DELIVERABLE REPORT

Thermal treatment for radioactive waste minimisation and hazard reduction

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The report has been reviewed by all THERAMIN Consortium members and, following amendment to address comments received, has been approved by THERAMIN WP5 leader. The THERAMIN coordinator has reviewed and approved the report for submission.

APPROVED FOR SUBMISSION:

This report has been approved by: Dr. Matti Nieminen, THERAMIN Project Coordinator.

Executive Summary

The **The**rmal Treatment for **Ra**dioactive Waste **Min**imisation and Hazard Reduction (THERAMIN) project was a European Commission (EC) programme of work partly funded by the Horizon 2020 Euratom research and innovation programme. THERAMIN ran from June 2017 to May 2020. Twelve European organisations (including nuclear waste management organisations, research institutes and consultancies), from seven European countries (Belgium, France, Germany, Lithuania, Slovakia, Finland and the UK), participated in THERAMIN, forming a community of experts on thermal treatment technologies and radioactive waste management and disposal. In addition, the project included an advisory group of waste owners / site operators and management organisations to provide an end-user view.

The objective of the project was to provide improved safe long-term storage and disposal of intermediate-level wastes (ILW) and low-level wastes (LLW) suitable for thermal processing. Work carried out within the project aimed to identify radioactive wastes that could benefit from thermal treatment, which treatment technologies were under development in participating countries, and how these could be combined to deliver a wide range of benefits. The work programme provided a vehicle for co-ordinated EU-wide research and technology demonstration, and consisted of five Work Packages (WPs): WP1 involved project management and coordination; WP2 evaluated the potential for thermal treatment of waste streams from across Europe; in WP3, the application of selected thermal treatment technologies to radioactive waste management was demonstrated and evaluated; in WP4, the disposability of thermally treated radioactive waste products was assessed; and WP5 concerned the synthesis of project outcomes and their dissemination to interested parties.

The links between the WPs are shown in Figure E.1. The key findings of each WP are summarised in this synthesis report, which acts as an entry point, providing context for the project deliverables developed in each WP and indicating where more detailed information can be found. A shorter summary of the key outcomes of the THERAMIN project has also been published[1].

The project has successfully demonstrated the applicability of six different thermal treatment technologies (SHIVA, In-Can Melter, GeoMelt®, thermal gasification, vitrification and hot isostatic pressing) to a range of waste groups representative of those identified in participating countries. The thermally treated products of these and other trials have also been characterised and these data used to undertake preliminary disposability assessments. Although further studies of long-term behaviour are required (e.g. leaching tests), to fully assess the suitability of the product for safe disposal, characterisation has demonstrated the removal of volatile components, organic complexants and water, which has benefits in terms

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[1] Scourfield, S., Kent, J., Wickham, S., Nieminen, M., Clarke, S. and Frasca, B., (2020). Thermal treatment for radioactive waste minimisation and hazard reduction: overview and summary of the EC THERAMIN project. *IOP Conference Series: Materials Science and Engineering*, 818, p.012001. https://iopscience.iop.org/article/10.1088/1757-899X/818/1/012001

of reducing the potential for gas generation, corrosion of storage and disposal containers and radionuclide transport rates within the disposal facility.

The project was supported and enabled by collation of a summary-level inventory of European radioactive wastes potentially suitable for thermal treatment and available thermal treatment technologies. Finally, THERAMIN developed a value assessment methodology that can be used to identify the benefits and challenges of thermal treatment compared to the baseline options, which can be used to inform strategic consideration of thermal treatment as the baseline for waste groups studied within this project.

The following areas of further work were identified:

- Maintenance and development of the community of thermal treatment specialists developed through the THERAMIN project.
- Gathering of further information to support comparison of thermal treatment technologies against the baseline for specific waste streams.
- Optimisation of thermally treated product composition to increase waste loadings and/or improve wasteform performance.
- Development of waste acceptance criteria for thermally treated products.
- Understanding of the long-term behaviour and chemical durability of thermally treated products.

