively. At $30 - 150$ years the main radionuclide is ^{63}Ni (and ^{59}Ni), which practically determines the total activity of steel plates. Depending on the level of uranium impurity of the metallic components, neutron-induced nuclear fission can also occur within the components, so that fission products are formed here through neutron activation. These fission products are firmly bound in the metal.

It is important to note that the results of preliminary simulation of the radionuclide concentrations in the nuclear fuel and the reactor construction materials usually are used to determine the optimal program for sampling. The measurements of the samples then are used to adjust parameters of the model and to determine an optimal segregation of RW into different waste streams. Preference in the determination of scaling factors using modeling methods (as in Figure 10) is also used in order to reduce the number of measurements as much as possible in difficult accessible (high activity,

difficult to reach) cases. However, in all cases it is necessary to validate used models and to measure sufficient number of specific activity concentrations of radionuclides.

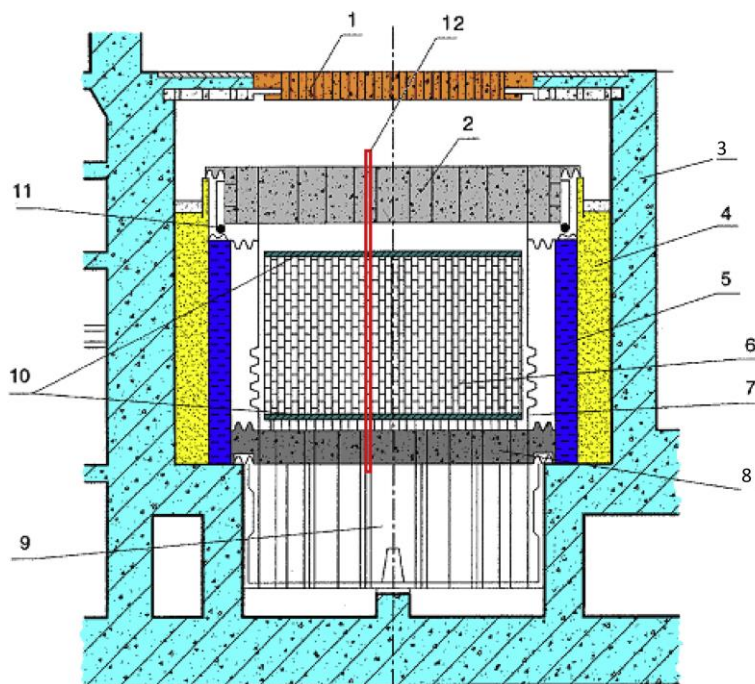


Figure 8 Cross-sectional cut of RBMK-1500 reactor vault [23], as in [21]. 1 - Central hall floor; 2 - Top metal structure with serpentinite; 3 - Concrete vault; 4 - Sand cylinder; 5 - Water tank; 6 - Graphite stack (core, top, bottom and side reflectors); 7- Reactor shell; 8 - Bottom metal structure; 9 - Reactor support structure; 10 - Shield and support steel blocks; 11 – Roller supports; 12 – Modeled segment [21].

6.4 Neutron activation and corrosion as well as fission products released to reactor coolant in different nuclear reactors

Reactor operations generate radioactive fission, activation, and corrosion products (see Table 9). The fission process produces radioactive fission products and neutrons in the core, which activate both the materials in the vicinity of the core as well as the coolant and hence the corrosion products present in the coolant. Although the radioactive inventory is contained in the fuel rod, during normal operation, radioactive products diffuse through the fuel rod cladding or leak through defects in the cladding and enter the reactor coolant system (RCS). These products could also pass from the RCS to the secondary coolant system through steam generator tube leaks [12].

- Fission products (FP) appear due to cladding defect: volatile (Cs, Ba, Sr, Ce, Pr, Zr, Ru, etc.) and actinides, which normally do not migrate. Amount depends on the length and position of the defect and the burn-up fraction of the defective fuel rod. FP are incorporated in the metallic oxides of the primary circuit surfaces.
- Activation products (AP) - chemical elements activated under the neutron flux (^{16}N , ^3H , etc.). Important nuclides for radiation protection - $^{110\text{m}}\text{Ag}$ and ^{124}Sb both due to defect of control rod cladding.
- CP come from the corrosion of the structure materials in contact with the primary coolant. The main CP responsible of dose rates are ^{60}Co and ^{58}Co , the others are ^{51}Cr , ^{54}Mn , ^{59}Fe . The prediction of the Co sources depends on the accuracy of transport, deposition and releasing mechanisms or kinetics of the nuclides and their soluble or particular forms. During measurement both ^{60}Co from corrosion and from activation of construction impurities is detected.

Besides the AP inside the construction materials, the concentration of AP in the surface contamination of the reactor components is due to the corrosion of the reactor materials and the solubility of the radionuclides in the coolant liquid, resulting in transportation inside the primary circuit

and deposition on the components. Furthermore, fission products are transported inside the primary circuit and detected in the surface contamination. For that reason, specific measurements are needed after dismantling of the components to distinguish activation products inside nuclear reactor construction materials and to evaluate the level of contamination.

Within the coolant systems, the activation of chemical elements present in water coolant forms activation products. The calculated levels of these products were determined using operating plant data.

Specific activation products include the following:

^{16}N production results from neutron reaction with ^{16}O . ^{16}N is a significant gamma emitter but has a short half-life. This chapter presents data for ^{16}N but, because of its short half-life, this product is primarily a concern within the containment area.

^{17}N production results from the neutron reaction with ^{17}O . ^{17}N is a short-lived neutron emitter and therefore is not a significant radiation source.

^3H is continuously produced during reactor operation from ternary fission in the fuel and from neutron activation of reactor coolant materials. In the reactor coolant, approximately two-thirds of the tritium activity results from the activation of ^{10}B in the soluble poison, and approximately one-third results from the leakage and diffusion of ternary fission tritium through the fuel clad. Less than three percent of tritium activity is produced from the activation of lithium and deuterium in the reactor coolant.

^{41}Ar production occurs by neutron activation of naturally occurring argon in air. ^{41}Ar production is primarily within the biological shield due to ventilation by air, but as short-living nuclide it is important only during reactor operation.

^{14}C is formed by the activation of carbon, nitrogen or oxygen. The primary reactions in a light water reactor are: $^{13}\text{C}(n,\gamma)^{14}\text{C}$, $^{14}\text{N}(n,p)^{14}\text{C}$, and $^{17}\text{O}(n,\alpha)^{14}\text{C}$. Reactor coolant corrosion products result from the in-core activation of impurities in the coolant fluid and activation of corrosion and wear products (including metallic fines). Corrosion products include isotopes of sodium, iron, cobalt and other miscellaneous nuclides. Corrosion product calculations are also based on operating plant data and are independent of fuel defect level [12].

Theoretical approach for determination of the specific activities of the metallic reactor constructions – pressure vessel and its internals, main piping, main circulation pumps, ion exchange resin filters – is also possible [43]. Historically many codes were developed for the Reactor Coolant System (RCS) radiological characterization dedicated to the determination of the relatively short lived beta and gamma emitting nuclides such as ^{51}Cr , ^{59}Fe , ^{54}Mn , ^{95}Zr during reactor operation and the long-lived nuclides $^{59,63}\text{Ni}$, ^{93}Zr , ^{90}Sr , ^{99}Tc , ^{129}I , $^{134,135,137}\text{Cs}$, $^{234,235,238}\text{U}$, $^{238,239,240,241,242}\text{Pu}$, ^{241}Am , $^{242,244}\text{Cm}$ for decommissioning stage. The decommissioning requires information about the activities of these long-lived radionuclides to be declared and compared with the radioactive WAC (see Figure 10 for nuclide vector determination steps).

The modelling of metallic constructions contamination processes via coolant of main circulation circuit (MCC) can be performed by sophisticated thermo-hydraulic codes [44] [45], which are used/adapted for analysed system in order to obtain the contamination level of the constructions both in normal operation, as well as during accidental states and in post closure stage. Specific activity of FP in reactor coolant depends on many factors, such as the number of operating defective fuel elements, reactor power history, fuel burnup and efficiency of reactor coolant purification system [46]. The processes of the release from fuel cladding defects, transport with coolant, and sedimentation on particles in the coolant or internal surfaces of the MCC take place. In addition, the sediments can dissolve or erode from MCC surfaces back to the coolant due to changing chemical or thermal-hydraulic conditions of the coolant. These processes are similar in BWR or PWR in general, although due to different water chemistry and physical processes to some extent various final contamination of MCC internal surfaces can be observed.

As an example of the codes dedicated to the modelling of the RCS internal contamination is the computer code OSCAR (Outil de Simulation de la Contamination en Réacteur), which was developed for Pressurized Water Reactors [44] [47], but also was adapted for modelling of the RCS internal surface contamination of VVER-440/230 reactors in [48].

The computer code OSCAR consists of two parts: the model OSCAR FP for fission products and actinides contamination calculation and the model OSCAR CP for corrosion products contamination calculation. These two parts use different physical models and RCS nodalization, therefore two different input files are used for FPs and CPs calculations. Despite of this, due to a similar general approach some input parameters are common (reactor power and power history, coolant flow rate, purification efficiency, core zoning). For OSCAR CPs calculations the thermal-hydraulic data, parameters such as fluid velocity, flow rate and fluid temperature are provided in OSCAR input. Furthermore, wall temperature is of major concern, since corrosion product equilibrium concentrations depend on the wall temperature gradient along the PC. In addition, material properties, such as metal composition and density of each part of the circuits have to be specified, since structural components are the source for specific corrosion products. Reactor operating conditions, which may impact contamination, have also to be provided in OSCAR input. These are information on power history, water chemistry and reactor shutdown. Application of the OSCAR appears to be a useful tool to determine preliminary scaling factors for the radiological characterization of systems, it can be useful for analysis of the distribution of surface contamination and to disclose the necessity of introduction of more than one scaling factor. Also, it provides the possibility to identify the parts of circuits differently contaminated by FPs and CPs. Therefore, calculation results can be used as a reference information for the determination of the recommended sampling places and preliminary number of samples to be taken from the contaminated equipment, taking into account the distribution profile of the nuclide activity in the circuits [48].

It has to be noted that radiological characterization and application of particular methods, such as scaling factor approach, or indirect (modelling) assessment methods are only possible if all equipment to be characterized is still in place as it was operated. Only under this condition it is possible to apply special modelling tools, which take into account the information of the physical processes governing the generation of radioactive waste [49]. After the equipment is dismantled and if information about its specific original place in the NPP is lost (or change of radioactive contamination occurred) the modelling is no more practicable as the information about contamination processes cannot be directly applied.

In any case, direct measurements are needed in order to validate the modelling results and to prove the derived nuclide vector for each radioactive waste stream.

7 Scaling factors approach

7.1 Key and Difficult-To-Measure (DTM) Radionuclides

Many of the important long-lived radionuclides contained in radioactive waste are DTM (see Table 9 for details) because they are low energy gamma, beta or alpha emitting nuclides. Identification of DTM nuclides using complex radiochemical analysis is costly, time consuming and not practical for large numbers of waste packages. This is especially important for LLW and VLLW, which are the most common classes of waste on the NPP site and for them decontamination and declassification would be the most preferred way before disposal. The internationally available experience [16] [50] presents the scaling factor methodology that, in many cases, can be applied for evaluation of the radioactive inventory of DTM nuclides in waste packages. This methodology relies on establishing a correlation (scaling factor) between the DTM nuclides and easy to measure (ETM) nuclides – or so-called key nuclides. The inventory of the ETM nuclides in a waste package can be derived based upon external gamma radiation measurements carried out on the waste package, and the DTM nuclides can be estimated from the inventory of the ETM nuclides using established scaling factors.

The selection of the key nuclides is performed considering the following criteria:

- Strong gamma emitter;
- Appears sufficiently often at measurable levels in waste;
- Sufficiently long half-life;
- Representative for either FPs or activated CPs or APs of fuel (alpha emitters);
- Representative for radionuclide generation and transport conditions.

Table 9. List of radionuclides to be declared for MRW

Decay	Nuclide	T1/2 (y or d)	Decay	Nuclide	T1/2 (y or d)
Gamma	⁶⁰ Co	5.2714 y	Beta	⁵⁵ Fe	2.73 y
	¹³³ Ba	10.51 y		⁶³ Ni	100.1 y
	⁵⁴ Mn	312.3 d		²⁴¹ Pu (²⁴¹ Am)	14.35 y
	⁹⁴ Nb	2.03E+4 y		⁹⁰ Sr (⁹⁰ Y)	28.79 y
	¹⁰⁶ Ru	373.59 d		⁹⁹ Tc	2.111E+5 y
	^{108m} Ag	418 y		¹²⁹ I	1.57+E7 y
	^{110m} Ag	249.79 d		¹⁴ C	5730 y
	¹²⁵ Sb	2.7582 y		³⁶ Cl	3.01E+5 y
	¹³⁴ Cs	2.0648 y	Alpha	²³⁸ U	4.468E+9 y
	¹³⁷ Cs(^{137m} Ba)	30.07 y		²³³ U	1.592E+5 y
	^{166m} Ho	1.20E3 y		²³⁴ U	2.455E+5 y
	¹⁴⁴ Ce	284.893 d		²³⁵ U	7.038E+8 y
	¹⁵² Eu	13.537 y		²³⁸ Pu	87.7 y
	¹⁵⁴ Eu	8.593 y		²³⁹ Pu	24110 y
	¹⁵⁸ Tb	180 y		²⁴⁰ Pu	6563 y
	¹⁵⁵ Eu	4.7611 y		²⁴² Pu	3.733E+5 y
			²⁴¹ Am	432.2 y	
			²⁴² Cm (^{242m} Am)	141 y	
			²⁴³ Cm	29.1 y	
			²⁴⁴ Cm	18.10 y	

To determine the activity concentrations of these DTM nuclides, each country usually selects the CP nuclide ⁶⁰Co as the key nuclide and determines the concentration of ⁶⁰Co by direct measurements on waste packages. Others, such as ¹⁵²Eu, ⁵⁸Co or ¹²⁴Sb, did not fulfil half the criteria listed. ⁶⁰Co is the only suitable radionuclide for activated CPs. The activity concentrations of the DTM nuclides are then calculated according to the functional relationship established between the activity concentration of ⁶⁰Co and the DTM nuclides such as ⁶³Ni. For FPs there is no major choice other than ¹³⁷Cs. Alternatives, such as ⁹⁴Nb, could fit for CPs, but usually is less frequently present in the waste samples. A suitable ETM for alpha emitters is ²⁴¹Am. It is used as key nuclide for alpha emitters and transuranium elements. However, the occurrence of alpha emitters in waste in measurable quantities is usually rather low. Therefore, the migration and occurrence similarities in RW of most alpha emitters to ⁶⁰Co led to the decision to omit a separate key nuclide for alpha emitters. Nonetheless, today total alpha activity is sometimes used as an additional parameter for RW storage and final repository decision.

7.2 Scaling factors in different metallic components of different nuclear reactor types and time evolution of scaling factors

In order to determine a representative nuclide vector for a defined set of metallic waste constructions or systems that will form a waste stream when dismantled, it is necessary to cover a larger part of the systems that make up the stream in order to increase representativeness and experimentally confirm the system division into streams. Also, in order to optimize the scope and cost of the project, technologically similar systems (i.e, systems that are interconnected because they are affected by flow of the same technological process liquid or gas, similar form, surface contamination peculiarities etc.) are combined into a group of systems that form a stream that will be characterized with one nuclide vector.

The calculated ratio of key and DTM nuclides activity concentrations should remain constant within some range of uncertainty for all the samples resulting from the same waste stream. In other words,

Scaling Factors can be used to distinguish one waste stream from another, enabling their integration and classification, as explained below.

