Deliverable 6.1: Review of treatment and conditioning processes and materials available or under development for challenging wastes

Work Package STREAM

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Authors	**Elena Torres Álvarez (CIEMAT), Michael Gracey, Mark Cowper (NWS), Quoc Tri Phung (SCK CEN), Borys Zlobenko (SIIEG·NASU), Federica Pancotti, Mafalda Guerra (SOGIN), Märten Kala, Janno Torop, Alan Tkaczyk (UTartu)**

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Executive Summary

This State-of-the-Art report provides a general analysis of the existing commercial or under-development technologies for the management of several types of common challenging wastes, in order to identify the R&D needs to be addressed within the framework of STREAM Work package (WP).

The purpose of this document is to stablish a baseline for the three technical tasks (task 3, 4 and 5) to avoid overlapping with previous projects and to identify the remaining challenges and uncertainties concerning the different management strategies studied in this work package.

This SotA is divided in seven main sections.

1. Scope of WP STREAM
2. Challenges in the treatment and conditioning of problematic waste streams
3. Waste treatment and conditioning processes in use
4. Needs of optimization of treatments and conditioning matrices under development
5. Scaling-up and industrial implementation of new treatments and conditioning processes
6. Challenges of new treatment and conditioning technologies
7. Gaps to be addressed in STREAM WP

Section 1 contextualizes the scope of STREAM within the EURAD roadmap and provides an overview of the technical tasks of the work package.

Section 2 summarizes the current problems faced in the management of certain challenging waste streams, including classification, treatment, conditioning and disposability aspects, as well as outlining the importance of compliance with existing Waste Acceptance Criteria (WAC).

Section 3 briefs about the current treatment and conditioning processes in use for waste management, highlighting international good practices and giving an overview of the available commercial treatments and conventional cementation systems in use.

Section 4 focuses on the needs of optimization of treatments and conditioning matrices under development. This chapter aims to provide a baseline for task 3, by detailing existing treatments and conditioning materials both, commercially available and under development. In this section, an analysis of the advantages and disadvantages of new methods over traditional processes is done as well as an assessment of the suitability for the most common challenging waste streams.

Section 5 deals with the scaling-up and the industrial implementation of the new treatments and conditioning processes. This chapter briefly describes the key aspects of the upscaling process based on the previous industry, Waste Management Organizations (WMOs) and waste producers' experiences.

Section 6 is focused on the challenges and remaining uncertainties that needs to be faced in the current context regarding treatment and conditioning of challenging wastes.

Finally, section 7 provides a summary of the knowledge gaps to be tackled in STREAM, based on the review of the EURAD and PREDIS Strategic Research Agendas (SRAs) and aims to correlate them with the proposed experimental work and the STREAM task where is going to be addressed.

Keywords

Treatment, conditioning, decontamination, low-carbon binders, scaling-up, disposability assessment

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Glossary

AACs	Alkali Activated Cements
AE	Accoustic Emission
ASR	Alkali-Silica Reaction
BFS	Blast Furnace Slag
CAC	Calcium Aluminate Cement
CBHs	Calcium Borate Hydrates
CBPCs	Chemically-Bonded Phosphate Ceramics
CFB	Circulating Fluidized Bed
COD	Chemical Organic Demand
COREMIX	Chemical Oxidation REduction using nitric permanganate and oxalic acid MIXture
CPC	Calcium Phosphate Cement
CRP	Coordinated Research Project
CSAC	Calcium Sulphate Aluminate Cement
D&D	Decontamination & Decommissioning
DEFAC	DEmilitarization FACility
DI	Domain Insight
DOE	US Department of Energy
DT	Digital Twins
DVB	Divinylbenzene
EBS	Engineered Barrier System
EDTA	Ethylenediaminetetraacetic acid
ESC	Environmental Safety caseCase
EU	European Union
FBSR	Fluidized Bed Steam Reforming
GAO	US Government Accountability Office
IAEA	International Atomic Energy Agency
LALNL	Los Alamos National Laboratory

LC^3	Limestone Calcined Clay Cement
LCA	Life Cycle Assessment
LCC	Life Cycle Cost
LILW	Low- and Intermediate Level Waste
LSC	Liquid Scintilliation Cocktails
MK	Metakaolin
MKPC	Magnesium Potassium Phosphate Cements
MPC	Magnesium Phosphate Cements
MS	Member States
MSH	Magnesium Silicate Hydrates
MSO	Molten Salts Oxidation
Mu-Tom	Muon Tomography
NDA	Non Destructive Analysis
NDT	Non Destructive Techniques
NEA	Nuclear Energy Agency
NWMS	Naval Surface Warfare Center
OPC	Ordinary Portland Cement
OSTI	U.S. Department of Energy Office of Scientific and Technical Information
PA	Performance Assessment
PS	PolyStyren
QMS	Quality Management System
R&D	Research and Development
RFID	Radio Frequency Identification
RLOW	Radioactive Liquid Organic Wastes
RSOW	Radioactive Solid Organic Wastes
RW	Radioactive Waste
S/S	Stabilization/Solidification
SciFi	Scintilliating Optical Fibre
SGS	Segmented Gamma Scanner

SIER	Spent Ion Exchange Resins
SiLiF	Silicon Lithium Fluoride
SMR	Small Modular Reactor
SotA	State-of-the-Art
SRA	Strategic Research Agenda
TBP	TriButyl Phosphate
TGS	Tomographic Gamma Scanning
TRL	Technology Readiness Levels
TRU	Transuranic
UV	Ultra Violet
VIM	Vacuum Induction Melting
WAC	Waste Acceptance Criteria
WAS	Waste Acceptance System
WF	Waste Form
WMO	Waste Management Organisation
WP	Work Package

European Commission (EC) Project Acronyms

CHANCE	Characterization of conditioned nuclear waste for its safe disposal in Europe (EC Project 2017-2022)
CORI	Cement-Organic-Radionuclide Interactions (Work Package of EURAD-1)
EURAD	The European Joint Programme on Radioactive Waste Management
PREDIS	The Predisposal management of Radioactive Waste
ROUTES	Waste Management Routes in Europe from Cradle to Grave
THERAMIN	Thermal treatment for radioactive waste minimization and hazard reduction

EURAD-2 Work packages Acronyms

ASTRA	Alternative Waste Management Strategies
FORSAFF	Waste management for SMRs and future fuels (FORSAFF)

ICARUS	Innovative Characterization Techniques for Large Volumes
L'OPERA	Long-Term PERformance of waste matrices
SUDOKU	Near-SUrface Disposal Optimization based on Knowledge and Understanding

1. Scope of WP STREAM

Work package STREAM (**S**ustainable **TREA**tment and I**M**mobilisation of Challenging Wastes), aims to develop new safe and sustainable management routes for waste streams without any previously identified or industrially-implemented treatment or conditioning route.

The topics addressed in the STREAM WP are identified in the EURAD Roadmap [1] under Theme 2 *Predisposal* and Theme 3 *Engineered Barrier Systems,* and more specifically within the Domains:

2.1.2. Identify parameters and metrics for waste acceptance criteria through whole life cycle (Waste Acceptance Criteria)

2.1.3. Assess potential technologies for the implementation phase, considering cost-benefit ratio and availability (Technology Selection)

2.2.2. Minimise the quantity and volume of radioactive waste through pre-treatment and treatment

2.2.3. Stabilise waste by conditioning prior to long-term storage (Conditioning)

3.1.3. Cemented LL-ILW (Cemented LL-ILW)

3.3.2. Backfill component under storage and disposal conditions (Backfills)

Scope of STREAM WP includes the design, optimization and industrial implementation of novel treatment and conditioning methods for most widespread problematic waste streams (Spent Ion Exchange Resins (SIERs), Radioactive Liquid Organic Waste (RLOW), sludges, evaporator concentrates, metallic wastes) [2]. This WP intends to expand the experience gained in past EU projects such as, PREDIS, ROUTES or THERAMIN, and will also benefit from other previous and ongoing international activities related to predisposal topics, like the IAEA CRP on *Geopolymers as an Immobilization Matrix for Radioactive Waste.*

Within EURAD-2, interaction with other WPs such as ICARUS, FORSAFF or ASTRA, for specific issues related to new management routes would be desirable, as well as with SUDOKU in disposability assessment-related aspects. Collaboration with WP L'OPERA has already been stablished and will include the delivery of samples and sharing of experimental data.

This work package is organized in 5 tasks (Figure 1), including three technical tasks:

- Task 1: Management/coordination of the work package
- Task 2: Knowledge Management
- Task 3: Study of treatment and conditioning methods
- Task 4: Scaling-up of treatment and conditioning methods
- Task 5: Deploying safe solutions achieving cost and environmental performances following the principles of circular economy

The main aim of task 1 is the overall management of the WP including scientific-technical coordination, monitoring and reviewing the WP progress. Task 2 covers knowledge management topics, including knowledge capture relevant for the SRA topic of this WP and knowledge transfer to the EURAD-2 community and beyond through the EURAD-2 KM programme.

Regarding the technical tasks, task 3 deals with the development and optimization of new treatments for the minimization of waste volume and secondary waste streams, including decontamination methodologies and thermal and Fenton-based treatments. Optimization of novel matrices, such as MKPC, geopolymers or low-carbon binders, for the conditioning of reactive metals and organic wastes are also within the scope of this task. Specifically, subtasks 3.1 and 3.2 attempt to provide with a suitable conditioning route for these types of wastes, by increasing the waste loading in the waste packages and therefore, reducing the environmental impact of disposal.

This WP has a strong emphasis on all aspects related to scaling-up (task 4) and industrial implementation of the processes and materials optimised in task 3 (task 5), having as one of its main aims the increase on their Technology Readiness Levels (TRLs). For that, STREAM tackles two key issues: disposability assessment and LCA/LCC analyses of the most promising processes and materials developed in this WP.

Sustainability and circular economy are key principles of the whole WP. As noted in the PREDIS Strategic Research Agenda (SRA) [3], a key theme for the implementation of these novel treatment and conditioning processes is the need for an integrated waste management approach, to enable the optimization of the whole waste lifecycle and to facilitate waste minimization and the drive to a circular economy. For that purpose, concepts such as primary and secondary waste volume minimization, use of recycled materials or the development of new low carbon footprint binders are underlying all technical tasks to lessen the impact on these novel treatment and conditioning technologies on the environment. Disposability assessment will be done according to the different types of disposal facilities and will consider issues relevant to Performance Assessment (PA) in the operational and post-closure stages such as, compatibility of resulting waste forms with existing Engineered Barrier System (EBS), diffusion and leaching processes or intruder/accident scenarios. Evaluation of the fulfilment of existing WAC by these novel waste forms or the need of development of new ones will also be addressed in task 5.

In the overall, and in alignment with EURAD and PREDIS roadmaps, STREAM aims to develop technological mature solutions for treatment and/or conditioning of the selected waste streams, considering environmental and economic aspects.

This WP aims in the last place to provide guidance about the most adequate treatment and/or conditioning matrix attending not only to the physic-chemical or radiological characteristics of each waste stream, but also to disposability-related aspects.

The purpose of this deliverable D6.1, is to review the current State-of-the-Art (SotA) in treatment and conditioning processes available or under development for challenging wastes, and identify the R&D gaps that need to be addressed in this work package to increase the TRLs of the proposed management routes.

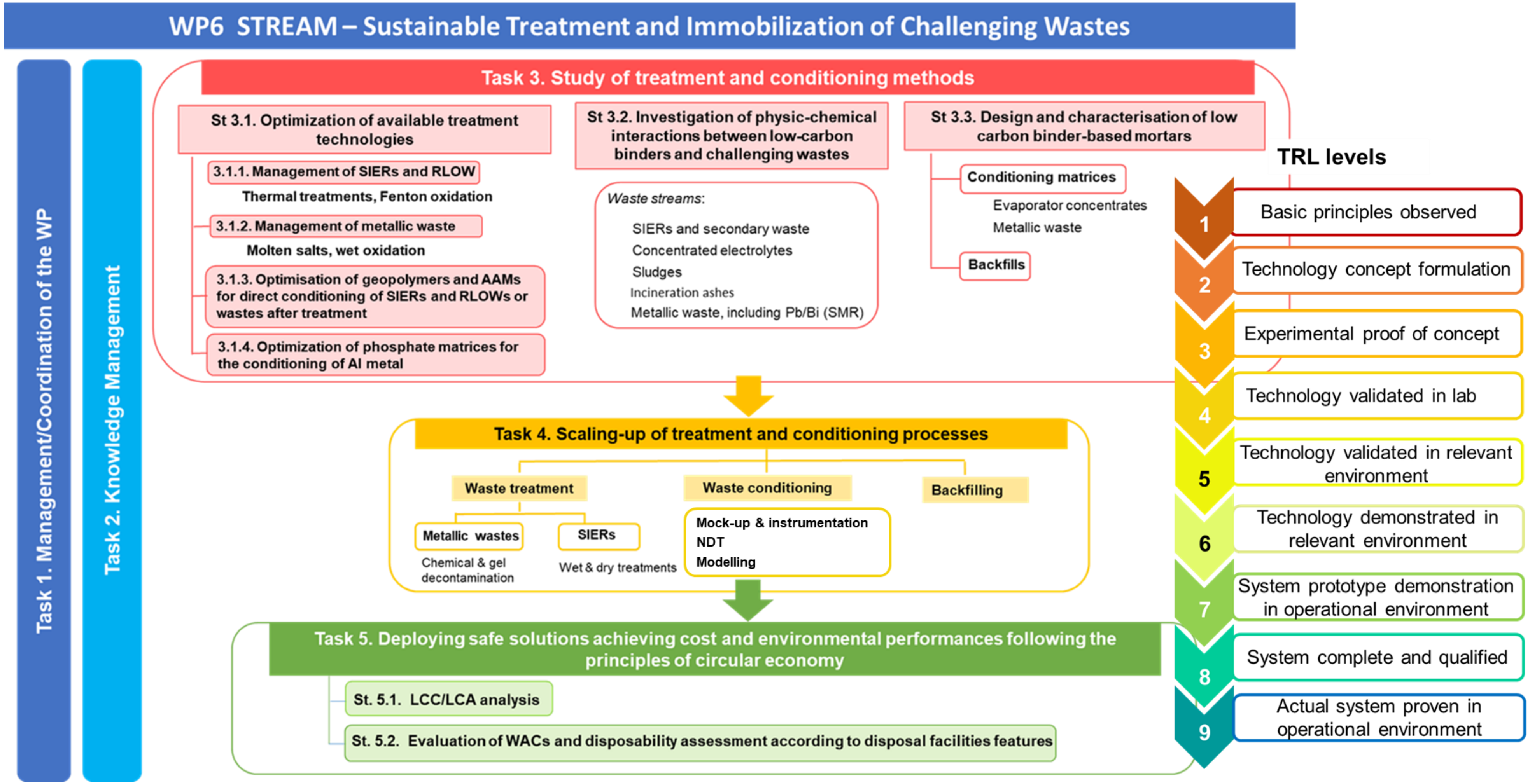


Figure 1 – Structure of STREAM Work package

2. Challenges in the treatment and conditioning of problematic waste streams

Radioactive wastes are diverse and varied in nature and it encompasses a broad range of radionuclides, half-lives, activity concentrations, volumes and physical and chemical properties. Other than radionuclides, waste may also contain other non-radiological contaminants of concern.

Due to this variety of composition, the choice of process(es) to be used for waste treatment and conditioning is quite complex and dependent on the level of activity, the type (form and characteristics) of waste and the overall strategy for waste management. Other issues, such as each country's nuclear waste management policy or its national regulations can also influence the approach taken [3].

For most common waste streams, technologies and practices for waste treatment and conditioning are well-defined. However, certain types of problematic waste streams (Figure 2) such as, organic wastes or reactive metals, still do not have optimised management routes [2].

Past and on-going R&D activities, and specifically STREAM WP, have been aimed to address these uncertainties still remaining to develop new safe and sustainable waste management routes for these types of challenging wastes.

Figure 2 – Reasons leading to consider a waste as challenging one [2]

2.1 Characteristics of waste streams studied in STREAM

For STREAM, a selection of most common problematic wastes was done on the basis of results and recommendations from previous projects, i.e. EURAD-ROUTES and PREDIS [2][3].

Six types of common challenging wastes have been chosen to be studied in this work package: Spent Ion Exchange Resins (SIERs), Radioactive Liquid Organic Wastes (RLOWs), reactive metals (Al, Be, Mg), sludges, evaporator concentrates and incineration ashes. Table 1 summarizes most relevant characteristics of each waste stream for their treatment and conditioning.

Additionally to these most widespread waste streams, a new type of waste from a GEN IV reactor has also been included in this WP, the Pb/Bi eutectic alloy, following the recommendations of the PREDIS Position Paper [4] on the new conditioning solutions for waste streams from new fuel types and advanced reactors/fuel cycles.

Type of waste stream	Source	Physic-chemical characteristics
Spent Ion Exchange Resins (SIERs)	Chemical and Volume Control Systems (CVCS), spent fuel pool cooling and clean-up system, treatment of liquid effluents system, steam generator blowdown, feedwater and condensate in BWRs	• Organic exchangers are primarily composed of polystyrene-divinylbenzene (PS-DVB) or acrylic-based polymers with sulfonic acid, quaternary ammonium, or chelating groups for ion exchange • Contaminants: radionuclides, heavy metals, and dissolved salts from water systems (i.e. borates)
Radioactive Liquid Organic Wastes (RLOW)	Including: lubricating oils, liquid scintillation cocktails, decontamination effluents or solvents from reprocessing activities	• Variable organic nature and wide range of contaminants depending on their origin • Poor chemical compatibility with traditional OPC matrices: thermal treatment or novel matrices as an alternative to cementation [5]
Reactive metals	Reactive metal waste, like Al, Be, and Mg, from nuclear power production or decommissioning activities	• Variable reactivity in alkaline media: hydrogen generation and risk of pressure build-up. • Critical dependence of pore alkaline pH [6][7]
Sludges	Primarily from effluent treatment processes like precipitation, evaporation and concentration	• Composition, activity concentrations and disposal route availability vary based on origin and treatment • One of the most difficult types of waste due to its chemical complexity, handling challenges, and long-term disposal issues [2]
Evaporator concentrates	Produced through an evaporation process. Evaporation is most effectively used for radioactive liquids with high concentrations of salts or other impurities	• The concentrate or bottoms product can range from 15 wt% solids to a virtually dry powder or cake, depending on the evaporator type and efficiency and on the chemical composition of the waste stream [8] • Contaminants: Cs-137, Co-60, Mn-54, borates, nitrates, alkali-hydroxides, and organic compounds (i.e. oxalates) [5]

Type of waste stream	Source	Physic-chemical characteristics
Incineration ashes	By-product of thermal treatment for LILW	• Properties depending on waste type and treatment conditions [9] • Generally, good compatibility with BFS-OPC and BFS-MK geopolymers [10]
Pb/Bi eutectic alloy	Used in liquid metal cooled reactors	• Contaminants from reactor corrosion and polonium generation arise during reactor operation [11]

Table 1 - Main characteristics of the wastes under study in STREAM.

2.2 Current problems for characterisation, treatment, conditioning and disposability

Characterisation is needed at any stage of the waste management: from the generation of the raw waste through to its collection, segregation, treatment, conditioning, storage, transportation and final disposal. The main properties that should be considered are the origin of the waste and its radiological, physical and, chemical properties. Characterisation needs to comply with the regulations to ensure proper handling and final safe disposal.

Treatment of radioactive waste involves operations intended to reduce the potential hazard of the waste and enhance safety in the long term (as one of a series of steps contributing to the safe predisposal management of radioactive waste). Due to the fact that radioactive waste is diverse and varied in nature and it encompasses a broad range of radionuclides, half-lives, activity concentrations, volumes and physical and chemical properties the choice of process(es) to be used for waste treatment is complex.