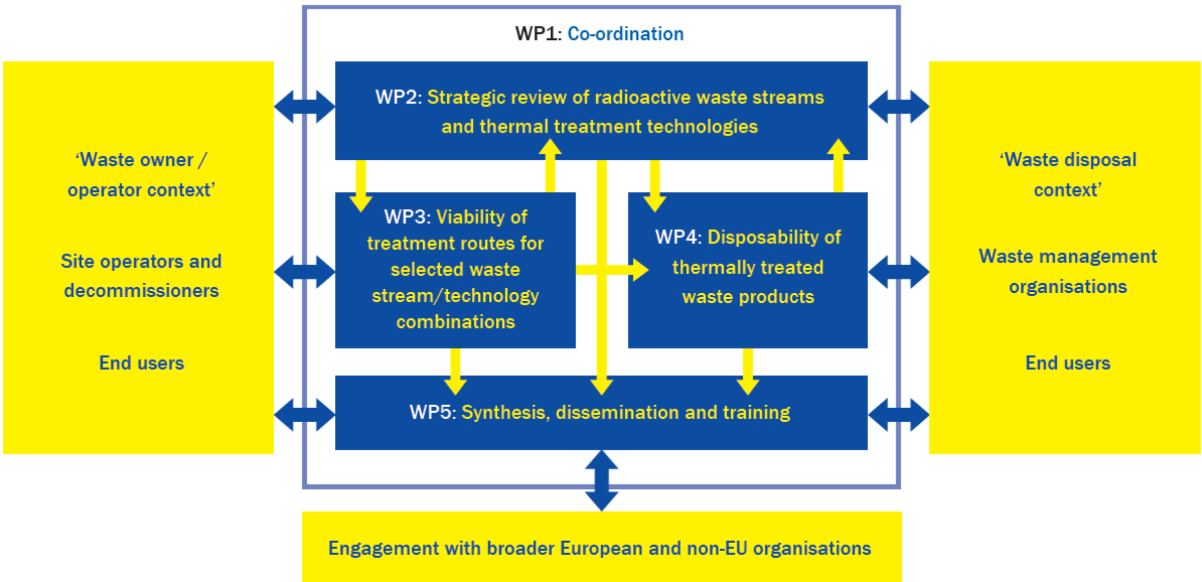

Figure E.1 Structure of the THERAMIN project.

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Acknowledgement

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THERAMIN Project Partners

Andra	Agence nationale pour la gestion des déchets radioactifs – France
CEA	Commissariat à l'énergie atomique et aux énergies alternatives – France
GSL	Galson Sciences Limited – UK
FZJ	Forschungszentrum Jülich GmbH – Germany
LEI	Lithuanian Energy Institute – Lithuania
NNL	National Nuclear Laboratory – UK
ONDRAF/NIRAS	Organisme National des Déchets RAdioactifs et des matières Fissiles enrichies – Belgium
Orano	Orano – France
SCK•CEN	The Belgian Nuclear Research Centre – Belgium
USFD	University of Sheffield – UK
VTT	Teknologian Tutkimuskeskus VTT Oy (VTT Technical Research Centre of Finland Ltd)
VUJE	VUJE a.s. – Slovakia

THERAMIN End User Group

Andra	Agence nationale pour la gestion des déchets radioactifs – France
AWE	The Atomic Weapons Establishment – UK
CEA	Commissariat à l'énergie atomique et aux énergies alternatives – France
EDF	Electricité de France – France
Fortum	Fortum Oy – Finland
INL	Idaho National Laboratory – USA
JAVYS	Jadrová a vyraďovacia spoločnosť - Slovakia
IGD-TP	Implementing Geological Disposal of Radioactive Waste Technology Platform
Nagra	Die Nationale Genossenschaft für die Lagerung Radioaktiver Abfälle – Switzerland
ONDRAF/NIRAS	Organisme National des Déchets RAdioactifs et des matières Fissiles enrichies – Belgium
RWM	Radioactive Waste Management Ltd – UK
Sellafield	Sellafield Ltd – UK
TVO	Teollisuuden Voima Oy – Finland

List of Acronyms

ALARP	As Low as Reasonably Practicable
ASR	Alkali-Silica Reaction
CSA	Centre de Stockage de l'Aube
EC	European Commission
EDX	Energy Dispersive Using X-Ray
GDF	Geological Disposal Facility
HIP	Hot Isostatic Pressing
HLW	High-Level Waste
ICV	In-Container Vitrification
IER	Ion-Exchange Resin
ILW	Intermediate-Level Waste
ISG	International Simple Glass
JEN	Jülicher Entsorgungsgesellschaft für Nuklearanlagen
KTE	Kerntechnische Entsorgung Karlsruhe
LL	Long-Lived
LLW	Low-Level Waste
LLWR	Low Level Waste Repository (UK)
MIND	Microbiology In Nuclear waste Disposal
NDA	Nuclear Decommissioning Authority (UK)
NPP	Nuclear Power Plant
PCM	Plutonium-Contaminated Material
PVC	Polyvinyl chloride
SEM	Scanning electron microscopy
SIAL	Sludge in an Inorganic Aluminosilicate matrix
SL	Short-Lived