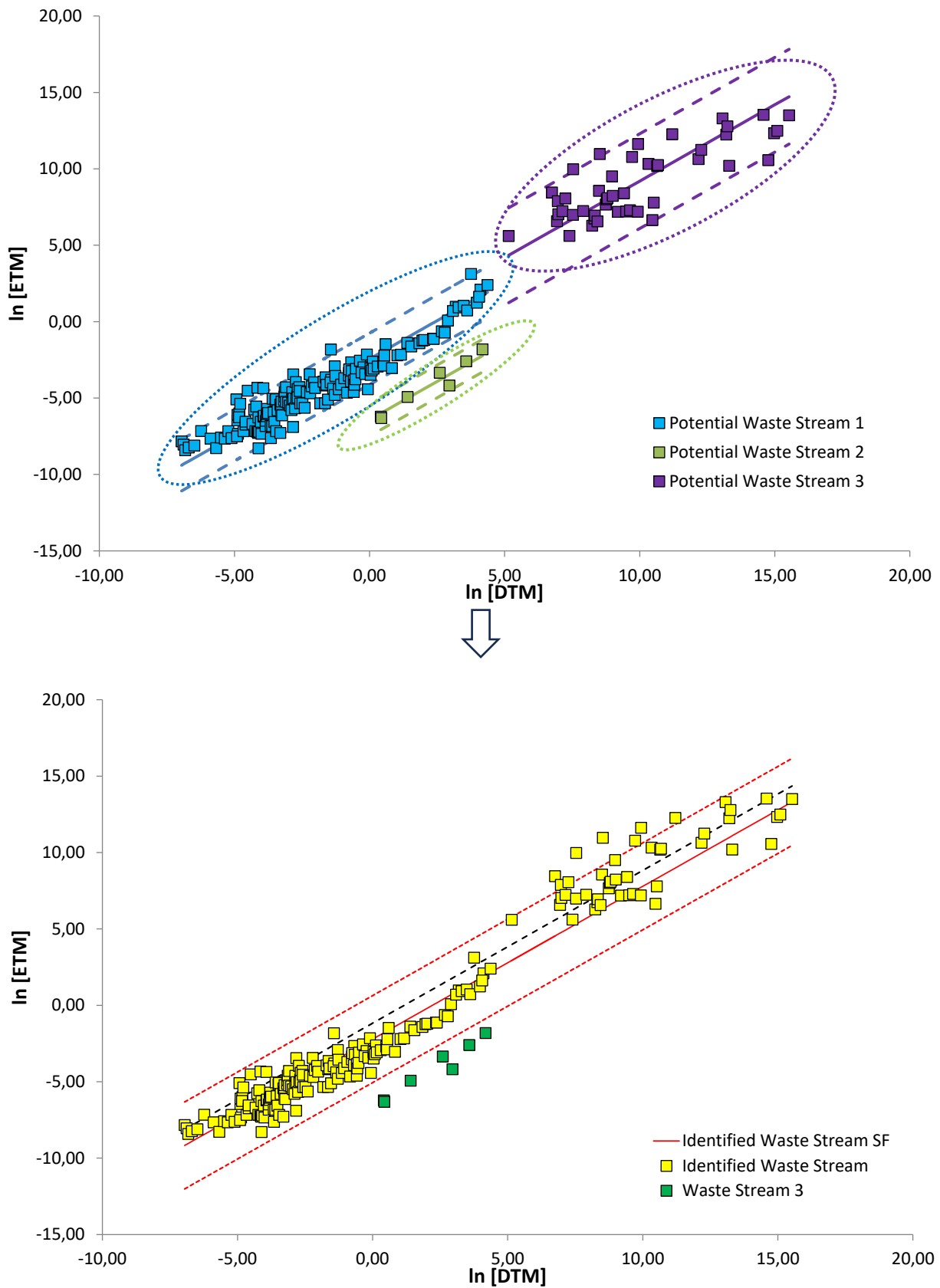


Figure 9 Waste stream integration.

When data collected from different metallic constructions of nuclear plant is available, the first step should be to consider them as potential different waste streams for each key-DTM selected pair. It is imperative to check the applicability of the scaling factor method for the particular RW stream by checking the correlation between concentrations of two radionuclides in this stream. If the correlation coefficient between concentrations of two radionuclides in the RW stream of MRW is less than 0.6 then this indicates that an additional analysis of the RW streams is required to redefine the assignment of RW to two streams.

A first log-log scatter graphical representation of collected data can give information about further grouping of the data (i.e waste streams) driven by the factors influencing correlation, mentioned in the previous section, may be necessary. The usage of the logarithmic concentrations allows one to include more diverse samples compared with the simple linear regression.

This might lead to a collection of candidate datasets, some of which could contain samples from different metallic constructions of the plant, that should be tested for correlation and compliance with the Scaling Factor model hypothesis in force.

Additional testing is necessary when a particular candidate dataset includes different potential waste streams to demonstrate that the data are all part of the same general population as it is sketched in **Error! Reference source not found.** That is, what based on a comparison of means and variance, it can be established that the data populations are not different with the desired confidence level.

As a result, new waste streams can be identified, and scaling factors derived from their datasets provided they pass all the tests.

During NPP operational life, the time stability of the calculated Scaling Factors may be roughly estimated through the study of their temporal change rate, which can be done by fitting the normalized activity concentration ratios versus time.

Fuel evolution due to reactor operation or other factors that could cause an appreciable change in the isotopic composition of the waste, require taking new samples to verify whether the SF are still valid, or an update is necessary.

Once a Scaling Factor is established, it may be tracked over time. Additional analysis of the waste streams identified by the methodology should be periodically incorporated into the datasets to check that data addition doesn't modify the value of the calculated SF.

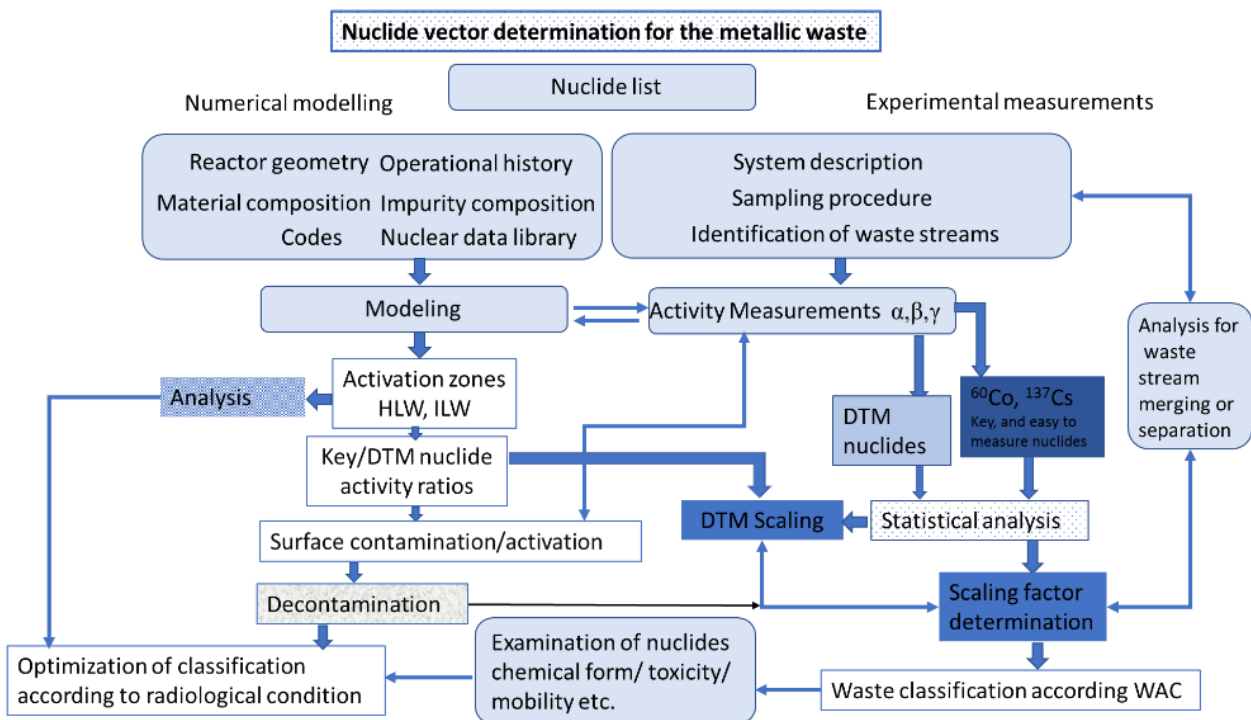


Figure 10 Chart of Nuclide vector determination and optimization.

Optimized NV is obtained by analyzing and systemizing the information about radioactive metallic waste streams, identifying the optimal list of relevant radionuclides (e.g., selection of ^{60}Co and ^{94}Nb

for ZrNb alloys), describing inter-correlations between key nuclides and difficult to measure nuclides (^{59}Ni , ^{63}Ni and other nuclides from the list of radionuclides to be declared for dedicated disposal site) including multivariate analysis of the already measured data at the sites and numerical analysis of activation and contamination parts for the waste streams as shown in the Figure 10 chart.

The optimization of NV can be done considering reactor construction activation zones, surface contamination determination, peculiarities of nuclides (waste form, matrix, radiotoxicity etc.) and also merging, separating different reactor systems according to measurements results and analysis. After the NV determination sequence, recommendation for optimized classification of the metallic waste in order to select the most appropriate decontamination techniques for clearance or declassification of the waste stream can be assessed. Especially, it is important to consider the possibility of clearance after implementation of a specific decontamination process (e.g. a sand blasting, melting). The validity of the nuclide vector is defined for a specific system and list of nuclides; also, the validity of the nuclide vector for relevant activity interval should be additionally justified either by measurements and/or theoretical considerations [16] [51]. As it can be seen in Figure 10 modelling branch can be particularly useful for the scaling factors determined by theoretical calculations (e. g. nuclear fuel composition calculations; material activation calculations, calculations of RCS piping internal contamination), but it should be validated by measurements. In particular, for optimised radioactive waste characterization the ratio between calculated and measured scaling factor should be not higher as several times for the relevant activity range in considered system (e. g., the RCS). This limit is set by the practical considerations and can be explained by the fact that waste with different nuclide vector has to be separated and treated individually as a separate waste stream. For practical reasons, it is desirable to have less waste streams in order to optimize the cost of waste management at NPP. However, reducing the amount of waste streams increases the conservatism of waste characterization and management: if lower activity waste is managed together with slightly higher activity waste (with increased amounts of radiotoxic nuclides) it leads to the overestimation of scaling factors and estimated activity of DTM nuclides in the lower activity waste part in the mixed waste stream. This finally results in the non-desirable overestimation of waste inventory, which leads to bigger volume of dedicated waste repository, where radioactive waste should be disposed off. That is why the iterative procedure of optimised nuclide vector approach is needed and should be applied every time in case of possibility to separate different (more and less contaminated, surface or volume activated) streams, to reconsider NV after decontamination of streams, take into account the time scale, etc.

The obtained NV should be checked for WAC of specific disposal facility and RW stream to be disposed off accordingly. In case of identification of possible improvement of RW acceptability to WAC of certain disposal facility (generic WAC to specific WAC), the final NV optimisation can be performed by means of, e.g., redistribution of RW streams, application of waste decontamination, etc. Also, if disposal and clearance requirements change an adaptation of RW characterization system can be easily done by applying the same NV optimisation approach [51] [16].

7.3 Factors affecting scaling factors

The correlation among activity concentrations of key and DTM nuclides, if exists, is mainly influenced by:

- The reactor type and component materials;
- The fuel performance history and plant operation conditions;
- The radionuclide production, transport mechanisms and physiochemical properties;
- The waste treatment and final waste forms.

Additionally, the following factors that affecting representative SF shall be taken into account:

- The number of measurements available to perform the SF calculation shall be sufficient.
- The data used to carry out the calculation must be representative of the studied stream.

The reliable determination of the nuclide composition is a key issue in the NPP decommissioning process. In order to assure the reliability of scaling factors the necessary amount of waste samples have to be taken, measured and evaluated afterwards. The laboratory measurement of radionuclide

activities is rather costly and time-consuming, therefore the optimal amount of measurements for the nuclide vector determination should be found.

Usually, only quantitative analysis of easy to measure nuclides called key nuclides (as ^{137}Cs , ^{60}Co) is carried out and the quantities of difficult to measure radionuclides are determined indirectly using the practically established ratios i.e. scaling factors to each waste stream. First, it is necessary to determine whether such a correlation exists, i.e. whether the scaling factor method is applicable. If such method is applicable, the gamma emitting nuclides are then estimated by measuring in waste packages and radionuclide composition is defined by applying the scaling factors.

Radionuclide generation and migration pathway from the source to the waste stream has to be taken into account for calculation of the correlation dependence of DTM and Key radionuclide activities. In the case when the pathway of analysed radionuclides is significantly different due to different origin of radionuclides (FP or AP), dissimilar chemical properties (e.g., ^{134}Cs , ^{90}Sr , actinides and key nuclide ^{60}Co), additional key nuclides (intermediate key nuclides) may be selected, whose calculated activities in nuclear fuel and the MCC coolant or in RW correlated better, e.g., $^{239+240}\text{Pu}$ in case of actinides and ^{137}Cs in case of FP. Then the scaling factor of the investigated radionuclide with respect to the intermediate key radionuclide is simulated using an appropriate model of radionuclide generation in the reactor: the nuclear fuel depletion model or the model for activation of the reactor construction materials, or contamination of MCC coolant respectively. The peculiarities of these models for simulation of the generation of radionuclides are described in Sections 6.3 and 6.4. If the specific activities of some radionuclides (for instance - actinides) can be determined using numerical simulations of the evolution of nuclear fuel in a reactor and the activation of reactor structures, then the relative activity A_x of DTM radionuclide can be determined by multiplying the relative activity of the auxiliary radionuclide (AR) by the ratio of the activities of these two radionuclides in the nuclear fuel (or activated material) [13]:

$$A_x = k_{x,p} \times k_{p,y} \times A_y \tag{1}$$

where: A_y is the activity of the AR, $k_{p,y}$ is the proportionality coefficient between the activities of the auxiliary and the key radionuclide, $k_{x,p}$ is the scaling factor between the activities of the DTM and AR.

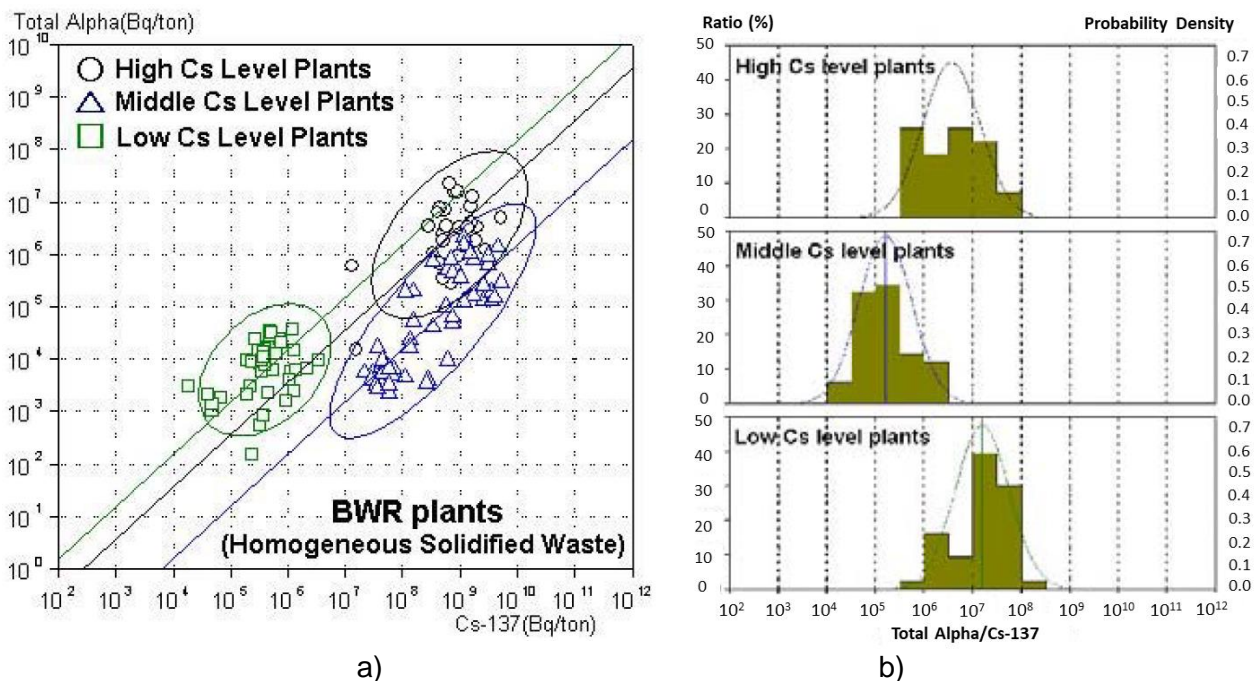


Figure 11 Effect of fuel failure on scaling factor (BWR, total alpha/ ^{137}Cs ; homogeneous solidified waste). (a) Scatter diagram; (b) histogram of total alpha/key nuclide [52] [53].