Waste stabilization by conditioning prior to long-term storage and disposal is a key activity of radioactive waste management. Radioactive waste conditioning consists of transforming the waste into packages suitable for transport, short or long-term storage and final disposal. Currently, the preferred solidification option involves a grout-based formula that flows to ensure complete encapsulation of the waste and container. In line with international best practice, as consignments are grouted upon receipt at a LLW repository, a consignor should assess the leachability of waste components, including hazardous materials, to understand their long-term performance in final disposal locations. Disposal sites have limits on hazardous and non-hazardous pollutants, as well as radioactive isotopes, which must be managed. Solidification must demonstrate the waste's durability and stability over the long term and under various environmental conditions. The solidified form requires careful consideration of waste movement and placement within solidified containers.

Guarantees of service are essential to encourage supply chain investment, maintaining robust systems for monitoring, transport, and disposal, which are critical for managing the complexities associated with solidified waste.

Extensive research is being undertaken to establish other solidifying matrices that may improve current practices, especially given the decline of core ingredients such as pulverized fuel ash. This research may advance the ability to solidify waste forms that are currently difficult to solidify by mixing with other materials, such as thermoplastic waste or low pH waste, or by using advanced solidification techniques like laser cladding [12] or vacuum induction melting (VIM) [13].

Table 2 summarizes the challenges and uncertainties in the characterisation, treatment, conditioning and storage/disposal of the different wastes included in STREAM.

	Characterisation	Treatment	Conditioning	Storage and disposal steps
Reactive metals (Al, Be, U, Mg..)	Characterizing reactive metal waste is challenging due to difficulties in obtaining precise list of activation products.	Reactive metals pose challenges for waste treatment and conditioning due to corrosion, which can cause waste form modifications (such as cracking), and changes in container mechanical properties.		Storage and disposal depend on the stability of waste packages.
SIERs	Challenges related to the characterization of SIERs is similar to sludge characterization. Techniques like gamma spectrometry and scaling factors help assess radiological inventories. Regarding chemical composition, MS noted chemical additives and corrosive products, but without specifics.	Increased activity after thermal treatment: While thermal treatments help reduce the volume of SIERs, they can also increase specific radioactivity. This complicates disposal and may require the use of alternative repository sites.	Challenges regarding stability issues with cement matrix: (1) challenges in maintaining the chemical and mechanical stability of the cement matrix when immobilising SIERs. (2) Contaminants interfering with Cement Hydration: Contaminants, particularly those from organic exchangers, can interfere with the cement hydration process, compromising the stability of the waste form. This can lead to issues such as swelling, gel formation, and reduced strength..	Main challenge: mechanical and chemical behaviour of waste packages, including potential swelling, corrosion, and complexing substances that increase radionuclide mobility.

	Characterisation	Treatment	Conditioning	Storage and disposal steps
Sludges	Uncertainties in radiological and chemical inventories and varying sludge volumes. Need for extensive characterization of legacy sludges.	Different immobilisation techniques are being tested, with cementation being the preferred option. Other methods like drying, high-density compaction or thermal treatment are also explored to address cementation challenges for specific sludges.		The main challenge in sludge disposal, aside from the availability of a final repository, is ensuring package compatibility with Waste Acceptance Criteria (WAC). Chemicals like sulphates and nitrates may interact with cement-based barriers and host rock, potentially mobilizing radionuclides. No definitive solutions exist yet, with treatment and conditioning being key focus areas.
Incineration ashes [14]	Characterizing thermally treated waste is complex due to its heterogeneity and the need for compliance with national and international disposal standards. Incineration ashes may reduce radioactive waste volume but can also concentrate or create new, harder-to-manage compounds.	Conditioning methods such as cementation and vitrification are effective in stabilizing incineration ashes, encapsulating hazardous substances, and reducing leachability, with the choice depending on the ash's radiological and chemical properties. Due to the variability in ash composition, disposal systems must be robust enough to handle waste with high concentrations of radionuclides or hazardous chemicals.		Incineration ashes present further challenges due to their diverse composition, requiring thorough radiological and chemical analyses to meet disposal criteria. Storing thermally treated waste is challenging due to concerns over physical stability and potential leaching. Long-term storage must ensure no environmental risk, requiring rigorous testing under simulated conditions. Disposing of incineration ashes is difficult due to the risk of leaching heavy metals and radioactive isotopes. Ensuring stable chemical and radiological characteristics over time is essential for safe disposal.
RLOW (Radioactive Liquid Organic Wastes): oils, LSC, decontamination effluents	Problems with the characterization of legacy wastes and complexing agent cocktails, including the ones resulting from plastic degradation.	Organic liquid wastes can be volatile, flammable, and toxic, requiring treatment and/or immobilization. Incineration of RLOWs is one of the options considered. However, certain waste streams are not compatible with incineration due to hazardous chemicals.		Retention of RLOWs in cement matrices is mostly physical, not chemical, making wastes vulnerable to leaching. Cementitious WF are susceptible to radiolysis, thermal, and microbial degradation that can lead to gas releases and cracking.

	Characterisation	Treatment	Conditioning	Storage and disposal steps
		Transport restrictions may not allow the waste to be transferred to appropriate treatment facilities. In order to overcome these limitations, immobilisation in Nochar polymer is an interesting option. Challenges for direct conditioning. Organic components interfere with cement hydration, causing delayed setting and porous matrices, particularly with polar solvents. Cementation also results in a volume increase. Currently, exploration of alternative treatment methods and geopolymers aims to reduce waste volume.		These issues limit the cementation of organic liquid wastes to a 10–12% loading. Complexing agents and chemotoxics present in some waste stream are banned or permitted within strict limits defined in the WAC.
Evaporator concentrates	Uncertainties in radiological and chemical inventories. Large volumes to characterise	Treatment technologies have focused on boron extraction mainly, in order to decrease borate contents and improve compatibility with OPC matrices [15] [16]	Alkali fission products (e.g., ^{85}Sr, ^{137}Cs) are highly soluble in cement, affecting only stabilization. In contrast, borates, sulphates disrupt hydration, altering setting time and strength. Mix designs for evaporator concentrates must account for borate effects, ensuring long-term durability under environmental and radiation exposure.	

Table 2 - Challenges and uncertainties in the characterisation, treatment, conditioning and storage/disposal of the challenging wastes included in STREAM (adapted from [2]).

2.3 Compliance with WAC

All waste sites and treatment/disposal sites must have a permit that describes the waste and its designated destination. This permit ensures that the facility operates within legal and environmental guidelines, specifying the types of waste it can handle and the methods of treatment or disposal.

An Environmental Safety Case (ESC) is a comprehensive document that demonstrates the safety of a disposal site. It includes detailed assessments of the potential impacts on human health and the environment, both during operation and after closure. The ESC is essential for obtaining regulatory approval and ensuring that the site meets all safety standards [17] [18]

The Waste Acceptance Criterion (WAC) defines the specific requirements that waste must meet to be accepted at a treatment or disposal facility. This includes physical, chemical, and radiological properties. The WAC ensures that only waste that can be safely managed and disposed of is accepted.

All sites must have a legal contract that outlines the agreement for the treatment and/or disposal of waste. These contracts ensure that all parties understand their responsibilities and the terms under which waste will be managed. They provide legal protection and clarity for both the waste generator and the disposal facility.

A Quality Management System (QMS) is a formalised system that documents processes, procedures, and responsibilities for achieving quality policies and objectives. In the context of nuclear waste management, a QMS ensures that all activities related to waste treatment, storage, and disposal are conducted in a controlled and consistent manner, meeting regulatory and safety standards [19] [20].

Governance arrangements refer to the structures and processes established to make decisions about the suitability of waste for treatment or disposal. This includes oversight by regulatory bodies, internal review processes, and stakeholder engagement to ensure that waste management practices are transparent and accountable.

An acceptance procedure details how the suitability of the waste form is assessed [21]. This includes a series of tests and evaluations to ensure that the waste meets the WAC and other regulatory requirements. Records of decisions made, along with justifications, must be maintained to provide a clear audit trail.

The final disposal location must be identified and approved. This involves selecting a site that meets all regulatory and safety criteria, often involving deep geological repositories for high-level waste. The site must be designed to isolate the waste from the environment for thousands of years.

Records must be maintained and accessible, detailing the contents of the waste, suitability declarations by consignors, and total and individual isotopic breakdowns of wastes. These records are crucial for tracking the waste throughout its lifecycle and ensuring compliance with regulatory requirements.

2.3.1 WAC definition

The link between WAC and the safety assessment for a facility is of primary importance. However, the scope of WAC does not necessarily have to depend entirely on the safety assessment of a facility [22]. It can also be linked with wider principles for waste management and supported by experience from waste operations. Regardless of their scope and the basis for their derivation, there must be a clear justification for how a suite of WAC has been developed and why each criterion is necessary [23].

Despite harmonisation is one of the biggest challenges regarding WAC, there are some observed commonalities [24]. In the case of LLW, radiation safety related criteria are incorporated in all programmes in terms of limiting surface contamination and dose rates to ensure safety while handling and transporting waste packages [25]. Specific activity limits of radionuclides are established based on the safety analyses of disposal facilities, and where it is relevant, activities of fissile and other selected radionuclides (^{226}Ra) are controlled as well. These limits are defined also in some preliminary WAC, apparently, based on international experience.

Additionally, the WAC of most countries address physic-chemical characteristics of the wastes or final waste forms as radioactive waste repositories are generally not intended for accepting hazardous, biodegradable and toxic materials. For safety reasons, the presence of some chemicals (aggressive, chelating) is not allowed or is strongly limited. Thus, disposal WAC can limit the risk of physical hazards (explosion, fire, …), putrescible materials (i.e., organic matter that is subject to decay through the action of bacteria and/or fungi), waste form stability, voidage, free liquids or prescribe the use of a certain type of container (or containers). Organic and reactive wastes that have the potential to produce gas, complexants and/or free liquids require treatment to convert them to a more passively safe form prior to conditioning and packaging, particularly where WAC preclude their acceptance for storage and/or disposal [26]. Gas generation, heat production, free liquids, fire resistance of solidified waste and their packages are other parameters taken into account. Table 3 summarizes the most common safety-related WAC.

Table 4 provides a simplified overview of waste acceptance requirements and parameters. The frequency of the use of various parameters in particular countries is visible from the table. Empty boxes do not necessarily indicate missing parameters, but just insufficient information about the actual status of the national Waste Acceptance System (WAS).

2.3.2 General requirements for Waste Forms

For disposal, solid or solidified RW is considered as the acceptable final waste form. Solidified final waste form should be compatible with the engineered system of the repository, if relevant, and with the host rock. This condition is usually not directly expressed as a special WAC, but the solidification matrix and the properties of the final waste form, such as stability, migration parameters, leachability, and strength resistance, are considered as inputs to repository safety assessment. Satisfactory results of safety assessment justify the use of the evaluated final waste form.

Solidification media must guarantee long-term stability of the final waste form by means of waste immobilisation, assuring transport parameters of the waste form, such as leachability, solubility, diffusivity and distribution coefficients, are as low as achievable. Not all solidification media are suitable for all types of waste: their improper use can lead to deterioration in waste properties during repository lifetime. Thus, the selection of an effective conditioning process is done on case-by-case basis while also considering WF performance during transport, storage and disposal. In the case of the challenging wastes under study in STREAM, conventional cementation does not seem a suitable option in terms of long-term durability and/or optimisation of waste loading. Table 2 summarizes main uncertainties related to their conditioning and causes of non-compliance with disposal WAC related to these categories of wastes.

The final waste form in most cases consists of solid or solidified waste and a container. However, individual solid pieces could usually be disposed of without container under conditions set down by WAC. Disposal of liquid waste is not desirable. In some countries, transport and disposal is totally banned, even if placed in containers with relatively long-life times, such as high integrity containers.

During the lifetime of the repository, new RW streams and/or new waste forms can arise, e. g. after technological changes in waste producer processes, or the use of new conditioning materials. In such cases, the new waste form must go through a waste form qualification process

[27]; it includes comparison with existing WAC, and in the case of non-compliance, development of new waste form specific set of WAC which has to be justified by new waste form oriented safety assessment.

	Parameters	Requirements
Radiation protection	Restrictions on dose rates	Dose rate at contact with and at a working distance
	Restrictions on fixed and/or removable surface contamination on waste packages	Maximum permissible contamination limits established for families of radionuclides (e.g., Bq/cm^2 of gamma radionuclides, beta radionuclides and/or alpha radionuclides)
Radioactivity	Restrictions on certain radionuclides	Maximum permissible limit for activity concentration of pre-defined radionuclides (e.g., maximum specific activity per waste package)
	Restrictions on fissile content	Maximum permissible limit for concentration or total amount of fissile radionuclides
Restrictions on content of waste packages	Restrictions on chemical or other hazardous constituents	Total ban on reactive chemicals, explosive and pyrophoric materials Maximum permissible limit for some materials such as corrosive materials per package
	Restrictions on biological, pathogenic, putrescible and/or infectious materials	Total ban on biological, pathogenic, putrescible and/or infectious materials
	Restrictions on free liquids	Total ban or maximum permissible limit of free liquids per package
	Restrictions on materials that present risks of explosion or ignition	Total ban or maximum permissible limit per package or accepted with restriction on case-by-case decisions (e.g. alkali metals, reactive metals, strong reducers…)
	Restriction on chelating compounds and chemotoxics	Total ban or accepted with restrictions
	Restriction on organic content	Total ban or maximum permissible limit per package
	Restriction on gas release	Upper limit on a per package basis (e.g., gas generation from corrosion, radiolysis and degradation of organic material)
	Restriction on heat generation rate	Total ban or maximum permissible limit per package of heat generation rate

Stability of the waste package	Chemical stability	Total ban or maximum permissible limit per package of reactive materials in the waste (including reactive/electropositive metals)
	Radiation stability of waste form	Specification of compatible materials to prevent or minimise possible damage to waste forms from radiolytic effects
	Physical stability of waste form	Specification or limits on void space in waste packages
	Durability of waste package/container	Specification or limit on materials to ensure adequate corrosion, fire, water, mechanical and impact resistance
	Mechanical properties of waste form	Specification of limits on mechanical properties such as minimum compressive strength and void space in the waste package.

Table 3 - List of most common safety related WAC [modified after [28]].

	Homogenity	*Dose rate*	*Surface contamination*	*Alpha specific activity*	*Beta/gamma spec.*	*Fissile element mass*	*Mechanical stability*	*Radiation & thermal & chemical stability*	*Package (damage, weight, type)*	*Physically hazardous materials: flammable,..*	*Void space*	*Built-up pressure in waste package*	*Gas generation*	*Heat generation*	*Organic content*	*Chemo-toxic waste*	*RW swelling*	*Free liquids*	*Reactive (electropositive) metals*	*Chelating / complexing*	*Leaching*
Belgium*		+	+	+	+	+	+		+	+			+	+	+		+	0	+	+	+
Bulgaria		+	+	+	+		+		+	0										0	+
Czech Republic		+	+	+	+	+		+	+	+			+			+		+		+	+
Finland**										+		+				+			+	+	+
France	+	+	+	+	+	+	+		+	+									+		+
Germany*		+	+	+	+	+						+						+	+		+
Greece		+	+	+	+	+		+	+	+			+	+	+	+		+		+	
Hungary	+	+	+	+	+	+	+		+	+			+					0	+	+	+
Italy*		+		+	+		+	+	+	0	+							+	0	+	+
Lithuania		+	+	+	+	+	+			+	+		+		+	+				+	+
Netherlands##		+	+	+	+	+	+	+	+	+	+		+			+		+		+	
Romania*	+	+	+	+	+		+		+	0	+		+	+	+	+			+	+	+
Slovakia		+	+	+	+	+			+	+			+								+
Slovenia*	+	+	+		+		+	+	+	+	+	0	+			+		+		+	+
Spain		+	+	+	+		+		+	0	+	0	0		+	0		0	0	0	
Sweden	+	+	+	+	+	+	+	+	+	0	+		+			+	+	0	+	0	
Switzerland		+	+	+	+		+	+	+	0	+		+	+			+	0		+	+
UK		+	+	+	+	+			+	+	+	0	+	+		+		+	+	+	+
Ukraine	+	+	+	+	+	+	+	+	+	+	+		+	+		+		+		+	

\+ in use

0 Banned item

* Generic/preliminary criteria, will be updated with the commissioning of a disposal facility

** WAC valid for FORTUM disposal facility

WAC for long-term storage

Table 4 - Parameters and requirements included in Waste Acceptance Criteria in different EU Member States [28]

3. Waste treatment and conditioning processes in use

3.1 International good practices

Several international instruments exist to support a safe and efficient management of radioactive waste. The IAEA and NEA provide Member States with guidance ([29] [30] [20] [31] [32] [33] [34] [35]) on radioactive waste management based on the principle of radioactive waste minimization. In addition, IAEA technical publications are available online ([36] [37] [38] [39] [8] [40] [41] [42] [43] [44] [45] [46]) to provide Member States with experiences and lessons learned on specific treatment and conditioning topics.

PREDIS Treatment Domain Insight (DI) [47] and PREDIS Conditioning DI [48] summarise the existing knowledge and approaches for radioactive waste treatment and conditioning, with the overall goal of minimising the quantity and volume of radioactive waste and put them in a form suitable for handling, transportation, storage and/or disposal.

The selection of the different approaches and technologies for waste treatment and conditioning is based on appropriate consideration of the characteristics of the waste and of the demands imposed by the subsequent steps in its management. Due to the fact that radioactive waste is diverse and varied in nature and it encompasses a broad range of radionuclides, half-lives, activity concentrations, volumes and physical and chemical properties (other than radionuclides, the waste may contain other hazardous elements (i.e. asbestos, mercury, beryllium, cadmium), the choice of process(es) to be used for waste treatment and conditioning is complex. The most appropriate treatment and conditioning options are those that lead to a waste form and package that meets the acceptance requirements of the disposal facility, whilst minimising waste volumes and doses resulting from these operations.

The PREDIS Treatment DI [47] discusses the good practices in waste processing, starting from the initial collection and segregation of waste up to the treatment technologies applied to the different types of radioactive waste. The PREDIS Conditioning DI [48] focuses on the conditioning processes that have been studied and developed to manage the radioactive waste produced by nuclear facilities. It explores the most commonly used matrices: hydraulic binders, bitumen, glasses and polymers and provides information on alternative composite matrices.

3.2 Available commercial treatments

The IAEA Technical Reports Series No. 402 [8] provides a description of the processes that are most commonly used for processing different types of radioactive waste. It examines processes for treating radioactive liquid aqueous and organic wastes (such as oils, scintillation fluids and miscellaneous solvents) and solid radioactive waste, including processes that are conventionally used, and those that are used for waste that requires special treatment considerations. The publication describes a wide range of contemporary matrix materials and processes for conditioning liquid and solid waste, including cemented waste. The chemical compatibility with selected waste and waste constituents is discussed. An account of compatible processing systems and equipment is included in the report and a dedicated section describes cementation processes for conditioning radioactive waste (e.g. ion exchange resins, precipitation sludges and evaporator concentrates) that may require prolonged storage, including waste feed compositions based on the operational experience of various countries and the sequential steps of the conditioning process.

The IAEA-TECDOC-1527 [46] provides an overview of the various thermal technologies and their applicability to various solid and liquid, organic and inorganic radioactive waste streams. Technologies are categorized as:

(i) **Pre-treatment processes:** whose end product typically requires further treatment (i.e. pyrolysis, steam reforming, calcinations, sintering, thermochemical treatment, and molten salt oxidation)

(ii) **Treatment processes:** which change the characteristics of the waste and may result in an end product which is, in itself, an appropriate waste form for disposition (i.e. incineration);

(iii) **Conditioning processes:** which result in an end product which is in itself a waste package suitable for handling, transport, storage and/or disposal (i.e. Vitrification, melting and plasma arc technologies). The publication focuses on those thermal technologies which are in use in one or more Member States and have been demonstrated to be proven, routinely used technologies.

