Deliverable 9.8: ROUTES – Review of characterisation of legacy and historical wastes

Work Package ROUTES

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Executive Summary

The ROUTES (Waste management routes in Europe from cradle to grave) work package (WP) is one of two strategic studies within the European Joint Programme on Radioactive Waste Management (EURAD). Its main objectives are to provide an opportunity to share experience and knowledge on waste management routes between interested organisations, to identify safety-relevant issues and their R&D needs, and to describe and compare different approaches to characterisation, treatment, and conditioning, as well as to identify opportunities for collaboration between member states (MS). The aim of Task 3 of ROUTES is to identify techniques for characterisation, to compare the methods applied in different countries, to identify knowledge gaps and give recommendations for future R&D. This report summarises the results gained in work on the second subtask of Task 3 on characterisation and segregation of legacy waste.

The information provided in this report result from a workshop held in February 2023 and was completed by additional information provided to the task leader after the workshop. The aim of the workshop was to systematically collect information on existing knowledge in handling of legacy waste (segregation, characterisation) and to design an integrated approach for the management of historical and legacy waste. This integrated approach should cover the aspects of retrieval, sampling, characterisation, treatment, conditioning, storage, and disposal.

One challenge of a general guideline of handling legacy waste is the variety of wastes, that may or may not fall under the category of “legacy waste” depending on the different member states legislation. To circumvent the lack of a globally accepted definition of the terms “historical waste” or “legacy waste”, this report adapted the definition of the ERDO working group and thereby developed a definition taking into account waste from nuclear accidents, the status of waste acceptance criteria (WAC) development in the different member states, as well as the technical and financial challenges of handling these wastes.

Legacy wastes can be divided into three categories: Unconditioned legacy waste, conditioned legacy waste and legacy waste of unknown status. Within these categories, certain attributes of legacy waste handling coincide as well as the associated challenges. **Unconditioned legacy waste** in general enables easy sampling, but with the challenge of representative sampling. These wastes are often mixed wastes, which need to be segregated before further management. Special care needs to be taken for reactive wastes, which need chemical speciation before further handling. Challenges also arise from high-volume waste (e.g., soil) and waste from nuclear accidents, which bare high loads of alpha-emitters. Most retrieved legacy wastes fall under the category of **conditioned legacy waste**. Characterisation of this waste is in general easier utilising non-destructive radioanalytical methods, as sampling might be difficult. As for the unconditioned legacy waste, chemical speciation is important for reactive wastes. The last category, **legacy waste of unknown status**, comprises all the waste with a general lack of information, where it is unknown if the waste has been treated and/or conditioned and characterisation results are not available or assignable to a specific waste batch.

Each waste category and waste type comes with certain specific **management issues**. However, common issues, irrespective on the specific type of legacy waste, relate to a lack of information on waste origin and history, resource shortages, or the problem of drum disintegration further complicating the waste management. In detail, the technical resource shortages refer to a lack of handling equipment specific to legacy waste, storage, and disposal capabilities, as well as a lack of WAC for these facilities. Additional shortages highlighted during the workshops for this report were financial and human resources.

These technical shortages also reflect on the research and development (R&D) and legislative recommendations highlighted in this report. As there are multiple overlaps of the waste type specific issues, the recommendations are structured by the legacy waste status, i.e., unconditioned and conditioned legacy waste and differ between legislative/organisational recommendations and R&D (technological) recommendations. For legislative recommendations, the WAC development for both interim storage and disposal of legacy wastes has often been stressed by the task participants. An

additional recommendation has been the safety and health risk assessment, including the monitoring of legacy waste in storage. For R&D recommendations, the long-term stability of wastes has been stressed, including resistance tests for legacy waste matrices of pre-conditioned legacy wastes and chemical interactions of mixed wastes (both unconditioned and conditioned).

During the preparatory work of this report in the WP, a model for an **integrated approach of radioactive waste management** (RWM) was developed, including the transitioning phase from legacy to regular radioactive waste, as well as the emphasis on the applicability of the individual management steps for small amounts of waste. This integrated approach for the management of legacy wastes includes the management steps retrieval, sampling, characterisation, treatment, and conditioning. The management of legacy waste comprises all steps from retrieval to conditioning. It should be highlighted, that characterisation is not necessarily a single step but accompanies the RWM routes throughout each step. For instance, the characterisation step could be used for retrieval, for segregation or classification, for treatment or for conditioning. After the final step of conditioning, the waste should be characterised (both physiochemical and radiological) and conditioned in a way that ensures a compliance of the waste with WAC and thereby being equal to general radioactive waste (RAW). For small amounts of waste, the RWM starts with the step “Sampling”, as the waste should already be accessible, sorted to some extent and basic information on the waste should be available.

This report focuses on the three steps “Segregation”, “Sampling” and “Characterisation” of legacy wastes and only includes generic information and guidelines on the other steps of the integrated approach for the management of legacy wastes. This decision has been taken on the grounds that for retrieval of legacy waste, the situation varies highly depending on the initial situation. This includes the status and type of waste, but also the location and accessibility of the specific legacy waste. The limiting framework for treatment and conditioning of legacy waste are significantly depending on the legal boundary conditions in the respective countries, e.g., existence of WAC for interim storage or final disposal, which also hinder specific handling guidelines.

The applicable technologies and methods for the management of legacy waste not only depend on the specific waste type, as it is the case for non-legacy radioactive waste. A specific characteristic of legacy waste is, that the variable of untreated and unconditioned waste versus already treated and/or conditioned waste is added. The difference in the approaches between unconditioned and conditioned legacy waste can be found mainly in the segregation and characterisation step. Additionally, in case of conditioned legacy waste, a reconditioning or repackaging might be necessary for final disposal.

Segregation of unconditioned legacy waste is mainly based on classification of the waste by dose rate and contamination levels, or by material/type of waste. The segregation by material facilitates, i. a., subsequent sampling and characterisation. As for conditioned waste, characterisation by destructive analysis (DA) might not always be applicable. If conditioned legacy waste can be segregated, options are non-destructive analyses (NDA), e.g., applying dose rate or x-ray imaging. Characterisation between unconditioned and conditioned waste differs in the applicable methods. For unconditioned legacy waste, samples can be obtained relatively easily and therefore DA methods are frequently used. As sampling might present a challenge for conditioned legacy waste, NDA is commonly used for characterisation.

This report is concluded by a set of case studies from Bulgaria, Cyprus, France, Germany, Portugal, Slovakia, Spain, and Ukraine, referring to distinct management steps of the integrated approach on radioactive legacy waste management.

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Glossary

DA – Destructive Analysis

DTM – Difficult-to-Measure

DSRS – Disused Sealed Radioactive Source

ERDO – European Repository Development Organisation

ETM – Easy-to-Measure

EURAD – European Joint Programme on Radioactive Waste Management

HLW – High Level Waste

HPGe – High-Purity Germanium

ICP-OES – Inductively Coupled Plasma Optical Emission Spectroscopy

ILW – Intermediate Level Waste

LILW – Low- and Intermediate Level Waste

LIMS – Large Inventory Member States

LLW – Low-Level Waste

LSC – Liquid Scintillation Counter

MS – Member State

NDA – Non-destructive Analysis

NORM – Naturally Occurring Radioactive Materials

NPP – Nuclear Power Plant

PG – Phosphogypsum

PIPS – Passivated Implanted Planar Silicon

RAW – Radioactive Waste

R&D – Research & Development

ROUTES – Waste Management Routes in Europe from Cradle to Grave

RWM – Radioactive Waste Management

SF – Scaling Factors

SIER – Spent Ion Exchange Resin

SIMS – Small Inventory Member States

UNGG – Uranium Naturel Graphite Gaz

VLLW – Very Low-Level Waste

WAC – Waste Acceptance Criteria

WMO – Waste Management Organisation

WP – Work Package

XRD – X-Ray Diffraction

XRF – X-Ray Fluorescence

1. Introduction

The ROUTES WP (Waste management routes in Europe from cradle to grave) is one of two strategic studies within the European Joint Programme on Radioactive Waste Management (EURAD). Its main objectives are to provide an opportunity to share experience and knowledge on waste management routes between interested organisations, to identify safety-relevant issues and their R&D needs, and to describe and compare different approaches to characterisation, treatment, and conditioning, as well as to identify opportunities for collaboration between member states (MS). The aim of Task 3 of ROUTES is to identify techniques for characterisation, to compare the methods applied in different countries[1], to identify knowledge gaps and give recommendations for future R&D. This report summarises the results acquired in the second subtask of Task 3 on characterisation and segregation of legacy waste.