One of the factors affecting the scaling factors is the type of reactor. The activity concentrations of DTM radionuclides formed during the activation of corrosion products are estimated by the concentration of ^{60}Co (activation-corrosion product) in waste packages. The concentration of ^{60}Co is determined directly by measurements, while the concentrations of other radionuclides are

determined on the basis of a functional dependence on the concentration of ^{60}Co . Since different types of reactors use slightly different materials, for example, BWRs are dominated by steel and cast iron, and PWRs are dominated by nickel alloys, the scaling factors are different for each type of reactor [52].

The appearance and presence of fuel defects during reactor operation affects the scaling factors very much. In Figure 11, the influence of the amount of Cs, which is related to the number and severity of fuel failures, on the scaling factor of ^{137}Cs and total alpha in light water reactor homogeneous solidified waste is shown. Depending on the leakage, three groups of reactors are distinguished according to the to the total $\alpha/^{137}\text{Cs}$ ratio, the same was observed for $^{90}\text{Sr}/^{137}\text{Cs}$ ratio [52] [53] [54].

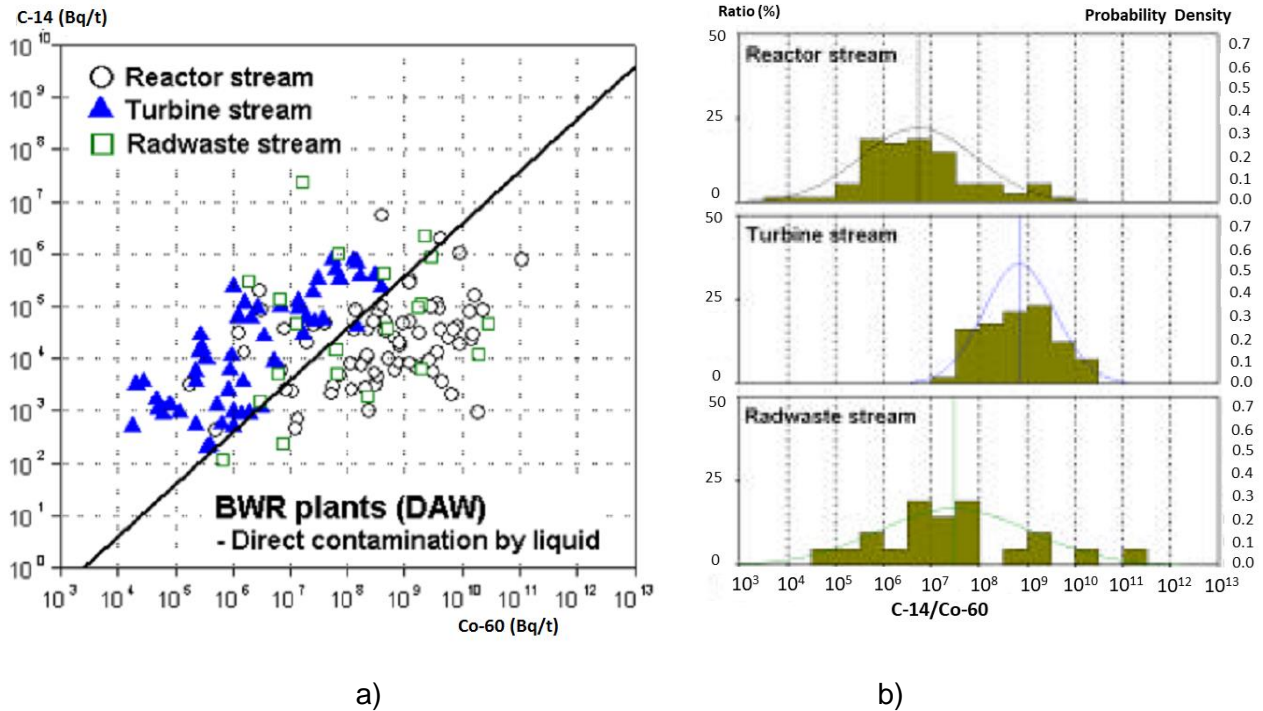


Figure 12 $^{14}\text{C}/^{60}\text{Co}$ scaling factor dependence on waste stream. (left) Scatter diagram of $^{14}\text{C}/^{60}\text{Co}$; (right) distribution diagram of $^{14}\text{C}/^{60}\text{Co}$ [52] [53].

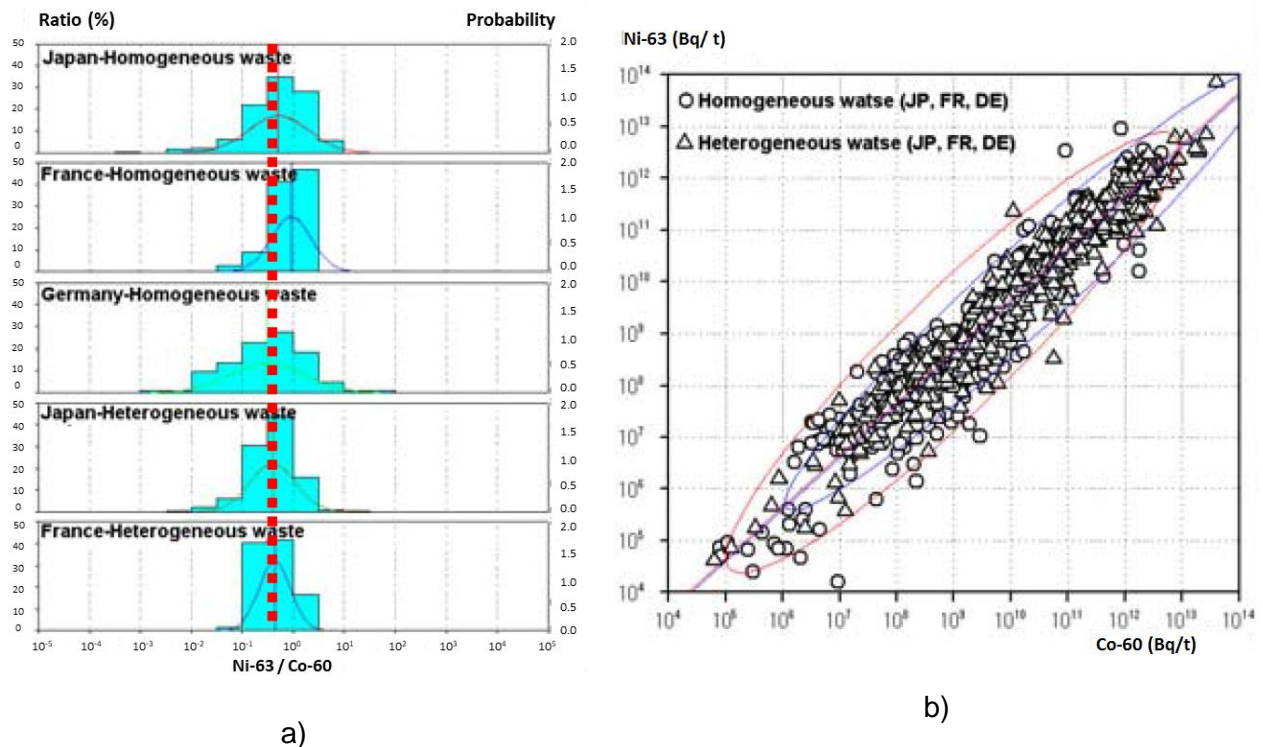


Figure 13 (a) Distribution and (b) scatter of $^{63}\text{Ni}/^{60}\text{Co}$ ratio for different radioactive waste streams generated at PWR plants from different countries [54].

The scaling factor is independent of the waste stream if activation corrosion products have similar physical and chemical characteristics (i. e. they are all insoluble metals). The exception is the $^{14}\text{C}/^{60}\text{Co}$ coefficient. For solid waste, it is necessary to determine the scaling factors according to the $^{14}\text{C}/^{60}\text{Co}$ ratio depending on the waste streams. In Figure 12 three waste streams for BWR reactors have been identified: reactor, MCC (turbine) and radioactive waste systems. For BWR reactors, the ^{14}C scaling factor for the turbine waste stream is different from the SF for the other two waste streams. This is because volatile ^{14}C and non-volatile ^{60}Co behave differently when entering the turbine. This feature of the BWR is due to the fact that water evaporates as it passes through the reactor core [52] [53].

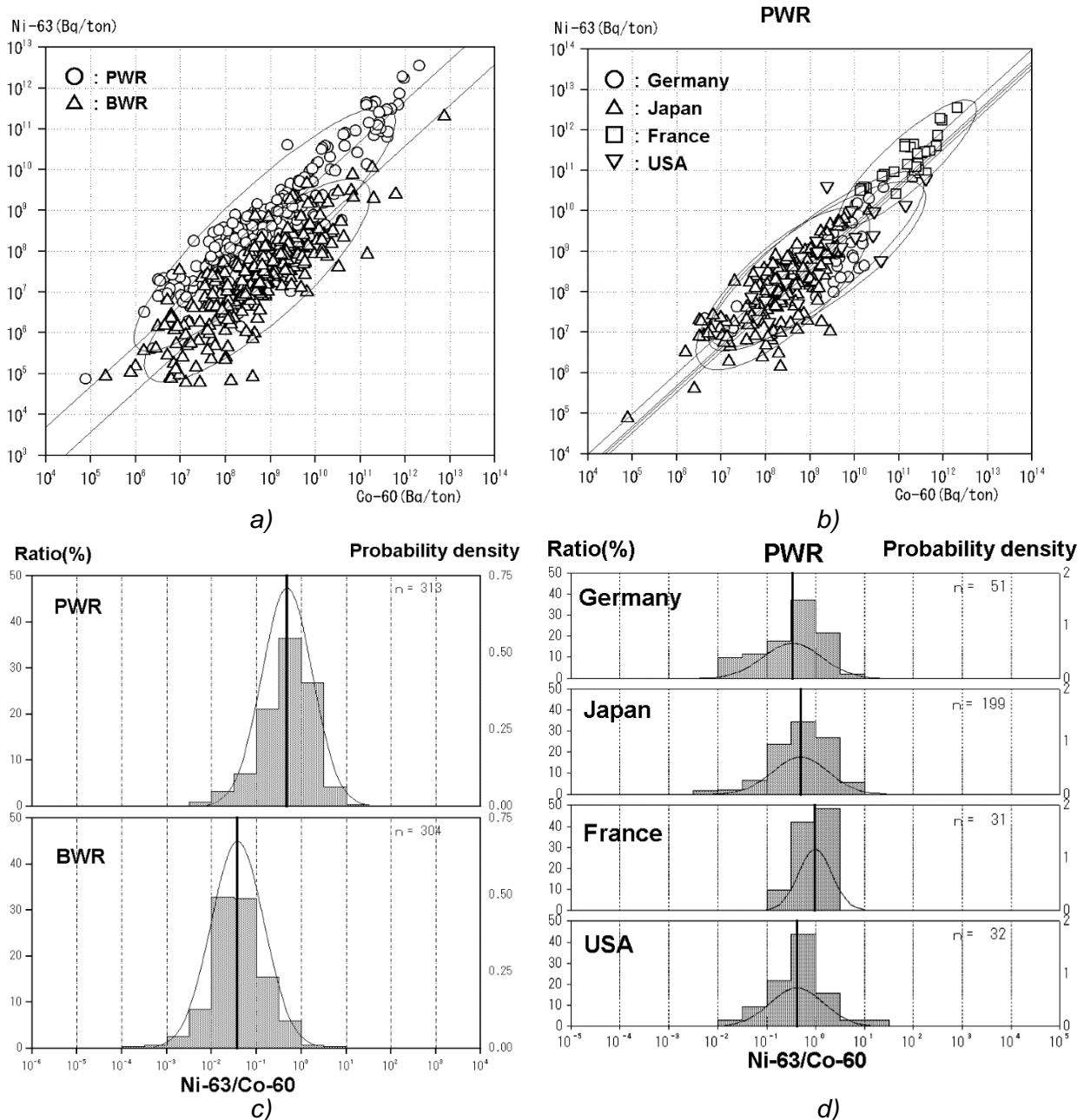


Figure 14 Comparison of $^{63}\text{Ni}/^{60}\text{Co}$ ratios for a BWR and PWR. Scatter (a) and distribution (c) diagram for different reactor type of $^{63}\text{Ni}/^{60}\text{Co}$; scatter (b) and distribution (d) diagram of $^{63}\text{Ni}/^{60}\text{Co}$ for PWRs in different countries [52] [53]

For homogeneous (activated) and heterogeneous waste (activated and contaminated), the scaling factor is the same if choosing ^{60}Co as a key nuclide, but it is important to take into account the

operating history (cases of fuel leaks) see Figure 13 for International comparison of homogeneous and heterogeneous waste [54]. When comparing the scaling factors for fission products and transuranic radionuclides with respect to ^{137}Cs , it was found that they differ for homogeneous and heterogeneous waste. This is due to the fact that ^{137}Cs is sufficiently soluble in comparison with ^{90}Sr or transuranic elements [52] [54]. The ^{60}Co scaling factor for corrosion products can be applied to any waste stream. However, the determination of ^{137}Cs scaling factors for fission products and transuranic radionuclides requires dividing the waste stream into at least two - homogeneous and heterogeneous waste stream. The scaling factors of radionuclides remain relatively constant throughout the life cycle, provided that the waste streams have been properly classified and the fuel leaks is also relatively constant [52] [53].

Depending on the reactor materials present in reactor core and/or main circulation circuit it is possible to distinguish PWR and BWR reactors according to the $^{63}\text{Ni}/^{60}\text{Co}$ ratio as presented in Figure 14 a) and c). BWR reactors are dominated by steel and cast iron, while PWRs are dominated by nickel alloys, so their scaling factors are also different. However, scaling factors for the same type of reactor in different countries varies slightly as can be noticed in Figure 14 b) and d) [52]

Solid waste can be contaminated with radionuclides directly or indirectly. Directly contaminated waste is constantly in contact with liquids and gases of the system (metal parts of equipment, pipes, liquids and air filter elements). Indirectly contaminated waste does not have permanent contact with liquids and gases, but is contaminated due to temporary (irregular) contact with directly contaminated waste or liquids/gases of the system (gloves, protective shields and other measures). Thus, this factor does not affect the scaling factors [52] [53].

7.4 Scaling Factor Determination methods

Scaling Factors can be calculated from direct measurements of radionuclide activity concentrations, through modelling codes or by a combination of both techniques as described in Figure 10. The role of sampling is different depending on the selected approach, i.e., it is used as an input for calculation or to validate the results of theoretical models.

For calculations based on experimental data there are two general methods: (1) arithmetical mean, which is highly influenced by wide range variations, thus, may provide skewed results, and (2) regression of logarithms, which can be fitted to linear relationships, (2a) the geometric or logarithmic mean, suitable when data ranges over several orders of magnitude, and (2b) non-linear relationships.

The final method can be a combination of (1) and (2), e.g., the geometric mean can be used as a first attempt, followed by a non-linear try if linear correlation cannot be verified.

Additionally, all these methods can include a weighting process with appropriate factors (based on variance estimation) to consider the dispersion of the data.

Also, when determining the NV, it should be noted that the contamination activity is dependent on the surface of the component and the neutron induced activity is dependent on the mass/volume of the component. Therefore, the determination of one nuclide vector for different components - even if they belong to one waste stream - may not be appropriate particularly if the characterization results of the specific waste stream will be used to make decisions about decontamination and clearance procedures. In this case, it may be useful to use two NVs and to calculate the activities for activation and contamination separately and then add them up for each component considering the component's surface mass. This also has advantages for sample evaluation and the subsequent representativeness of the samples.