The EURAD - Deliverable 9.12 [49] provides examples of facilities having possibility to treat or having treated foreign waste including examples of commercial and mobile facilities.

3.3 Traditional OPC-based binders and conventional conditioning systems for cementation

As reported in the EURAD Cemented LL-ILW DI [50], the cementation of radioactive wastes is an effective and cost-efficient conditioning technique that utilizes readily available materials. Cementation has a great flexibility to accommodate various waste types, including solids and liquids. Once hardened, cement becomes a durable solid with high compressive strength and low permeability, making it suitable for the safe storage and disposal of radioactive waste.

Key functional requirements for cemented wasteforms include radionuclide confinement to prevent environmental contamination, structural integrity to minimise radionuclide leaching and maintain the stability of storage/disposal systems, and long-term performance to align with safety assessments regarding waste management options.

Ordinary Portland Cement (OPC) is the most widely used type of cement. OPC binders are classified into several standardised categories depending on the binder composition and mineral addition content: CEM I, CEM II, CEM III, CEM IV and CEM V [51]. The specific binder formulations for conditioning radioactive waste are influenced by the waste's physical and chemical characteristics, as well as the acceptance criteria of the relevant storage and disposal facilities. The main parameters to be considered include:

- waste loading: typically, in the range 25–45 weight % [43]: but this range may vary depending on the type of waste and its radiological emission,
- waste chemistry: directly linked with the cement formulation to minimise the formation of secondary phases that can compromise the durability of the waste packages,
- cement durability: it must exhibit low dissolution rates in water to prevent the release of radioactive and chemical constituents, ensuring compliance with waste acceptance criteria,
- radiation stability: high radiation tolerance is essential to maintain the physical integrity of the cemented LILW,
- natural analogues: to provide insights into the long-term behaviour of cemented waste,
- environmental compatibility: as this impacts the integrity of the wasteform over extended periods.

IAEA-TECDOC-1701 [44] provides some examples of research and practices on the use of cementitious materials to immobilise low and intermediate level waste. It discusses chemical pre-treatment options and the use of blended cements containing mixtures of fly ash, slag or silica fume or the use of sorbents such as zeolites or bentonite clay to increase waste loading and provide short-term improvements.

Modern Portland cements have been in use for 150 years, leading to a well-established understanding of cement behaviour and chemical processes within cementitious materials. The field of cemented low-level and intermediate-level waste is considered mature, but there are still some challenges and ongoing innovations that need to be considered:

Availability of Materials and Environmental Impact Reduction: Modern OPC powders are produced finer and react more quickly, and this can complicate the controlled setting time needed for nuclear wasteforms. Moreover, cement industry is undergoing significant changes to reduce its carbon footprint, leading to the development and standardization of new types of cements that still need to be assessed for their chemical compatibility with existing waste streams.

Cement-Waste Interactions and New Cement Formulation Challenges: Certain waste constituents can adversely affect OPC, hindering hydration and altering solid properties. For instance, complexing agents like EDTA can limit calcium availability, while organic ion exchangers and certain salts can disrupt hydration. To address these issues, alternative cement formulations are being developed, including calcium aluminate cements, calcium sulphoaluminate cements, magnesium phosphate cements, and geopolymers, which enhance binding capacities and reduce negative interactions. While alternative systems, such as alkali-activated cements (AACs), show promise for durability there is still need for further optimization and better understanding of their long-term properties.

4. Needs of optimization of treatments and conditioning matrices under development

4.1 Treatment and decontamination

Safe, efficient, and cost-effective processing methods are not readily available for treating and conditioning all types of “problematic” radioactive wastes. Currently, several treatment technologies are available. Appendix A summarizes most relevant aspects for technology selection: advantages over conventional methods, needs of optimisation, project phase and TRL.

Waste treatment involves multiple phases, each employing specific techniques tailored to the nature and radioactivity level of the waste. The initial phase, pretreatment, encompasses sorting, size reduction, and decontamination to prepare the waste for subsequent processing. These processes aim to diminish waste volume, transform waste into a more stable form, and mitigate, eliminate, and contain hazardous components. Treatment approaches can be broadly categorized based on their operational characteristics: thermal techniques (e.g., incineration, pyrolysis, induction melting, and plasma melting), chemical techniques (e.g., wet oxidation and acid digestion), and mechanical techniques (e.g., compaction, super-compaction, shredding, and cutting). It is important to note that not all treatment methods are suitable for every waste type, and the selection of the most appropriate technique is influenced by various technical and non-technical factors. Some common methods can be applied to diverse mixed radioactive and hazardous wastes. Table 5**Erreur ! Source du renvoi introuvable.** provides a general comparison of different types of thermal and chemical treatment technologies.

Technology	Advantages	Wastes	Limitations
Thermal treatments	Well developed, commercial technology. High volume reduction for combustible waste. Residual ash amenable to immobilization, stabilization in agents.	Aqueous liquids, organic liquids, inorganic/organic sludges, ion exchange solids, organic solids, metals	Need to limit off–gases (generates secondary waste). Poor public acceptance for some treatments (e.g., incineration).
Chemical treatments	Reduced volume of off-gases such as dioxins, furans and mercury. Potential for high volume reduction for some types of waste. Can destroy many organic compounds and convert inorganic compounds into more stable form.	Aqueous liquids, organic liquids, inorganic/organic sludges, organic/inorganic solids, metals	Waste stream specific. Low volume reduction. Expensive corrosive resistant materials required.

Table 5 - Thermal and chemical treatment technologies for radioactive waste management

4.1.1 Thermal treatments

Safe management of radioactive waste is challenging in minimising environmental impact. However, deploying thermal treatment technologies can significantly improve volume reduction, waste passivation, and organic destruction. However, technologies' applicability and benefits to the identified waste streams need to be considered prior to industrial implementation [52].

Numerous technologies for thermal treatment of radioactive waste are currently available or under development worldwide (*Figure 3*). These technologies can be applied to a wide variety of radioactive waste streams, including non-standard waste types with specific waste management challenges. Thermal treatment can result in significant volume reduction and allow the reduction of organic contents. Nevertheless, formation of complexing agents may enhance potential radionuclide migration in the repository. Thermal treatments, in-use or under development, include incineration, pyrolysis, gasification, calcination, underwater plasma incineration, hydrothermal oxidation, and an induction metal melter [53]

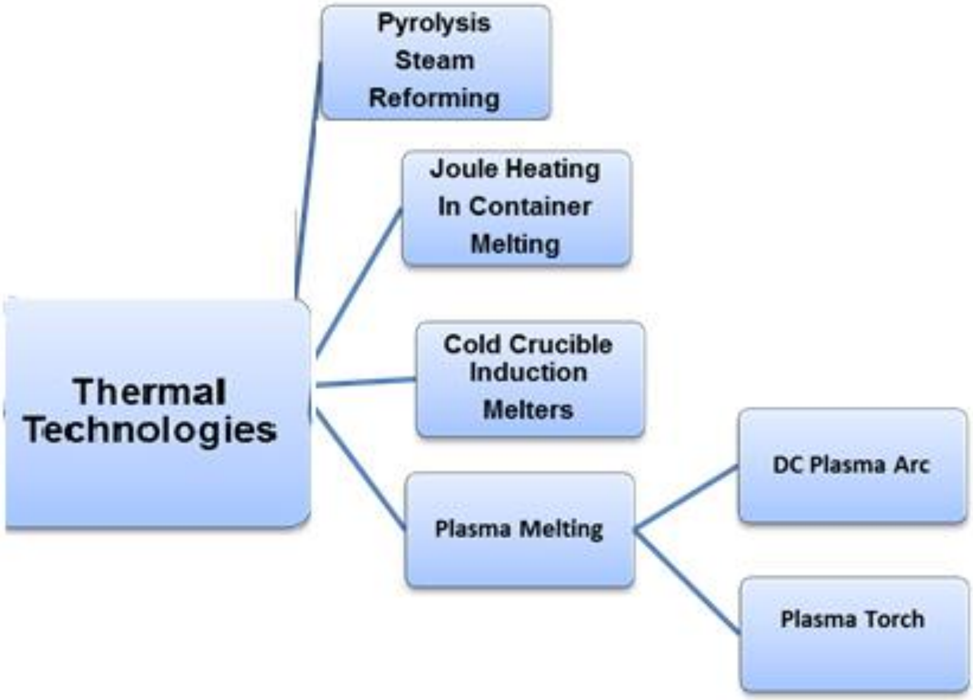


Figure 3 - Various thermal technologies applied to the treatment of "problematic" waste streams

Hereunder, a selection of most-widely used thermal treatments and most promising alternatives under development is provided. A description of the technologies and main outcomes of THERAMIN and PREDIS projects, as the most recent EU projects dealing with this subject, is also given below. EU-THERAMIN project was aimed to evaluate thermal treatment technologies' applicability and achievable volume reduction to a broad range of waste streams (ion exchange resins, soft operational wastes, sludge, organics and liquids). Disposability of ash and residues was assessed as well [53]. In the case of PREDIS, thermal treatment was considered as a previous step prior to immobilization [54]. Nevertheless, both projects proved the benefit of thermal treatment in reducing the volume and destroying the organic compounds.

4.1.1.1 Incineration

Incineration is a well-proven technology with very high-volume reduction of processed waste (for dry solid waste and a small percentage of wet waste). The main characteristics of the incineration process are here summarized:

- High throughput process.
- Process continuity (i.e., the process can operate continually 24 hours/day.
- It is susceptible to waste composition.
- Investment requires a relatively high capital cost.
- Public acceptance and licensing are difficulties.

Among the disadvantages of this technology, it should be pointed out the need of meeting environmental requirements for discharges as well as not being a cost-effective alternative for the treatment of small amounts of waste.

Concerning its application to the treatment of radioactive wastes, spent organic ion exchange materials can be incinerated in combination with other combustible waste or in an incinerator solely dedicated to that purpose. The incineration of spent resins in an oxygen rich atmosphere result in the oxidation of the initial feed material and produces a volume reduction factor ranging from 30 to 100. The final volume reduction factor depends on several factors such as the activity concentration in the incineration residues or the conditioning method.

Despite being a well-proved and reliable technology currently in-use in several disposal facilities, incineration still poses some challenges such as the loss of volatile elements, e.g. B or Cs, or the need of a special regime for the treatment of alpha-bearing waste [46].

4.1.1.2 Gasification-based thermal treatment

The gasification process involves heating waste in a low-oxygen environment where it undergoes partial oxidation and combustion [55]. The process converts the organics in the waste into a synthetic hydrocarbon gas (syngas) composed primarily of CO, H, and CH_4. The produced gas contains significantly less unwanted SOx and NOx than is produced by incineration. Any non-combustible components are left as a glassy ash residue with low organic content. The syngas can be burned for heating, electricity generation, or powering the treatment plant.

Non-combustible materials, such as metal, cement, and glass, are unsuitable for this technology as they can harm the system's performance (reducing the temperature). Wastes containing significant quantities of volatile radionuclides are neither good candidates for this technique. Additionally, off-gas treatment is expected to be required, resulting in the generation of associated secondary wastes that will need further management.

VTT Technical Research Centre (Finland) has proposed using their gasification plant [8] [56] to treat low-level wastes, but the system is still at the pilot scale. Inactive trials have been completed within the European Commission THERAMIN and PREDIS projects. Their results have shown very efficient removal of organic matter from IERs and significant volume reduction factors. Technical maturity is TRL 6 – technology demonstrated in the industrial environment. Tests have been undertaken on simulant waste at scale, although no tests on active waste at scale have been undertaken. Within the PREDIS project, the State Institution “Institute for Environmental Geochemistry” of the National Academy of Sciences of Ukraine (SIIEG NASU) deployed a coupled plasma-gasification process to thermally treat a surrogate spent cationic IER doped with Cs [54]. Development work focused on optimising the reaction temperature to ensure the conversion of the mobile and volatile Cs species into inorganic and thermally stable compounds.

4.1.1.3 Pyrolysis

Pyrolysis is one of the most attractive thermal method for waste processing. It is characterised as a low-temperature flameless process (compared with high-temperature thermal methods, e.g., incineration) in which the organic material is heated in a reducing atmosphere to leave a carbonaceous product or char. Being a flameless technology, pyrolysis has the advantage of lower operational temperature and, of critical importance in the nuclear context, an enhanced safety profile. Thus, pyrolysis features as an innovative technology in a recent IAEA report on the thermal processing of radioactive wastes [57], [58], being considered, and even practised, as a method for the treatment and conditioning of radioactive organic wastes such as solvents, oils and especially spent ion exchange resins derived from the operation of nuclear facilities.

Pyrolysis is a flameless dry distillation process that uses high temperatures to break down the organic components of waste into new gases, liquids, and solids (pyrolysates) under oxygen-free or oxygen-deficient conditions [59]. The organic fraction is not combusted (oxidised); complex organics are fractured into smaller components. The main outputs of pyrolysis are low-mass hydrocarbons (methane, ethane, benzene and toluene), tars, and a carbon-rich char [46].

Pyrolysis has lower operating temperatures than incineration, allowing it to retain relatively volatile radionuclides in the solid residue. CEA's (Commissariat à l'énergie atomique et aux énergies alternatives, France) IRIS process provides an example of a pyrolysis process designed for radioactive waste treatment [60].

Constraints include non-combustible waste, which is unsuitable. Halogenated plastics can be managed (as in the IRIS process) but require additional engineering. The system produces secondary gases that must be filtered and burned before discharging the atmosphere. The filters and scrubbing fluid from the off-gas system must be replaced periodically and disposed of appropriately.

Technical maturity is TRL 9 – proven technology in commercial use for radioactive waste treatment. Combining low-temperature pyrolysis and high-performance plasma treatment (HPPT) of the off-gas could be a novel solution for organic matrix nuclear wastes and provide economic and safety advantages for countries with low and medium-scale inventories. Moreover, this approach can be valuable for some industrially problematic organic wastes as a safe, economical, and environmentally friendly alternative. The PREDIS WP6 SIIEG, NASU show lab-scale study results related to the pyrolysis of IERs [61]. Studies of the influence of different operating parameters on the removal of model compounds using an inductively coupled plasma flow reactor are shown. The SIIEG NASU research group have worked with partners for several years to develop a hybrid gasification process of IER with incineration by plasma torch and gas cleaning technologies for different applications. The plasma thermal treatment process is a fundamentally new technology of a multiloop circulating gasifier, which provides a complete thermal decomposition of RSOW (Radioactive Solid Organic Wastes) at high temperatures in the reactor without oxygen. Ecological safety is the primary requirement imposed on modern RSOW processing technology. Adopting the multiloop circulation gasifier principle in waste processing allows for the significant fulfilment of this requirement, given the more profound destruction of waste. Furthermore, toxic volatiles in the pyrolysis gas are after-burned at a high temperature using a plasma torch and completely decompose. After gasification, ash is loaded into steel containers for further encapsulation. The first results of a bench scale arrangement combining both technologies are presented in [62].

4.1.1.4 Plasma techniques

Plasma technology has become a valuable technique for the thermal treatment of various chemical, mixed radioactive, and hazardous wastes [63] [64], [65], [66]. The method can also be applied to the treatment of spent resins. In this technique, radioactive waste, including spent intermediate-level radioactive waste (IERs), is fed into a plasma furnace; waste packages filled with resins are inserted directly into the furnace. The melting capacity depends on the design of the specific facility; for example, a plasma torch melter in Switzerland has a melting capacity of approximately 50 kg/h.

The volume reduction for spent resins ranges from 15 to 30%, depending on the amount of additives used. The treatment process is relatively costly and becomes economical only when operated at high throughput rates, rather than as a facility dedicated solely to treating IERs. The use of non-thermal inductively coupled plasma for treating IERs has been investigated in Japan [29]. Plasma technology offers a highly effective way of treating this type of waste. Main advantages of this technique are the high-volume reduction factors achieved and the obtention of a residue that is free from organics, liquids, and moisture, and generally meets the storage and disposal WAC. By means of a plasma beam of approximately 5000°C, the inorganic materials are melted into a glassy slag containing most of the radioactive isotopes, while the organic material is gasified, oxidized and purified in an off-gas cleaning system.

4.1.1.5 Molten salt oxidation (MSO)

Molten salt oxidation (MSO) is a flameless thermal desorption process [67][68][69]. Waste is introduced into a bath of molten salts, typically at temperatures between 500 and 950°C. An advantage of MSO over conventional incineration is that acidic gases, produced, for example, by the decomposition of halogenated organics, react with the carbonate melt and are retained as a salt (this property also allows MSO to be used as part of an off-gas management system as an alternative to wet scrubbing [70]). This oxidises the waste's organic constituents, producing carbon dioxide, nitrogen, and water, significantly reducing the burden of filtering and scrubbing compared to incineration or pyrolysis.

The end product is an organic-free salt residue that captures radionuclides, metals, and other inorganics. The formation of stable salts inhibits the production of acid gas emissions. This necessitates the discharge of salts once they are 'spent' (typically at ash contents of about 20 wt%). When the salts are cooled, the inorganic components, including radionuclides, are bound within the crystallised salts.

The CV Řež in Czechia operates an experimental MSO reactor focusing on spent ion exchange resins and scintillator oils. Within PREDIS [61], the MSO process was used at CV Řež to treat spent IER surrogates. A two-stage MSO process was used where IERs were fed into a sodium carbonate salt bed in one vessel. The off-gas is routed through a second sodium carbonate salt bed in another vessel to oxidise flue gases and fully capture fly ash. Constraints are wastes where C-14 or tritium are significant radionuclides. Secondary waste from MSO off-gas is relatively clean, so only small amounts from the off-gas system are expected. Technical maturity is TRL 6 (*technology demonstrated in the industrial environment),* and small-scale tests have been undertaken on radioactive waste. Industrial-scale plants for inactive waste are at DEFAC (S. Korea) [71] and NSWC Indian Head (USA) [72].

4.1.1.6 Metal melting

Metal melting is a high-temperature technology, where the scrap metal is heated above its melting temperature and during this process different elements and their radioactive isotopes are redistributed between the ingot, slag and dust [46]. The distribution of contaminants during melting is a complex process that depends on the elemental properties (chemical composition, solubility of an element in the molten metal, density of oxides, composition and basicity of the slag former) as well as on furnace properties (melting temperature, furnace type). Some elements chemically similar to iron, such as cobalt, nickel, chromium, zinc and manganese, mainly remain within the melt. More volatile elements, such as caesium, iodine or tritium, leave the melt and are transferred to the off-gases, or to the slag. Transuranic elements as well as some fission products can be readily oxidized and will be transfered to the slag. The advantages of metal melting application should be briefly summarized as follows [73][74][75].

- Decontamination of melted scrap metal is achieved by effective separation of radionuclides from the metallic waste and their distribution to the slag and dust/fumes. Decontamination factor therefore depends on the radionuclide present as a contaminant.
- After melting, the ingot (or end product) can be unconditionally or conditionally released into the environment or in case of remaining activity, can be stored for some period while radioactivity decays to releasable levels.
- The end product is typically homogenous and remaining activity is bound in the metal.
- Melting provides high volume reduction, thus the disposal capacities are preserved.
- Remaining activity bounded in the metal can be precisely determined.

Metal melting is an extensively proven technology, with commercial treatment facilities available (e.g. Studsvik melting facility, CENTRACO and INFANTE facilities in France, ECOMETs in Rusia and CARLA facility in Germany).

4.1.1.7 Fluidized Bed Steam Reforming (FBSR)

Novel thermal technologies that are already being, or are likely to be, deployed in future include those using pyrolysis, in which organics are destroyed in the absence of air. This is more environmentally compliant than incineration (which destroys organics in the presence of air) and can be performed in calciners, drums, or by Fluidized Bed Steam Reforming (FBSR) [76].