The information provided in this report results from a workshop held in February 2023, and was completed by information provided to the task leader after the workshop. The aim of the workshop was to systematically collect information on existing knowledge in handling of legacy waste (segregation, characterisation) and to design an integrated approach for the management of historical and legacy waste. This integrated approach should address retrieval, sampling, characterisation, treatment, conditioning, storage, and disposal. This previously listed themes are discussed in multiple chapters of this study.

Chapter 1 provides information about the initial situation in the different MS. As there is no broadly accepted definition of the term “historical and legacy waste”, this chapter begins with a definition of the terms that will apply for this report and the information provided within. The chapter concludes with a collection of the existing knowledge on legacy waste in the participating MS.

The following chapters, Chapter 1 and Chapter **Erreur ! Source du renvoi introuvable.**, present the identified management issues of legacy wastes and recommendations for R&D activities. All this information has been collected during the workshop underlying this report.

The last and 0th chapter presents an integrated approach for the management of historical and legacy wastes and tackles the management steps retrieval, sampling, characterisation, treatment, and conditioning. The presented information is completed by examples from different MS.

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[1] Participating countries were Bulgaria, Cyprus, France, Germany, Greece, Portugal, Slovakia, Spain, and Ukraine.

2. Methodology

This task investigates the segregation and characterisation of legacy waste, as well as of small amounts of waste. To this aim, the existing knowledge about legacy waste and the experience in handling legacy waste have been systematically collected and analysed. This approach will cover as many perspectives as possible, but without claiming to be exhaustive. The existing knowledge has been shaped into an integrated approach for the management of legacy waste, including retrieval, segregation, sampling, characterisation, treatment, and conditioning. Based on the provided information by MS, issues related to the management of legacy wastes have been gathered, analysed and R&D recommendations have been derived.

The information provided is this report has been collected through information provided by country representatives, e.g., via a questionnaire, and two workshops with the task participants:

- In a first workshop, the general aim and the distinct foci of this report have been discussed and decided on. Specifically, the structure and potential issues, such as the different applied definitions of the term "legacy waste", have been discussed. This first workshop resulted in the provision of the structure of the report, the content of the follow-up questionnaire and the content of the second workshop.
- The questionnaire, shared following the first workshop in preparation of the second workshop, provided valuable information of the status of legacy waste and legacy waste management in the countries participating in this task. The questionnaire was separated into three parts:
    - Part 1 – Definition of historical and legacy waste
    - Part 2 – Systematic collection and analysis of existing knowledge about historical and legacy waste
    - Part 3 – Designing an integrated approach for management of historical and legacy waste

  In Part 1, the participating MS were asked to provide their country's definition of historical and legacy waste. In Part 2, the participating MS shared their information on existing legacy waste types including origin, amount, key radionuclides, waste status (e.g., unconditioned, conditioned, undefined), identified management issues, experience in segregation and characterisation of legacy waste, as well as R&D recommendations depending on the legacy waste status "unconditioned", "conditioned", and "undefined". In Part 3 of the questionnaire, the MS stated for all steps of the integrated approach for the management of legacy waste (i.e., retrieval, segregation, sampling, characterisation treatment, and conditioning), if experience is available and if the MS could provide a case study for single or multiple steps of the integrated approach. All information provided via the questionnaire was included in the discussions throughout the second workshop.
- The second and last workshop of this task aimed at discussing the legal status of legacy waste and national legacy waste management. Based on this information, a common understanding of legacy wastes has been decided on and a universally applicable integrated approach on legacy waste management has been developed.
- Based on the steps of the integrated approach on legacy waste management, case studies have been provided by task participants. Within these case studies selected information, challenges,

and successes of legacy waste management in participating member states have been described.

3. Understanding legacy waste across Europe: Inventory, Legal Framework and Definition

3.1 Overview of the regulatory framework for legacy waste

The current situation of legacy wastes in the MS is quite diverse, as is the information available about these wastes. One factor for these wide ranges comes from the different legislations on legacy waste across Europe.

The objective of the NEA Expert Group on characterization Methodology of Unconventional and Legacy Waste (EGCUL) is to support organizations in the management and characterization of wastes generated at post-accident and legacy sites. They published a report [1] that focused on the characterisation of radioactive waste in terms of safe management, considering both radiological and non-radiological risks. The EGCUL emphasizes in the above cited study, that legacy waste is radioactive waste created by previous nuclear activity or others involving radioactive materials, that does not conform with current regulations. These are often wastes created early in a country's nuclear program that were not managed in accordance with contemporary waste management and environmental protection regulations. Some legacy wastes, for example, were held in vaults and silos with no information documented about the waste's origin or properties, and with little thought given to how the waste would eventually be collected. Furthermore, waste pre-treatment, treatment, and conditioning for storage and disposal were not considered. As a result, managing such wastes, ensuring safe storage in their present facilities, identifying, and implementing optimized waste processing options, and determining where and how the wastes are to be disposed of, is challenging. In the USA, legacy wastes are “wastes generated during the development, production, and testing of nuclear weapons for the Manhattan Project and the Cold War. They can contain a wide range of radioactive materials and/or hazardous chemicals” [2].

Within the scope of this task, each participating member state (MS) shared the definition of what constitutes a legacy for their nation. Hence, some countries, such as Bulgaria, Cyprus, Spain, or Ukraine only define specific waste types or origins as legacy waste. Indeed, the WMO (ENRESA) classifies waste in Spain as legacy waste, also referred to as non-typified waste, which was produced before the establishment of a RWM methodology. For Ukraine, only three distinct waste kinds inherited from the Soviet era (nuclear industry, Chernobyl accident, or Soviet military program) are covered.

In contrast, other countries, such as France or Slovakia, do not have a legal definition for “legacy waste”. In the case of France, legacy waste encompasses two legal definitions:

- heritage or historical waste (produce before 1990) from research, production, and fuel reprocessing activities that have not yet been processed nor packaged. Some of this waste is still in raw form or, in some situations, may require reconditioning [3]. According to this definition, legacy waste would account for just 2 to 3% of total inventory in France [4]. This waste offers a technical and financial challenge due to technological constraints and the expense of management (retrieval, treatment, conditioning, and packing) [5]. Furthermore, French regulations require that all long-lived intermediate-level waste (ILW-LL) generated prior to 2015 be conditioned by the end of 2030 (The Environmental Code, Article L. 542-1-3).
- "orphan" waste i.e., waste for which the institution /person responsible is unknown or has failed to act, Andra can assure the management of "orphan" radioactive waste, within the context of its missions outlined by Article L. 542-12 of the Environmental Code.

Finally other countries, such as Germany, Greece or Portugal, have a very wide definition of legacy waste. For these countries, legacy waste is defined under national programs. For Greece and Portugal, all radioactive wastes that have not been effectively defined or handled and for which no characterisation are available falls into the legacy waste category. In Germany, legacy waste is defined as any waste that was conditioned before April 3, 2007.

3.2 Categorizing Legacy Waste: Inventory and types

Legacy waste covers basically all waste types, the amounts range from single drums to kilotons of material, the age ranges from waste originating from the beginning of the nuclear age to the early 21st century (depending on the country-specific definition of legacy waste) and covers the waste forms raw or unconditioned to conditioned, as well as unknown/undefined.

Based on the information obtained, three categories of legacy wastes were identified:

- **Unconditioned legacy waste:** This category of waste is generally easy to sample but presents a representative sampling issue. These wastes are often mixed wastes and need to be segregated before being managed. Special care needs to be taken for reactive wastes, such as co-precipitation sludge from effluent treatment, which need chemical speciation before further handling. This category also includes high-volume waste such as soil and waste from nuclear accidents, which bare high concentration of alpha-emitters.
- **Conditioned legacy waste:** This waste category is valid for most retrieved legacy wastes. Characterisation of this waste is generally easier using non-destructive radioanalytical methods, as sampling might be difficult. As for the unconditioned legacy waste, chemical speciation is important for reactive wastes.
- **Unknown legacy waste:** In this category all legacy wastes to which there is no information available on the waste history or possible prior treatment and/or conditioning are included.

Waste inventories differ greatly between countries (*Table 1*) and may not always have the same influence on the country regardless of whether they are LIMS or SIMS.

Country	**Quantity of Legacy waste**	**Examples of legacy waste inventory**
Bulgaria		Textiles, metals, polymers, mixed wastes. Conditioned legacy waste in interim storage.
Cyprus	Amount: approx. 300 kilotons	Phosphogypsum from fertilizer industry, unconditioned.
France	2 to 3% of total inventory in France	STE2 sludges, resins, graphite, and magnesium waste.
Germany	Approx. 100.000 m^3 of conditioned wastes.	Retrieved wastes from Asse II mine.
Greece		Raw wastes. Origin: Research reactor, laboratories. Mixed wastes in drums.
Portugal	Approx. 800 drums	220 L Drums containing historical waste not identified or records seem to have been lost (most from medical applications). Status of waste: conditioned.
Slovakia	69.000 m^3 of soils	Decommissioning from NPP A1, wastes from unknown producers. E.g., graphite (unconditioned), potassium dichromate (conditioned).
Spain		Sludge (conditioned) with unknown radiochemistry.
Ukraine		Type A – radioactive waste disposed in Soviet times at RADON storage facilities. Type B – radioactive waste from Chernobyl accident, stored at different sites. Type C – radioactive waste from Soviet military programme.