7.5 Characterization of waste streams of MRW

Classification of the MRW streams with different radionuclide compositions generated in the decommissioning of NPP from the results of measurements, modelling and historical data should be done after quantification of the activities and radionuclide composition for every waste stream according to NV. At the first step, contaminated/activated and non-contaminated parts of constructions of the NPP should be determined using results of preliminary radiological survey and

characterization according to NV. At the second step, volumes of contaminated parts of the constructions should be determined using information of NPP documentation. At the third step, the masses of contaminated parts of the constructions should be determined using the volumes of the constructions and data on material type, using available NPP documentation and historical information or by expert decision. At the fourth step, mass flow from decommissioning of contaminated parts or waste packages in the storages should be segregated into waste streams of different radiological categories according to NV radionuclide composition and activity concentration (VLLW, LLW, ILW, etc.).

8 Sampling procedure

8.1 Estimation of sampling places

For determination of activated/contaminated metallic construction (as well as other materials) of NPP two methods are used:

- Gamma dose rate measurements in all the premises of NPP to determine the dose rate distribution on the metallic surfaces,
- Determination of specific activity of nuclides in samples from representative places.

Radiological survey is implemented during the several campaigns. Every campaign can include a few steps, if necessary. The first campaign provides preliminary data on gamma dose rate and nuclide composition variation in contaminated areas. It includes in-situ survey of easy-to-access areas as well as review of available historical knowledge. The second campaign is targeted to confirmation of preliminary results and total detailed characterization of all areas if preliminary results are confirmed. If the results are not confirmed during the second campaign, then additional campaigns should be organized. Therefore, the measurement of the gamma dose rate by portable hand-held measuring instruments at the points of selected grid can be a good indicator of the local contamination/activation level. In the increased gamma dose rate places the more detail investigation with fine grid around the area should be done. The final contamination/activation zone is determined by its radionuclide composition and the contamination adhesion (fixed or loose) by using portable gamma spectrometers or other nondestructive technique (see Chapter 9.2). α and β/γ surface contamination can also be present in the investigated zone, so measurement of the total α and β/γ surface contamination should be performed. The preliminary contamination zone can be divided into smaller final zones and laboratory samples should be taken. If the laboratory measurements of the samples from this zone and/or surface contamination type in some areas of this zone show that radionuclide composition in various parts of the preliminary zone are not the same, additional samples should be taken in order to have representative sample. For the determination of representative sampling places also reactor operation/historical data on radioactive waste management should be used. Historical data show that special attention during sampling should be paid. Different samples (smear and scratch samples) of all kinds of materials (from the surface) should be taken. The sampling could be carried out in several campaigns.

Laboratory measurements of both ETM and DTM nuclide specific activities in the samples from the selected zone are used for scaling factors determination. The adequacy of the measurement results for determining nuclide vectors should be also analyzed (more detail in subchapter 8.3). Depending on the result of this analysis as well as on the analysis of the results of gamma spectrometry, total α and β/γ surface contamination in-situ measurements, a decision on the necessity of additional samples as well as the specific activity of these samples needed to be collected during the additional sampling campaign could be done.

8.2 Sample preparation and registration

The determination of gamma, beta and alpha activity in a laboratory samples should include preliminary preparation of samples, radiochemical destruction of the samples for the measurement of beta and alpha activity, determination of the yield of radionuclides in radiochemical separation procedures, measurements of the sample activity by nuclear spectroscopy.

Every original sample should be weighted by precision balances, after that, the sample should be divided into specimens for analysis by different nuclear spectroscopy methods. Every specimen should be weighted. Detailed measurement technique and samples preparation for radiochemical destruction for specific radionuclide measurement is described in Chapter 8.1.

For determination of the gamma emitters (such as ^{60}Co , ^{59}Ni , ^{94}Nb , ^{137}Cs , ^{152}Eu , ^{154}Eu , ^{241}Am) the gamma spectrometry is used (see Chapter 9.2 for detail).

To reduce human error two persons should participate in the performance of every measurement. The standard deviation of the mean should be used as a statistical uncertainty of the measurement results, while the uncertainty from the calibration certificate should be applied to obtain the standard uncertainty of the measurement result which is calculated as a square root of the sum of all known uncertainties (standard uncertainty components).

The measurement description, location, time, used equipment, results and uncertainty as well as traceability should be depicted in signed measurement protocols.

8.3 Estimation of representativeness and number of samples

Since the method of SF is semi-empirical, its accuracy is also determined by the number of available data. A sufficient number of data is such that beyond that data statistics improve so little that it is not worth investing more to improve. This also can be expressed quantitatively. The correlation coefficient describes the degree of dependence between the two radionuclides under consideration. As the number of data changes, so does the correlation coefficient. Reliable confidence bands are also taken into account, e.g. 0.95. The required number of samples for required correlation coefficients is determined as follows:

Table 10: Dependence of the correlation coefficient on the number of samples

Correlation coefficient	0,6	0,7	0,8	0,9	0,95
Number of samples	40	35	30	25	20

The criterion presented in Table 10 is applicable for the determination of scaling factors according to equation (2) when there are sufficient number of samples. This method of calculating the geometric mean is widely used.

$$SF = \sqrt[n]{y_1 / x_1 \times y_2 / x_2 \times \dots \times y_n / x_n} \quad (2)$$

However, if number of samples is insufficient, scaling factors can also be estimated. However, in this case, if the database would be additionally supplemented with new data, larger fluctuations of the scaling factors are possible. Therefore, it is necessary to calculate the upper limit of the scaling factor for the established confidence limit. When the number of data is less than that specified in the Table, but correlation between DMT and EMT radionuclides is established and its distribution is lognormal, equation (3) is used to calculate the scaling factors:

$$SF_{CL} = SF + t_{0,95} \frac{s}{\sqrt{n-1}} \quad (3)$$

where: x – key nuclide specific activity, y – DTM nuclide specific activity, s – sample variation, n – number of samples, $t_{0,95}$ –significance level 5 %.

For determination of nuclide vectors, it is necessary to collect samples from a wider range of activities, thus the parameters of the statistical analysis are determined more reliably and the measurement uncertainties of individual points have less influence. The collected samples will undergo explicit measurement procedure using available nuclear spectrometry methods, where detection limits are considered. Therefore, it is not appropriate to select very low activity samples,

as the low activity nuclides may not be detected due to limited sensitivity of the spectrometric instruments and the limited time available for each measurement. It is also inappropriate to select highly active samples in order not to create preconditions for contamination of measuring devices and to limit the radiation exposure of laboratory staff.

The objective of the sampling process is to obtain representative and accurate values of key and DTM nuclides activity concentrations to characterize the potential waste streams by the Scaling Factor Method. In this context, “sample” means collecting portions of the potential waste stream and obtaining the activity concentrations in the laboratory.

“Error” is defined as the difference between the actual activity value (which remains unknown) and the laboratory value, in terms of statistical expectancy - E:

$$E[A_V - L_V]^2 = (A_V - E[L_V])^2 + E[L_V - E[L_V]]^2 = (A_V - \mu_{L_V})^2 + S_{L_V}^2 \quad (4)$$

where: A_V – Actual activity value, L_V – Lab activity value, μ_{L_V} – expected value of Lab activity, $S_{L_V}^2$ – Variance of Lab value.

The first part of the right hand is the trueness (“bias/skewness”), a term related to systematic error, which gives an idea of the amount of information contained in the sample:

$$Skewness^2 = (A_V - \mu_{L_V})^2 \equiv Sk^2, \quad (5)$$

and the second one is the uncertainty, related to precision (low uncertainty means high precision) and reproducibility, which reflects random fluctuations introduced by the measurement process:

$$Uncertainty^2 = S_{L_V}^2 \equiv U^2 \quad (6)$$

Therefore, analysis of Representativeness/Accuracy, R, not only means analysis of uncertainties, but trueness should be also considered in this process:

$$R^2 = Sk^2 + U^2 \quad (7)$$

Thus, sampling should be carried out with the aim of eliminating bias and reducing the variance.

Assuming that all the information required to apply the Scaling Factor method is contained in the results of the lab analysis, the mean of the lab value tends toward the actual value, showing null skewness/high trueness. Therefore, in these cases uncertainty would be the only term to account for representativeness and trueness should be ensured by following appropriate procedures.

A good way of improving trueness is by mean of sampling correctness:

- Any part of the waste stream (batch/lot) has the same probability of being selected for sampling regardless its location.
- All samples are taken in the same way.

Assuming sampling correctness is ensured, the sampling error may be defined as the variance of the following non dimensional quantity [55]:

$$Sampling\ Error = Variance \left[\frac{A_V - L_V}{A_V} \right] \quad (8)$$

All operational solid radioactive waste, generated at the NPP, should be collected and sorted according to the type and class and should be loaded into the containers. Results of the gamma spectrometric measurements of waste from selected containers are performed regularly from different waste collection points before downloading it into the storage section.

In general, it could happen, that completeness and reliability of available historical data on waste are low. Because of incompleteness and insufficient reliability of the historical NPP spectrometric data it cannot be used directly for the evaluation of the MRW nuclide vector scaling factors. In such a case, well planned sampling and measurements are needed to be done. All available historical data serves as initial information for development of the optimal sampling program and the primary radioactive waste nuclide composition analysis.

The optimal number of samples (i. e. minimal sufficient number) to be taken from each section for the forthcoming evaluation of the nuclide vector scaling factors can be determined by using modelling of dispersion of radionuclide specific activity ratios. The number of samples for the evaluation of the average scaling factor with desired reliability depends on the scattering of the activity ratios from sample to sample.

During the operation life of the facility, the more common waste are ion exchange resins, evaporator concentrates and sludges. Heterogeneous waste, compactible and non-compactible, are produced to a lesser extent. On the other hand, during decommissioning stage, the more frequent waste type is heterogeneous waste (metals, debris, tools, components, rubble, etc.).

When carried out appropriately, sampling of liquids provides good accuracy due to the homogeneity of the waste, whereas sampling of heterogeneous waste requires additional sampling optimization strategies with the aim of saving costs while improving confidence.

8.4 A basic sampling optimization approach

A basic sampling optimization approach should take the following items into consideration:

- Classification of samples by both their origin and the simplest or easiest lab analyses, such as dose rate, total alpha or total beta.
- For each sampling area, generation of composite samples covering the previously defined classification (of dose rate, total beta, total alfa, in-situ gamma spectrometry), ensuring that they are homogeneous in relation to the measured parameter.
- Radiochemical analysis of the minimum number of composite samples needed to cover the identified range of values, such that their activity concentrations are large enough to above the minimum detectable activity (MDA).
- Storage of samples to support future optimisation analyses and a suitable quality assurance programme.

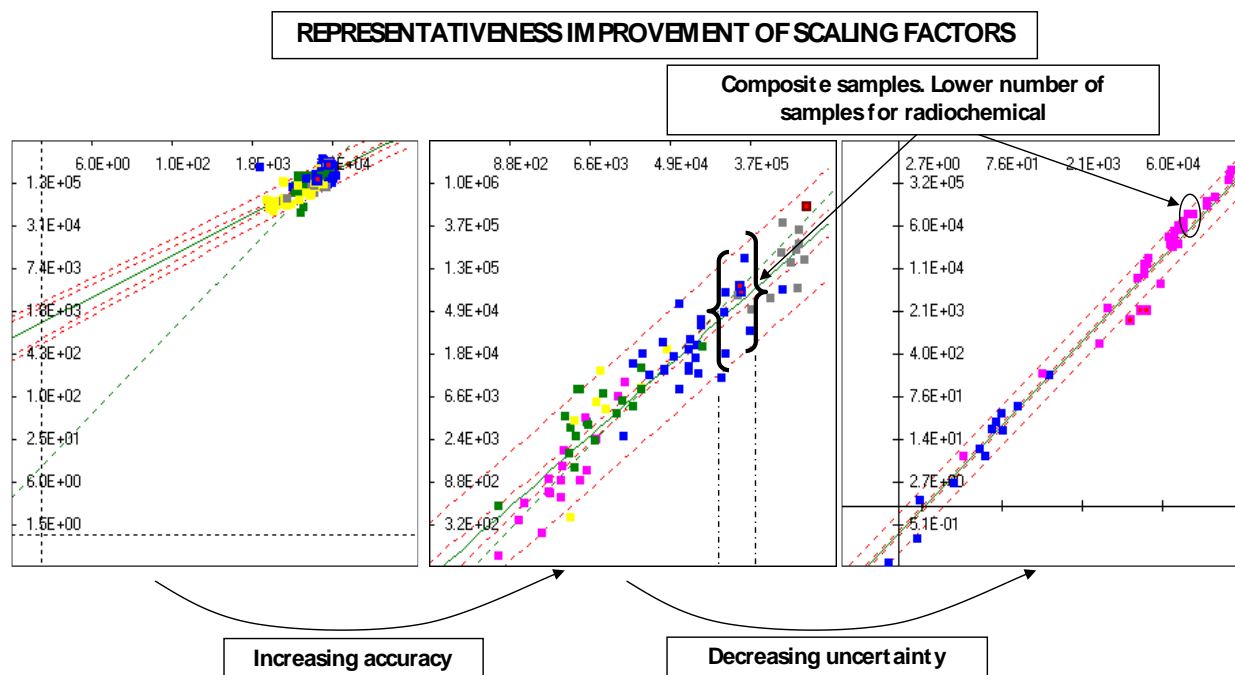


Figure 15 SF Accuracy & Uncertainty improvement by the manner of sampling

The common approaches to achieve accuracy in sampling are illustrated in Figure 15. They are:

- **Composite sampling:** Each sample should be a combination of several samples taken randomly from different areas or parts of the waste, ensuring that the values of their key parameter (dose rate, total beta, total alpha, key nuclide activity concentration) are similar. That is to say, once the whole data range of the sampling parameter is considered, samples with similar parameter values can be combined, creating on composite sample for each class of the data range.
- **Accumulated sampling:** In many cases it is not possible to collect enough samples in the same campaign. In this case, samples will be taken periodically over future campaigns and data produced over time to obtain a representative SF. These samples should cover the widest possible range of activities, like the actual range.

9 Measurement techniques for MRW characterization

The most accurate way to determine scaling factors is to measure radionuclide concentrations directly. However, it is not very easy to determine the concentrations of all radionuclides important for safety in this way, because the specific activities of some long-lived radionuclides are not high enough, or because of the presence of other radionuclides, the continuum is high under the peaks of radionuclides of interest and therefore, the sensitivity is lower, or there are no medium or high-energy gamma lines in their decay schemes or their fraction yield is low, and also due to the absorption of ionizing radiation in waste and packaging materials. Such difficult-to-measure radionuclides include most transuranic radionuclides, which are alpha emitters, fission products, such as ^{90}Sr , ^{99}Tc , which emit exclusively beta particles, activation products, such as ^{59}Ni , ^{63}Ni , ^{55}Fe , which also undergo radioactive decay by beta radiation. To determine the specific activity of difficult-to-measure radionuclides, complex methods of measurement and sample preparation are applied. Most often for scaling factors determination in MRW, after chemical treatment, e.g. separation of the radionuclide to be determined, α -, β - and γ -spectrometric methods are used by performing a series of measurements. To ensure reliable results, the measured specific activity values should fall within the reliable range of values and satisfy the Grubbs test. The upper and lower limits of the scaling factors are obtained from the confidence interval of the correlation function as it was already discussed in chapter 8).

Often mass spectrometry methods are needed to determine all isotopes of radionuclide (Pu, I). In addition to non-destructive gamma spectrometry, other non-destructive techniques are used, such

as: gamma camera, muon spectrometry, neutron interrogation, x-ray etc. depending on the possibilities and the specific task. For determination of some radionuclide's activities, both destructive and non-destructive analysis methods can be used. Non-destructive analysis methods are usually applied when the radiation intensity of the investigated material is sufficient for precise analysis, e.g. spent nuclear fuel claddings [56]. Passive methods are based on direct applications of X-ray, gamma, neutron spectrometry. In addition to passive methods, active methods are used, when the material is activated by an intense pulse of neutrons or gammas and the radiation of newly formed materials is observed [57]. Destructive analysis methods are usually important in low radionuclide concentrations cases. They are based on the preparation of a special sample for analysis, often separating the analyzed chemical element from a mixture of other elements and preparing samples of specific geometry and chemical properties. These methods are characterized by a low detection limit and selectivity. In subchapters 9.1 and 9.2 the destructive and non-destructive techniques will be discussed in more detail.