A commercial facility to continuously process radioactive wastes by pyrolysis at moderate temperatures in a hydrothermal steam environment was built by Studsvik in Erwin (Tennessee, USA) [77]. This facility uses FBSR technology to pyrolyse ^{137}Cs and ^{60}Co organic resins from commercial nuclear facilities.

Applying FBSR, in which a bed of granular material exhibits fluid-like properties, to nuclear waste requires two fluidised beds. The first (the Denitration and Mineralisation Reformer) operates in a reducing environment to evaporate the liquid nuclear waste stream, destroying organics, reducing nitrates, nitrites and nitric acid to nitrogen gas and forming a stable solid waste product. The second FBSR (the Carbon Reduction Reformer) operates in an oxidising environment, and its function is to gasify carbon fines, oxidise CO and H_2 to CO_2 and water, and convert trace acid gases to stable alkali compounds by reacting these acids with the bed media comprising calcium carbonate and/or calcium silicate particles.

The addition of bulk aluminosilicates to the fluidised bed results in the production of phases such as sodalite ($Na_8[Al_6Si_6O_{24}](Cl_2)$), which can accommodate waste species in their cage-like crystal structures including I, Cs, Sr and Mo.

This technology can process various solid and liquid streams, including wastes containing organic ion exchange resins, charcoal, graphite, sludge, oils, solvents, and cleaning solutions. When clay is added as a mineralising agent, the feldspar holds minerals (sodalite, nosean and nepheline) formed by a nanoscale reaction with the clay. The phases formed to act as hosts for high Cl, I, F, ^{99}Tc, and SO_4 alkali (Na, K, Cs) bearing wastes [76], and organics are destroyed, creating steam and CO_2. The mineralisation occurs at moderate FBSR temperatures because the FBSR operating temperature is in the range in which most clays become amorphous at the nanoscale level, e.g. kaolin, bentonite (montmorillonite), and illite. The clays lose their hydroxyl (OH^-) groups at the FBSR temperatures, destabilising the octahedral (6 nearest neighbouring atoms that form an octagon) Al^{3+} in their structure and becoming amorphous. The alkali in the waste “alkali activates” the unstable Al^{3+} cation to form new mineral phases, and the fluidising agent, steam, catalyses the mineralisation. Many of these mineral phases only form without steam at temperatures >1200°C.

Treatment methods	Features	Limitations	Secondary waste
Incineration	Well-proven technology. Very high-volume reduction of processed waste (for dry solid waste and a small percentage of wet waste). High throughput process. Process continuity (i.e., the process can operate continually 24 hours/day). It can be used for both solid and liquid wastes.	It is susceptible to waste composition. Investment requires a relatively high capital cost. Public acceptance and licensing are difficulties. The environmental requirements for discharges need to be met. Disposing of small amounts of waste is generally not economical. A special regime is required for the treatment of alpha-bearing waste.	Off gases Scrubbing Solutions
Gasification	Gas emissions contain significantly lower SOx and NOx than in the case of incineration Non-combustible components remain in a glassy ash residue with low organic content. Syngas can be burned for heating, electricity generation, or powering the treatment plant.	Not suitable for non-combustible materials (i.e. metals, cement, glass) or wastes containing significant quantities of volatile radionuclides	Off gases Scrubbing Solutions
Pyrolysis	Extensive commercial experience in processing high organic content waste (e.g. cartridge filters, charcoal, IE resins, plastics, waste with high organic content). Low process temperatures. Retention of volatile species in the ashes. Retention of radioactivity in the pyrolyser residue is > 99.99%. Low gas flow rates (compared to incineration). Insignificant NOx production. The end product is easily managed. The processes can be heated externally, thus minimising gas flows requiring radiological control.	Limited experience processing inorganic waste with a stabilised (monolithic) end product. Extensive waste pretreatment is required. Sensitive to waste composition and feed Sodium/potassium bearing waste might cause operational problems for fluidised bed pyrolysers.	Off gases Scrubbing Solutions

	It can usually treat the waste as generated (i.e., no prior segregation is necessary) for solids, liquids, and metals. The process temperature is up to 1800oC, which allows the melting of waste. The final waste form is robust, free of organic material and suitable for long-term storage and disposal. Volume reduction factors for metallic waste can range from 6:1 to 10:1, while for other combustibles, they can rise as high as 100:1. Less production of certain flue gases.		
Plasma techniques	It can usually treat the waste as generated (i.e., no prior segregation is necessary) for solids, liquids, and metals. The process temperature is up to 1800ºC, which allows the melting of waste. The final waste form is robust, free of organic material and suitable for long-term storage and disposal. Volume reduction factors for metallic waste can range from 6:1 to 10:1, while for other combustibles, they can rise as high as 100:1. Less production of certain flue gases.	Limited full-scale plant experience The process is expensive to construct and operate. The demands on an off-gas treatment system are greater than waste incineration. High maintenance to replace plasma torches: a special regime is required to treat alpha-bearing waste.	Off gases Scrubbing Solutions
Metal melting	Extensively proven technology for waste metals such as ferrous metals (carbon steel and stainless steel), aluminium, lead, copper and brass. High volume reduction, typically from 5:1 to 20:1 The end product is typically homogeneous and stable, with the remaining activity content bound in the metal.	Pre-sorting is usually required due to the different metals dedicated to melting furnaces and different melt temperatures. Melting mixed metal components (such as small electric motors) is usually not economical.	Off gases Slag

	The end product has the potential to be reused or recycled within the nuclear industry or after clearance within the conventional metal industry.		
Molten salt oxidation	Emerging technology. Alternative to traditional incineration of organic waste. Complete destruction of organic material. Lower operating temperature. Low levels of gaseous emissions. Costs are relatively low.	Limited commercial experience. Requires specialized techniques for adequate conditioning of the salt product. Limited to waste programmes	Salt residues

Table 6 -The main features and limitations of thermal treatment methods

4.1.2 Chemical treatments

The nuclear industry has access to a wide range of chemical treatments. The choice of treatment is highly dependent on the specific task, chemical nature of the contaminant, and the characteristics of the underlying material. Table 7**Erreur ! Source du renvoi introuvable.** provides a summary of the key features of currently used chemical treatment technologies.

Technology	Applicable waste streams	Temperature/ Pressure	Emissions	Secondary wastes
Electrochemical oxidation	Organic liquids, cellulosic, some plastics	40–60º C/ Ambient pressure	CO_2, CO, NO_x, HNO_2	Depleted acid, inorganic sludge
Photo oxidation	Dilute liquids, no solids	25–40º C/ Ambient pressure	CO_2, CO	Organic by–products
Acid digestion	Organic liquids, cellulosic	200ºC/ Low Pressure	CO_2, CO, NO_x	Sludge
Precipitation	Aqueous liquids	25–40º C/ Ambient pressure	None	Sludge/precipitate
Direct chemical oxidation	Organic liquids, cellulosic	200ºC/ Low Pressure	CO_2, CO	Sludge
Catalytic chemical oxidation	Organic liquids and sludges	200ºC / 100 psi	CO_2, CO	Sludge, spent catalyst
Chemical neutralization	Acidic or alkaline liquids	25–40º C/ Ambient pressure	None	Neutralized liquid

Table 7 - Comparison of chemical treatment technologies

4.1.2.1 Fenton and Fenton-like oxidation

Fenton and Fenton-like wet oxidation of radioactive wastes is one of the alternatives considered for the destruction of organic materials for around 40 years due to the attractive option to significantly reduce solid waste volumes and potentially re-categorise wastes for acceptance into national repositories. The presence of organics, especially organic ligands, is sometimes restricted due to concerns over flammability, degradation of organics over time and potential increased mobility of radionuclides [78], [79], [80].

Extensive trials on the implementation of wet oxidation have been undertaken in Italy, Sweden, Japan, the USA, and the UK. Laboratory-scale research, pilot plants, and multinational trials have determined that volume reduction and adequate destruction of certain materials are achievable. Wastes targeted in these trials include ion-exchange resins, waste reprocessing solvents, sludges, and decontamination effluents. Typically, wastes have been treated in batch or semi-batch processes, with remnant solids sent for cementation and liquid effluents disposed of via other routes.

Many of these national trials have focussed on specific challenging wastes. The most extensive testing at varying levels has been conducted in the UK, with several scale pilot plants and mobile plants treated with various ion-exchange resins, sludges, cellulosic waste, and liquid organics, including active trials utilising real waste. On the back of these research programmes worldwide, several full-scale wet-oxidation plants were planned for construction, usually utilising conventional $Fe–H_2O_2$ Fenton chemistry. None of the proposed facilities were ever constructed beyond pilot plant scale, being more conventional treatment facilities preferred (e.g. incineration or cementation).

This technology presents new opportunities and remaining challenges to become a viable industrial treatment process. Although the volume of solid wastes is reduced, Fenton reactions require large volumes of H_2O_2 to degrade the solid resins, resulting in an equally large volume of secondary liquid wastes generated. These liquid wastes have often been overlooked in plans for large-scale pilot plants.

Strict environmental discharge limits for radioactive material makes necessary further processing of these wastes either via an enhanced Fenton process or in additional treatment facilities. Further processing is also required for solids generated, with larger plants typically envisaged to operate at the cementation facility's head-end. Due to these secondary wastes, a longer-term understanding of any final waste-waste form interactions is required (e.g. between precipitated sulphates and cement), or research into alternative waste forms (i.e. glass or ceramic materials) for wastes generated. Development of mobile treatment facilities seems especially interesting for wet oxidation methods. For these mobile units, an original hope for wet-oxidation facilities was for smaller, mobile plants that could treat varied wastes across different sites.

On a more positive perspective, the ability of Fenton wet oxidation to degrade material at lower temperatures is particularly interesting for wastes containing volatile radioisotopes, for which high-temperature processes may require extensive off-gas systems. Opportunities exist for optimising degradation reactions with varied catalysts (including metal ions not favoured during typical wastewater treatment, such as copper and chromium), optimising the quantity of H_2O_2 utilised to reduce secondary wastes, co-treatment with other organic materials or integration into an alternative waste immobilisation process (e.g. head-end of a hot isostatic pressing system).

Improvements and modifications to the traditional Fenton process have been realised to increase reaction kinetics, maintain catalyst reactivity, and/or further reduce remaining organics in treated solutions. Routes to achieving these include using alternative homogeneous catalysts, utilising heterogeneous catalysts, and using more complex setups such as photo- and electro-Fenton [81].

Overall, the Fenton-like oxidation of organic IERs can be very effective, with reported reductions in organic carbon of >98%, with the capability to use a source of Fe, H_2O_2 and some pH control. Resulting liquids are usually neutralised, resulting in a sulphate-rich product and the precipitation of many radionuclides. Shorter treatment times typically result in a higher residual organic content and a higher presence of lightweight organic species, which appear relatively resistant to Fenton oxidation.

Degradation of IERs using Fenton-like reactions has consistently found near-boiling temperatures to produce the most rapid and efficient degradation of resins. Temperatures lower than 90°C have consistently resulted in higher final chemical oxygen demand (COD) values and sometimes incomplete resin degradation [82] [83][79]. The effect of pH on the degradation of IERs is not clear-cut. Due to the high organic loading during decomposition of these materials and the interplay between acidic organic products and sulphuric acid/ ammonia generated during decomposition, the pH can alter significantly during degradation.

Several national nuclear research institutions and companies worldwide have trialled larger-scale applications of Fenton processes to specific nuclear wastes. This has resulted in several pilot and small-scale plants utilising Fenton chemistry, all using homogeneous catalysis. Substantial interest was garnered from the 1980s to the 2000s, with strong interest from Sweden, Italy, the UK, the USA and Japan. None of these trialled systems appears to have entered whole commercial operation outside of limited testing, although significant experience and knowledge have been acquired from these research programmes. Nevertheless, in Italy, there is a Fenton treatment facility located in Nucleco site for treating radioactive liquids coming from medical, research and industrial applications, but not wastes from their national nuclear program. The potential for lower temperature processing, with a significant reduction in resultant solid wastes, could provide another technology in the toolbox of treatment options.

4.1.3 Decontamination processes

Radioactive metallic waste comes from multiple sources and are categorised as Low-Level Waste and Intermediate-Level Waste depending on its associated radioactivity. Usually, decontamination techniques are employed to mitigate radioactive contamination, aiming to remove the radioactive oxide layer where radionuclides are often physically adsorbed.

Conventional decontamination techniques, such as mechanical and thermal methods, often face limitations in effectively removing radionuclides. Other techniques, such as chemical and laser, are also effective but generate secondary wastes that are difficult to manage. Therefore, development of innovative decontamination approaches that are, both efficient and environmentally friendly, has become an issue of increasing interest. Challenges to address include varying waste volumes, radioactivity levels, and related materials. Prevalent chemical techniques, particularly conventional washing with specific chemical solutions, prove commonly effective.

Electrochemical and/or gel decontamination methods can be employed favorably for specific surfaces, providing notable advantages, including the reduction of secondary waste. These approaches contribute to efficient decontamination and address concerns related to waste generation.

4.1.3.1 Chemical Gels

The chemical methods are easy to apply and present an increased contact time, which improves the removal efficiency. In addition, they can reach remote and hidden areas. However, to achieve maximum effectiveness, repeated applications may be required.

The innovative decontamination process using chemical gels aims to overcome problems associated with chemical-based decontamination techniques, such as reagent baths, foaming solutions, or solvents. The gels are commonly prepared by dispersing thickening agents, such as silica or alumina particles, in solution, forming a gel-like suspension. The excellent rheological properties of decontamination gels allow them to be sprayed and remain attached to the surface. This enables the implementation of this technique over large surfaces at a large scale. The gels crack after drying, forming non-dust flakes where the contaminants are trapped and are easily removed by brushing or vacuum cleaning (Figure 4).

It offers the advantages of safe handling, high penetration, and small volumes of secondary waste [84][85]. The commercial DeconGel™ 1101 was used to remove ^{60}Co and ^{137}Cs radionuclides during the decommissioning of a nuclear research reactor. Decontamination was more efficient for nonporous materials, and the decontamination factors could reach more than 90%. The newly developed polyvinyl alcohol-borax complex-based spray coating contains adsorbents (Prussian blue, bentonite, and sulfur-zeolite) to decontaminate ^{137}Cs-contaminated surfaces [86]. The gel-like coating adhered to vertical surfaces with a ^{137}Cs removal efficiency of 56.9%, compared with 27.2% for DeconGel. In addition, the coating could be easily removed by rinsing with water, leaving no residue.

Moore et al. [85] prepared a nitric-acid-laded polymer hydrogel with high removal of ^{137}Cs and ^{90}Sr on stainless steel. It displayed high removal efficiencies of 91.61% for painted cement, 97.05% for aluminium, 94.1% for stainless steel, and 53.5% for cement, 2.3 times higher than that of commercial Decongel. The adsorbent could be separated from the used hydrogel by a simple magnetic separation, reducing waste disposal costs.

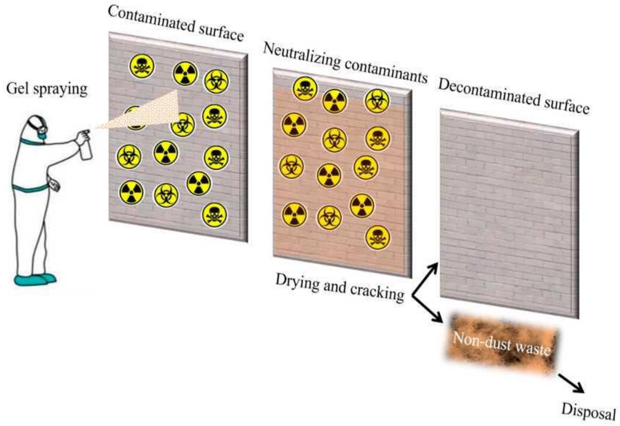


Figure 4 - Flow chart of chemical gels technique [31]

4.1.3.2 Magnetic gel decontamination methods

Magnetic gel decontaminants are advanced materials that combine magnetic nanoparticles with polymer networks to create systems for metal removal from contaminated environments. These systems' technology readiness level (TRL) depends on carefully controlling and optimising multiple parameters throughout development. The key areas requiring optimisation include the synthesis conditions of the magnetic components for the gel network formation parameters and the operational conditions during decontamination. Success in raising the TRL requires systematic optimisation across physical properties (like porosity and magnetic response), chemical characteristics (such as binding affinity and selectivity), and application parameters (including contact time and pH conditions). Understanding these interdependent factors is crucial for developing effective decontamination systems transitioning from laboratory success to practical field applications.

Using magnetic nanoparticles in decontamination processes represents a significant advancement in treatment technology. A key benefit is that these particles can be easily retrieved from suspension using magnetic fields, eliminating the need for conventional filtration steps that would otherwise be required to separate treatment materials from the processed water [87]. Additionally, the ability to manipulate these particles using magnetic fields enables more efficient solid-liquid separation during the decontamination process. While this technology is relatively new regarding radioactive metal decontamination applications, its potential to streamline treatment processes and reduce operational complexity makes it a promising approach for practical implementation.

Critical material and preparation considerations must be addressed when developing magnetic sorbents for radioactive metal decontamination. The synthesis process should prioritise simplicity, convenience, and cost-effectiveness from an industrial implementation perspective while avoiding harsh conditions [88].

The selection of precursor materials must align with environmental sustainability goals, mainly focusing on ecologically benign components that follow the CHON principle (primarily carbon, hydrogen, oxygen, and nitrogen) to enable complete incineration at end-of-life with minimal secondary waste generation.

The resulting magnetic sorbent must demonstrate chemical and radiological stability to maintain its effectiveness across multiple adsorption-desorption cycles without degradation of its core components, coating, or functional groups [88].

The material should also achieve a high preconcentration factor to maximise efficiency in practical applications. This will enable the concentration of radionuclides from large-volume solutions into smaller, more manageable quantities for safe disposal. The optimisation of sorbent dosage represents a critical trade-off in magnetic gel decontamination processes. While increasing the sorbent material improves overall metal ion recovery, it decreases sorption capacity per unit of sorbent [89].

This inverse relationship creates two distinct operational scenarios depending on the primary goal of the treatment process: when the main objective is to achieve maximum decontamination with minimal residual concentrations, a higher sorbent dosage is necessary; conversely, if metal recovery and concentration are priorities, a lower sorbent dosage is more appropriate [90]. In practice, a moderate sorbent dosage of around 0.5 g/L has been found to effectively compromise these competing objectives in the optimisation of magnetic gel decontamination processes [91].

4.1.3.3 Vacuumable decontamination gels

Vacuumable decontamination gels are already used regularly in nuclear decommissioning. A typical example of such gels is the so-called ASPIGEL 100E. This gel is based on an acidic, Ce^{4+}-containing formulation and is well suited for removing fixed contamination in several tens of µm of the material. Another example is the ASPIGEL 400, a basic formulation for decontaminating aluminium alloy materials. The gel is sprayed to form a homogeneous layer of the desired thickness (0.5–1 mm). Upon drying (2–48 hours, depending on the formulation and climatic conditions), the gel shrunk, forming cracks and flaking. After drying, the dry gel entraps the contaminants, and the formed flakes are easily removed using brushing and/or vacuuming.

The whole process considerably limits personal exposure, and the secondary waste generated is in a solid form and is, therefore, easy to handle and manage for disposal. The gel formulation can be adapted for stainless steel or concrete applications.

PREDIS project Deliverable 4.3 [92] describes the development of new vacuumable gel formulations and implementation processes performed within the PREDIS project, which aimed to increase their range of applications.