Table 1 – Amount and Examples of Legacy waste inventories in different MS

3.3 Definitions of legacy waste

Since the previous workshop in May 2022, it became clear that there is a broad range of definitions concerning “legacy waste” or “historical waste” among the various member states. Therefore, the different applicable meanings of the member states were presented and debated, before the task’s participants decided which definition of legacy waste should be used within this task. The final definition was based on the legacy waste definition of the European Repository Development Organisation (ERDO) but includes some minor adaptions. The agreed-on definition for legacy waste in ROUTES Task 3.2 is:

“Radioactive waste produced as a result of a historical nuclear activity and industrial/ medical activity for which there is no sufficient characterization from a physicochemical and radiological point of view. This waste may or may not be conditioned, does not approvably comply with WAC, if already available, and represents a technical and financial challenge due to technological constraints and management costs (recovery, treatment, conditioning).”

This definition considers the waste from nuclear accidents, the status of WAC development in the different member states, as well as the technical and financial challenges when dealing with this type of waste.

4. Identification of management issues

Management has been discussed and current issues with the management of legacy waste has been summarised. Additionally, R&D recommendations have been elaborated (**Erreur ! Source du renvoi introuvable.**).

Based on the challenges in the participating member states with respect to the management of legacy waste, a summary of the most common management issues has been composed.

General issues, independent on the specific type of legacy waste, relate to a lack of information on waste origin and history, resource shortages, as well as the problem of drum disintegration further complicating the waste management. In detail, the technical resource shortages refer to a lack of

- handling equipment specific to legacy waste,
- storage and disposal capabilities,
- WAC for these facilities.

Additional shortages highlighted were financial and human resources.

The legacy waste type specific issues are summarised in *Table 2*. One issue highlighted is the monitoring of legacy waste, that implies the risk of gas releases. This waste is, e.g., organic waste releasing hydrogen due to radiolysis or NORM waste emitting Radon. Mixed waste might as well imply the risk of hydrogen generation if the constituents include organics. An additional issue highlighted for conditioned waste concerns the separation of waste from the matrix.

Waste type	**Identified management issue**
Organic waste	• Monitoring (e.g., gas release) • Management of liquids
Metals	• Characterisation methodology missing (mixed legacy metal wastes)
Graphite	• Assessment of Wigner energy in the context of WAC • Ignition risk of mixed graphite & magnesium wastes (fuel cladding)
Chemicals	• High content of alpha RN • Chemical reactions (e.g., exothermic)
Sludge	• Missing or incomplete information on the origin of waste • Characterisation of cemented sludges • Chemical reactions (e.g., exothermic)
DSRS	• Unknown activities of non-retrieved sources
NORM (e.g., PG)	• Erosion of Disposal sites • Monitoring • High amounts of waste
Mixed	• Monitoring • Segregation for high activity materials

Waste type	**Identified management issue**
	• Waste history
Accident waste	• High load of alpha emitters

Table 2 – Identified management issues of unconditioned legacy waste

5. Integrated approach for the management of legacy waste

During the preparatory work of this report in the WP, a model for an integrated approach of RWM was developed, including the transitioning phase from legacy to “regular” radioactive waste, as well as highlighting the applicability of management steps for small amounts of waste. This integrated approach for management of historical and legacy waste is shown in *Figure 1*.

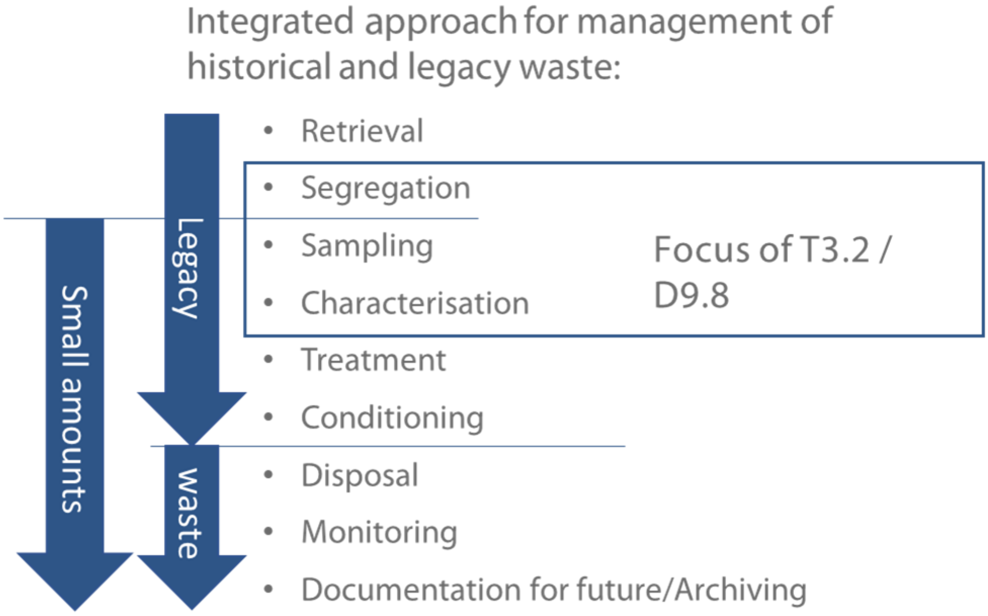


Figure 1 – Integrated approach for management of historical and legacy waste.

The management of legacy/historical waste comprises all steps from retrieval to conditioning. In case of characterisation, it should be highlighted that characterisation is not necessarily a single step but accompanies the RWM routes throughout each step. For instance, the characterisation step could be used for retrieval, for segregation or classification, for treatment and for conditioning of the waste. After successfully going through these steps, the waste should be characterised (both physico-chemical and radiological) and conditioned in a way that ensures a compliance of the waste with WAC and thereby complying with the preconditions applicable to general RAW. For small amounts of waste, the RWM starts with the step “Sampling”, as the waste should be accessible, sorted to some extent and basic information on the waste should be available.

During the discussions at Task 3.2’s first [MS110] and second [MS161] workshops, it was decided that this report, deliverable D9.8, will focus on the three steps “Segregation”, “Sampling” and “Characterisation” of legacy wastes and only includes generic information and guidelines on the other steps of the integrated approach to legacy waste management. This decision has been taken on the grounds that for retrieval, the situation varies highly depending on the initial situation. This includes the status and type of waste, but also its location and accessibility. The limiting framework for treatment and conditioning of legacy waste is significantly depending on the legal boundary conditions in the respective countries, e.g., existence of WAC for interim storage or final disposal, which also hinder specific handling guidelines.

The information presented in this report are based on the experience of the participating member states. *Table 3* summarises the provided case studies by different participating member states on the single steps of the integrated approach. This table visualises the focus of the work on legacy wastes in the different countries, highlighting the importance of segregation, sampling, and characterisation during management of legacy radioactive waste.

	Bulgaria	**Cyprus**	**France**	**Germany**	**Portugal**	**Slovakia**	**Spain**	**Ukraine**
Retrieval				x		x		x
Segregation	x		x	x	x			
Sampling		x						
Characterisation							x	
Treatment								
Conditioning								

Table 3 – Case studies of the single steps of the integrated approach, provided by participating member states.

5.1 Introduction to the integrated approach

The applicable technologies and methods for the management of legacy waste not only depend on the specific waste type, as it is the case for non-legacy radioactive waste. A specific characteristic of legacy waste is, that the variable of untreated and unconditioned versus already treated and/or conditioned waste is added.

The highlighted difference in the approaches between unconditioned and conditioned legacy waste are mainly in the segregation and characterisation steps. Additionally, in case of conditioned legacy waste, a reconditioning or repackaging might be necessary for final disposal.

Segregation of unconditioned legacy waste is mainly done by radiation or contamination level and/or by material/type of waste. The segregation by material facilitates i.a. subsequent characterisation. Characterisation of conditioned waste by destructive analysis (DA) might not always be applicable. If conditioned legacy waste can be segregated, options are non-destructive analyses (NDA), e.g., applying dose rate or x-ray imaging.

Characterisation between unconditioned and conditioned waste differs in the applicable methods. For unconditioned legacy waste, samples can be obtained relatively easy and therefore destructive analysis methods are frequently used. As sampling might present a challenge for conditioned legacy waste, non-destructive analysis is commonly used for characterisation.