SNETP	Sustainable Nuclear Energy Technology Platform
THERAMIN	Thermal treatment for radioactive waste minimisation and hazard reduction
TRL	Technology Readiness Level
VLLW	Very-Low-Level Waste
VNS	Veolia Nuclear Services
WAC	Waste Acceptance Criteria
WMO	Waste Management Organisation
WP	Work Package
XRD	X-ray Diffraction

1 Introduction

1.1 Background to the THERAMIN Project

The **Ther**mal treatment for **Ra**dioactive Waste **Min**imisation and Hazard Reduction (THERAMIN) project was a European Commission (EC) programme of work partly funded by the Horizon 2020 Euratom research and innovation programme. The THERAMIN project ran from June 2017 to May 2020. Twelve European nuclear waste management organisations (WMOs) and research and consultancy institutions from seven European countries participated in the project.

1.1.1 Scope and Objectives of the THERAMIN Project

The overall objective of THERAMIN was to provide improved safe long-term storage and disposal of intermediate-level wastes (ILW) and low-level wastes (LLW) suitable for thermal processing. The work programme provided a vehicle for coordinated EU-wide research and technology demonstration, designed to provide improved understanding and optimisation of the application of thermal treatment in radioactive waste management programmes across Europe, and to move technologies higher up the Technology Readiness Level (TRL) scale.

1.1.2 THERAMIN Project Structure

The THERAMIN project was carried out in five work packages (WPs).

- **WP1: Co-ordination.** This WP included project management and coordination and was led by VTT.
- **WP2: Strategic review of radioactive waste streams.** This WP evaluated the opportunities for thermal treatment of particular waste streams across Europe; this WP was led by GSL.
- **WP3: Viability of treatment routes for selected waste stream/technology combinations.** In this WP, the application of selected thermal treatment technologies to radioactive waste management was demonstrated and evaluated; this WP was led by NNL.
- **WP4: Disposability of waste products.** In this WP, the disposability of the thermally treated radioactive waste products was assessed; this WP was led by Andra.
- **WP5: Synthesis, Dissemination and Training.** This WP was led by GSL and enabled synthesis of the project outcomes and their dissemination to other interested organisations.

The key findings of each WP are summarised in this synthesis report, which acts as an entry point, providing context for the project deliverables developed in each WP and indicating where this detailed information can be found (listed in Table 1.1).

Table 1.1 Deliverables produced during the THERAMIN project (available on the THERAMIN website, except for those shown in red, which are confidential and have not been published).

Deliverable Number	Deliverable Title	Reference
	Work Package 2	
D2.1	Database of European radioactive wastes suitable for thermal treatment.	[1] GSL (2018)
D2.2	European radioactive wastes suitable for thermal treatment.	[2] GSL (2018)
D2.3	Current Status of Thermal Treatment of Radioactive Waste Streams in the European Union.	[3] GSL (2018)
D2.4	Thermal Treatment Viability Matrix.	[4] GSL *et al.* (2020)
D2.5	THERAMIN Value Assessment.	[5] GSL (2020)
	Work Package 3	
D3.1	Matching of waste streams to appropriate technology demonstrators	[6] NNL *et al.* (2018)
D3.2	Test report covering all WP3 demonstrations	[7] NNL *et al.* (2019)
D3.3	SHIVA and IN-Can Melting technologies and demonstration	[8] CEA (2019)
D3.4	Gasification based waste treatment	[9] VTT (2019)
D3.5	GeoMelt Technology	[10] NNL *et al.* (2019b)
D3.6	Hot Isostatic Pressing (HIP) demonstration	[11] NNL and USFD (2019)
D3.7	VICHR technology	[12] VUJE (2019)
	Work Package 4	
D4.1	Waste Acceptance Criteria and requirements in terms of characterisation	[13] ANDRA *et al.* (2018)
D4.2	Characterization of thermally treated waste products	[14] ANDRA *et al.* (2019)
D4.3	Safety case implication studies	[15] ANDRA *et al.* (2019)
D4.4	Synthesis report of WP4	[16] ANDRA (2020)