New gel formulations were initially developed, inspired by the COREMIX (Chemical Oxidation REduction using nitric permanganate and oxalic acid MIXture) process. Their decontamination capability has been compared to a commercial product, such as Aspigel 100E from the FEVDI Company. Although the COREMIX-based gels were less efficient, they offer an alternative in situations where the commercial product cannot be used (e.g. due to waste acceptance compatibility, etc.) or for soft surface decontamination operations.

Magnetic gels were developed in order to overcome the limitations of conventional methods, especially for small objects with complex geometries (such as pipes, valves or pumps) or limited access surfaces. A direct relationship between the number of ferromagnetic particles, the rheological and spreading properties, and the gel decontamination efficiency was highlighted, paving the way to developing even more optimised formulations. An additional advantage of this type of products is the possibility of remote application, which reduces occupational radiological risks [93].

In order to improve the efficiency of gel decontamination processes, several operational parameters need to be optimised:

- **Reagent Concentration Control**: The concentration of chemical reagents must be carefully optimised to ensure effective decontamination while maintaining cost efficiency in Chemical Oxidation Reduction Decontamination (CORD) processes.
- **Contact Time Optimization:** To achieve optimal results, the duration of contact between the decontamination agents and the contaminated metal surface needs to be precisely controlled.
- **Gel Viscosity Management:** The viscosity of inorganic gel treatments must be carefully controlled to ensure proper coverage and treatment effectiveness.

- **Surface Adherence Properties:** The gel's ability to properly adhere to metallic surfaces is a critical parameter that requires optimisation to ensure effective contaminant removal.
- **Temperature Control:** To ensure optimal decontamination efficiency, the reaction temperature should be monitored and controlled.
- **pH Level:** The solution pH needs to be maintained within appropriate ranges to facilitate effective metal ion removal and ensure gel stability

4.1.3.4 Strippable Coating

Several decontamination methods combine chemical and mechanical or a hybrid of the two. Strippable coatings use chemical and adhesive methods to remove the contamination from the surface and again require mechanical coating peeling [94].

Wang et al. [95] prepared a strippable coating using acrylate emulsion as the primary film-forming agent and lauryl sodium sulphate as a surfactant. The decontamination rate reached 92.26% for uranium dust on the concrete surface with a 2.5 kg m^2 dosage. Pozo et al. [96] explored the feasibility of applying chitosan gels with or without Fe_3O_4 nanoparticles to deal with radioactive contamination. A removal efficiency of 85% was achieved for non-compactable waste contaminated with uranium.

The strippable coating method has neither airborne contamination nor secondary liquid waste. However, it is best suited for minor decontamination activities and only works for easily removed contaminants.

4.1.3.5 Future perspectives

Each technology has advantages and disadvantages in specific scenarios *(*Table 8*). The* selection of proper technologies mainly depends on the type of facilities, involved isotopes, the activity level of the equipment and parts, and the physical/chemical properties of the materials used to decontaminate. The selection must consider safety, efficiency, cost-effectiveness, waste minimisation, and industrialisation feasibility [97].

Among the chemical methods, chemical gels are suitable for decontaminating complex shapes and vertical and overhead surfaces. In addition, they can enhance other decontamination agents' efficiency by allowing them to stick to surfaces and improving contact time.

However, the formulation of colloidal gels is complex, and no gel type is helpful for all contaminants. Therefore, novel and versatile gels must be developed to enlarge their application field.

No single technology can address all kinds of problems. Combining various decontamination methods often has better results. Therefore, a reasonable and practical combination of these decontamination methods has become the main direction for developing and applying decontamination technologies for decommissioning nuclear facilities in the future.

Techniques	**Advantages**	**Disadvantages/Limitations**
Chemical gels	Easy application and increased contact time. It can reach remote and hidden areas. Minimal secondary waste generation.	It may require repeated applications to achieve maximum effectiveness. The formulation of colloidal gels is complex.
Strippable coating	Produce a single solid waste. No airborne contamination.	The spray gun nozzles clog.

	No secondary liquid waste.	Best suited for smaller decontamination activities. It only works for easily removed contaminants.

Table 8 - The comparison of different decontamination techniques [40]

4.2 Novel conditioning matrices

In the past decades, non-Portland clinker alternative binders have emerged as an interesting alternative to Portland cements for the immobilization of radioactive wastes. The evolution of cement industry under way to reduce its carbon footprint has led to the development and standardization of new cements (CEM II/C-M, CEM VI, belite-calcium sulfoaluminate cements, Geopolymers and Alkaly activated binders, Phosphate cement, Magnesium phosphate cements, LC3) which may offer new prospects for nuclear waste management.

These alternative or non-traditional cements differ significantly from OPC binders in terms of composition, raw materials or nature of reaction products (Figure 5). The history and state of knowledge concerning these cements varies greatly from one type to another. For this reason, information on these cements is divided into two types: those with a history of use in construction and those which are long known but not used in structural applications.

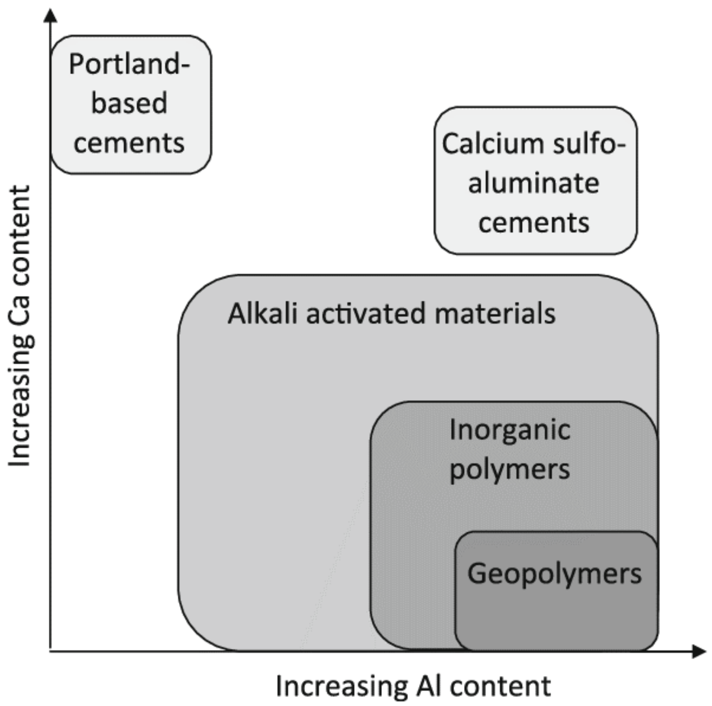


Figure 5 - Classification of different subsets of AAMs, with comparisons to OPC and calcium sulfoaluminate binder chemistry [98].

CAC and CSAC have a relatively long history of use: for CAC, of more than 100 years but for sulfoaluminate types, only since the 1970s. Both types have high heats of hydration and liberate much of this heat within the first 24 hours of hydration, so the initial hydration exotherm needs careful management. They may be used to form grouts, mortars and concretes and are compatible with most mineral aggregates except those containing gypsum or other calcium sulphates.

Phosphate cements have been traditionally used for medical applications. This type of cements have a wider chemical and structural composition compared with other binders, depending on the nature of the raw materials and the activators used. MPC/MKPC cements have been extensively studied for waste conditioning applications [6], including the immobilization of reactive metals, highly-saline effluents or U-contaminated ashes. A commercial MKPC matrix named **Ceramicrete™** is already available and has been applied to the conditioning of a wide range of problematic LILW, including Pu-contaminated ashes, heavy metals and Ra wastes or salt cakes. **Ceramicrete™** is not properly considered a cementitious matrix. It is included among the chemically-bonded phosphate ceramics (CBPCs), in which the acid component is orthophosphoric acid or a soluble orthophosphate and the resulting ceramic is an insoluble orthophosphate [99]

M-S-H cements are based on the interaction of MgO or $Mg(OH)_2$ with amorphous silica, resulting in the formation of a M-S-H binder gel. This type of mineral matrix is relatively new among other cementitious materials for RW solidification but has already received increasing attention.

Geopolymers and Alkali-Activated Materials have gained global interest as an alternative to standard OPC blends. This is due to geopolymer's high strength, adaptable gel network, and overall low environmental impact. Recent research has extensively explored formulation development in this field, resulting in various potential waste immobilization formulations. Metakaolin and/ or slag-based systems have demonstrated to be robust solutions, able to stabilise diverse waste streams (RLOW, SIERs, thermally-treated wastes) [100] [101].

However, there are remaining issues that need to be addressed such as, the establishment of future waste form testing protocols or the assessment of long-term durability of geopolymer waste forms under relevant disposal conditions. These two subjects will be tackled in the coming years in the IAEA CRP 2405 [102] and in the framework EURAD-2 L'OPERA WP [103], respectively.

Currently, there are two geopolymer-based conditioning matrices that are commercially available: **SIAL™** and **DuraLith™**. SIAL™ is already accepted by the Slovak and Czech regulatory bodies for disposal in their respective repositories. It is currently used for the conditioning of SIERs or mixed SIERs/sludges with waste loadings up to 20% wt. (on a dry basis) in 200l-drums. DuraLith™ was initially patented for the stabilization of ^{129}I and ^{99}Tc present in the secondary liquid streams from Hanford Waste Treatment Plant. New research seems to be oriented to extend its application to other waste streams.

In recent years, limestone calcined clay cement (LC^3) has been developed as a promising blended cement with a comparable performance as OPC but a much lower carbon footprint . It is composed of OPC clinker (as low as 50%), calcined clay, limestone and a small certain of gypsum for proper sulphation. The wide availability of the raw material (limestone and low-grade clay), and compatibility with current OPC manufacturing process and application codes, makes LC^3 a seamless sustainable alternative to OPC [104]. Currently, initial lab work has been conducted to demonstrate its suitability for the immobilization of borated waste streams and highly-saline solutions [105].

Type of cement	Waste stream
Calcium Aluminate Cement (CAC)	Wastes containing: Liquid borates, radioiodide, Cs and heavy metals (Cr, Cd, Zn, Mg, Sn)
Calcium Sulfoaluminate Cement (CSAC) *(i.e. belite-CSA)*	IERs Reactive metals (Al and U) Sludges with high sulphate and borate contents
Magnesium Silicate Hydrate Cements (M-S-H cements)	Magnox Sludges (as precursor)

Type of cement	Waste stream
Phosphate cements	
Magnesium phosphate cement (MPC)/Magnesium Potassium Phosphate (MKPC)	Reactive metals (Al, U) Solutions containing chloride, nitrate, nitrite, sulphate Borated wastes Evaporator concentrates Incineration ashes Contaminated concrete from decommissioning activities
Calcium phosphate cement (CPC)	Spent adsorbents
Geopolymers and Alkali Activated materials	SIERs and other RSOW Evaporator sludges RLOW (oils, LSC, greases, TBP/Kerosene...) Thermally-treated wastes (ashes, slags..) Borate-rich wastes

Table 9 - Alternative binders used for the conditioning of problematic wastes

4.2.1 Advantages of new conditioning matrices over OPC cementation

Alternative binders can exhibit many desirable properties for the immobilization of radioactive wastes. This may include enhanced chemical resistance in aggressive environments and enhanced chemical tolerance to problematic and complex waste streams [106].

These novel binders would allow the implementation of simplified cementation process routes, as they generally show faster curing than OPC binders and eliminate the need for waste pre-treatment. Their higher fluidity could also lead to potentially higher waste loadings. Additional advantages of these cements are lower gas and water permeability and an improved durability [107].

Another interesting characteristic for waste conditioning is their higher efficiency for physical and chemical immobilization of radionuclides and heavy metals due to ion exchange and adsorption capacity of zeolitic-like reaction products. This would enable the use of alternative binders as adsorbents and chemical additives [108].

The introduction of these alternative binders can also provide resilience and security of cement powders and supplementary materials, as the evolution of cement industry to reduce its carbon footprint has resulted on a risk for the supply of cementitious materials currently in use for LILW conditioning. [50].

Table 10 summarizes the characteristics of most representative types of alternative binders and their advantages over conventional OPC binders.

Type of cementitious material	Alternative binder	Characteristics	Advantages over OPC binders	Country of commercial application for RW S/S
Calcium Aluminate Cement (CAC)		Fast hardening, high strength, low permeability, high freeze-thaw, corrosion resistance	Tolerance of waste components retarding the setting and hardening (i.e. B) Enhanced chemical binding of a wide variety of metals and RNs by ion exchange (zeolitic reaction products)	France (for immobilization of hazardous non-radioactive waste encapsulation)
Calcium Sulfoaluminate Cement (CSAC)			Avoid pre-treatment Lower corrosion rates for reactive metals and gas generation (corrosion, radiolytic) Suitable for wastes containing of setting retarders (B, Zn…)	
Magnesium silicate cement (M-S-H cement)		Control of initial pH and reduced shrinkage	Significant potential for encapsulating certain problematic waste streams containing Al [109]	
Phosphate cements	Magnesium phosphate cement (MPC)/Magnesium Potassium Phosphate Cement (MKPC)	Fast setting, high early strength, adhesive properties, low water demand and drying shrinkage, high temperature and chemical resistance	Compatibility with a wide range of wastes H_2 radiolytic generation 2-3 times less than in OPC Low corrosion rates for reactive metals	Russian Federation, USA **(Ceramicrete™)**

Type of cementitious material	Alternative binder	Characteristics	Advantages over OPC binders	Country of commercial application for RW S/S
	Calcium phosphate cement (CPC)			
Alkali-activated cement	Alkali-activated slag cement (AASC)	Fast setting, high strength, low porosity and high temperature and chemical resistance	Compatibility with a wide range of challenging wastes Increased waste loadings (i.e. SIERs up to 25%) Suitable for wastes containing of setting retarders (B, Zn…)	Ukraine
	Geopolymer			Czech Republic, Slovak Republic **(SIAL™),** Slovenia, France, USA **(DuraLith™)**,
LC³		Low porosity, enhanced durability, low carbon footprint	Reduced alkali-silica reaction- Resistant to chloride and sulphate attack- Lower gas and water permeability [110]	

Table 10 - Most representative types of alternative binders: characteristics and advantages over conventional OPC binders (modified after [108]).

4.2.2 Challenges to address and compatibility with different waste streams:

Properties and short/medium/long-term performance of these alternative binders need to be further studied to fill existing knowledge gaps. Three main gaps have been identified in existing literature:

- Management of exotherm reactions during setting/hardening processes
- Cement-waste interactions: included in the scope of STREAM in subtask 3.1 and 3.2
- Disposal assessment, including aspects such as:
    - Compatibility with existing EBS in disposal facilities
    - fulfilment of existing WAC or the need to develop new ones

In general, in all cases, the reactions involved in the setting/hardening process are exothermic. These may affect the scaling-up approach proposed, as large-volume drums of cemented waste forms may exhibit a substantial temperature rise. This issue is of major importance since the solid phase composition in materials based on CSACs, CACs, magnesium phosphate and alkali-activated binders are temperature-dependant.

Another key issue to address when evaluating the suitability of a conditioning matrix for a specific waste stream is understanding cement–waste interactions, and their impact on waste form performance in the short and long-term. This will allow to limit adverse chemical reactions that may jeopardize the durability of the waste form.

Evaluation of chemical stability of the waste-matrix system under realistic disposal conditions, including interactions with other engineered barriers in the near-field, is a critical task for the acceptance of these alternative binders for waste conditioning [108].

Long-term durability of waste forms under realistic disposal conditions is a fundamental issue that needs to be address prior to acceptance of a new formulation, and though it is not under the scope of STREAM WP, it will be addressed in the framework of EURAD-2 in WP7 L'OPERA.

4.2.2.1 Expected waste-matrix interaction

Interactions between different cement systems and waste streams have been studied in several national and international R&D programs [44][111][112]. Waste-matrix systems have proved to be complex, and further efforts are required to fully understand and quantify these types of interactions.

Results obtained have shown that by controlling the internal chemistry, microstructure of the matrix and the hydration products formed, through incorporation of reactive admixtures, choice of curing temperature and moisture content, water /cement ratio, systems may be developed selectively to enhance immobilization of a specific waste component or group of components.

Additional research is still needed to elucidate physical and chemical effects of waste ions on the cement structure during solidification, the formation of exotic hydration products, the speciation of waste ions and the resistance of the constituent solid phases to degradation. These interactions must be understood because apparently slight changes in matrix chemistry could result in significant change in immobilization capacity and therefore, can promote radionuclide leaching [44].

In traditional OPC systems, it is well-known the adverse effect of certain chemical compounds on setting and hardening of the waste forms:

- complexing agents such as EDTA or boron can interfere with Ca availability
- organic ion exchangers can take up water under high pH conditions and expand in the wasteform
- and zinc salts and borates can inhibit hydration.

To overcome these limitations, alternative formulations have been developed to improve the matrix binding capacity for selected radionuclides and to reduce the reactions between waste constituents and the cement hydrates [50].

Table 11 summarizes the currently-in-use OPC formulations and the proposed alternative binders for the conditioning of waste streams included in STREAM.

Waste stream	**Conventional Cementitious matrix**	**Alternative binders under development or already commercially available**
Spent Ion Exchange resins	Slag-Portland blends	Calcium Sulfoaluminate Cements (CSAC) Geopolymers (including commercial products **SIAL™**) Magnesium Phosphate Cements
Sludge and concentrates generated from treatment of LLW	OPC, with or without additives	Calcium Sulfoaluminate Cements (CSAC) Magnesium Phosphate Cements (MPC) **LC³** Geopolymers
Incineration ashes	OPC, with or without additives	Magnesium Phosphate Cements (MPC) Geopolymers
Mixture of sludge and ion exchange resins	OPC, slag-Portland cement	Geopolymers (including commercial products **SIAL™)** **LC³** Magnesium Phosphate Cements (CSAC)
Intermediate level liquid waste	Slag-Portland blands, OPC with vermiculite	Magnesium Phosphate Cements (MPC) Geopolymers
Secondary waste generated during treatment of solvents from reprocessing	OPC	Geopolymers
Evaporator concentrate containing boric acid	OPC	Calcium Aluminates Cement (CAC) & Calcium Sulfoaluminate Cements (CSAC) Magnesium Phosphate Cement (MPC) Geopolymers (**DuraLith™)**

Table 11- Waste streams studied in STREAM and examples of conventional Portland formulations and alternative binders used for conditioning [50]

For STREAM, a selection of the most common problematic operational wastes has been made, including SIERs, liquid organic wastes, evaporator concentrates, sludges or reactive metals. An extensive work has been focused on the treatment and conditioning of these type of challenging wastes, but some uncertainties are still remaining, especially concerning the compatibility of these new binders with several types of challenging wastes [6][100][101].

4.2.2.2 Spent Ion Exchange Resins

Spent Ion Exchange Resins (SIERs) usually contain as major contaminants fission products (i.e. Cs, Sr), corrosion products, borates, nitrates and alkali components. For direct immobilization of in OPC matrices, SIERs content is typically kept below 10%wt., though the use of additives (i.e. silica fume, blast furnace slags) may allow to increase waste loading up to 20%.

Bead resins have a deleterious effect on the cementitious matrix as contaminants can retard setting (i.e. borates, Zn). That is the case of SIERs used for the treatment of borate streams. Resins composed of salts of short chain water soluble organic acids (i.e. acetic, formic, picolinic acids) can also interfere with hydration reactions. Additionally, higher content of anionic resins may increase bleeding during curing of the waste forms. Apart from the chemical interactions, SIERs may also influence the mechanical performance of the waste form, as osmotic swelling of resins can lead to the formation of microcracks. Water saturation of the SIERs previously to cementation can be done to avoid this mechanical effect [5].

4.2.2.3 Liquid Organic Wastes

Physic-chemical characteristics of Radioactive Liquid Organic Wastes (RLOW) are highly-dependent on the chemical composition of the effluent or the nature of the by-products generated during the hydrolysis and radiolysis of the organic compounds. Most typical RLOWs are contaminated lubricating oils, liquid scintillation cocktails and solvents from fuel reprocessing and effluents generated in decontamination activities.