A case study presenting an integrated approach for the management of liquid, organic legacy waste is provided by Slovakia It summarises the handling of the heat transfer fluids Chrompik and Dowtherm after the shut-down of A1 NPP in Jaslovske Bohunice. Additional, short case studies for the single steps of the integrated approach have been prepared by different participating member states of ROUTES Task 3.

Management of legacy RAW coming from NPP A1 – Case Study of Slovakia

Principal source of legacy radioactive waste (RAW) in Slovakia is the nuclear power plant A1 (A1 NPP) in Jaslovské Bohunice. The A1 NPP, a gas-cooled, heavy water moderated reactor, was shut down after an accident, which took place in 1977. Due to the accident, the fuel in the reactor core was significantly damaged and fission nuclides spread to the primary circuit and partly also to the secondary circuit. The dose rate in certain sections of the primary circuit and suction pipes of the turbo-compressors reached up to 3 Gy/h. Still in 2003, the contamination levels of the inner surfaces of the primary circuit ranged from 1 kBq/cm² to 10 MBq/cm².

Challenges related to decommissioning of the reactor are physical and radiological characterisation in the field of high dose rates, fragmentation and transport of large size, heavy weight and activated or heavily contaminated systems, structures, and components. Processing and treatment of these specific types of RAW represent additional challenges due to high specific activity, content of alpha radionuclides and/or aggressive chemical properties. In some cases, RAW with different properties are mixed, therefore a preceding segregation is required. In many cases, a special type of installations had to be developed for the processing and treatment of these wastes. New advanced techniques and methods for characterisation, dismantling and fragmentation should be applied whenever needed. Preferably remote-controlled equipment and procedures should be applied for certain operations.

One of the most challenging type of liquid RAW coming from NPP A1 is Chrompik, an aqueous solution of potassium chromate and dichromate. Chrompik had been used as liquid heat transfer medium for cooling of the spent fuel assemblies during the operational time of A1 NPP. The specific activity of this waste is up to 1 GBq/dm³. For conditioning, the clear liquid phase of Chrompik is vitrified into a glass matrix. However, a sludge phase of Chrompik cannot be vitrified due to technical reasons and thus must be segregated by decanting from the clear liquid phase and solidified using geopolymers. Due to the high specific activity and content of alpha radionuclides, this kind of RAW will be disposed of in a deep geological repository.

Another problematic waste type is Dowtherm, which has also been used for the cooling and storage of the spent fuel elements at the A1 NPP. Dowtherm is a eutectic mixture of diphenyl and diphenyloxide and was developed to prevent corrosion of the spent fuel elements. As treatment, this liquid, organic waste is burned in an incineration facility at temperatures from 750°C to 950°C, which is followed by an afterburning at temperature of 1100°C in the post-combustion chamber. The ashes arising from the incineration process are mixed with paraffin, encapsulated into 200 L steel barrels and inserted in a fibre reinforced container to produce waste package approved for near-surface disposal in Slovakia.

5.2 Retrieval of legacy waste

Processes for the retrieval of legacy waste highly depends on the initial situation. This includes the status and type of waste, but also the location and accessibility of the waste. Therefore, due to the nature and universal approach of this report, no specific information on retrieval can be given. Three short case studies have been prepared by member states on their specific waste retrieval project.

The German case study describes the retrieval of legacy waste from the Asse II mine, a deep geological repository used until the 1970s and which should be cleared. The case study provided by Ukraine discusses the retrieval of legacy waste in the Chernobyl exclusion zone.

Retrieval of waste from the mine ASSE II – Case Study of Germany

The former potash and rock salt mine ASSE II in northern Germany served as a research facility with the aim to create and maintain a final repository for different types of radioactive waste, including SIERs, sludge, evaporator concentrates, scrap metal and metallic components, organic resin and others (contaminated tools, plastics, ashes and bitumen), [1]. Between 1967 and 1978, over 100,000 containers filled with LLW and over 1,000 drums containing ILW were stored in 13 different chambers on different levels of depth within the mine, [2]:

- *11 chambers on the 750 m level;*
- *one chamber on the 725 m level;*
- *one chamber on the 511 m level.*

While the contents of the deeper chambers solely comprise of LLW (both drums with and without additional concrete shielding on the exterior of the drum), the content of “ELK 8a/511” consists of ILW containers mainly, [3, p. 43]. The containers used in “ELK 8a/511” are drums with a nominal volume of 200 l. Material conditioning originally included encasing identical types of material in either concrete or bitumen, but the conditioning status of some drums remains unknown, [1]. The documentation of the exact distribution and methods of treatment (e.g., exact amount and variety of materials within one single drum) lacks detail and reliability, [3, p. 37].

Due to continuous inflow of water at different locations of the mine, German federal legislation demands the retrieval of the deposited waste, [2]. This retrieval poses major challenges.

One chamber (725-m-level) is still accessible, while visual and physical contact is impossible for the remainder of chambers. As a result, the status of these chambers is unknown [1]. Therefore, planning the retrieval is a challenge, among other things, due to many missing facts: Whether it is the structural integrity, radiation dosage levels or the position and geometrical orientation of containers (Use of different techniques during deposition (chaotically, horizontally, and vertically) [2]). The factual situation found within the chambers during exploration efforts can always negate the basis for specific retrieval plans [2].

Additionally, the condition of the drums is unsure as well. Mechanical impact of salt rocks falling from the chambers’ back or pressure from overlaying backfill material may have affected the condition and the integrity of drums, [4]. Other constraints that complicate the retrieval are the possible decay of drums due to corrosion [4], legal restrictions (mining and radiation legislation) and the requirements of coordinating the different subprojects that make up the entire retrieval and shut down project of ASSE II, [2].

The planning efforts consider different scenarios based on calculations and reports from the time of deposition [4] to clarify and gather the missing data described beforehand. Furthermore, to complete the missing information on the chambers’ conditions, different investigations with endoscopic cameras and geological measurements (e.g., borehole X-rays, seismography) have been conducted [5] or are part of future endeavours, [6]. The current planning process is adjustable and flexible and consists of a multitude of optional possible retrieval methods, [4]. The results of the currently ongoing planning process show that a new system of underground tunnels will be necessary (“ASSE V”), which will be connected to the existing underground shafts and tunnels of the former mine, [2].

Every retrieval option shares the following key feature: To ensure personnel safety, remote controlled machinery and vehicles will conduct the main work within the chambers, [3]. These works include measures to improve the chambers’ structural integrity and the actual retrieval of the waste containers in the different chambers. The handling of drums resorts to different grippers that are attachable to the RC vehicles, [4]. Shielded containers allow the safe transport to the surface where further treatment of the legacy wastes takes place afterwards, [2].

Retrieval of waste in the exclusion zone – Case Study of Ukraine

Radioactive waste temporary localisation site (RWTLS) in the areas adjacent to Chernobyl Exclusion Zone (CEZ) were created during mitigation of the Chernobyl accident. They contain waste from decontamination of settlements (topsoil, roofing materials, construction waste, etc.), as well as waste from decontamination of equipment. A total of 53 RWTLS were created around the CEZ. Wastes localised in RWTLS belong to the categories of very low-level and low-level radioactive waste. The main radionuclides contained in these wastes are ^{90}Sr, ^{137}Cs, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am.

The RWTLS "Pisky-1" site was selected as a pilot facility for remediation. The overall storage area is estimated at approximately 124 m², containing an estimated waste volume of approx. 187 m³. The trench of RWTLS "Pitsky-1 was covered with a protective top layer of local sandy soil up to 0.6 m thick, but no engineered barriers were in place for the retention of radionuclides. Drainage ditches are arranged along the facility perimeter. A radiation survey of the RWTLS "Pisky-1" site established that the disposed radioactive waste consisted mainly of construction debris (roof tile and slate) and contaminated soil.

The main challenge was that Ukraine had not performed such works earlier, and therefore there was no methodological basis and well-trained personnel for that. It was also necessary to take into account special siting conditions and the lack of information on the characteristics of the RWTLS "Pisky-1".

Preliminary characterization of the site was provided using a gamma camera to identify hot spots located near the surface. Gamma dose rate mapping was also performed, and four wells were drilled to a depth of 2.5 m to allow sampling at 50 cm intervals. Those samples were analysed in a laboratory to confirm activity concentrations and detect possible contamination spread below the storage trench. Georadar and metal detector surveys were used to identify large objects as well as metallic objects and to confirm the boundary of the site. Groundwater monitoring was also undertaken to confirm assumptions around hydrogeology and for groundwater analysis.