9.1 Destructive techniques

The analysis of α - and β - emitters requires complex methods. Radiochemical separation methods are used to prepare the samples for determination of the specific activities. After the radiochemical preparation [58] [59] [60], the samples are measured by α -, β - spectrometry, mass spectrometers or beta counters as appropriate.

α - spectrometric method is used mostly for the relative and absolute concentrations of uranium and transuranium determination in MRW (see Table 9 for detail) in specially prepared samples. Transuranium radiochemistry technologies, used for sample preparation, separating the chemical element of interest from a mixture of other elements, are reviewed in [61]. The ideal α - source should be a monolayer film, but in reality, this is not the case, and the scattering of α - particles determines the resolution. The samples are dissolved in a liquid and deposited by electrolysis, vacuum sublimation, direct evaporation, microdeposition, and other methods [62]. The most popular methods are electrolysis and microdeposition. The latter method is faster. The simplest way to prepare a sample for α - spectrometry is electrolysis [58] [62]. Preparation of samples on nuclear membranes is presented in [63]. This method is technologically simpler and cheaper, but less homogeneous thin layers are obtained, compared to electrolysis methods - the recorded shape of the α -line in the spectrum is asymmetrical, the resolution is worse, but it is successfully used to study active samples. Due to the low cost, simple operation and high efficiency, semiconductor α -spectrometers, whose resolution can reach 11-15 keV, are the most widely used [64]. The main drawback of α -spectrometry is that the classic semiconductor α -spectrometer cannot distinguish between ^{239}Pu and ^{240}Pu radiation, as well as ^{238}Pu and ^{241}Am radiation. However, methods have been proposed to address this limitation. The work of [65] shows that by using reference sources of ^{239}Pu and ^{240}Pu , correlations can be obtained between the number of pulses in specific spectral intervals and the activity ratios of ^{240}Pu and ^{239}Pu , by which this ratio can be determined in samples. By applying mathematical deconvolution, it is possible to distinguish the Cm isotopes [66]. ^{238}Pu and ^{241}Am radiation can be separated using a liquid scintillator spectrometer and a special scintillator cocktail [67] [68]. The spectral peaks are separated due to the different chemical properties of plutonium and americium. The scintillation spectrometer can also determine the ratio of α - and β - activities [69] or estimate the amount of ^{241}Pu [70]. ^{241}Pu content can be estimated also by using α - spectrometric method as in [70].

Mass spectrometry (MS) is an expensive, destructive, but accurate and sensitive method of isotopic analysis. The results of the analysis do not depend on the radiation properties of the material [71] [72] [73]. The samples for analysis are decomposed and specially prepared. The advantage of the mass spectrometry method for plutonium isotopic analysis over alpha spectrometry is the short analysis time (up to 2 h for ICP-MS, whereas for alpha spectrometry, it takes ten times longer) and the possibility to identify all isotopes. However, the mass spectrometric methods are not absolute – resolution and the detection level is limited by the background and the possible interferences with other ions or their complexes [74]. It should also be noted that these methods do not distinguish between isobars. For ^{238}Pu analysis, this is important because the sample always contains natural

impurities of ^{238}U , and ^{241}Am is always detected together with ^{241}Pu . ^{238}U not only interferes with ^{238}Pu , but also creates a background for neighboring isotopes, especially for the analysis of ^{239}Pu ($^{238}\text{U}^1\text{H}^+$ ion is formed) [74]. MS is divided into several types according to the ionization process of the investigated element. ICP-MS (inductively coupled plasma) [71] [74], LA-MS (laser ablation) [75], SS-MS (spark source), TIMS (thermal ionization) [76] [77], AMS (accelerator) [78], RIMS (resonance ionization) [79] are used for actinoid analysis. The relative error in determination of isotopic ratios by TIMS spectrometer can reach 0.002 %, usually 0.1-0.5 % [80] [81]. The analysis takes a long time - up to several hours, and the detection limit is much lower than the gamma spectrometric methods [76]. ICP-MS and related methods are more widely used [72] [71] [82], they are faster, but the detection limit is worse, they are sensitive to various isobars. RIMS [79], AMS [78] methods are more selective in terms of isobars.

In **β spectrometry** the scintillation detectors are very widely used - they are almost the only high-resolution devices that can be used to observe the energy distribution of β particles. The sensitive material of the spectrometer can be inorganic (NaI(Tl), CsI(Tl)) crystal, organic materials (anthracene, stilbene). Liquid scintillator β spectrometers are often used for low activity concentration samples including environmental radioactive samples. A β particle entering the scintillating material create photons registered by several photodetectors (pulses of background radiation are recorded by only one detector - and thus eliminated). Such measurements require special sample preparation. At the same time, the registered α particles have higher energies and do not interfere with the separated β particle energies (for example, ^3H $E_{\text{max}} - 18$ keV, α particle in the order of MeV). As concerning the sample preparation, for example, for ^{14}C determination two methods may be used: sample decomposition by using a mixture of inorganic acids to obtain a clear solution and later measurement by using LSC technique [83], or - combustion in pure oxygen at high temperature (600-1000°C) to convert carbon to gaseous form. CO is converted to CO_2 which is collected by trapping in an alkali solution before measurement of ^{14}C activity by Liquid Scintillation Counting (LSC) or gas activity detectors.

In order to maintain the homogeneity of the sample and reduce labor costs, radiochemical procedures can be used to determine multiple radionuclides from a single sample [84] [85] [83]. Usually combination of different measurement techniques both non-destructive and destructive are used to determine specific activities of radionuclides from the same sample.

9.1.1 Samples preparation

For the characterisation of MRW (e.g., stainless steel, carbon steel and various alloys) by means of destructive assays it is necessary to distinguish between radionuclides on surface (contamination) and those that have resulted from activation within the mass of the metal. For different cases, the sample treatment will also be different. Surface decontamination involves removing a layer of a few millimetres from the surface of the sample, where most of activity is concentrated. This technique is used when the contamination is on one side of the metallic sample, indicated during the sampling phase. Complete dissolution of the sample can be performed in case of volume activation of the entire sample.

An important step that should be performed before and after sample destruction is a gamma spectrometry measurement. This gamma measurement allows obtaining:

1. the gamma radionuclides (and prediction of associated beta and alpha radionuclides which can be expected).
2. the sample activity due to contamination, comparing the total sample activity before and after surface layer removal.
3. a validation of non-destructive gamma spectrometry measurement made (if any), thanks to the results ensured by destructive analysis technique later on.

Once a solution of sample is obtained and measured by gamma spectrometry, part of it is used for the determination of the radionuclides of interest. The radionuclides to be investigated for waste classification are linked to the history of the waste and thus to the processes it has undergone and which generated it. In the next paragraph details will be provided for the determination of several different radionuclides. All aspects concerning radiochemical determinations were developed based on procedures described in references [86] [87] and [88].

9.1.2 Determination of ^{55}Fe and $^{59+63}\text{Ni}$

Among the characteristic radionuclides due to activation processes in metals, ^{55}Fe and ^{63}Ni are certainly ones to be considered and are important for correlation with ^{60}Co for the purpose of calculating SF. The process of separating iron and nickel is divided into two phases: an element extraction phase using columns filled with selective resins, and a purification phase that is carried out by precipitation and subsequent filtration (in the case of nickel) or centrifugation (in the case of iron).

The yield of the extraction process is obtained through an initial analysis of the amount of stable iron or nickel (known titre carrier) in the solution prior to extraction, which is compared with the final analysis of iron or nickel downstream of extraction and purification.

The column used for the extraction is filled with an ion-exchange resin and conditioned in a hydrochloric environment that is selective for iron while allowing the elution of nickel. Then, by changing the environment from hydrochloric to nitric 8M, the iron retained by the resin is eluted. After creating a basic environment, a complexing agent is added to the beaker containing nickel allowing the formation of nickel flakes. After centrifugation, or filtration, the precipitate is purified by repeated attacks of nitric acid and hydrogen peroxide. Once purified, the sample is ready for liquid scintillation measurement.

Instead, ammonia 30% is added to the beaker containing Fe until the pH is basic. This causes the formation of red flakes of iron hydroxide. After centrifugation, the precipitate is purified by repeated attacks of nitric acid and hydrogen peroxide. Once purified, the sample is ready for liquid scintillation measurement.

The extracted solutions of ^{55}Fe and $^{59+63}\text{Ni}$ to be subjected to liquid scintillation are diluted in the scintillating cocktail, in vials for scintillation, and through electronic transition mechanisms an energy transfer of the ionising particle takes place, resulting in the emission of light in the form of photons that are collected by photomultiplier tubes (PMT).

The range of the emission energy of ^{55}Fe is from 0 to 6 keV, unless there is a shift to lower values due to chemical 'quenching'. The range of the emission energy of $^{59+63}\text{Ni}$ is from 0 to 66 keV, unless there is a shift to lower values due to chemical 'quenching'.

Quenching is the reduction of the number of photons reaching the photocathode thus decreasing the counting efficiency, proven by characteristics of the sample and the scintillation cocktail.

The activity of the unknown samples is subtracted by the activity of the 'blank', i.e. a solution prepared and counted under the same conditions as the sample but which does not contain the radionuclide. Before the process of analysis, the instrument is calibrated by counting a set of "quenched" standards plotted with certified sources of ^{55}Fe and $^{59+63}\text{Ni}$ having known activity in order to generate the efficiency curves necessary to correct the measurement result of the samples under analysis.

9.1.3 Determination of ^{90}Sr and ^{90}Y

Radionuclides list originating from fission processes, which can be correlated with Cs, certainly includes ^{90}Sr and ^{90}Y .

The method described below is based on the measurement of yttrium by the Cherenkov effect after more than 21 days passed since the contamination process took place: after such time, in fact, these two radionuclides, strontium and yttrium, are in secular equilibrium, so it is possible to measure ^{90}Y and then trace the activity of strontium.

For the extraction of strontium and yttrium, an aliquot of the source solution is taken to which the yttrium carrier is added.

The yield of the extraction process is obtained by an initial analysis of the amount of stable yttrium in the solution before extraction, which is compared with the final analysis of yttrium after extraction and purification.

In order to remove Fe and other interferents in solution, the sample undergoes a series of purifications through the addition of oxalic acid and ammonia. In this way, at a pH between 3 and 5, yttrium precipitates as oxalate while Fe and other interferents remain in solution. To assist the precipitation of oxalates, about 1 ml of calcium carrier is added.

The precipitate containing yttrium oxalate is subjected to repeated attacks, at elevated temperatures, of concentrated nitric acid and hydrogen peroxide until the oxalates are completely eliminated. Excess ammonia 30% is added to the yttrium-containing solution until a totally basic environment is obtained that will allow the precipitation of yttrium in the form of hydroxide. After centrifugation and

discarding the supernatant, the precipitate containing yttrium hydroxide is dissolved in 6M nitric acid and is ready for liquid scintillation measurement.

The activity concentration of ^{90}Sr (parent) is determined by measuring the activity of ^{90}Y (daughter) cause the two radionuclides are in secular equilibrium. The activity of ^{90}Y is determined by the Cherenkov effect, by measuring the solution extracted in the vial in the absence of scintillating liquid, using the production of light emitted by a charged particle with a velocity greater than that of light in the medium. The region of the spectrum in which the signal falls ranges from 0 to 25 keV.

The activity of the unknown sample is subtracted by the activity of the 'blank', i.e. a solution prepared and counted under the same conditions as the sample but which does not contain the radionuclide. Upstream of the analysis process, the instrument is calibrated by counting a standard traced with a $^{90}\text{Sr}/^{90}\text{Y}$ solution having known activity in order to extrapolate an efficiency value necessary to correct the measurement result of the sample under analysis.

9.1.4 Determination of ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu

Among the radionuclides of radiological importance for the characterisation of radioactive waste, the determination of plutonium isotopes is of great interest.

For plutonium extraction, an aliquot is taken from the solution to which ^{242}Pu tracer is added for recovery yield evaluation. The use of ^{242}Pu as a yield standard is given by the unlikelihood of finding it within the sample to be measured [87]

In order to purify the sample from interferent elements, a series of washes with oxalic acid and ammonia are performed. In this way, at a pH between 3 and 4, the plutonium precipitates as oxalate, leaving the interferents in solution. To assist the precipitation of oxalates, about 1 ml of calcium carrier is added. The precipitate containing plutonium is subjected to repeated attacks of concentrated nitric acid and hydrogen peroxide at high temperatures until the oxalates are completely eliminated. Once all oxalates have been eliminated, the sample is taken to 8M nitric acid and sodium nitrite is added to reduce the oxidative state of the plutonium, which will allow it to be selectively extracted during the chromatographic separation process [89]. Once the extract containing plutonium has been brought into a weakly nitric environment, the sample is taken for liquid scintillation determination of ^{241}Pu [90]. The remaining sample is ready to be electrodeposited and on a steel plate and measured by alpha spectrometry (see section 9.1.6) [91].

The sample to be measured, following the radiochemical extraction of the plutonium, is diluted in the scintillation cocktail, inside a light-transparent vessel (vial), and through electronic transition mechanisms an energy transfer of the ionising particle takes place, resulting in the emission of light in the form of photons, which are collected by PMT.

The range of the emission energy of ^{241}Pu is from 0 to 20.8 keV, unless a shift to lower values occurs due to chemical 'quenching'.

The activity of the unknown sample is subtracted by the activity of the 'blank', i.e. a solution prepared and counted under the same conditions as the sample but which does not contain the radionuclide. Upstream of the analysis process, the instrument is calibrated by counting a set of 'quenched' standards traced with a certified source of ^{241}Pu having known activity in order to generate an efficiency curve necessary to correct the result of the sample measurement under analysis.

9.1.5 Determination of ^{241}Am , ^{242}Cm and ^{244}Cm

In addition to Plutonium isotopes, among the alpha emitters of interest for waste characterisation and to be used for correlation is certainly the determination of americium and curium isotopes.

For the extraction of the americium and curium isotopes, an aliquot is taken from the stock solution to which the ^{243}Am tracer is added for evaluation of the recovery yield. In order to purify the sample of interferents, a series of washes with oxalic acid and ammonia are performed. In this way, at a pH between 3 and 5, the americium precipitates as oxalate, leaving the interferents in solution. To assist the precipitation of oxalates, about 1 ml of calcium carrier is added. The precipitate containing americium is subjected to repeated attacks of concentrated nitric acid and hydrogen peroxide at high temperatures until the oxalates are completely eliminated. Once all the oxalates have been eliminated, iron carrier is added to the sample, which will act as an adjuvant for the precipitation of the alpha hydroxides by the addition of 30% ammonia. The precipitate is brought into a hydrochloric environment and passed through a chromatographic column that will elute americium and curium while retaining the other alpha emitters. Once the extract containing americium has been brought

into a weakly nitric environment, the sample is ready to be electrodeposited and subsequent measurement.

9.1.6 Measurement alpha radionuclides

The sample prepared by electrodeposition is counted by alpha spectrometry using the detection of alpha particles by a silicon semiconductor detector, the active volume of which consists of the emptying region of a p-n junction, polarised inversely. The instrument returns a spectrum of counts as a function of the energy of the alpha particle, and thus allows unambiguous qualitative and quantitative determination of the radionuclide [91]

The alpha spectrometer is calibrated in energy and efficiency using a source with known activity of ^{241}Am , ^{239}Pu and ^{244}Cm . These 3 radionuclides are used because their emission energies are distinct. The typical emission energies for alpha spectrometry of ^{241}Am (5.486 MeV) overlap with those of ^{238}Pu (5.499 MeV), and it was therefore necessary to carry out a separate radiochemical extraction of the two elements to be analysed upstream.