Deliverable Number	Deliverable Title	Reference
	Work Package 5	
D5.1	External Communication Plan	[17] GSL and USFD (2019)
D5.2	Technical Training School held at CEA's Le Visiatome Centre Marcoule, June 2019	Presentations and posters available on the THERAMIN website [18]
D5.3	Conference Proceedings from the THERAMIN Conference – Manchester, February 2020	[19] USFD and GSL (2020)
D5.4	THERAMIN Project Synthesis Report (this document)	GSL *et al*. (2020)
D5.5	Project Summary Report (submitted as a paper within the conference proceedings)	[20] GSL *et al*. (2020)
D5.6	Plan for exploitation of the results of the THERAMIN project	[21] VTT (2020)
D5.7	Preliminary business plan	[22] VTT (2020)
D5.8	THERAMIN website	[23]

In addition to the formal deliverables listed in Table 1.1, dissemination of knowledge gained from the THERAMIN project and training of early career researchers was achieved through the following activities coordinated by WP5:

- A final THERAMIN project conference in Manchester in February 2020, which was attended by over 80 researchers and industrialists from 10 countries, including contributions from non-EU countries.
- Training placements for early-career researchers and individuals from countries without active thermal treatment plants, hosted by VTT, the University of Sheffield and CEA.
- Annual THERAMIN project newsletters for timely dissemination of key results.
- Presentation of two posters, one produced early in the project describing the project scope, and the other summarising the key outcomes of the project, for use by project participants at international conferences.

1.2 Objectives of the Synthesis Report

The objective of this report is to collate the final THERAMIN project outputs into an overall synthesis report suitable for a technical readership. The report seeks to:

- Act as an entry point for understanding the content and nature of other project deliverables.
- Identify common wastes and common challenges across EU countries, to encourage mutual sharing of experience in the management of specific waste types.
- Identify generic advantages and challenges of thermal treatment technologies and their applicability to specific waste streams.
- Set out understanding of the characteristics and properties of thermally treated wasteforms and their viability for geological disposal, and implications for the safety case.
- Establish preferred routes and potential for international sharing of thermal treatment technologies and facilities to open management routes and opportunities across Europe, including identification of remaining uncertainties, further required R&D, and next steps.

1.3 Structure of the Synthesis Report

The remainder of this report is set out as follows:

- Section 2 describes the approach used to collate a summary-level inventory of European radioactive wastes potentially suitable for thermal treatment, and to make a strategic analysis of the drivers and benefits of applying thermal treatment techniques to these wastes. This section also summarises the availability and level of maturity of thermal treatment technologies in European countries. This work was carried out as part of WP2 of the THERAMIN project.
- Section 3 provides details of the treatment technology demonstrators that were matched with waste streams identified in WP2 (Section 2), followed by an overview of the demonstration of the following thermal treatment solutions: the SHIVA process, the In-Can Melting test, gasification-based waste treatment, GeoMelt® technology, Hot Isostatic Pressing (HIP), and vitrification of Chrompik. This work was carried out as part of WP3 of the THERAMIN project.
- Section 4 reviews the Waste Acceptance Criteria (WAC) of interest for thermally treated wastes in each participating country and identifies the characterisation requirements relevant to these wastes. This section also summarises the characterisation tests carried out on the products of thermally treated wastes and discusses safety case implications. This work was carried out as part of WP4 of the THERAMIN project.

- Section 5 summarises the viability of technologies to treat different waste types, and provides a methodology for assessing the benefits and challenges of thermal treatment of different waste types, based on the work carried out in THERAMIN. This work was carried out as part of WP2 of the THERAMIN project.
- Section 6 summarises the project outcomes and identifies remaining uncertainties, further required R&D, and next steps.

2 Work Package 2: Strategic Review of Radioactive Waste Streams

Under Work Package 2 (WP2) of THERAMIN, strategic reviews of thermal treatment technologies and European radioactive waste streams for which thermal treatment could bring benefits have been undertaken. WP2 was structured in five linked tasks:

- Task 2.1: development of a European database of radioactive waste groups suitable for thermal treatment (THERAMIN project Deliverable D2.1 [1]).
- Task 2.2: implementing a strategic analysis of the benefits of thermal treatment of the waste groups identified and the risks and barriers that exist to such treatment (THERAMIN project Deliverable D2.2 [2]).
- Task 2.3: summarising the availability and maturity of different thermal treatment technologies (THERAMIN project Deliverable D2.3 [3]).
- Task 2.4: constructing a summary matrix of waste groups and applicable thermal treatment technologies (THERAMIN project Deliverable D2.4 [4]).
- Task 2.5: undertaking a value assessment of the strengths and challenges of selected waste group / thermal treatment combinations (THERAMIN project Deliverable D2.5 [5]).