An analysis of practical experience in setting criteria for remediation throughout Europe, Asia and the USA was undertaken, to support the setting of remediation criteria for RWTLS "Pisky-1". Examples of established detailed remediation criteria are:

- *PIMIC "Lenteja" by CIEMAT in Spain with 100 μSv/year,*
- *"l'Orme des Merisiers" in France remediated former waste storage site, and*
- *the American sites Hanford, Rocky Flats, Fort Dix, and Brookhaven with 150 μSv/year.*

The main dose criterion for remediation was established at 100 μSv/year for a person located in the area after remediation. The justification of the criterion took account of world experience in remediation of such sites, the level of contamination in the surrounding area and optimization of the volume of materials to be disposed. Based on this criterion and using conservative assumptions the remediation criterion for the specific activity of ^{137}Cs was established.

Radioactive waste removal was performed layer by layer, with each layer being around 25 cm thick. Prior to each layer removal, the surface was scanned to detect any radiological hot spots. Any identified hot spots were prioritized for removal and packaged separately.

Waste excavated from the trench was placed in large bags that were transferred to a buffer storage area for characterisation and sorting. Each bag was subject to gamma dose rate measurement. Packages with activities above the remediation criterion were defined as radioactive waste and transferred to a temporary storage facility. Waste bags below the remediation criterion were transferred to the site storage area for later use in trench backfilling.

Following the completion of the remediation activities, monitoring was performed, and activity concentrations were confirmed to be below the end-state criterion. The radiation characteristics of the RWTLS "Pisky-1" site after its remediation was:

- *Gamma radiation dose rate: 0.08 to 0.17 μSv/h;*
- *Beta-particles flux density: 1 to 3 β-particles per (min·cm²);*
- *Cs-137 activity in soil samples: below 0.11 Bq/g.*

5.3 Segregation of legacy waste

For unconditioned mixed legacy waste, segregation is commonly done by dose rate, e.g., in Ukraine in the scope of safe waste management prior disposal. Another approach is the physical segregation of materials based on the following treatment, e.g., by liquid/solids or organics/inorganics. Metals might be segregated into contaminated and activated metals. Contamination can be detected either via swipe samples or by detection of ^{137}Cs using gamma spectrometry. Activation can be assumed if ^{60}Co is

Segregation by physical and chemical properties – Case Study of Bulgaria

The integration approach for management of the RAW covers a wide spectrum of different types and categories of radioactive waste. A particularly important place in this spectrum occupy the legacy waste at least due two reasons: First, legacy wastes exist in almost all European countries because they result from the operation not only of nuclear facilities in the so-called nuclear countries having nuclear power plants. They are also common in other countries, some of which have research nuclear reactors, and in almost all countries because of the use of radioactive sources in medicine, industry, agriculture, etc. Second, legacy waste in the general case is very different as types and mixes of partially or completely decomposed biodegradable waste, plastic wastes, textiles, metals, glass and other components, generated for years or decades, and dumped in designated locations (in some cases uncontrolled, even forgotten).

This case study considers mainly the legacy wastes related to the operation of NPPs and/or decommissioning of these. As characteristic basic legacy waste, for example related with VVER-1000 could be listed: Textiles (contaminated work clothes, etc.), metals, construction waste, wadding, Rubber, polymers, shavings, wood, paper, other wastes, and mixed wastes.

The approach for segregation by physical and chemical properties of the legacy waste identified in Bulgaria is based on the classification "radioactively contaminated and uncontaminated", "primary and secondary", and "solid and liquid".

Concerning radioactivity, the existing legacy waste could be categorised applying the classification of the IAEA Guide "Classification of Radioactive Waste" [7]. By this, the existing legacy waste can be classified as exempt waste, very short-lived waste, or very low-level waste.

The Council of Ministers of Bulgaria has approved a regulation for the safe management of radioactive waste in 2013, which is in accordance with the IAEA Safety Guide "Classification of Radioactive Waste" GSG-1. The Bulgarian regulation uses the following classification for RAW:

1) Category 1 - with low activity, not in need of measures for radiation protection and high level of isolation and containment. Category 1 is sub-divided into:

• Category 1a – Waste that meets the levels for release from regulatory control under the Act on the Safe Use of Nuclear Energy (ASUNE) of Bulgaria.

• Category 1b – Very short-lived waste containing mainly radionuclides with short half-life (not more than 100 days), whose activity decreases below the levels for release from regulatory control as a result of appropriate storage on site for a limited period of time (usually not more than several years).

• Category 1c – Very low-level waste with levels of specific activity exceeding by a minimal value the levels for release from regulatory control under the ASUNE and with a very low content of long-lived radionuclides, which represent a limited radiological risk. For this category of waste, the application of specific measures for radiation protection or for isolation and containment is not required.

2) Category 2 – Low-level waste and intermediate level waste require reliable isolation and retention, but do not generate heat. Category 2 is sub-divided into:

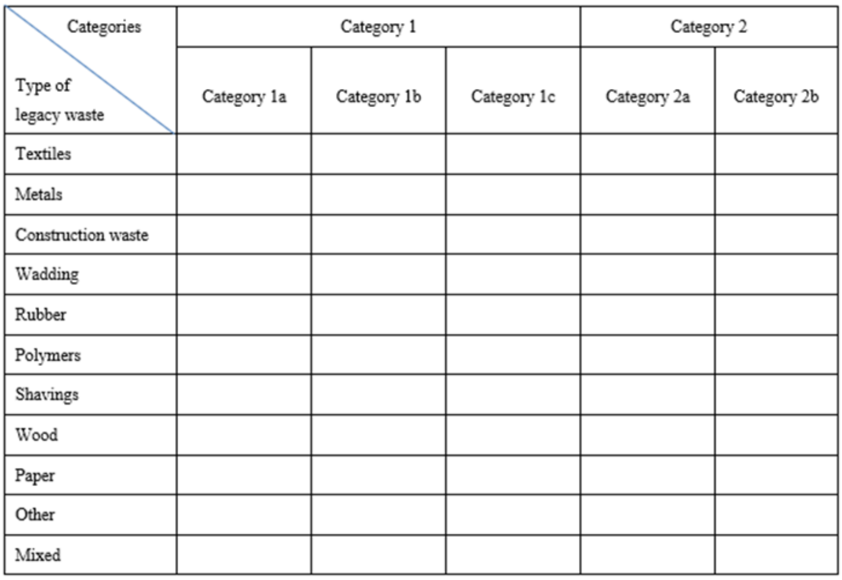
• Category 2a – Low- and intermediate level waste containing mainly short-lived radionuclides (with a half-life not longer than that of Cesium-137) as well as long-lived radionuclides at significantly lower levels of activity, limited for the long-lived alpha- emitters at 4.106 kBq/kg for each individual package and a maximum average value for all packages in the respective facility of 4.105 kBq/kg. For this category of radioactive waste a reliable isolation and containment is required for up to several hundred years.

• Category 2b – Low- and intermediate level waste containing long-lived radionuclides at activity levels of long-lived alpha emitters, exceeding the limits of category 2a.

3) Category 3 – High-level waste, which needs isolation and containment, as well as heat removal during storage and disposal.

In practice, legacy wastes in Bulgaria are in different sub-categories mainly of Category 1, in some cases in Category 2.

The following segregation approach for the legacy waste is recommended in Bulgaria (Table 4):



Categories / Type of legacy waste	Category 1			Category 2	
	Category 1a	Category 1b	Category 1c	Category 2a	Category 2b
Textiles					
Metals					
Construction waste					
Wadding					
Rubber					
Polymers					
Shavings					
Wood					
Paper					
Other					
Mixed					

Table 4 – Segregation approach for legacy waste. The segregated legacy waste from each type/ category should be stored in separate barrels for further management. Regarding the segregation of legacy waste, specific and significant challenges are:

- *The definition and prioritisation of segregation criteria, valid for the long-term management and acceptable in the different European countries. One particular challenge is the prioritisation of waste properties to be determined, i.e., the radiological, toxic, chemical or physical properties;*
- *Maintaining an up-to-date inventory of the segregated legacy waste, considering the time behaviour of the physical, chemical, radiological and other properties;*
- *Identification of difficult-to-measure radionuclides in the legacy waste;*
- *Timely characterisation of legacy waste, e.g., on producer's site.*

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detected and contamination can be excluded. The application of neutron spectrometry might be useful depending on legacy waste.

Within the case study provided by France, the segregation approach of UNGG fuel elements, uranium metal rods with magnesium and graphite cladding, from the La Hague reprocessing plant UP2-400 is presented.

A guidance on how the wastes from the operation and decommissioning of NPPs are segregated by physical and chemical properties in Bulgaria is given. A second case study is provided by Germany, on how large amounts of contaminated soil is sorted by dose rate, to enable the separation soil feasible for exemption.