RLOWs can interact with the cementitious matrix by covering the anhydrous cement grains and preventing the reaction with water. This will lead to set retardation as well as to an increase in the matrix porosity. This effect is more intense in the case of polar solvents [113].

Another issue of concern related to this type of wastes is the greater susceptibility of RLOW-waste forms to leaching, as the immobilization of the liquids is mainly physical and no chemical bonding is generally observed. Liquids are usually trapped in the pores and void space of the matrices and no structural bonding occurs.

Gas generation is another point to take into consideration since it can lead to cracking of the cementitious waste form. Organic liquid wastes are highly susceptible to degrade under storage/disposal conditions by several mechanisms: radiolysis, thermal and microbial degradation.

Understanding microbial activity and its impact on waste integrity is essential for long-term management. This understanding is crucial to prevent the development of honeycombs in the solidified matrix, which may lead to stack collapse if placed incorrectly in a container stack. This is even more relevant for polymeric or bitumen waste forms. In the case of cement-stabilised WFs, biodegradation testing is generally included in waste form qualification protocols for WFs containing carbonaceous materials, such as oils or SIERs.

All these problems limit RLOW waste loading to 10%wt. in cementitious waste forms, if direct immobilization is used. In order to overcome partially the previously exposed problematic, several additives and admixtures have been used including clay, silica- and calcium-based additives and emulsifiers. Use of two-step conditioning achieves significant improvements of waste loading. Pre-emulsification and pre-impregnation can lead to waste loadings up to 50%.

In PREDIS project, WP5 was devoted to the immobilization of RLOWs in geopolymers. Results pointed to geopolymer as a promising alternative for oil conditioning. However, further work needs to be done to assess the long-term behavior of the waste forms in disposal environment [100].

4.2.2.4 Reactive Metallic Wastes

Consideration should be given to managing the hydrogen evolution of certain reactive metals, especially unstable when using a high pH grout. OPC-based systems are not adequate for the immobilization of reactive metallic wastes, such as aluminum, beryllium or magnesium, since they are likely to react with the cement paste.

Interaction of these metals with the cementitious matrix results in the corrosion of the metal surface. High corrosion rates lead to a significant hydrogen generation, that may cause the cracking of the waste form and an overpressure in the waste package that can lead to the failure of the waste package. Corrosion control is especially relevant for the encapsulation of Al-containing wastes. Use of low-pH matrices, such as MPC, has proved to contribute to reduce corrosion rates of reactive metals and therefore, limiting hydrogen generation [6] [114]. However other alternative binders for Be and Mg such as geopolymers are alternative binder under consideration.

Another interesting alternative is the use of inert gas blankets [115], using gases such as carbon dioxide or others that would drive out the hydrogen prior to reaching the flash point, thereby preventing the creation of an explosive environment within a container or work area. Alternatively, increasing ventilation is another option to be consider to avoid the risk of ignition or explosion.

4.2.2.5 Evaporator concentrates and concentrated solutions

This waste stream is characterized by a significant heterogeneity of its radiological and physic-chemical characteristics. Concentrates can contain a great variety of salts and even organics.

Borates, phosphates, sulphates, nitrates, fluorides and organic compounds are known for their impact on the hydration reactions that can lead to set-retarding or on the contrary, can accelerate the hydration process. Boron is a well-known setting retarder whereas nitrates tend to accelerate hydration reactions.

Depending on the saline content and chemical composition, waste incorporation to the paste can affect significantly the microstructural and mechanical properties. Criteria for selection of immobilization matrix

Cementitious matrices are widely applied in the nuclear industry for the conditioning of LILW basically due to:

- Compatibility with a wide range of different waste streams
- High binding capacity for radionuclides
- Improved chemical, thermal and radiological stability
- Good compatibility with cementitious EBS in disposal facilities.

The design of a conditioning matrix aims to produce a durable waste form with reliable long-term performance under storage and disposal conditions. For the choice of the matrix and its later optimization, several considerations should be taken into account:

1. Quality requirements and standardization of raw materials
2. Compatibility with the physic-chemical characteristics of the waste stream: i.e. components that influence hydration reactions
3. Factors affecting the cementation process: setting time, workability, viscosity, etc.
4. Optimization of the formulation: cement/water ratio, use of additives, optimised waste loading
5. Finally, type of disposal concept and the required safety functions for the waste forms

In general, compressive strength, permeability, irradiation behavior or leaching resistance are used as performance indicators during the design of the cementitious waste forms to assess solidification, stabilization and hydraulic performances [113][116]. Resulting waste forms must fulfill WAC defined for the disposal facility, on the basis of its safety case [18][27].

5. Scaling-up and industrial implementation of new treatments and conditioning processes

Addressing the challenge of radioactive waste management requires robust, scalable, and commercially viable methodologies for processing and treating diverse forms of waste, including organic substances, metals, concentrated residues, and sludge. The progression from experimental techniques developed in laboratories to large-scale industrial applications involves overcoming a multitude of obstacles, encompassing technical complexities, regulatory requirements, and financial considerations.

5.1 Previous experiences from industry, WMOs/waste producers

Generally, the selection of the process(es) for the (pre)treatment of radioactive wastes is based on the level of activity and the classification of waste. Nevertheless, waste management policies and national regulations can also influence the adopted approach.

Treatment processes such as compaction and incineration are widely extended. Chemical processes are also of common use for the (pre)treatment of several waste streams on an industrial scale, either in-plant or in treatment facilities.

5.1.1 Examples of thermal treatments

Table 12 summarises the incinerators at industrial scale to treat liquid and solid radioactive wastes available across European countries. Figure 6 shows an example of off–gas system of the CILVA incinerator located at Belgoprocess (Belgium) to treat solid and liquid wastes.

Country	Facility	Capacity	Notes
Belgium	CILVA, Research Center Belgoprocess	80 kg/h solid 50 kg/h liquid	Commercial treatment: solids, liquids and ion exchange resins
France	Cyclife France	2500 ton/yr solid 2000 ton/yr liquid	Commercial ILW & LLW treatment facility
Netherlands	COVRA, Vlissingen–Oost	60 kg/h solid 40 l/h liquid	Two incinerators, one for liquids, one for animal carcasses & other solids
Slovakia	Jaslovske Bohunice BSC	50 kg/h solid 10 kg/h liquid	Used in campaigns for LLW
Spain	ENRESA El Cabril	50 kg/h total solid & liquid	Located at LLW disposal facility
Sweden	Cyclife Sweden AB	License limit 600 ton/yr Solid and liquid	Can also treat activated carbon

Table 12 - Incinerator facilities to treat liquid and solid waste in Europe

Figure 6 - View of the off–gas system of the incinerator. (Courtesy of Belgoprocess).

Other thermal treatment technologies currently on a pilot scale are the ELIPSE facility at one of the CEA sites (France) and the CFB gasification pilot plant in VTT (Finland). The ELIPSE method employs more potent plasma (45 kW) immersed in an aqueous solution (submerged sprayed plasma arc). This high-power plasma facilitates the decomposition of organic liquids at a rate of up to 5 L/h (Figure 7). This process can handle a diverse range of organic components, including chlorine, fluorine, phosphorus, and scintilliation cocktails. Liquids containing TBP/dodecane or chloroform mixtures can be processed at a rate of 2 L/h for durations ranging from 1 to 30 h. The efficacy of the destruction was assessed by examining the total organic carbon in the remaining solution, which consistently approached 99%. Substantial advancements and demonstrations have been achieved in recent years, positioning the process at a technology readiness level near six on the TRL scale. However, this method still requires further technical enhancements before it can be implemented in industrial and commercial operations.

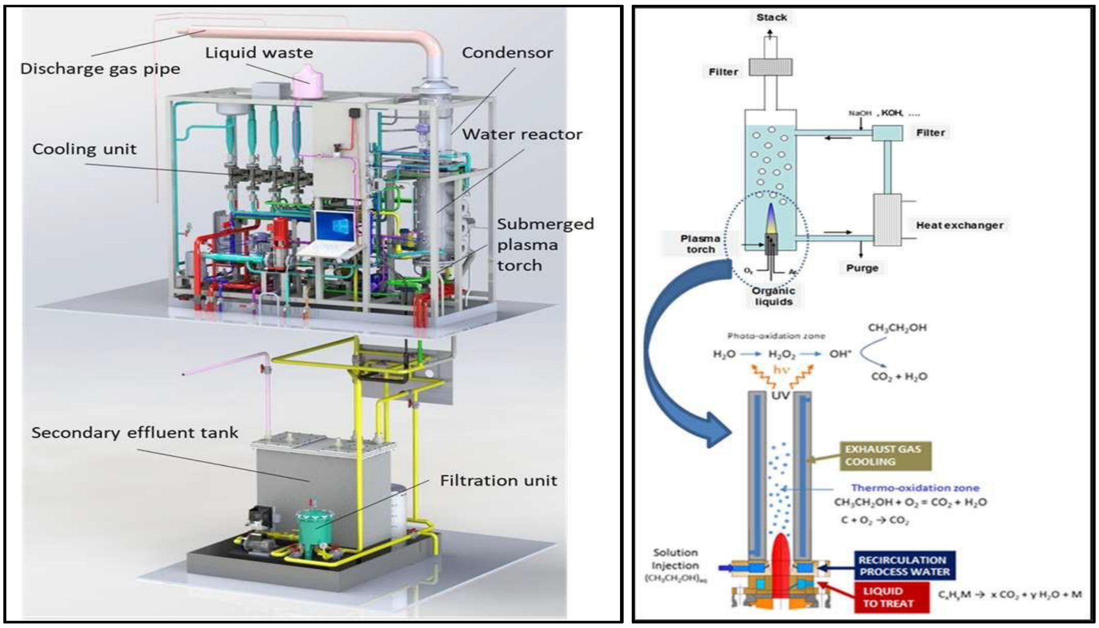


Figure 7 - Schematic view of the elimination of liquids by plasma in water (ELIPSE) process. (Courtesy of CEA).

At the Technical Research Centre of Finland Ltd. (VTT), researchers have been advancing on the upscaling of thermal gasification technology for the treatment of spent ion exchange resins [117]. This technique, known as circulating fluidized-bed gasification (Figure 8), is also capable of reducing the volume of low-level operational waste containing organic matter, provided the waste is crushed beforehand. The current test facility is specifically designed for processing spent ion-exchange resins; therefore, modifications to the feeding systems are necessary to accommodate other waste types.

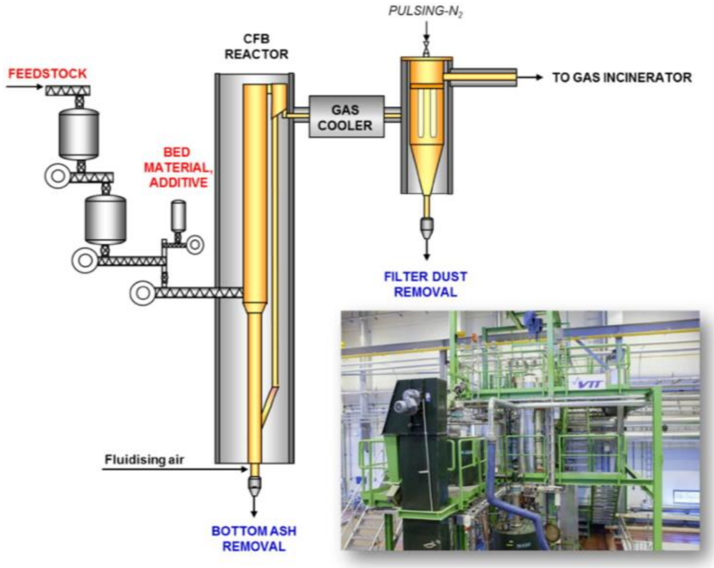


Figure 8 - Pilot scale Circulating Fluidised–Bed (CFB) gasification test rig. (Courtesy of VTT).

5.1.2 Examples of chemical treatments

The nuclear industry has access to a wide range of chemical treatments. Chemical neutralization or precipitation are procedures of common use for several types of waste streams generated in different activities of the nuclear cycle. The choice of treatment is highly dependent on the specific task, chemical nature of the contaminant, and the characteristics of the underlying material. Table 13 provides a summary of the key features of currently used chemical treatment technologies.

Technology	Applicable waste streams	Temperature/ Pressure	Emissions	Secondary wastes
Electrochemical oxidation	Organic liquids, cellulosic, some plastics	40–60º C/ Ambient pressure	CO_2, CO, NO_x, HNO_2	Depleted acid, inorganic sludge
Photo oxidation	Dilute liquids, no solids	25–40º C/ Ambient pressure	CO_2, CO	Organic by–products
Acid digestion	Organic liquids, cellulosic	200ºC/ Low Pressure	CO_2, CO, NO_x	Sludge
Precipitation	Aqueous liquids	25–40º C/ Ambient pressure	None	Sludge/precipitate
Direct chemical oxidation	Organic liquids, cellulosic	200ºC/ Low Pressure	CO_2, CO	Sludge

Technology	Applicable waste streams	Temperature/ Pressure	Emissions	Secondary wastes
Catalytic chemical oxidation	Organic liquids and sludges	200ºC / 100 psi	CO_2, CO	Sludge, spent catalyst
Chemical neutralization	Acidic or alkaline liquids	25–40º C/ Ambient pressure	None	Neutralized liquid

Table 13 - Comparison of chemical treatment technologies

5.1.3 Waste conditioning

Waste conditioning, a crucial step in the preparation of materials for storage or disposal, involves several key techniques. This report concentrates solely on cementation/polymerization techniques utilizing innovative or low-carbon binder systems. A survey conducted during the PREDIS project (results as of February 2021), involving 11 organizations from Belgium, the Czech Republic, Denmark, Estonia, Finland, France, Italy, Spain, Sweden, and Switzerland, revealed that only a few entities have implemented new conditioning processes using blended cement systems at an industrial scale [118]. As illustrated in Table 14, blast furnace slag is the primary supplementary cementitious material employed in the conditioning matrices.

Name	Conditioning process	Container/ drum material	Shape	Dimensions	Waste class	Closure system	Pre-treatment	Conditioning matrix
Solidified waste package	Homogeneous	Concrete with stainless steel reinforcement	Cylindrical	Height: 1,3 m; Diameter: 1,3 m; Wall thickness: 105 mm; Inside volume: 1 m^3	ILW	Concrete lid cast after waste solidification	Evaporation, Cs separation	Cement and blast furnace slags
Resins from power plant water treatment	Homogeneous	Steel	Cylindrical	Height: 0.9 m, Diameter: 0.6 m	LLW	Lid, screwed or clamping ring	-	Sulphate resistant portland cement, additional zeolites for sorption
Solidified waste package	Homogeneous	Concrete with stainless steel reinforcement	Cylindrical	Height: 1,3 m; Diameter: 1,3 m; Wall thickness: 105 mm; Inside volume 1 m^3	LLW	Concrete lid cast after waste solidification	Evaporation, Cs separation	Cement and blast furnace slags
Scrap and interchangeable parts from reactor operation and maintenance Smaller metal. Can contain steel, Al, Cu,	Heterogeneous	Steel drums	Cylindrical	Height: 0.9 m Diameter: 0.6 m	LLW	Screwed lid or clamping ring	-	Sulphate resistant portland cement or special light weight cement

stainless steel or mixtures of these								
Steel 400 l drum	Heterogeneous	Galvanized steel	Cylindrical	400 l	LLW / ILW	Seam folding	Incineration, pre- or super-compaction	BFS cement

Table 14 - Low-carbon binder used for conditioning as an outcome from PREDIS's survey

Despite extensive laboratory research demonstrating its effectiveness, industrial-scale application of geopolymers (or alkali-activated materials in general) for radioactive waste treatment remains limited [119][120][121]. Currently, SIAL®, which was developed by Jacobs®, is the most widely used commercial geopolymer matrix. This technology has been employed in various countries, including Slovakia, the Czech Republic, Japan, and Taiwan, with plans for its implementation in the UK and France. In 2021, the national utility ČEZ opted for Jacobs' SIAL® geopolymer encapsulation technology to safely solidify 250 metric tons of low- and intermediate-level radioactive sludge at the Dukovany Nuclear Power Plant (Figure 9). The process involves encapsulating the sludge, previously stored in tanks at the facility, into 200-litre drums for long-term storage. The treatment was conducted in an onsite unit.

Figure 9 - On-site unit to condition ion exchange resin using SIAL® geopolymer at Dukovany NPP, Czech Republic (Courtesy of Jacobs).

5.2 Upscaling process

Upscaling radioactive waste processing and treatment operations requires a methodical approach to ensure dependability, effectiveness, and adherence to safety protocols. Upscaling involves increasing the size of the operations from the laboratory to the industrial scale, which is essential for efficiently managing substantial quantities of waste. This process encounters technical obstacles such as preserving process stability with larger equipment and financial hurdles, including substantial initial investments. Regulatory adherence is vital for guaranteeing that safety standards are upheld. The methodology encompasses laboratory-scale development, pilot testing, and the design of full-scale facilities with ongoing monitoring for continuous enhancement. Scaling up is crucial for handling large volumes of waste efficiently while addressing the associated technical, economic, and regulatory challenges.

5.2.1 Key steps and considered factors

To upscale a treatment and conditioning process, a systematic approach is necessary:

- **Process selection:** Identify an appropriate method based on waste characteristics. For HLW, vitrification is often chosen; for LLW, cementation/polymerisation or incineration may suffice. Waste characterization, including composition and activity levels, is critical to inform this choice. When it comes to cementation/polymerization, the selection of the binder needs to be done on the basis of the waste characteristics (e.g., geopolymer for borate waste) and then optimise formulations through lab-scale testing, considering waste loading and curing conditions.

- **Lab-scale development**: Develop and characterize the process on a small scale, understanding chemical reactions (e.g., glass formation in vitrification), and physical transformations. This stage ensures feasibility and identifies the initial parameters.
- **Pilot plant testing:** Conduct tests at a larger scale to optimise the process, identify issues such as heat transfer inefficiencies or equipment scaling limitations, and optimise the processes. For example, the Hanford Site conducted pilot tests for vitrification, which demonstrated the continuous processing of tank waste. This step identifies the technical issues and optimises the process.
- **Design and construction:** Design the full-scale facility and select equipment (e.g., furnaces for vitrification, mixers for cementation) capable of handling increased volumes. For example, MPC's rapid setting of MPCs requires efficient mixing and pouring systems. Safety measures such as shielding and remote handling must be integrated. Environmental considerations, such as minimizing release, were also addressed.
- **Regulatory engagement and economic analysis:** Collaborate with regulatory bodies to establish standards, as the IAEA does for geopolymers. This includes demonstrating compliance by extensive testing and documentation. Conduct cost-benefit analyses considering initial investment and long-term savings. For instance, MPC's lower energy requirements of MPC compared with vitrification could reduce operational costs.
- **Operation and continuous improvement:** Implementation of the scaled-up process with continuous monitoring for quality control (e.g., glass durability tests) and efficiency. Adjustments may be required based on operational data to ensure compliance and safety.

5.2.2 Challenges of upscaling

5.2.2.1 Treatment upscaling

Upscaling treatment processes presents several challenges:

- **Technical challenges:** Scaling up requires maintaining process stability and efficiency. For vitrification, ensuring glass quality (durability and stability) at larger scales is complex, with equipment such as furnaces needing to handle increased volumes while managing high temperatures (up to 1100°C for vitrification). Material handling of radioactive waste requires specialized equipment and remote operations to minimise worker exposure, particularly for HLW. Variability in waste streams can affect process consistency, which requires flexible systems.
- **Economic challenges:** The initial investment for large-scale facilities is significant, with vitrification plants having high operational costs owing to energy consumption and the need for qualified personnel. Economic viability is crucial, especially for locations with stable, large waste volumes, as smaller or variable streams may not justify investment.
- **Regulatory challenges:** Compliance with local, national, and international standards is essential, with strict requirements for emissions (e.g., incineration gases must meet standards) and waste form stability. Regulators impose rigorous disposal criteria, and upscaling often requires additional permitting, increasing complexity, and a timeline.