^{242}Cm and ^{244}Cm can be directly measured by alpha spectrometry thanks to their well-distinguished emission energies (6.069 MeV and 6.112 MeV for ^{242}Cm and 5.762 MeV and 5.804 MeV for ^{244}Cm) and to the fact that only ^{241}Am is present as interferent but has a very different energy (5.486 MeV). As for ^{239}Pu and ^{240}Pu , due to the similar emission energy (≈ 5.15 MeV), it was not possible to separate the contribution of one from the other by alpha spectrometry, so ^{239}Pu was measured by ICP-MS to determine the activity of the individual radionuclide (see section 9.1.7).

9.1.7 ICP-MS

Mass spectrometry is an analytical technique based on the ionization of a molecule and its subsequent fragmentation into ions of different mass-to-charge (M/Z) ratios. It is therefore a destructive method of analysis [92].

Samples placed for analysis are ionized, selectively, inside the instrument using a plasma flashlight. Then, under high vacuum conditions, the ions are separated, via a quadrupole, by the creation of magnetic fields. Ions interested in analysis, based on their mass-to-charge ratio, pass through the quadrupole and reach a counter that produces a signal proportional to their concentration.

Mass spectrometry analysis thus consists of the ionization of molecules and the separation of the different ions produced and, consequently, the transformation of a signal related to the concentration of these ions. Thus, in this way, it is possible to perform an isotopic analysis of the elements present in the sample reaching concentrations in the order of ppb (parts per billion).

The solution containing $^{239}\text{Pu}+^{240}\text{Pu}$ is therefore analysed in ICP-MS, their isotope ratio determined. This value is then applied to the total activity value, determined in alpha spectrometry, thus obtaining the activity of the individual isotopes.

9.2 Non-destructive techniques

Non-destructive techniques used for radiological characterization rely on the detection of radiations emitted by the radioactive sources present in radioactive waste, with gamma-rays and neutrons being the main types of radiation detected. Eventually, they can be complemented by other non-destructive evaluations, such as the x-ray radiography or neutron radiography to evaluate other characteristics of the waste.

The general aspects presented hereafter can find comprehensive references in [56] (including its addendum published in 2007) and [61], and in all the references listed in these documents. Specific references will be added where relevant.

9.2.1 Gamma spectrometry

The gamma-spectrometry technique consists in the detection of photons emitted in spontaneous nuclear decays, like alpha and beta-decays. Such gamma rays are emitted with a rate and energy spectrum that is unique for each radionuclide. This uniqueness provides the basis for most gamma-ray assay techniques: by counting the number of photons emitted with a specific energy, it is possible to determine the number of nuclei that emit that characteristic radiation, thus the activity of the radioactive element.

Nuclear decays are often accompanied also by x-rays, which are photons emitted during rearrangement of the atomic electron structure rather than changes in nuclear structure. The x-rays energies are unique to each element but the same for different isotopes of one element. Although x-ray spectrometry is a measurement technique used in nuclear field, it has a niche application field (and far smaller than gamma spectrometry) and it is usually limited to laboratory analysis (due to the low energy of x-rays). Therefore, only the gamma spectrometry is discussed hereafter.

Detecting a photon in gamma spectrometry implies not only the generation of a signal to be associated with it, but also these signals has some property proportional to the photon energy (which is often completely released in the detector used). A modern gamma spectrometry detector uses electrical signals whose amplitude is proportional to the energy released by the photons; a voltage difference is applied to generate the electric field in the detector active volume which then permits the collection of the generated charges (i.e., the generation of a current).

State-of-the-art gamma spectrometers used semiconductor materials (mainly High Purity Germanium (HPGe)) as active volume, which ensure the optimum energy resolution (i.e., the width (FWHM) of a peak in the spectrum corresponding to a monoenergetic gamma ray emission) keeping the detection efficiency (i.e., the number of photons reaching the detector which are completely absorbed and contributes to a full-energy peak) adequate. However, their cost and operational limits (in particular, the need to be cooled down to $-176\text{ }^{\circ}\text{C}$ to reduce the electronic noise) leave room to the development and application of other kind of detectors, like the low-cost high-efficiency NaI(Tl) scintillators (which have a poor-energy resolution) or solid-state CdZnTe (CZT) (cost comparable to HPGe, efficiency and energy resolutions slightly worse than HPGe, but they don't need cooling and can be used also for other measurements scopes – like gamma imaging). Figure 16 reports some representative spectra for comparing energy resolution of these crystals.

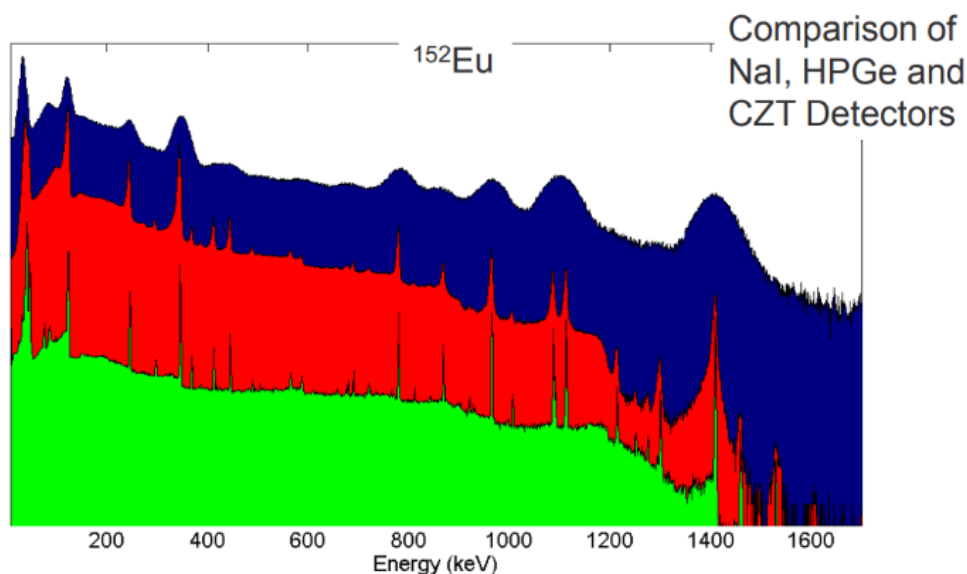


Figure 16 – Energy spectrum of a ^{152}Eu source acquired with different detectors: NaI (in blue), CZT (in red) and HPGe (in green). Credit to Prof. Zhong He.

Once a photon arrives at the detector active volume (i.e., the volume to which the voltage difference is applied), it can undergo three different processes depending upon its energy and the detector's material:

- Low-energy photons are directly absorbed via photoelectric effect: the gamma ray interact with a bound atomic electron losing all its energy (and then ceases to exist), used to overcome the electron binding energy and then transferred to the freed electron as kinetic energy (the momentum balance associated to the atom requires a neglectable amount of energy). The electron is stopped quickly in the active volume generating a small output pulse proportional to the energy of the original photon. Being the photon energy completely absorbed, the pulse generates a peak in the gamma spectrum which has the same energy of the original photon: this peak is called photoelectric peak or full-energy peak.

For mid-energy photons, conservation of energy and momentum allows only a partial energy transfer from the photon to the electron when the electron is not tightly bound enough for the atom to absorb the recoil energy. This process is called Compton scattering. In a Compton scattering, the photon continues its path in the material, undergoing to further Compton scattering or to photoelectric absorption, while the freed electron travels with a kinetic energy equal to the difference between the energy lost by the gamma ray and the electron binding energy (which is usually neglectable compared to the energy lost by the photon). The directions of the photon and electron depends on the amount of energy transferred, which can vary within a range of values. The electron is quickly absorbed in the detector active volume, similar to the photoelectric effect, and the resulting pulse generates a continuum gamma spectrum (because its kinetic energy can assume a range of values and not just one or more discrete values). However, if the scattered photon interacts in the active volume in such a way that is completely absorbed, the overall energy released in the crystal (and thus the generated pulse) is equal to the energy of the incoming photons. This case produces the full-energy peak in the gamma spectrum. In cases, where the gamma ray releases only a portion of its energy in the active volume, the corresponding event generates a pulse, which contributes to the continuum of the spectrum produced by the electron absorption: This portion of the spectrum is generally referred to as the Compton

- A gamma ray with an energy higher than 1.022 MeV can interact with the strong electromagnetic field in the vicinity of a nucleus to create an electron-positron pair (and then disappear) when it is under the influence of the strong electromagnetic field in the vicinity of a nucleus. This process has a threshold energy equal to 1.022 MeV because that it is the minimum energy required to create the electron-positron pair (each with only their rest mass and no kinetic energy). If the gamma ray energy exceeds the threshold, the excess energy is shared between the electron and positron as kinetic energy. The electron is quickly absorbed in the detector active volume. After losing its kinetic energy, the positron annihilates with an electron, releasing two gamma rays with energy of 0.511 MeV. These photons can undergo further interactions discussed previously, or may eventually escape from the active volume. If both annihilation gamma rays are absorbed in the detector, the interaction contributes to the full-energy peak in the spectrum; if one of them escapes from the crystal, the interaction contributes to what is called single-escape peak at 0.511 MeV below the full-energy peak; if both annihilation photons escape, the interaction contributes to the so-called double-escape peak located at 1.022 MeV below the full energy peak. The relative heights of the three peaks depend on the energy of the incident photons and the size of the detector.

All the pulses generated by the gamma rays reaching the detector undergoes to the amplification process and are then digitalized and 'sorted' by dedicated electronics, called Multi Channel Analyser (MCA): for each pulse a count is added to the corresponding channel, which is matched with the correspondent energy. The correspondence between the pulse amplitude and the energy (which is linear for HPGe detectors) depends upon the amplification chain set and thus has to be established experimentally by means of known emissions.

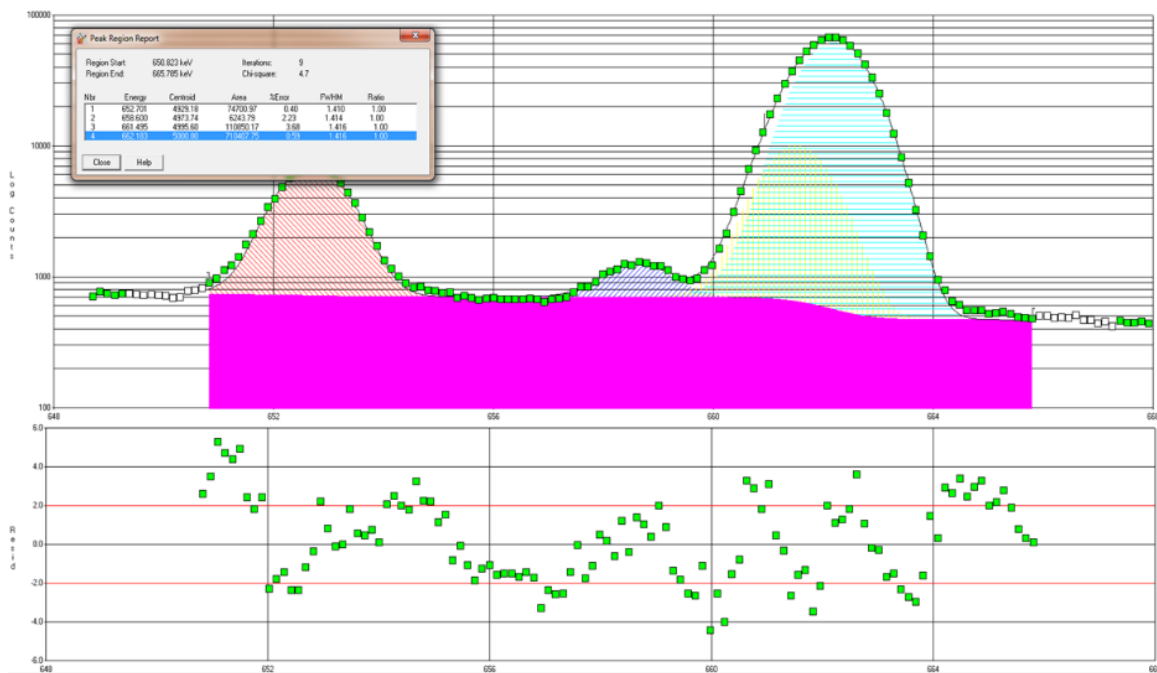


Figure 17 – Example of peaks identification and area calculation in a complex region including 4 emissions (distinguishable by the different colour). The residual distribution (the plot at the bottom) laying in the 2-sigma region (red lines) testify the goodness of the fit.

Once a spectrum is collected, several standard algorithms are available to identify the peaks present and calculate their net area as presented in Figure 17 (i.e, the area corresponding to the peak after removing the contribution from the continuum) [93] [94].

A peak is identified with a net area (S), matching with a radionuclide among those expected in the measurement and for which branching ratio Y is known from literature and thus the activity of the radionuclide is calculated as:

$$Act [Bq] = \frac{S}{T [s] \cdot Y \cdot \epsilon} \tag{9}$$

where: T is the measurement live time; ε is the measurement efficiency and Y is the branching ration; S is net area of peak.

Depending upon the situations, specific corrections must be introduced to account for phenomena like the decay during the measurement, the cascade effect, the events pile up, the dead time.

The measurement efficiency ε is the most critical parameter for modern gamma spectrometry characterization systems. It represents the probability that a gamma ray emitted by the item assayed reaches the detector and contributes to the full-energy peak. As such, the efficiency is depending on many parameters:

- Detector characteristics, which are typically known by the user thanks to the data provided by the manufacturer or through direct experimental campaign.
- Measurement setup, namely collimation and relative position of the detector with respect to the item to assay, which are known by the user.
- The physical and radiological characteristics of the item to assay, i.e., the distribution of material in the item and the distribution of radioactivity in the material. Those distributions are often unknown by the user, who can ‘guess’ them, evaluate them by means of other techniques (x-ray imaging, gamma-ray imaging, tomography, etc...) or use some qualitative descriptions inevitably introducing some bias in the results.

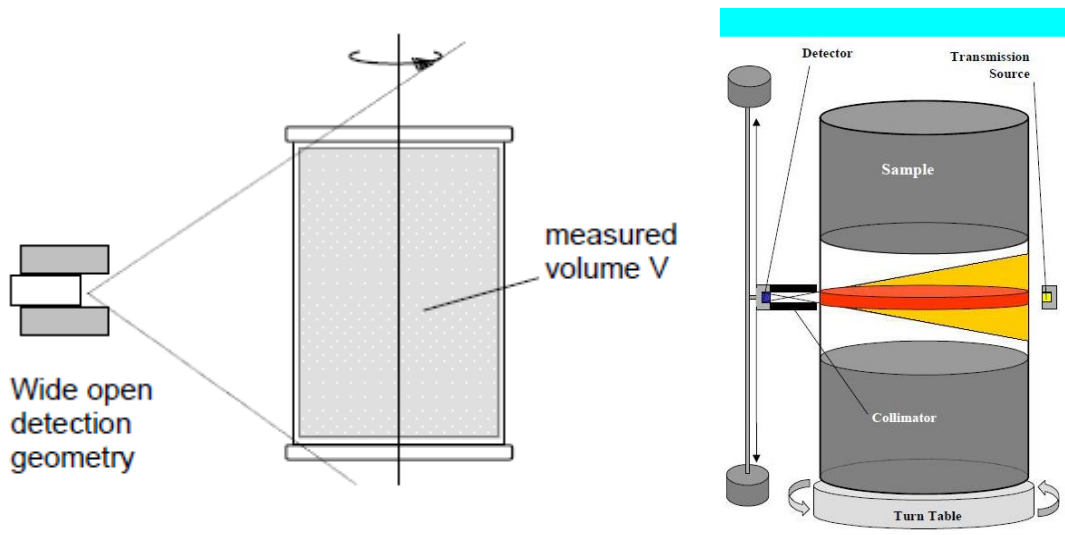


Figure 18 – Example of non-destructive assay (NDA) gamma spectrometry setup: on the left the open-geometry, on the right the SGS (similar for TGS).