Segregation of mixed conditioned legacy waste might not always be applicable and comes in general with high costs. After being examined (x-ray tomography, gamma and neutron spectrometry, etc.), some conditioned solid waste can be "deconditioned" in dedicated facilities, e.g., in France. In general, the segregation is supported by gamma spectrometry and neutron spectrometry, if applicable. For large amounts of waste, the application of scaling factors (SF) might be useful.

An example for challenging segregation of liquid organic legacy waste is liquid Dowtherm (an organic heat transfer fluid) mixed with inorganic wastes. A general method used in this case is decanting.

For unconditioned solid mixed legacy wastes, segregation is done in general by physical segregation (e.g., segregation of graphite in Slovakia or other decommissioning wastes). For liquid mixed wastes, segregation can be done by mass balance (e.g., segregation of sludge in Spain), decanting (e.g., segregation of potassium dichromate in Slovakia) or segregation of organic and inorganic liquids.

Segregation of UNGG fuel elements and magnesium characterization – Case Study of France

The former UP2-400 reprocessing facility began operations in 1966 and has been permanently closed since January 1, 2004. The four INBs are now engaged in operations involving the recovery and conditioning of old waste (RCD) as well as dismantling. UP2-400 has dry storage silos for structural waste from first-generation reactor defueling (UNGG). This waste held is mostly structural waste from UNGG fuel refining (Chinon 1, 2 and 3, St Laurent 1 and 2, Bugey and Vandellos). A UNGG fuel consists of a uranium metal rod encircled by a magnesium cladding and fitted with two magnesium pins, all of which are put into a graphite cladding. The assembly is supported radially by magnesium centering pins and axially by a graphite saddle linked to the sleeve by stainless steel saddle wires at one end.

Orano has deployed a sorting system in a shielded cell using a 3D localization system, numerous magnesium counting cameras and a mechanical automated sorting system. This system was designed to optimize the packaging while controlling the pyrophoricity and radiolysis risk. Large waste is recovered using a grapple, with segregation of the wastes (UNGG waste, aluminium waste, and small granulometry waste) and magnesium quantification. Each "bundle" of waste is subjected to a magnesium measurement. A pattern recognition system installed on the conveyors identifies and quantifies the magnesium objects among the other waste. The device allows to control in real time the magnesium surface.

Segregation of contaminated soil by dose rate – Case Study of Germany

Throughout the process of the deconstruction or the decommissioning of nuclear facilities, large amounts of possibly contaminated soil can occur. In these cases, the examination of large quantities of bulk materials is necessary and as a result, an efficient and secure way of separating contaminated soil from non-hazardous soil is therefore favourable. For example in Germany, the need to analyse large amounts of salt during the retrieval of radioactive wastes from the former potash and rock salt mine ASSE II [7] led to the development of a measurement device that serves now for the analysis of large quantities of homogeneous bulk materials [8]. These bulk materials can differ in terms of consistency (loose sands, grit, soil, rocks and stones), chemical structure (silica-based minerals, organic soils, salt, rubble and deconstruction debris) and possible presence of radioactive nuclides [8].

For the analysis, large piles of bulk material have to be transformed into one continuous material stream, which is subsequently distributed onto a conveyor belt [7]. A continuous surveillance of this material stream with different measurement equipment (gamma spectrometry, α- and β-detection) is then possible [8]. Thus, the material is evaluated "on line", whilst moving along with the conveyor belt. If a particular portion of material on the belt exceeds the defined maximum values of contamination (dosage, radiation, etc.), control mechanisms can separate this part of the material flow, [7]. For example, by transferring it to another conveyor belt which transports the contaminated bulk material to a specially shielded container. Retesting the contaminated material allows to determine the degree of contamination [7] (e.g. the bulk material could be characterized as LLW or ILW). This determination serves as a basis for the decision on how to dispose of the material correctly.

If the remaining material proofs inconspicuous throughout the series of measurement and detection stations, the conveyor belt system piles up this uncontaminated material in another spot, (1). The further treatment of this material as e.g. non-hazardous soil or backfill material in underground applications is then possible.

This procedure allows the fast examination of large quantities with a high reliability. In the wake of a decommissioning project of a former nuclear facility in Belgium, the system has proven its functionality and reliability with a throughput of up to 100 tons of soil per hour [9].

5.4 Sampling of legacy waste

One general difference between sampling of legacy and non-legacy waste that should be highlighted is that for sampling of legacy wastes more protective measures need to be taken. Additionally, if the segregated legacy waste is heterogeneous, more samples need to be taken to allow a representative characterisation of the waste. The inclusion of statistical techniques, such as reported in D3.7 of the INSIDER project, should be considered [6].

Common sampling methods for unconditioned metallic legacy wastes are swipe tests, as well as solid samples by drilling.

For unconditioned graphite waste, solid samples are generally taken by drilling. The sampling process for graphite is challenging, if samples need to be taken from the reactor core prior to the decommissioning of the reactor. Additionally, if no information is available about the "position" of the graphite in the reactor (after decommissioning of a reactor), more samples need to be taken to enable a reliable characterisation.

For unconditioned sludge the samples are liquid. Special care must be taken to ensure representative sampling if sludges are heterogeneous, e.g., due to sedimentation.

Sampling of unconditioned disused sealed radioactive sources (DSRS) generally consists of swipe tests. For aged sources the access to the source itself might become challenging due to corrosion effects on the shielding.

Solid samples of unconditioned naturally occurring radioactive materials (NORM) is generally taken by drilling (e.g., phosphogypsum, PG). In case of PG, representative sampling might be hindered due to a change of the radionuclide inventory based on rainwater leaching and cavities in a PG stack. A case study by Cyprus presents the approach for the sampling by drilling and sampling of stack fluids, as well as chemical and radioanalytical characterisation of PG in stacks.

For unconditioned solid wastes, samples can be dissolved to enable a wide range of radioanalytical methods. In case of conditioned (solid) waste, samples in general must be taken by drilling or stripping of material.

Sampling and characterisation of phosphogypsum in stacks – Case Study of Cyprus

The phosphogypsum (PG) waste in Cyprus originated from the production of fertilizer in the late 80's until the early 90's by an industrial complex located at a coastal area in the southern part of the island. The PG produced (~300 000 tons) as a by-product was disposed in a lagoon excavated adjacent to the sea. This disposal area (~50 000 sqm) is divided in three sub-areas (i.e., the one in front of the sea, the second with the old/dried PG and the third with the lastly disposed PG).

In the frame of an environmental impact assessment, PG sampling was carried out by drilling down to five meters and included all three sub-areas. Samples were obtained at certain depths and where possible, stack fluids were collected from the cavities formed below the PG body.

The PG characterisation included the analysis of the chemical composition and the radiology utilizing:

- *Inductively coupled plasma optical emission spectroscopy (ICP-OES) after dissolution,*
- *Carbon/Sulphur analysis,*
- *X-ray diffraction (XRD) and x-ray fluorescence (XRF) analysis),*
- *Gamma dose survey, radionuclide levels by gamma-spectroscopy (HPGe),*
- *Radon emanation measurements,*
- *Liquid scintillation counter (LSC) and*
- *Alpha-spectroscopy using passivated implanted planar silicon (PIPS) detectors of the alpha radionuclide content in seawater samples, in stack fluids and in the collected PG samples (after total PG dissolution).*

In the meantime, the disposal site has been covered with a plastic line soil and vegetation. Future studies should focus on the long-term chemical stability of PG under the given conditions, the erosion of the PG front facing the sea and the evolution of the cavities formed below the PG body.

5.5 Characterisation of legacy waste

As highlighted in [6], numerous member states are having difficulties managing historic waste, which comprises both conditioned and unconditioned waste. The challenges originate mostly from categorization issues, since most nations must manage past radioactive waste without understanding its origin or radionuclide content, and in certain cases, the waste streams have been intermingled. These uncertainties may be related to the indirect quantification of specific radionuclides (e.g., ^{14}C in graphite waste) or to techniques for detecting specific species such as activation products or complexing substances, but one of the main areas of concern is the strategy to be implemented to recover unconditioned waste when its characterization is uncertain.

Independent of the specific legacy waste type, it should be highlighted that the general equipment used for characterisation is not always applicable, due to the unknown radionuclide inventory of legacy wastes. This unknown also makes a step-by-step approach necessary, and in general more single steps are necessary for a reliable characterisation of the legacy waste. study of historical information might pose helpful to determine the waste origin and specific characteristics, but the possibility of legacy waste being mixed wastes from different facilities should not be excluded.

For the characterisation of unconditioned organic legacy waste, both liquid and solid, in general the same characterisation approaches as for non-legacy radioactive wastes are applicable. The same applies to unconditioned disused sealed radioactive sources (DSRS). Special care needs to be taken during handling when characterising high activity organic wastes.