5.2.2.2 Conditioning upscaling (with focus on innovative binder systems)

Upscaling of conditioning processes using alternative binders still faces many challenges, including a lack of standardized testing protocols, variability in waste composition affecting formulation, and the need for long-term durability studies. The application has not been implemented on a large scale, with ongoing research to facilitate its practicality [122].

In general, upscaling the conditioning process using these innovative binder systems involves several challenges, summarized as follows.

Technical challenges:

- **Scaling process consistency:** Maintaining quality and performance at larger scales, as seen in the need for standardized testing protocols for geopolymers. Variability in waste streams can affect the process stability.
- **Equipment and handling:** Large-scale production requires specialized equipment, especially for MPC, because of its exothermic reaction and remote handling of high-activity waste in hot cells. However, the geopolymer matrix requires more storage for raw materials and precise control of the activator dosages. The mixer often requires higher power to handle a more viscous geopolymer matrix.
- **Long-term durability:** Ensuring that waste forms remain stable over extended periods, with geopolymers and low-carbon cements requiring further durability studies [123] [124].

Regulatory challenges:

- **Approval for new matrix:** Obtaining regulatory approval for geopolymers, low-carbon cement, and MPC, given the lack of established standards, as highlighted by IAEA's efforts to benchmark testing protocols [102].
- **Compliance with standards:** Meeting stringent requirements for emissions, leachability, and waste form stability may delay implementation [125].

Economic challenges:

- **High initial costs:** Significant investments are required for new facilities. Ensuring economic viability, especially for smaller waste volumes, is a key issue for industrial implementation.

In summary, scaling up and industrial implementation of new treatment and conditioning processes for radioactive waste requires interdisciplinary collaboration, technological innovation, and adherence to strict regulatory frameworks. By leveraging advancements in materials science, process engineering, and automation, industries can achieve safer and more efficient long-term waste-management solutions. Drawing upon previous experiences from organizations such as the International Atomic Energy Agency and national waste management agencies, continued collaboration and knowledge sharing will be crucial to further refine these technologies and ensure their successful deployment on an industrial scale.

5.2.3 Case study at Hanford Site's Vitrification Plant

The Hanford Site (https://www.hanfordvitplant.com/about-project), a major nuclear waste cleanup site, is an upscaling vitrification site to treat 56 million gallons of waste. The Waste Treatment Plant, under construction since 2002, aims to immobilize waste in glass, but has encountered significant challenges, including cost overruns and technical difficulties, highlighting the complexities of large-scale implementation.

The Hanford Site, with 56 million gallons of radioactive waste in 177 underground tanks, exemplifies upscaling challenges. The Waste Treatment Plant under construction since 2002 aims to vitrify HLW and immobilize it in glass for safe disposal. The construction faced delays due to technical issues, paused in 2012, and resumed in 2022 with over $200 million spent, yet challenges like waste stream variability and facility design remain. The U.S. GAO highlighted in 2023 that DOE's analysis of alternatives was limited, potentially missing cost-saving options like mixing waste with concrete instead of glass, which could save tens of billions. The plant is not yet operational, with completion projected beyond 2084 at a cost of about $341 billion. The lessons learned include the need for independent validation of analyses and early addressing of technical risk. This case study illustrates the complexities of upscaling with significant cost and schedule implications and underscores the importance of robust planning and regulatory oversight.

5.3 NDT and monitoring for Quality Assurance of waste packages

This report provides a comprehensive exploration of the current state of the art in monitoring radioactive waste packages using innovative non-destructive techniques, focusing on methods that do not damage or alter the waste package. The analysis is based on recent research and technical reports and aims to offer detailed insights for stakeholders in nuclear waste management, including regulators, facility operators, and researchers.

Non-destructive techniques are preferred for monitoring because they allow for repeated assessments without compromising the integrity of the package, which is critical for long-term storage and disposal. These techniques are particularly important for verifying the content, checking for leaks, and ensuring structural integrity, especially in high-radiation environments where direct human intervention is limited. Non-destructive techniques involve direct analysis of materials by observing their spontaneous emission of nuclear radiation or using external probes, such as gamma rays and neutrons, without physically altering the package. Common methods include gamma-ray spectroscopy, neutron counting, and tomographic imaging, which are used for both characterization and ongoing monitoring. The focus is on innovative and advanced applications, particularly those developed or enhanced in recent years.

5.3.1 Tomographic Gamma Scanning (TGS)

TGS is a sophisticated non-destructive assay (NDA) method that uses gamma-ray tomography to create a low-resolution three-dimensional image of the radioactive material distribution within the waste package. This technique improves upon the Segmented Gamma Scanner (SGS) by performing emission and transmission scans, yielding both spatial and activity information. The images were used to make accurate point-to-point attenuation corrections, enhancing the assay accuracy for heterogeneous waste. TGS is particularly valuable for drummed waste, with field applications demonstrated at sites such as Rocky Flats for uranium-contaminated waste and LANL for heat-source plutonium, as summarized in a 2014 OSTI report [126].

5.3.2 Gamma Ray Spectroscopy

Gamma-ray spectroscopy is another critical NDA technique that identifies and quantifies gamma-emitting radionuclides by analysing the energy spectrum of the emitted gamma rays. This method is essential for determining the types and amounts of radioactive materials present, aiding in waste classification and compliance with regulatory thresholds, such as the alpha activity threshold of < 0.37 MBq/kg (10^{-2} Ci/t) for characterization of waste packages at Andra, France [127]. It is often used in combination with TGS to provide detailed isotopic information, thereby enhancing the overall characterization process. Recent applications include sensitive imaging of actinide materials in shielded radioactive wastes, as noted in a recent publication [128].

5.3.3 Neutron Counting

Neutron counting measures neutron-emitting radionuclides such as plutonium isotopes using passive or active methods. Passive neutron counting detects spontaneous fission neutrons, whereas active methods such as neutron interrogation induce fission to measure the response. This technique is crucial for assessing transuranic (TRU) waste and is often combined with gamma-ray methods for comprehensive analysis [129].

5.3.4 Combined NDA Techniques

The integration of multiple NDA methods, such as TGS, gamma-ray spectroscopy, and neutron counting, is a growing practice to achieve more accurate and comprehensive characterization. The combined use of neutron multiplicity counting, calorimetry, gravimetry, and gamma-ray spectroscopy has been used to detect partial material defects in waste packages, demonstrating the synergy of these methods [130]. This approach is particularly useful for addressing the challenges posed by the waste matrix density, elemental composition, and distribution heterogeneities, as noted in a technical report [131]

5.3.5 Techniques developed and used in PREDIS WP7 project [132]

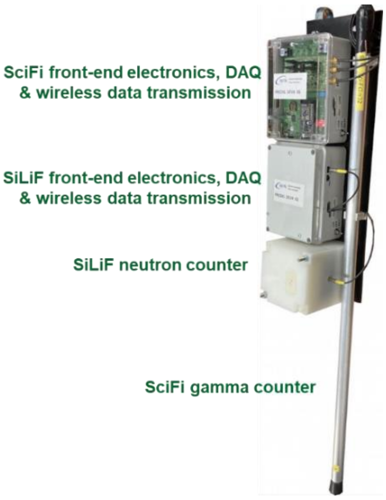
Table 15 summarizes the technologies developed and tested within the WP7 of the PREDIS project.

Technology	**Technology developers**
Scintillating optical Fibre (SciFi) gamma radiation monitoring	INFN, Italy
Silicon Lithium Fluoride (SiLiF) neutron radiation monitoring	
Sensorised Long-range Radio (LoRa) wireless sensor network	UniPi, Italy
Acoustic Emission (AE) for measuring ASR	Magics and SCK CEN, Belgium
Non-contact ultrasonic scanning	NNL, UK
RFID embedded sensors	BAM, Germany VTT, Finland
Muon tomography (Mu-Tom)	INFN, Italy

Table 15 - List of technologies developed and tested within the WP7

5.3.5.1 SciFi (gamma) and SiLiF (neutron) radiation monitoring

Compact flux detection instruments, known as SiLiF neutron counters and SciFi gamma-ray counters, have been developed for external mounting on waste drums. These devices were optimised and evaluated using INFN in PREDIS WP7 (*Figure 10*). The SiLiF neutron counter incorporated a semiconductor detector comprising a silicon diode with a 6LiF neutron converter layer on both sides. The SciFi gamma ray counter features a scintillating fiber with silicon photomultipliers at each end, all encased in an aluminum tube. The proposed monitoring approach involves affixing four SciFi and SiLiF sensors to the cemented drum to enable continuous monitoring throughout the pre-disposal stage.



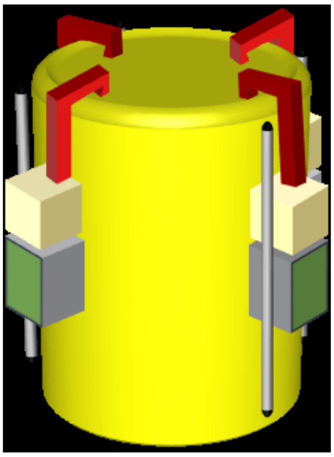


Figure 10.Complete monitoring unit consisting of one SiLiF detector and one SciFi detector with the related electronics (left). A possible arrangement of four radiation monitoring units on a cemented drum during its pre-disposal phase (right).

PREDIS project assessed the viability of extended-term monitoring using radiation detectors placed around radioactive waste containers. This evaluation encompassed initial laboratory experiments, computer-based simulations, and a two-month data gathering phase. The demonstration utilized a cement-based mock-up drum containing a 165 MBq ^{137}Cs gamma source. In addition, the practicality of neutron detection was examined using a PuBe neutron source in a separate test. These findings suggest that this monitoring approach could soon prove to be an invaluable method for the early identification of irregularities or potential interference with drums during their pre-disposal stage in actual industrial settings.

5.3.5.2 Sensorised LoRa Wireless Sensor Network [132]

UniPi has created and evaluated an advanced system for assessing the longevity of waste forms under storage and repository conditions. This system incorporates LoRa technology to facilitate the extended monitoring of radioactivity levels in waste containers and to measure the surface radiation intensity and internal structural integrity. This approach employs passive gamma and neutron counting, and offers ongoing surveillance. By analyzing changes in fluence over time, alterations in the waste matrix structure can be detected, reducing the inconsistencies and human errors typically associated with waste package management. The Wireless Sensor Network proposed by UniPi comprises three integrated levels, as illustrated in Figure 11 -

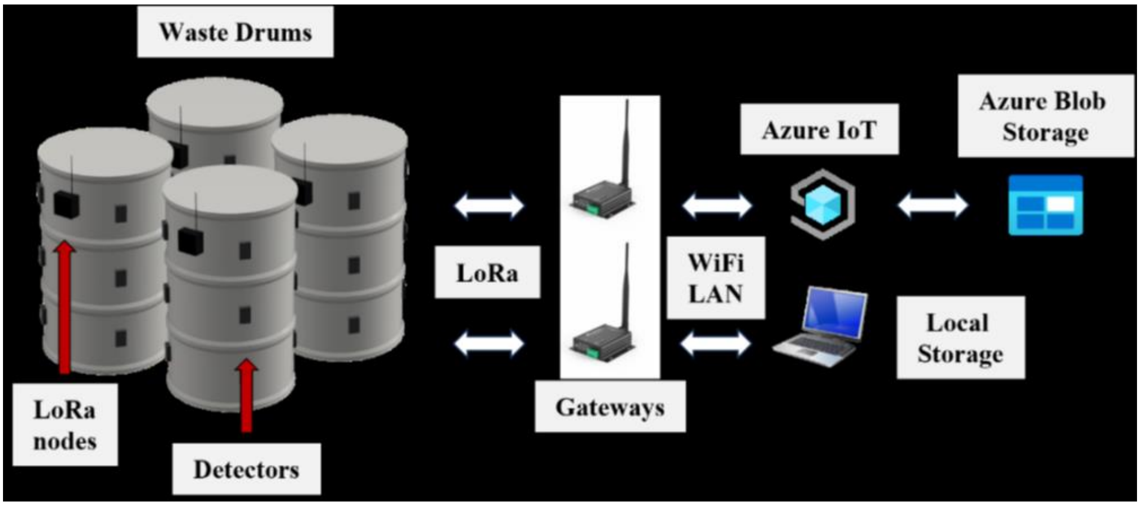


Figure 11 - LoRa radiation monitoring framework for radioactive waste drums

The system, which employs LoRa technology for its energy-efficient, cost-effective, and long-range attributes, underwent initial characterization at UniPi's Laboratory of Nuclear Measurements. It then proceeded to field trials at Nucleco's Rome site and a three-month demonstration at the UJV as part of the PREDIS project. These evaluations confirmed the technology's ability to perform detailed activity monitoring and anomaly detection within containers, showcasing its suitability for remote, automated monitoring without Internet connectivity. The tests assessed crucial aspects, such as radiation detector accuracy, wireless data transmission reliability, battery longevity, and the capacity of the system to identify structural changes within the drum matrix. This validation process established the efficacy of the system for long-term radioactive waste drum surveillance and its adaptability to various operational requirements.

5.3.5.3 Sensor Acoustic Emission for measuring ASR [132]

Acoustic Emission (AE) technology, developed by Magics and SCK CEN, is a non invasive monitoring method employed to evaluate the condition of waste containers. One instance where disturbances might occur in a waste drum is during the alkali–silica reaction (ASR) in cement-based packages. This process results in the creation of a gelatinous substance that expands, generates stress, and potentially causes concrete fractures. The AE technique utilizes highly responsive piezoelectric sensors (Figure 12) placed on the exterior of the package. When a crack forms, the resulting elastic stress wave travels through the material and can be detected using an AE sensor. To identify these AE occurrences, a continuous waveform must be processed. Once events are detected, a cumulative tally of events can be produced as a function of time.

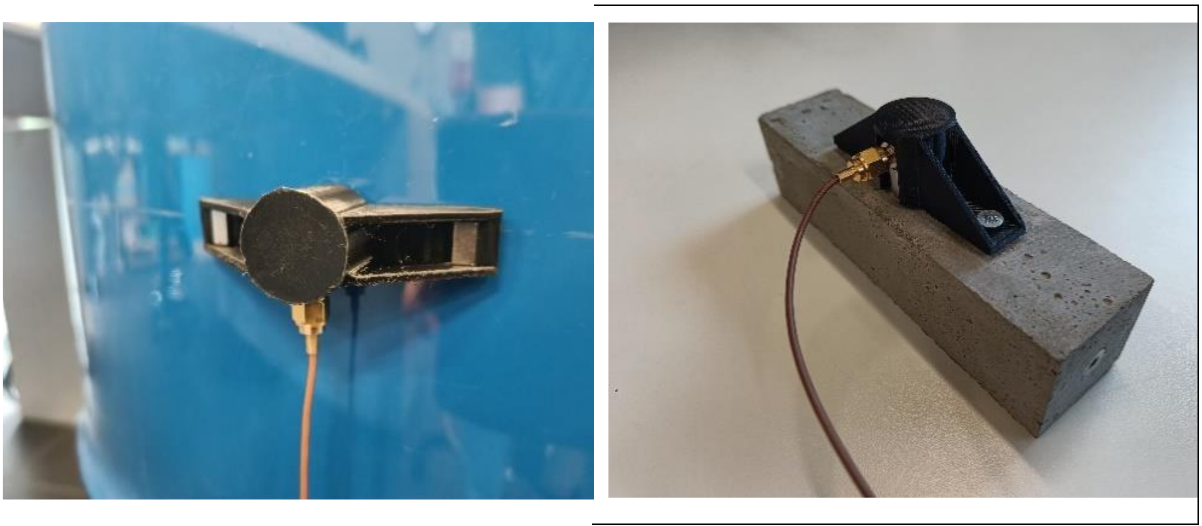


Figure 12 - AE sensor attached to a waste drum (left) and placed on top of a concrete sample for testing (right)

Laboratory-scale experiments conducted in PREDIS demonstrated the viability of using AE to monitor concrete expansion. The initial results indicated a relationship between the total number of acoustic events and the expansion in certain concrete compositions and environmental settings. However, drum-scale experiments failed to reveal distinct expansion signals. Consequently, it was decided to delay the processing and analysis of the gathered AE data until noticeable changes were observed.

5.3.5.4 Muon Tomography [132]

The Mu-Tom technique is a novel and promising non-destructive approach for examining the internal structure of cemented drums. The INFN conducted tests on this method in the PREDIS project. This technique utilizes cosmic ray-generated muons, which are highly penetrative particles that are capable of passing through matter without being absorbed. As muons traverse materials, their trajectories are influenced by multiple Coulomb scatterings. The scattering angle distribution depends on the density, atomic number, and thickness of the material. By examining these scattering angles, Mu-Tom enables the exploration of radioactive waste drum content without using destructive methods. The system employed two muon detectors positioned approximately 3 m apart (Figure 13). This technique can produce three-dimensional images and scan objects at various horizontal levels using a 3D reconstruction algorithm, although the current apparatus has not been optimised for vertical coordinate measurements.

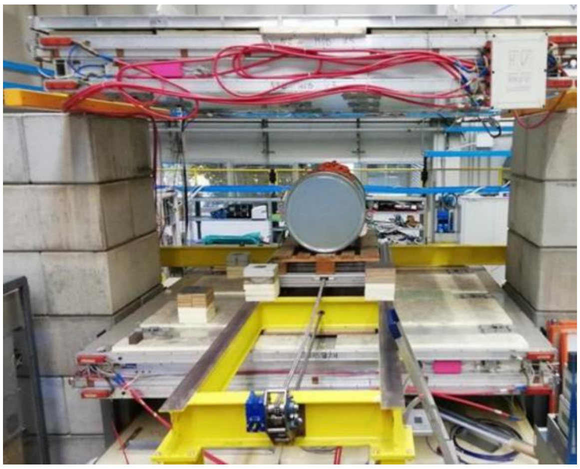


Figure 13 - A picture of the mock-up produced by UJV and installed in the INFN Padova Mu-Tom demonstrator. The two muon detectors are above and below the waste drum.

PREDIS conducted experiments using a simulated waste drum created by a UJV, which contained metallic components encased in concrete to mimic an actual waste container. These trials effectively showcased Mu-Tom's ability to identify metal objects within substantial concrete blocks, representing a notable improvement in non-destructive examination methods for structural evaluation. The subsequent stage of this investigation will focus on broadening data collection to further assess and refine Mu-Tom's capabilities. Through the accumulation and examination of additional information, researchers aim to improve the precision and responsiveness of the technology for detecting and characterizing metallic elements within concrete structures.

5.3.6 Digital twins and waste package monitoring

Digital twins (DT) are virtual representations of physical objects or systems that mirror their real-time status via data integration. In waste package monitoring, digital twins are used to track the condition of radioactive waste packages, such as radiation levels, temperature, and structural integrity, without physical intervention. This is particularly crucial for managing low- and intermediate-level radioactive waste during the pre-disposal phases, including treatment, conditioning, and extended interim storage. Traditional monitoring methods often involve direct measurements, as discussed above, which can pose risks to operators owing to radiation exposure. Digital twins offer a solution by enabling remote real-time monitoring and predictive analysis, reducing human intervention, and enhancing safety [133].