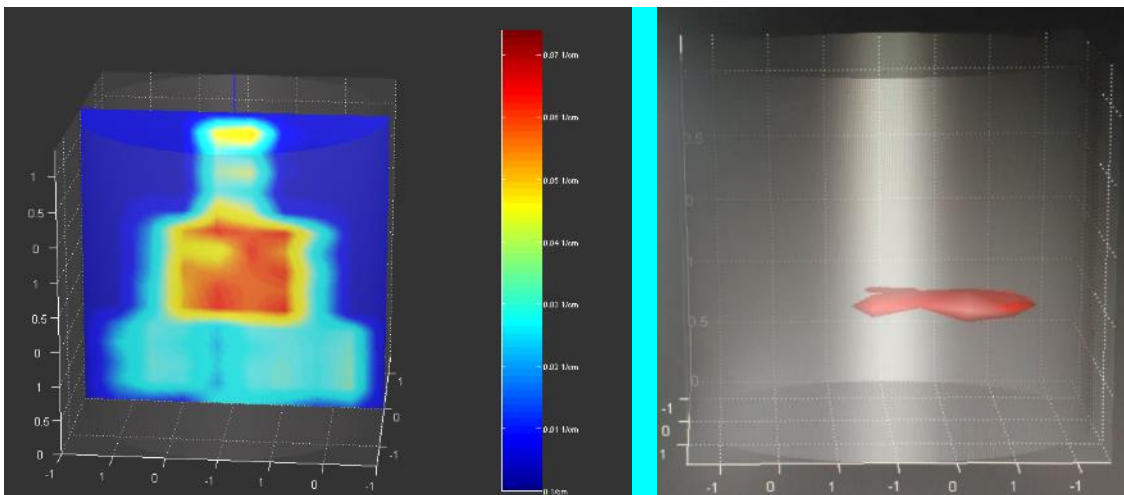


Figure 19 – Examples of TGS reconstruction: the matrix distribution on the left, the radioactivity distribution on the right.

The efficiency can be determined experimentally through measurements or calculated by means of Monte Carlo simulations [95], mathematical calculations or a combination of both approaches [96]). The need to make the efficiency evaluation more accurate and precise and thus to improve the gamma spectrometry results reliability, as well as the possibility to obtain complementary information than the mere activity quantification, has arisen the development of different measurement setup and ways to deal with the efficiency evaluation. Three main modalities are nowadays used (Figure 18):

- Open geometry: the item is measured entirely with only 1 measurement. This approach can be applied only to relatively small items (typically up to a drum or a 1 m³ box), or to items with very homogenous activity and density distributions.
- Segmented Gamma Scanning (SGS): typically applied to drums, but the ‘philosophy’ can be applied in general cases, and it’s the one also used in this work for the development of the new gamma spectrometry system (see chapter 10).
- SGS involves measuring the item at different positions to gather information relative to smaller parts of the item, reducing the uncertainty. This process helps assess item characteristics that affect efficiency and thus final results. Fixed systems, typically used for drums, often use a known radioactive source to evaluate the transmission of the drum and then apply localized corrections for the matrix distribution.
- Tomographic Gamma Scanning (TGS) operates similarly to SGS but involves acquiring more data, providing additional degrees of freedom. This expanded dataset enables the

reconstruction of the 3D distribution of both material and radioactivity within the scanned item (refer to Figure 19). Due to the complex hardware requirements and computational demands, TGS is typically employed for objects with simpler geometries, such as cylindrical drums.

- Tomographic Gamma Scanning (TGS): similar to SGS, but involves acquiring more data, providing additional degrees of freedom, which permits the reconstruction of the 3D distribution of material and radioactivity within the item (Figure 19).

9.2.2 Gamma-ray imaging

The gamma-ray imaging is a technique based on gamma ray detection, and provides information about the direction from which the detected photons are coming. In order to do this, state-of-the-art gamma imaging systems (an example in [97]) uses 2 main algorithms (Figure 20):

- For low energy photons, which undergo photoelectric effect in the detector, a moving coded-aperture mask with well-known pattern of holes is used to select the possible incoming directions [98]. The extension of the mask dictates the portion of the solid angle that the imaging system can use for reconstruction.
- For mid-high energy photons, the mask becomes less effective (as gamma rays are less attenuated) but photons undergo Compton scattering and electron/positron pair production. These events can be traced in the crystal if segmented electronics for signal read out are applied to the active volume of the detector [99]. All directions can be reconstructed by this algorithm, meaning that the entire solid angle is determined with each measurement.

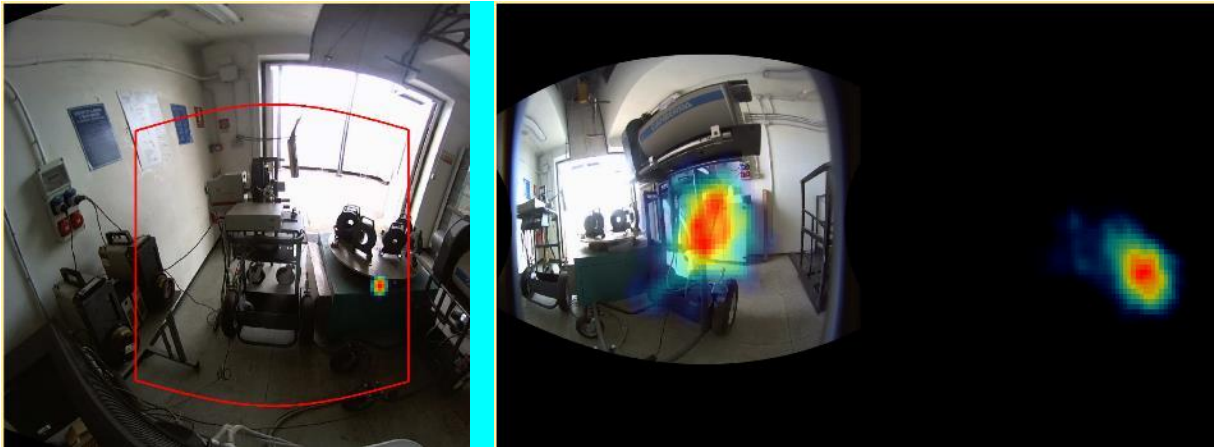


Figure 20 – Examples of gamma ray imaging reconstruction. On the left, an ^{241}Am source is imaged by means of the coded mask (visible as the red-contoured area). On the right, 2 ^{133}Ba sources are reconstructed: one is in the forward direction respect to the detector, the other is in the backward direction.

Most of modern gamma ray imaging systems are based on CdZnTe crystals, which can provide nice energy resolution allowing not only to create image for single photo-peaks, but also to use the system for quantitative gamma spectrometry like HPGe [100] [101].

Imaging information can help in defining the radioactivity distribution in a item: making multiple measurements around the item can permit to 'triangulate' the signals and thus to get a rough 3D reconstruction of radioactivity distribution [102].

9.2.3 X-ray imaging

X-ray imaging is an active technique that involves the use of an X-ray emitting system to assess the absorption of the signal by the item to assay [103]. The technique is applied in many different fields, like security or safety inspection, but can be used also to complement other radiological characterization procedures [102]. X-ray imaging can be used to make digital radiography of an item (an example in Figure 21), providing information about its density distribution.

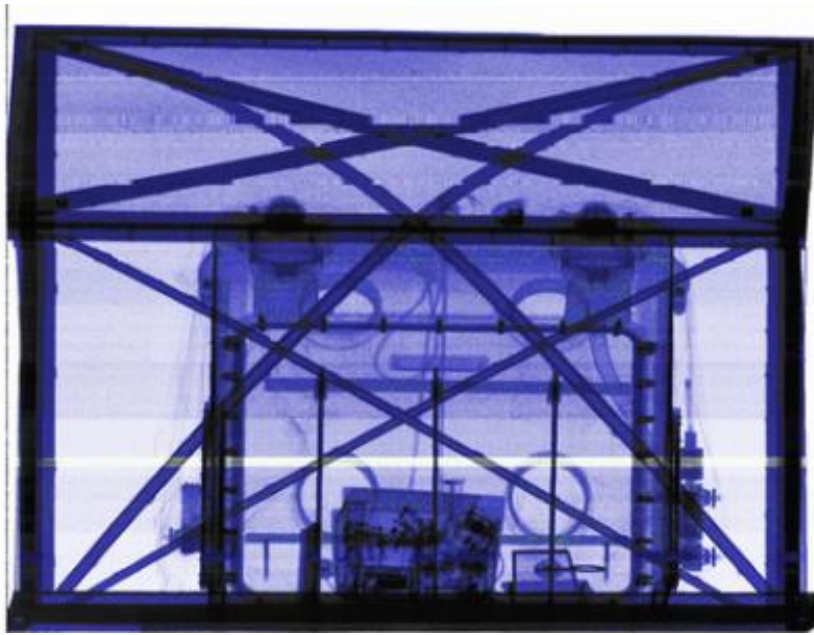


Figure 21 – Application of x-ray imaging to make a digital radiography of a metallic box containing a glove box with other components.

9.2.4 Neutron counting

The neutron counting is another main player in non-destructive characterization, in particular for radionuclides undergoing to nuclear fission, which is the main process generating neutrons. In a nuclear fission the electromagnetic repulsion in the nucleus is comparable to the attractive nuclear force and there is a probability for the nucleus to be deformed and then to separate into two fragments which are unstable and emit neutrons, gamma rays and beta particles. In order to make this probability high enough, the nucleus must have a high atomic number, belonging the fact to the Special Nuclear Material (SNM) list.

Compared to gamma rays, neutrons have a much higher penetration power, making them detectable even for high-density item which absorb the gamma rays it emits; moreover, neutrons are less influenced by matrix and radioactivity distributions in the item to assay, thus the results are less affected by such sources of uncertainty. However, neutrons have not a clear, distinguishable signature which can be used to match a certain signal to a precise radionuclide: this means that specific (and complex) algorithms must be used dealing with other characteristics of the fission decay (e.g., the multiplicity of neutrons emitted in a fission and their time correlation) to correlate the number of neutrons to the amount of radioactivity. Even in this case, neutron cannot be distinguished among the different radionuclides emitting them, thus only a total 'equivalent' amount of radioactivity is quantifiable. In order evaluate from this 'equivalent' amount the mass (or activity) of each single neutron emitter some complementary data are needed, like the isotopic composition in case of plutonium radioisotopes: such data can be known to the user, or can be determined by other assay techniques, like gamma spectrometry [66].

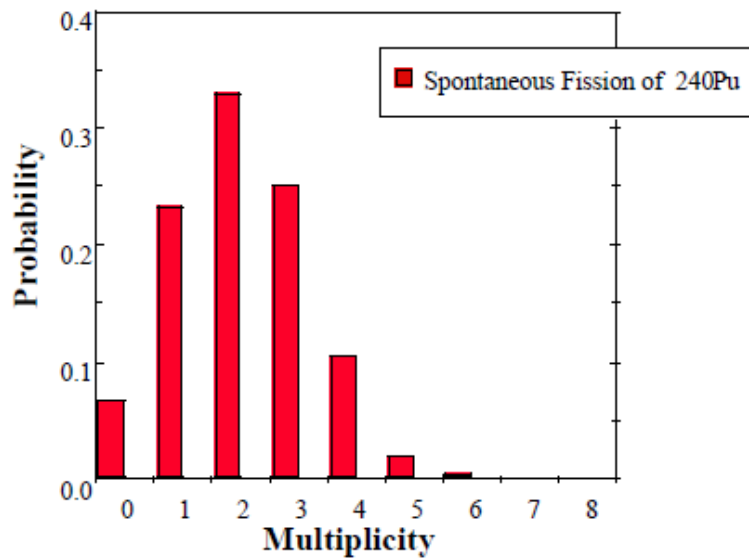


Figure 22 – Neutron multiplicity distribution for ²⁴⁰Pu spontaneous fission (adapted from ortec-online.com).

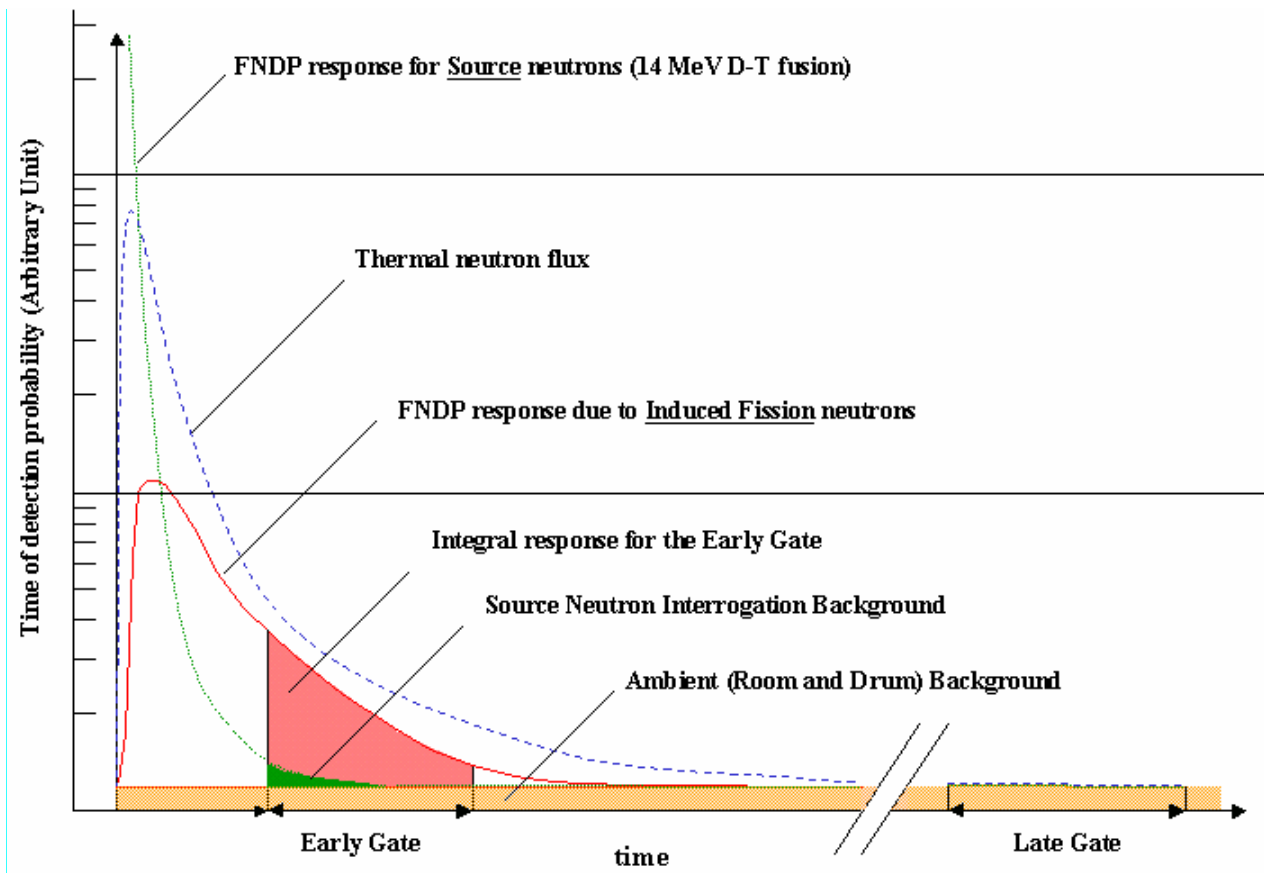


Figure 23 – FNDP (Fast Neutron Detector Package) Signals during an active neutron interrogation assay.

A passive neutron counting system is counting the neutrons reaching its detectors, which are usually gas filled detectors (like ³He tubes) covered with different possible materials useful for cut out from the count some part of the energy spectrum of the emitted neutrons. Because a fission reaction releases on average more than one neutron (Figure 22), different detectors must be used to being able to distinguish between signals coming from different neutrons. In order to avoid the several possible sources of spurious signals (e.g., neutrons from (α, n) reaction, or cosmic rays miming signals in detectors like neutrons), total counting of neutrons is nowadays being substituted by the

coincidence analysis (i.e., looking at neutrons arriving close in time) or by the multiplicity analysis (looking at all possible combinations of signals which can be correlated in time according to a fission reaction), where the latter being more precise but applicable only when the amount of nuclear material is enough. Many variations of these algorithms have been developed over the years, with different ranges in which each one provides the best results. During the algorithms tuning phase, the system calibration has to be performed, i.e., must be determined the relationship between the amount of a certain SNM and the signal it generates in the system according to the quantities analysed by each algorithm.