During the characterisation of unconditioned metallic wastes, the scaling factor methodology has only a very limited applicability, as most of the legacy waste arose from multiple origins and waste streams.

Current issues for the characterisation of metallic legacy waste are the unknown quantity of radionuclides and the lack of an adequate characterisation methodology.

For most unconditioned graphite legacy waste some historical information is available, e.g., the reactor history, facilitating the characterisation process for this waste. One option to further optimise characterisation are neutron activation simulations (e.g., before decommissioning of legacy sites).

Characterisation, treatment, and conditioning of packages containing sludge – Case Study of Spain

The legacy waste currently stored at "El Cabril" repository, inter alia consisting of 37 packages containing sludge in asphalt, will be managed by Enresa, the Spanish WMO, following the below described procedure:

1) Localisation of the packages in the warehouses in order to determine their viability of accessibility for the different tasks to be performed;

2) Identification of waste package status:

- *220 litre package introduced in 400 litre drum without concreting or,*
- *220 litre package reconditioned in 400 litre drum already concreted;*

3) Gamma spectrometry measurements for all packages independent of their status (introduced and/or reconditioned);

4) Classification of the waste packages as very low-level radioactive waste (VLLW) or low- and intermediate level radioactive waste (LILW), depending on the measurement result;

5a) Further management route for VLLW:

- *Packages identified as VLLW and which have not been reconditioned yet, i.e., 220 litre package introduced in 400 litre drum without concreting, will be transported to "El Cabril" laboratories for extraction of specimens and subsequent leaching tests for chemical products.*
- *If specimens cannot be extracted due to the nature of the bitumen/asphalt, a material sample is taken for granular residue testing.*
- *If the waste is identified as toxic or hazardous, the 220 litre drums are concreted in the 400 litre overpack drum.*
- *If the waste is non-toxic and non-hazardous, the 220 litre drum will be removed from the 400 litres overpack drum, provided that the 220 litre drum is physically intact, and directly broad to the disposal site for VLLW.*
- *The packages of 220 litres already reconditioned, i.e., concreted in the 400 litre overpack drum, will be stored directly in VLLW cells.*

5b) Further management route for LILW: LILW packages are considered as non-compliant packages for disposal at existing sites. LILW packages already reconditioned, i.e. packages concreted in a 400 litre overpack drum, are put into storage in a shielded unit. Unconditioned 220 litres, i.e., drum introduced but not concreted in a 400 litre overpack drum, will be extracted from the 400 litre overpack drum, provided that the 220 litre drum is physically intact, prior to their introduction into the storage unit.

6) Prior to the storage of drums independent of their specification, the corresponding acceptance documentation for storage is updated.

The characterisation of legacy, unconditioned chemicals, sludges and spent ion exchange resins (SIER) is challenging, as general characterisation methods might not be applicable. Additional challenges arise during the handling of aggressive, highly contaminated liquids (e.g., highly contaminated acids from chromic-acid-baths) and the chemical characterisation of legacy sludges from water treatment facilities. For all these three unconditioned legacy waste types, WAC might not be defined yet and there is a lack of knowledge and practice.

The characterisation of unconditioned naturally occurring radioactive material (NORM) depends highly on the specific type of material. As an example, the characterisation of phosphogypsum is based on the determination of Radium, Thorium, and the Radon emanation of the material. Additional chemical characterisation is necessary due to the possibility of other toxic chemicals in phosphogypsum.

A special type of legacy waste is unconditioned waste from nuclear accidents. One challenge might be a large quantity of waste, such as in Slovakia with approx. 69,000 m³ of contaminated soil at the NPP A1 site. Additionally, high levels of contamination (including alpha contamination) should be expected. Lastly, mixed legacy waste originating from nuclear accidents presents a challenge. A case study presenting the national approach on legacy waste characterisation for different waste types has been prepared by Portugal.

Planning of legacy waste characterisation – Case Study of Portugal

Instituto Superior Técnico (IST), the School of Engineering of the University of Lisboa, operated a 1 MV pool-type research reactor since the 1960s at its Campus Tecnológico e Nuclear. The research reactor was shut down in 2016 and the irradiated spent fuel shipped to the USA in 2019 in the framework of a MoU between Portugal, USA and IAEA. At the same Campus, the Radiation Protection and Safety Laboratory of IST also operated since 1960 the only radioactive waste storage facility of Portugal, a surface type facility dedicated to the storage of very low and low-level waste (VLLW, LLW) as well as small amounts of intermediate level waste (ILW). The radioactive waste in this storage facility is classified as very low, low- and intermediate level waste type (VLLW, LLW and ILW). The radioactive waste generated in Portugal consists mainly of materials from past Uranium and Radium mining and milling activities, spent and/or disused sealed radioactive sources, smoke detectors, lightning rods, contaminated scrap metal, depleted Uranium from aircraft counterweights, materials contaminated with unsealed sources produced from the applications of ionizing radiation in the fields of medicine, industry and research.

Following the publication of Decree-Law n. 156/2013, of November 5th, that transposes the European Council Directive 2011/70/EURATOM, IST prepares an inventory of the RW stored at its facility and submits to the Regulator annually. Records registered after the year 2000 are reliable but there is a strong uncertainty relative to the waste collected before, considered as legacy waste that requires characterization and classification.

Clearance and exclusion levels as set by the European Council Directive 2013/59/Euratom (and General Safety Requirements Part 3) were adopted by the Portuguese legislation as Ministerial Order n. 44/2015 presently superseded by Ministerial Order n. 138/2019.

A first National Programme for the Management of Spent Fuel and Radioactive Waste (2015-2019) was approved by the Government and published in the official journal as the Resolution of the Council of Ministers n. 122/2017, on September 7th, following the strategic environmental assessment and public consultation. More recently, a second Programme was prepared and published in the official journal as well as Resolution of the Council of Ministers n. 129/2022, on December 20th.

The national programme specifically considers the following activities: the characterisation and identification of legacy waste; eventual preparation of clearance processes for waste that no longer require storage at such a facility; restoration of enough room for waste of concern and corresponding available volume for storage and the improvement of the overall inventory of radioactive waste.

IST is motivated to perform the characterisation of the legacy wastes stored at its facility for which there are no records (or records could not be found), proceed to treatment, and optimize the available volume of the facility for storage of radioactive waste.

The characterisation of legacy radioactive waste at IST will depend on the waste type inside the 220 litre drums and consists mainly of dose rate measurement (at contact and at 1 m distance), the assessment of the activity concentration by gamma spectroscopy for gamma emitters, and swipe test analyses by liquid scintillation counting (LSC) for alpha and beta emitters. The characterisation will start in the near future and following the characterisation process, it is expected that some of the material will be cleared by the regulator.

For the handling and characterisation of conditioned legacy waste, the applied processes differ by country. France has identified a problematic interaction between waste and matrix in some cases, resulting in challenges for the encapsulation of legacy waste (STE2 sludge conditioning is under development, the matrix considered are bituminizing, cementing or calcination/vitrification). Depending on the legacy waste, Germany utilises technologies such as active neutron interrogation or tomography to identify conditioned legacy waste inventories. In Slovakia, conditioned legacy wastes are inserted into a special fibre concrete (concrete reinforced with steel fibres). Containers devoted to disposal are measured by means of gamma scanners. The stepwise approach of Spain in characterising legacy sludge conditioned in asphalt matrices is presented in a case study.

5.6 Treatment & conditioning of legacy waste

The treatment and conditioning strategies for legacy wastes depend significantly on the national legislation for interim storage and final disposal (e.g., WAC). Therefore, this section only highlights selected examples and challenges of legacy waste treatment.

A common treatment procedure for legacy unconditioned organic liquid wastes is the incineration prior to disposal. One challenge of the incineration of waste is the risk of exceeding legal activity limitations of facilities after treatment due to the activity concentration during the processing. A case study on the treatment and conditioning of Dowtherm, a synthetic organic heat transfer fluid (polychlorinated biphenyls) designed for liquid phase, is presented by Slovakia.

At the La Hague site in France, unconditioned graphite legacy waste is cut and stored below water to prevent the risk of magnesium self-ignition of the magnesium and graphite cladding of fuel elements. The future encapsulation of the graphite legacy waste presents a challenge.

For legacy unconditioned sludge, the mixture of sludges may pose a risk due to chemical reactions (incl. exothermic reactions) based on difficult chemical compositions of sludges in the waste.

Treatment of DOWTHERM – Case Study of Slovakia

A special and very problematic type of radioactive legacy waste is the organic heat transfer fluid Dowtherm, which was used at the NPP A1 in Jaslovké Bohunice, Slovakia. This power plant was in operation during the period from 1972 to 1977. The A1 NPP, a gas-cooled, heavy water moderated reactor, was shut down after an accident, taking place in 1977, when the fuel in the core of the reactor was significantly damaged and fission nuclides were spread to the primary circuit and partly to the secondary circuit. The material Dowtherm was developed and used for the cooling and storage of the spent fuel elements (metallic natural uranium). From the chemical point of view, it is a eutectic mixture of diphenyl and diphenyloxide and was designed to prevent corrosion of spent fuel elements.