In this context, a DT is a dynamic data-driven model that integrates real-time sensor data, simulation models, and advanced data processing techniques. It allows for the continuous monitoring of waste package parameters, such as gamma and neutron emissions, temperature, and pressure, and can simulate various scenarios to predict future behaviour. This technology supports decision making for safe handling, transport, and acceptance into final disposal facilities, as well as identifying packages requiring remedial treatment. Key technologies enabling digital twins include the Internet of Things (IoT) for sensor data collection, machine learning for data analysis, and 3D visualization for operator interaction. These technologies facilitate cyber-physical integration, where the virtual model updates in real time based on physical measurements, providing a comprehensive view of the status of the waste package.

The state-of-the-art in digital twins for waste package monitoring includes several innovative applications, particularly highlighted in recent projects such as PREDIS. The project aimed to develop, adapt, and demonstrate digital twin technology for waste package monitoring, as detailed in Work Package 7 (WP7), "Innovations in cemented waste handling and pre-disposal storage." The PREDIS project proposed new methods for managing and analysing data from digital twins, including signal processing and data learning techniques [134]

Despite advancements, challenges remain, including the high initial cost of developing digital twin systems, the need for extensive data for training machine learning models, and the integration of diverse data sources. Literature on digital twins for radioactive waste disposal, including waste package monitoring, is sparse, indicating the need for further research and validation.

6. Challenges for new treatment and conditioning technologies

Though for most common waste streams, technologies and practices for waste treatment and conditioning are well defined, certain types of problematic waste streams such as, organic wastes, reactive metals, legacy wastes or even wastes from future fuel cycles and reactors, still do not have a mature and consolidated management route.

New waste streams from D&D operations can pose significant challenges for their treatment and/or conditioning, not only because of their radiological or physic-chemical characteristics but because of the large volumes to treat. Some of these types of waste will not have an identified management route or will require adaptations of existing conditioning techniques [3] [116].

Another issue of increasing interest in nuclear industry is the identification of suitable waste **management routes** for new waste streams generated in the operation of new reactors, **SMR**s and **GEN IV** is essential for their future licensing [4].

Development of new conditioning matrices is mainly driven by the need of finding an adequate conditioning technology for problematic wastes that currently do not have an adequate management route. However, other aspects such as sustainability or the need to secure raw materials used for waste cementation are issues of increasing interest in the current context.

Sustainability of novel management routes is a key point to be assessed as a previous step to industrial implementation [3]. Implementation of principles such as waste minimization or recycling of materials will increase efficiency, reduce volume of waste to be disposed and reduce costs. Applying the principles of circular economy to nuclear decommissioning and waste management will lead to more cost-effective alternative whilst improving environmental sustainability by minimizing waste volumes and optimizing disposal volume.

In that sense, **development of new low-carbon footprint cements** can also aid to lessen environmental impact of conditioning processes. However, there are still uncertainties that need to be addressed. Design and optimisation of alternative cementitious systems (geopolymers, AAMs,..) are aimed to overcome the existing limitations of traditional OPC systems. And though, promising results have been obtained in previous projects [100] [101], there are still relevant issues for their industrial implementation that need to be studied. Further research is needed in issues such as, matrix-waste interactions, disposability assessment or WAC compliance.

New conditioning materials must also exhibit certain flexibility to adapt to **variable volumes** and **variability of physic-chemical characteristics** of non-conventional waste streams, including legacy wastes.

Security of the supplies of cement powder and supplementary materials (**fly ash, blast-furnace slag**) is currently that the nuclear industry is facing. Evolution of cement industry to reduce its carbon footprint has resulted on a risk for the supply of cementitious materials currently in use for LILW conditioning. Changes in physic-chemical characteristics of OPC powders can be a challenge for the manufacturing and qualification of the waste forms [50]. However, this transformation of cement industry has led to the development of novel cements (CEM II/C-M, CEM VI, LC^3, belite-calcium sulfoaluminate cements…) that may offer new prospects to design cement-based matrices and backfill materials with reduced environmental impact.

7. Gaps to be addressed in STREAM WP

Certain types of challenging waste streams require specific conditioning solutions that have not yet been developed or need TRL increase. This may be the case for some ashes, reactive materials, reactive metals, non-incinerable organic materials, sludges or chemo toxic substances. The methods for converting or transforming these wastes into accepted waste forms still need to be studied [3].

STREAM WP is focused on the development and optimization of new or existing treatment and conditioning methodologies for LILW for which no adequate or technological mature solutions are currently available. including metallic wastes, liquid organic wastes, SIERs, sludges or evaporator concentrates. Additionally, to conventional waste streams, conditioning of wastes from new reactors and fuel cycles have also been included in this WP according to the recommendations of the PREDIS Position Paper on EURAD SRA [4].

STREAM scope is aligned with the RD&D needs identified in the treatment and conditioning domains in the PREDIS and EURAD SRAs and addresses most of the common activities of interest specified for areas related to treatment and processing, decontamination and conditioning technologies.

This work package aims to advance on the knowledge generated in previous international initiatives on predisposal topics, like THERAMIN, PREDIS or EURAD-ROUTES projects, in order to increase TRLs of technologies under development in former projects up to TRL 7 (prototype demonstration).

Treatment and processing technologies have reached a high level of maturity and even commercial applications are already available, especially in the case of thermal treatments (e.g. Studsvik FBSR, Belgoprocess Plasma treatment,…). However, not all waste streams can undergo conventional thermal treatments due to their small volumes or their physic-chemical or radiological characteristics. Therefore, further work is required for achieving the industrial implementation of alternative processes, particularly aiming at improving performance, safety, waste minimization and cost-reduction.

In the case of decontamination technologies, gels and foams have attracted considerable attention. A large amount of work has been done in PREDIS project [93]. However, optimization of decontamination solutions is still necessary to improve the efficiency of the processes and to minimise the volume of primary and secondary wastes.

The need of the design and optimization of alternative binders have been extensively investigated in previous national and international projects. Some new cements, such as MPC [6] are ready to move to real-scale demonstration (real scale containers). However, other novel conditioning matrices, like geopolymer and AAMs, still need further optimization (i.e. increase of waste loading), especially for certain challenging wastes.

Other low-carbon binders, such as CEM II/C-M, CEM VI, LC3, belite-calcium or sulfoaluminate cements, can offer new prospects for waste immobilization whilst minimizing environmental impact. Nevertheless, their immobilization capacity and resulting waste form performances still need to be evaluated. Additional work needs to be done to elucidate waste-matrix interactions in the short/medium/long-term and their impact on waste form performance under realistic storage/disposal conditions.

For increasing the maturity level of these novel conditioning materials, aspects such as scale-up or standardization and qualification must be evaluated prior to an industrial implementation phase. Disposability assessment, including the suitability of existing WAC for new waste forms or the need to develop new ones, is a key issue for increasing TRL.

Long-term performance of waste forms is closely related to the definition of WAC for disposal facilities [135]. In STREAM, use of non-destructive control techniques and the development of simulation models has been proposed to support understanding package stability/performance through both long-term storage and final disposal. Despite no experimental work will be done to assess long-term durability of the new waste forms, this WP will benefit from results obtained in WP L'OPERA. Collaboration with this work package is already on-going and additionally to the exchange of data, both experimental and modelled, sharing of samples is foreseen.

Table 16 summarizes the knowledge gaps identified and the correlation with the proposed research activities in STREAM

These identified gaps will be addressed in the three technical tasks of STREAM. Task 3 (*Study of treatment and conditioning methods*) will deal with issues related to the optimization of treatments and alternative binders for the conditioning of the selected wastes. Task 4 (*Scaling-up of treatment and conditioning methods*), on the other hand, will address the challenges of scaling-up, including the development of new monitoring and modelling tools to assist the manufacturing and performance of large scale prototypes. Finally, task 5 (*Deploying safe solutions achieving cost and environmental performances following the principles of circular economy*) will cover some of the remaining uncertainties referred to the suitability of existing WAC and qualification testing protocols for the resulting waste form.

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EURAD Roadmap Domain	***Identified gap(s)***	***R&D activity in STREAM***	***(Sub)task in STREAM WP***	***Expected outcome***
2.1.2. Identify parameters and metrics for waste acceptance criteria through whole life cycle (Waste Acceptance Criteria)	Need to define new WAC for novel wasteforms considering treatment and/or conditioning and storage/disposal phases.	Analysis of existing WAC, considering operational and post-closure assessment (engineer barriers compatibility, diffusion/leaching, short-mid-term behaviour, intruder/accident scenarios….)	5.2. Evaluation of fulfilment of WACs and disposability assessment according to disposal facilities features	Assessment of the suitability of existing WAC to novel-binder WFs (and adaptation if necessary)
2.1.3. Assess potential technologies for the implementation phase, considering cost-benefit ratio and availability (Technology Selection)	Need of mature treatment and conditioning technologies that allow to minimise waste volume or can provide an optimised management route for certain wastes	Demonstration of the upscaling feasibility of treatment and conditioning processes developed in task 3 by a combination of large-scale testing	4.1. Demonstration of up-scaling feasibility of treatment and conditioning processes.	Provide guidance to endusers for the selection of the most suitable treatment and/or conditioning technology considering maturity of technology, cost-effectiveness and environmental aspects.
		LCC/LCA analyses of the most promising processes/materials	5.1. Technical and economic requirements related to the treatment and conditioning matrices	
2.2.2. Minimise the quantity and volume of radioactive waste through pre-treatment and treatment	Improved waste treatment routes, including methods of decontamination and improved segregation, that allow waste minimization and reduction of secondary waste streams,	Design and optimisation of thermal and chemical treatment: i.e low-gradient thermal treatment and heterogeneous Fenton wet oxidation processes	3.1. Optimization of available treatment technologies and conditioning matrices based on alternative binders.	Optimised chemical and physical methods of treatment, including decontamination solutions
		Scaling-up of chemical and gel decontamination processes using surrogates and real radioactive samples	4.1. Demonstration of up-scaling feasibility of treatment and conditioning processes.	Scaling-up to industrial prototyping of the most promising technologies

EURAD Roadmap Domain	***Identified gap(s)***	***R&D activity in STREAM***	***(Sub)task in STREAM WP***	***Expected outcome***
2.2.3. Stabilise waste by conditioning prior to long-term storage (Conditioning)	Development of new cementitious matrices for: challenging wastes without suitable conditioning alternative, to increase waste loadings in waste packages, to improve WF performance under disposal environment	Optimisation of formulations already developed in previous projects, i.e. PREDIS	3.1. Optimization of available treatment technologies and conditioning matrices based on alternative binders.	Guide for the selection of the most appropriate low-carbon binder according to the composition of the waste. Optimisation of waste loading Improve waste form performance
		Development of low-carbon conditioning matrices, assessing the chemical compatibility of these cements with selected waste:	3.2. Development of low-carbon conditioning matrices, assessing the chemical compatibility of these cements with selected waste	
2.3.2. Evaluate potential for improving and optimizing implementation phases with new technologies, to improve costs and environmental impact while maintaining safety and accounting for potential accident scenarios (optimization	Optimization of treatment and conditioning technologies to improve costs and environmental impact	LCA/LCC of most promising methods after optimisation and scaling-up	5.1. Technical and economic requirements related to the treatment and conditioning matrices	Selection of most adequate technology for different waste streams regarding technical, economic and environmental aspects
3.1.3. Cemented LL-ILW (Cemented LL-ILW)	Assessment of physicochemical properties and behaviour of waste packages under storage and disposal conditions	Development of monitoring and NDT methodologies to assess waste package performance	4.1. Demonstration of up-scaling feasibility of treatment and conditioning processes.	Evaluation of real-scale waste package performance
		Modelling thermal and volume stability of the waste package: validation and calibration of codes	4.2. Development of numerical models to simulate and predict the stability of cemented/geopolymer waste packages	Modelling as a tool for assisting scaling-up: identification of most relevant processes and constraints/boundaries for scaling up

EURAD Roadmap Domain	*Identified gap(s)*	*R&D activity in STREAM*	*(Sub)task in STREAM WP*	*Expected outcome*
3.3.2. Backfill component under storage and disposal conditions (Backfills)	Need for more sustainable backfill materials, that allow to minimise waste volume by incorporating recycled materials	Design and characterization of low-carbon binder-based mortars backfill materials, incorporating recycled or secondary aggregates	3.3. Design and characterization of low-carbon binder-based mortars	Development of new backfill materials, considering the option of using recycled materials (especially from D&D operations)

Table 16 – Summary of knowledge and technological gaps to be filled by STREAM WP

Appendix A. Review of treatment and decontamination technologies: industrially-implemented and under development

Type of treatment	**Advantages over traditional routes**	**Processes/parameters that need to be optimised to increase TRL**	**Project phase** *(pilot phase, prototype, lab scaling-up, lab)*	**TRL**
Thermal treatments				
Incineration	Well proven technology. Very high volume reduction of processed waste. Can be used for both solid and liquid wastes	It needs to meet the environmental requirements for discharges. Generally not economical for small amounts of waste. Special regime is required for treatment of alpha bearing waste	Technology deployed for industrial-scale	**TRL 9**
Pyrolysis	Pyrolysis is one of the most attractive thermal methods. It is characterised as a low-temperature flameless process (compared with high-temperature thermal methods, e.g., incineration) in which the organic material is heated in a reducing atmosphere to leave a carbonaceous product or char	Constraints include non-combustible waste, which is unsuitable. Halogenated plastics can be managed (as in the IRIS process) but require additional engineering. The system produces secondary gases that must be filtered and burned before discharging the atmosphere.	Technology in commercial use for radioactive waste treatment. Pyrolysis facilities used: Pyrolysis/incineration Studsvik's, IRIS process (CEA - Valduc, France)	**TRL 9**
Plasma	Plasma technology offers a very effective way of treating this waste with a high-volume reduction factor, is free from organics, liquids, and moisture, and meets the acceptance criteria for safe storage and disposal. Volume reduction factors range from 6 for mainly mixed with metal waste to more than 100 for primarily organic waste	Constraints are unsuitable for wastes containing significant quantities of volatile radionuclides. Secondary waste includes high-efficiency particulate Air (HEPA) filters, slag/sludges, and aqueous solutions, all of which may require subsequent treatment.	Technology deployed for industrial-scale radioactive waste treatment	**TRL 9**

Type of treatment	**Advantages over traditional routes**	**Processes/parameters that need to be optimised to increase TRL**	**Project phase** *(pilot phase, prototype, lab scaling-up, lab)*	**TRL**
Melting	Technology for waste metals as ferrous metals (carbon steel and stainless steel), aluminium, lead, copper and brass. High volume reduction, typically from 5:1 to 20:1. End product has the potential to be reused or recycled within the nuclear industry or after clearance within the conventional metal industry	Pre-sorting is usually required due to dedicated melt furnaces and differences in melt temperatures of the different metals Secondary waste: off gases, slag. Complicated off-gas treatment. Processes of predecontaminated scrap	Technology deployed for industrial-scale radioactive metals	**TRL 8-9**
Molten salt oxidation	Molten salt oxidation (MSO) is a flameless thermal desorption process [2]. Waste is introduced into a bath of molten salts, typically at temperatures between 500 and 950°C. An advantage of MSO over conventional incineration is that acidic gases, produced for example by the decomposition of halogenated organics, react with the carbonate melt and are retained as a salt	Constraints are wastes where C-14 or tritium are significant radionuclides	Technology demonstrated in the industrial environment, and small-scale tests have been undertaken on radioactive waste. Industrial-scale plants for inactive waste are at DEFAC (S. Korea) and NSWC Indian Head (USA)	**TRL 4- 6**
Chemical treatments				

Type of treatment	**Advantages over traditional routes**	**Processes/parameters that need to be optimised to increase TRL**	**Project phase** *(pilot phase, prototype, lab scaling-up, lab)*	**TRL**
Wet oxidation (Fenton)	Fenton oxidation can be classed as an advanced oxidation process, in which H2O2 and a source of Fe2+ ions are utilised to produce hydroxyl radicals in situ, which go on to decompose organic materials. The Fenton process is furthermore of great interest due to rapid Fe/H2O2 reactions, relatively cheap. Cu is a choice for non-Fe Fenton-like oxidation processes, although acting in a potentially different mechanism to Fe. One advantage of copper is the apparent wider pH range over which Cu is an active Fenton-like reagent, with particular efficiency closer to neutral pH.	The ability of Fenton wet oxidation to degrade material at lower temperatures is of particular interest for wastes containing volatile radioisotopes, for which high-temperature processes may require extensive off-gas systems. Due to the continued presence and generation of organic radioactive wastes, and a drive towards safe, final disposition of nuclear wastes, Fenton and Fenton-like wet-oxidation research will likely continue to offer solutions and opportunities for nuclear waste management. The potential for lower temperature processing, with significant reduction in resultant solid wastes could provide another technology in the toolbox of treatment options.	Extensive trials on the implementation of wet oxidation have been undertaken in Italy, Sweden, Japan, USA and the UK. Laboratory scale research, pilot plants and multinational trials have determined that volume reduction and adequate destruction of certain materials are achievable	**TRL 4- 6**
Acid digestion	The acid digestion process is a method that traditionally utilizes the dehydrating action of concentrated sulphuric acid to carbonize solid organic materials and nitric acid to oxidize the carbon	Constraints is waste sizing (by e.g. shredding) prior to digestion for best efficiency. Treatment of halogenated organics is possible but results in the production of hydrochloric acid which requires additional handling. Liquid wastes require further treatment. Suitable methods include neutralisation of the acid solution followed by encapsulation or drying.	The process has been studied in laboratory and bench-scale test units. Acid-digestion unit in Hanford. A 100-kg/d test unit has recently been constructed	**TRL 9 Industrial scale facilities none remain in operation**

Type of treatment	**Advantages over traditional routes**	**Processes/parameters that need to be optimised to increase TRL**	**Project phase** *(pilot phase, prototype, lab scaling-up, lab)*	**TRL**
Supercritical water oxidation	Thermal treatment process in which oxidation occurs in supercritical water with at pressures and temperatures above its critical point (P>22.1 MPa and T>374°C). Supercritical water acts as a very effective solvent and is completely miscible with oxygen and organic liquids, allowing for a very fast and complete oxidation reaction. The process products are a gaseous exhaust (CO2, O2, N) and an aqueous liquid effluent composed of water and salts.	Solid waste must be size reduced. The liquid effluent requires further treatment (some form of dewatering and immobilisation) to be suitable for disposal.	Technical maturity has been implemented for the treatment of radioactive liquid organic waste, although there has only been pilot scale treatment of solid radioactive organic waste	**TRL 6**
Decontamination processes				
Chemical methods				
Chemical gel	Suitable for large surfaces A small amount of applied chemicals High efficiency of activity removal Suitable for intervention within plants	Significant amounts of secondary waste Corrosive and toxic reagents may need to be handled in order to obtain high DFs Not usually effective on porous surfaces	Laboratory scale research, Gossard et al. “Method using ferromagnetic gel”. Patent # WO2022184996. COREMIX gel,	**TRL 4**

Type of treatment	Advantages over traditional routes	Processes/parameters that need to be optimised to increase TRL	Project phase *(pilot phase, prototype, lab scaling-up, lab)*	TRL
EASD® Gel	EASD® Gel is one technology variation that utilises a gel-based electrolyte	The chemical reagent is nitric acid, has formed the basis of the electrolyte-media used in the EASD® Gel technology. It has study compatible with a variety electrolyte solutions and different metallic waste (e.g., stainless steels, nickel-alloys, lead etc.).	Laboratory scale research. Improved decontamination efficiency Spent gel is stored in a plastic container prior to neutralisation and disposal. Potential waste reduction	**TRL 3-4**
Foams	Suitable for large surfaces and items of excessive weight Improve the contact between surface and chemicals Short lifespan (15–30 min) Low secondary waste production if reagent is reused	Spraying requires direct operator intervention and cannot be used for closed volumes (vessels, cavities). Higher exposure of workers. Lifetime of foam is limited. Care must be taken when flushing	Laboratory scale research.	**TRL 3-4**

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