The same passive neutron counting system can become active if equipped with a 'neutron probe', like a deuterium-tritium (D-T) neutron generator: it emits a monochromatic 14 MeV neutron flux which can induce the fission reaction in fissile radioisotopes (like ^{235}U and ^{239}Pu), in particular if the neutrons are thermalized before reaching the item to assay in order to increase the fission cross section. In this case, it's said that the nuclear material is 'interrogated' by the neutron probe.

As the neutron flux from the neutron probe passes through, the neutron signal coming from the induced fissions increases and decreases back to environmental values once the 'interrogation' is concluded (Figure 23). An early gate, counting the gross neutron yield, is started as the 'interrogation' probe is reasonably close to zero, and this then close when the signal from fissions is expected to be close to environmental background data. Then, a late gate is opened corresponding to a region in time when only background signals are expected to be present: this background is subtracted to the gross signal measured in the early gate to calculate the net signals coming from the induced fissions, which is used to compute the 'equivalent' mass of the fissile SNMs by means of a dedicated calibration.

9.2.5 Calorimetry

Calorimetry is used as NDA technique for determining the power output of heat-producing nuclear materials: the heat is generated by the decay of radioactive isotopes within the item. Because the heat-measurement result is completely independent of material and matrix type, it can be used on any material form or item matrix. Calorimetric assay is the determination of the mass of radioactive material through the combined measurement of its thermal power by calorimetry and the knowledge of its isotopic composition (by external information or by other measurements). Calorimetric assay is routinely used as a reliable NDA technique for the quantification of plutonium and tritium content. Calorimetric assay is a very precise and accurate technique, being even better than destructive assay (DA) techniques in certain cases. However, its application field is very limited to high amount of material (like hundreds of grams of plutonium as minimum) and quite small items (maximum diameter of few tens of centimetres).

The radionuclides for which calorimetry is tailored as a NDA technique are out of the scope for low level radioactive metallic waste, thus this technique will not be described deeper.

10 A non-destructive gamma spectrometry method for characterization of radioactive metallic components

As it was discussed above, one of important tasks for successful decommissioning process of power, research reactor or other nuclear facilities is the optimization of MRW management by applying sorting and segregation of MRW. For this purpose, different approaches may be used for radiological characterization. Full-scale measurements covering the estimation of activity of all radionuclides in all metallic structures are costly and might be hardly feasible. In chapter 8 it was discussed in detail, that the neutron activation calculations can be used for DTM nuclides determination in SF method for intermediate and high level waste, but modelling accuracy depends on the input data - such as material data (composition and distribution of impurities), neutron flux and energy, nuclear data libraries - and on the methodology of the process and the simulation codes. For low and very low-level waste, the modelling of FP and CP transport via MCC is even more complicated and could be less accurate. Taking into consideration the modelling uncertainties, in all cases validation of the calculations is needed.

In reality, both the neutron activation and the surface contamination terms might be present and contribute to metallic structure activity. Their input to waste composition and corresponding results

of activity measurement may vary depending on the reactor type, the operation history and MCC peculiarities. The theoretically determined scaling factors may need to be validated and corrected using the available measurement data.

Most of long-lived DTM radionuclides contained in the radioactive waste are low energy gamma or pure beta or alpha emitters [16] and require radiochemical methods to separate the various radionuclides for measurement. For successful application of the SF method, one needs to establish a correlation between easily measurable gamma emitting nuclides (key nuclides) and DTM nuclides. The activities of DTM nuclides are then estimated by measuring the gamma emitting nuclides and applying the scaling factors. The key radionuclides are usually ^{60}Co and ^{137}Cs , which are measured by non-destructive gamma spectrometric method. ^{60}Co is mostly activation product produced by the $^{59}\text{Co}(n,\gamma)^{60}\text{Co}$ reaction in the 100% abundant stable cobalt isotope ^{59}Co with a cross-section of 18.7 barns [104]. On the other hand, ^{137}Cs is a fission product and due to its high-water solubility is easily transported and settled as surface contamination.

Gamma-ray spectrometry dealing with low activity of different samples is a fundamental part of radiological characterization programmes. For efficient characterization of low-level metallic waste, the determination of surface contamination is of great importance if applying the simple and non-destructive γ -spectrometry measurements. In the frame of the PREDIS project, T4.5 several approaches of non-destructive gamma spectrometry have been proposed and tested for characterization of radioactive metallic components by using (efficiency calibrated) modelling and different available measurement techniques, based on a scintillation detector NaI(Tl) and a HPGe in approach I and a HPGe and a CeBr₃ scintillation detector in approach II.

10.1 APPROACH I

A non-destructive gamma spectrometry technique by using MCNP6.1 simulations is proposed for interpretation of the resulting gamma-ray spectra of the radionuclides in activated components exhibiting contamination. In particular, NaI(Tl) and HpGe simulation spectra were produced, based on defined activities of the key radionuclides in the activated components (i.e. spectra for point sources, as well as for an activated metallic slab and a metallic slab with surface contamination). The quantitative agreement between the simulation and experimental spectra justifies the validity of the simulation method procedure for spectra production for point sources as well as for both surface and volume gamma sources. The simulation spectra show that not only the gamma peaks are characteristic but also the continuum is representative for the radionuclide distribution (activity distribution between volume and surface) in the measured sample (item). Metallic slabs with nominal activities inside their volume as well as on the surface were prepared and analysed by the proposed technique. The simulation spectra are compared with the experimental spectrum to determine the activation as well as the contamination of the slabs. The results were in good agreement with the nominal values. Therefore, the proposed method could validate the results for gamma emitters of neutron activation calculations in activated components exhibiting contamination.

10.2 APPROACH II

The aim of this approach was to investigate the γ -spectra of ^{60}Co (activation) and ^{137}Cs (surface contamination) laboratory-made metallic samples in different shielding geometries in order to develop a methodology for separation of surface contamination and volume activation from the spectra analysis. The analysis was performed by using both MCNP6 modelling spectra of sample-detector system and experimental spectra of different geometry laboratory-made metallic samples with ^{60}Co and ^{137}Cs sources acquired by HPGe and by CeBr₃ [105] detectors. Inter-comparison of modelling and experimental γ -spectra, peak intensity and peak/Compton ratio dependence for laboratory-made samples of ^{60}Co and ^{137}Cs in different thickness of iron plates (i.e. different shielding conditions) was carried out.

MCNP modeling of different source cases: planar source shielded with different metal plates and volume source, have revealed that surface contamination ^{137}Cs source can be distinguished if compared with reference source case by using modeling and experimental γ -spectra analysis The

analysis concerns the peak intensity and Compton(backscatter)/peak ratio or the shape of the spectrum. This proves possibility of separation of surface contaminated and activated metallic waste samples.

Inter-comparison of modelling and experimental γ -spectra, peak intensity and Compton (backscatter)/peak ratio dependence for lab-made samples of ^{60}Co and ^{137}Cs in different iron (plates/thickness) shielding conditions have shown a very good consistency of experimental and modelled results for spectra obtained with HPGe detector. As concerning in-situ measurement with CeBr_3 , despite the fact, that CeBr_3 is characterized by 10 times lower energy resolution it is still suitable for in-situ measurements of metallic radioactive waste for identification of surface and volume contamination. But the consistency of experimental and simulation results is worse in Compton (backscatter) region comparing to HPGe case, due to complicated geometry of the laboratory environment. Therefore, for the CeBr_3 detector more detail modelling of the measurement geometry is needed in order to validate the modelling results and allow usage of the models for investigating of γ -spectra of real surface contaminated or volume activated metallic samples with CeBr_3 detector.

This analysis provides valuable insights into the determination of surface and volume activity in different thicknesses of shielding metal by using HPGe and CeBr_3 detectors, and consequently can improve the characterization of activated and contaminated materials assay with better accuracy.

11 Proposed setup for characterization of low activity metallic segments with low measurement uncertainty

From the decommissioning of a NPP, a large amount of low activity metallic waste, which belongs to the classes VLLW, EW is produced ([18], [106], [107]). The segregation of waste is optimized by a more accurate radiological characterization and the selection of the management routes (i.e. decontamination processes and clearance/declassification procedures) is made more effective, increasing the volume of metals for recycling.

After dismantling, in-situ characterization is carried out to classify and package the generated metallic segments [108]. This is usually achieved by using portable devices to measure dose rate or total counts. Then, the packages that are usually 1-2 m³ are monitored by the non-destructive gamma spectrometry or by plastic scintillators for assessment of the activity and selection of the management route. The measurement uncertainty in this case is high, sometimes higher than 100%. The uncertainty is mainly due to the inhomogeneity of the density as well as the uneven distribution of activity within the package. For localization of the activity spots and more accurate estimation of radioactivity in waste containers, the sectorial gamma scanning [109] can be used. This technique is time-consuming and not suitable for the huge amount of waste produced from decommissioning. Recently, a system of plastic scintillation detectors [110] as well as the use of a portable large field-of-view gamma camera [111] are proposed for localization of gamma-ray emitting sources (activity inhomogeneity or activity spots) and therefore, for more accurate determination of radioactivity in waste containers. The problem of high uncertainty during measurement of waste in containers was also highlighted at the 12th International symposium release of radioactive materials – Provisions for clearance and exemption, Frankfurt, November 2022 [112] [113].

This work proposes a new non-destructive gamma spectrometry setup, which can be used after dismantling and segmentation of the reactor components to segregate the low activity metallic waste into: those to be managed as radioactive waste; those to be decontaminated and cleaned or declassified; the amount for direct release. The measurement parameters were defined for more accurate evaluation of the waste activity and therefore for more effective selection of the decontamination techniques and clearance procedures. The aim was the determination of the optimum parameters of the measurement to crucially reduce the measurement uncertainty (e.g., less than 30%), while the sensitivity for key radionuclides (^{60}Co , ^{137}Cs) is sufficient for acceptable measuring time (i.e., comparable to that of the usually used methods, characterization of 100 kg of metallic waste in 1-2 min).

For the proposed non-destructive gamma spectrometry setup, the efficiencies of the measurement for several source geometries (i.e., straight & convex pipes, metallic slabs and convex surfaces, screws and bolts, flanges grids, etc.) were evaluated by simulations performed by the MCNPX code. The bias due to activity inhomogeneity for several segments geometry was also determined [114].

The models were validated by using volume sources of nominal ^{137}Cs activity prepared at NCSR. When the geometry of the segments cannot be accurately simulated (e.g., a quantity of screws, bolts, grids), simplified geometries are proposed (i.e., slabs with homogeneous density distribution). Moreover, sensitivity analysis against the parameters that influence the measurement efficiency were carried out (i.e., diameter and wall thickness of pipes).

The final stage of this work is the detailed design and construction of the measurement infrastructure for industrial use (TRL 8), based on the proposed non-destructive gamma spectrometry setup. The infrastructure for industrial use will include the structure, the mechanical part, the sensors, the detectors, the collimators as well as the appropriate software and interface. In the frame of the PREDIS project, the TRL5 was achieved. So far, the feasibility of the proposed methodology was proved, and the technology was developed for demonstration at the Lab. The development of the technology under the PREDIS project includes the design of the specific collimators.

The results showed that by the proposed setup based on HPGe 20% detectors, crucial reduction of the measurement uncertainty (less than 25%) of metallic waste arisen from decommissioning was achieved, while the sensitivity is sufficient. Determination of 0.1 Bq/g of the key radionuclides ^{137}Cs and ^{60}Co in metallic waste of total mass more than 100 kg can be achieved in less than 2 min.

The proposed method is more accurate than the usually used methods while is equally time efficient. Metallic pipes are the most common geometry of metallic waste from decommissioning. Usually, the waste are put in steel containers of 1-2 m³. The 2 m³ container with metallic waste is measured in 15 min. Let's assume that the container is filled with pipes (i.e. 18 m of 30 cm diameter or 50 m of 20 cm diameter or 200 m of 10 cm diameter). By the proposed method, 18 m of 30 cm diameter pipes can be measured in 8 min, 50 m of 20 cm diameter pipes in 14 min and 200 m of 10 cm diameter in 27 min.

It should be mentioned that the proposed set up for measurement of metallic waste after dismantling can be used effectively to select the management route as well as the decontamination and clearance procedure. Nevertheless, it could be also used to demonstrate that the activity is below the clearance values. If clearance is not possible and decontamination not reasonable, characterisation of metals as radioactive waste can be realised. Therefore, the proposed set up offers "three at one".

12 Summary and future plans

This deliverable significantly contributes to the understanding about characterization of MRW in different kinds of reactors taking into account modelling, measurements, Nuclide Vector (NV) determination.

The outcomes of the T4.5.1 subtask:

- Provided the state-of-art scientific information on MRW classification, possible management routes and procedures for declassification of waste or clearance. Established the list of DTM nuclides for Metallic Radioactive Waste (MRW) streams and key nuclides, which allow MRW characterization by using Nuclide Vector (scaling factor) method.
- Modelling of nuclear fuel composition calculations, material activation calculations, calculations of RCS piping internal contamination can be particularly useful for the scaling factors determination, but it should be validated by measurements.
- Scaling factor determination methods and the iterative procedure of optimised nuclide vector approach have been introduced and should be applied every time in case of possibility to separate different (contaminated or volume activated) streams, to reconsider NV after decontamination (e.g. a sand blasting, melting) of streams, taking into account the time scale, etc.

Regarding the waste of low activity the aim of characterization is to decide if decontamination will be efficient as well as to select the most effective decontamination techniques and clearance methodologies. An innovative technique for MRW characterization, which can be used to distinguish activation from contamination on metallic components or waste have been carried on. Moreover, a semi-empirical technique for optimization of sorting of low activity metallic waste arisen from the

dismantling and cutting of NPP components have been introduced. The outcomes of the T4.5.2 subtask are:

- Combination of gamma spectrometry measurements and MCNP Monte Carlo simulation allows distinguishing of surface contamination from volume activation by the spectrum shape or photo peak intensity and photo peak/Compton ratio of γ -spectra (of key nuclides ^{137}Cs and ^{60}Co) analysis. This was examined by using NaI, CeBr₃ and HPGe detectors for different known activity laboratory-made samples.
- Proposed a new technique, which concerns the monitoring of metallic segments after dismantling and cutting, aiming at reduction of the measurement uncertainties related to the density and activity distribution. The method allows determination of activities of ^{137}Cs and ^{60}Co at the level of specific clearance for recycling in 1-2 min (amount of MRW ~100 kg in each measurement) and allows crucial reduction of uncertainties related to activity and density inhomogeneities (i.e. lower than 30 %). The proposed method can be used effectively to select the management route as well as the decontamination and clearance procedure. Furthermore, it could be also used to demonstrate that the activity is below clearance values. If clearance is not possible and decontamination is not reasonable, characterisation of metals as radioactive waste can be realised. Therefore, the proposed set up offers “three at one”. The technique was developed at TRL5 level.
- The procedure for optimized scaling factors determination by applying described gamma spectrometry technique was demonstrated taking into account discrimination of surface contamination from volume activation of the analyzed metallic samples, and key nuclide ^{60}Co measurement on-line, facilitating Nuclide vector determination for specific RMW stream. The technique was developed at TRL3 level.

The future plans:

- The problem of how to translate the sampling uncertainty (relatively high) to package uncertainty, where both, the mass and the similarity of the package content to the sampling space from which the SF is defined, could be different. Package mass is much greater than the sampling mass, therefore the package uncertainty should be lower. The implication of considering conservative Confidence interval in packages is the risk of classification change/mistake. However, the total inventory is well controlled due to the large number of disposed/stored packages that make the relative total uncertainty be relatively low. All these features are going to be addressed in ICARUS EURAD2 project. The methodology to quantify the uncertainty/accuracy in packages that make easiest the control and negotiation with regulatory body/agency etc. will be defined.
- The innovative technique, which can be used to distinguish activation from contamination on metallic components as well as the technique for optimization of sorting of low activity metallic waste arisen from the dismantling and cutting of NPP components are going also to be addressed in ICARUS EURAD2 project.

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