The treatment of this kind of organic liquid RAW is based on an incineration procedure in a specialised facility at temperatures from 750 – 950°C, followed by an afterburning at temperatures of 1100 °C in the post-combustion chamber. In the following, the ash arising from the incineration process is mixed with paraffin, encapsulated into 200-liter steel barrels, and inserted in a fibre-reinforced container to produce waste package approved for near-surface disposal.

Although the most challenging type of liquid RAW coming from NPP A1 is Chrompik (an aqueous solution of potassium chromate and dichromate with specific activities of up to 1 GBq/dm³), DOWTHERM treatment (incineration) was a real technological challenge for about 20 years.

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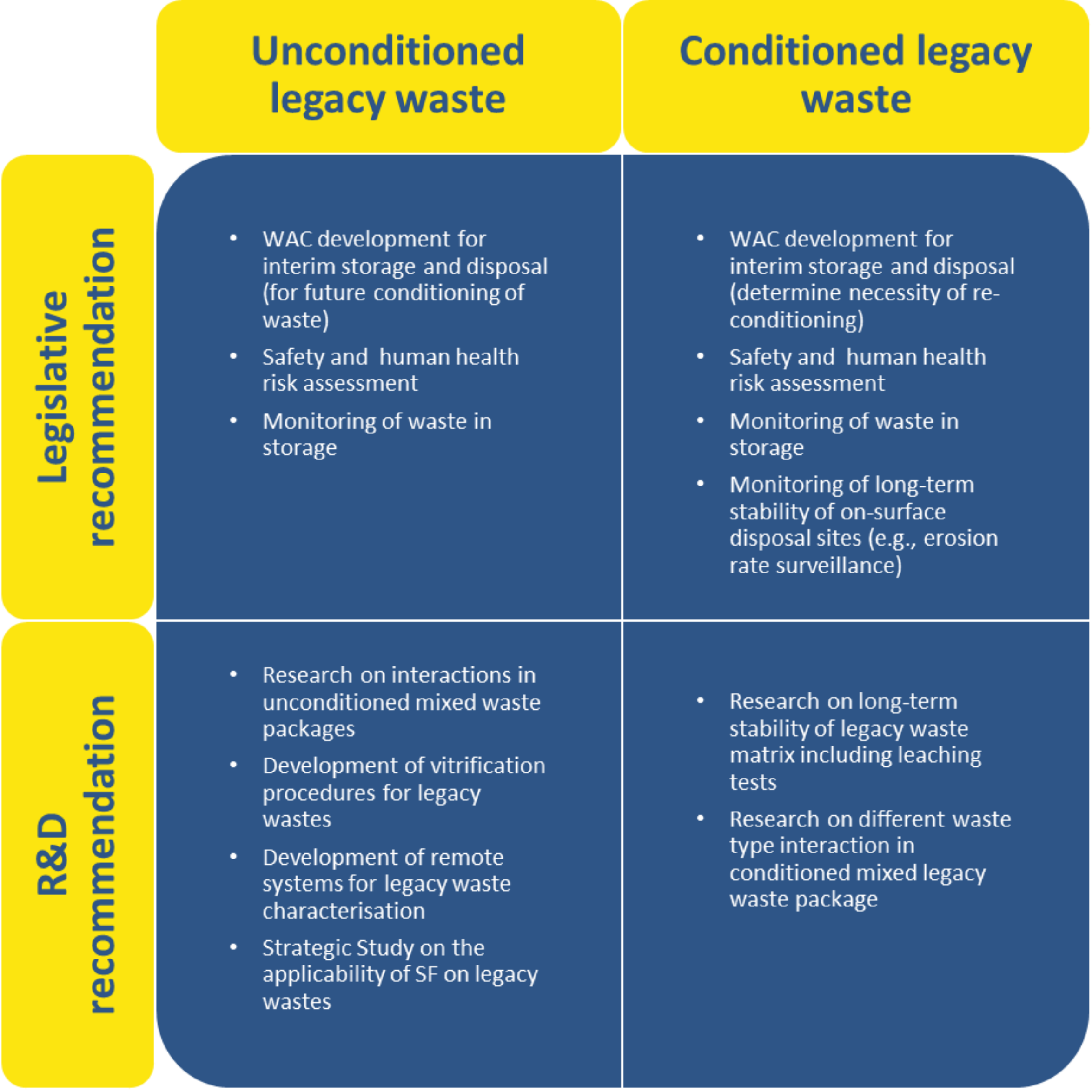
6. Recommendations

Based on the identified management issues for different types of legacy waste, R&D and legislative recommendations have been identified.

As there are multiple overlaps of the waste type specific issues, the recommendations are structured by the legacy waste status, i.e., unconditioned and conditioned legacy waste and differ between legislative/organisational recommendations and R&D (technological) recommendations. These are shown in *Figure 2*.



	Unconditioned legacy waste	Conditioned legacy waste
Legislative recommendation	• WAC development for interim storage and disposal (for future conditioning of waste) • Safety and human health risk assessment • Monitoring of waste in storage	• WAC development for interim storage and disposal (determine necessity of re-conditioning) • Safety and human health risk assessment • Monitoring of waste in storage • Monitoring of long-term stability of on-surface disposal sites (e.g., erosion rate surveillance)
R&D recommendation	• Research on interactions in unconditioned mixed waste packages • Development of vitrification procedures for legacy wastes • Development of remote systems for legacy waste characterisation • Strategic Study on the applicability of SF on legacy wastes	• Research on long-term stability of legacy waste matrix including leaching tests • Research on different waste type interaction in conditioned mixed legacy waste package

Figure 2 – Identified recommendations for legacy waste management

In terms of legislation, recommendations were developed for both conditioned and unconditioned legacy waste, relating to waste acceptance criteria (WAC), safety and human health issues, and waste monitoring. Both for the unconditioned and conditioned legacy waste management, it should be highlighted, that waste acceptance criteria (or generic WAC) need to be developed to ensure targeted characterisation, treatment, and conditioning (or re-conditioning, if applicable) of legacy wastes. Additionally, safety and risk assessments in legacy waste management including handling and storage, should be promoted. Lastly, both for unconditioned and conditioned legacy waste the legislative recommendation on legacy waste monitoring in storage, as well as for legacy sites has been raised.

Regarding the latter, the long-term stability of on-surface disposal sites with respect to the degradation and erosion of the covering material should be monitored.

Several pathways have been recommended for R&D needs, depending on whether the waste is in unconditioned or conditioned form. R&D recommendations for unconditioned legacy waste are:

- Research on interactions in unconditioned mixed waste packages;
- Research on vitrification procedures for legacy wastes;
- Development of remote systems for legacy waste characterisation, especially for high activity legacy waste;
- Strategic study on the applicability of scaling factors (SF), a methodology exploiting known ratios of DTM (Difficult-to-Measure) and ETM (Easy-to-Measure) radionuclides to facilitate characterisation, on legacy waste characterisation;

R&D recommendations for conditioned legacy waste management are:

- Research on long-term stability of existing legacy waste matrices, including leaching tests, with the aim of determining the necessity of re-conditioning;
- Research on different waste type interaction in conditioned mixed legacy waste packages.

Lastly, waste type specific gaps and associated research and development recommendations developed within the frame of this project are summarised in *Table 4*.

Legacy waste type	**Gap**	**R&D recommendation**	**Priority**	**Comment**
Organics	Management of Liquids	Further R&D on incineration / thermal treatment of organic liquids, especially with high amount of RN	Medium – High	
		R&D on innovative (polymer) matrices with regard to long-term safety	High	R&D on-going
Graphite	Magnesium content in graphite	Conditioning matrix for graphite with magnesium	Medium – High	R&D on-going
	Lack of conditioning process	R&D on (polymer) matrices	Medium – High	R&D on-going
Sludge	Chemical reactions (e.g., exothermic) during handling and storage/disposal	R&D on (polymer) matrices	Medium	
DSRS	(Re-)Characterisation of conditioned DSRS	Strategic Study (StSt) on best equipment for the NDA of conditioned wastes	Low – Medium	
NORM	High amounts of phosphogypsum NORM waste	R&D for treatment with aim to reuse of material, e.g., in cement industry	Medium – High	R&D on-going
Mixed	Chemical reactions	Research on waste interaction	Medium – High	
		Research on decontamination of wastes	High	

Table 4 – Waste type specific R&D commendations for the legacy waste management

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