Many of the findings of WP2 were commercially sensitive, therefore the deliverables from these tasks (D2.1, D2.2, D2.3 and D2.4) are confidential; only D2.5 is available in the public domain. A high-level summary of the findings from these tasks are summarised in this report in the following sections:

- Task 2.1 and Task 2.2 are summarised in Section 2.1.
- Task 2.3 is summarised in Section 2.2.
- Task 2.4 and Task 2.5 were conducted throughout the project, utilising the outputs of other work packages, and are described in Section 5.1 and Section 5.2, respectively.

2.1 Strategic Analysis of Waste Streams

A Europe-wide review was undertaken to gather inventory information on radioactive waste streams that could benefit from thermal treatment. The information was gathered by means of a questionnaire completed by THERAMIN Partner and End User Group countries (Belgium, Finland, France, Germany, Lithuania, Slovakia, Switzerland, UK). In addition, the European Commission (EC) encouraged inclusion of Ukraine, and its inventory was also captured in the waste database. Waste information was also provided by the EC project Microbiology In Nuclear waste Disposal (MIND), which resulted in the addition of the following countries: the Czech Republic, the Netherlands, Spain, and Sweden.

The waste streams identified were categorised according to waste type and composition into 14 generic waste groups (Table 2.1) and compiled in an electronic database for presentation and data analysis (Task 2.1, THERAMIN Deliverable D2.1 [1]). Each waste group may have specific issues associated with treatment, processing, packaging or transport, although for some groups the issues are common. The categorisation into generic groups was based on commonalities in the waste stream properties and composition. There are common waste types that occur in several countries, such as sludges, ion-exchange materials, cement-conditioned wastes, and bitumen-conditioned waste, whilst others were identified as present in fewer countries (e.g. polymer-conditioned waste and non-organic liquid wastes).

The database was used to generate volume information for the generic waste groups across the countries reviewed. Figure 2.1 provides a summary of the breakdown of wastes (both existing and future arisings) covered in the database, excluding Ukraine, which would dominate if included because its inventory, which represents the national inventory, is volumetrically much larger than those of other countries. It is noted that the estimated volumes of waste are those that have been deemed potentially suitable for thermal treatment by country contacts filling out the questionnaires. They do not represent the total inventories in the listed countries. In addition, the listed countries in Figure 2.1 are those that have identified a particular waste as a potential candidate for thermal treatment, noting that thermal treatment may be an alternative to other treatment options, rather than an optimized solution. Other countries not listed may hold similar waste but manage it via an already established route.

The database was also used to enable a strategic analysis of the benefits of thermal treatment. The analysis considered the drivers for thermal treatment, and the risks and barriers to applying this treatment. For each country, the analysis described the risks if the wastes are left untreated and potentially without a management route, and identified those wastes for which thermal treatment is likely to bring significant benefits. This analysis was supported by a brief description of the national context and programme status in each participating country. The database and strategic analysis are documented in a report (Task 2.2, THERAMIN Deliverable D2.2 [2]).

Table 2.1 Generic waste groups defined in [1].

Generic waste group	Description
Alpha waste (including PCM)	Material contaminated with alpha-emitting radionuclides (e.g., plutonium, uranium, etc.). This waste includes plutonium-contaminated material (PCM).
Bitumen-conditioned waste	Wastes that have been conditioned in a bitumen matrix. The nature of the original raw waste is varied.
Cement-conditioned solid waste	Wastes that have been conditioned in a cementitious matrix. The nature of the original raw waste is varied.
Filters	Filters are used to remove radionuclides and particulates from contaminated air or other media. Example filters include: HEPA, charcoal filters, and cartridge filters used to remove radionuclides and particulates from active effluent.
Graphite	Waste graphite from decommissioning of reactors that used graphite as part of the reactor design. This could include core graphite or graphite debris from the fuel assemblies.

Generic waste group	Description
Hazardous or chemotoxic waste	Wastes which have chemotoxic properties (e.g., Be, Cd, Hg) or which are hazardous (e.g., asbestos).
Inorganic ion-exchange material	Ion-exchange materials used for the removal of soluble radionuclides (e.g., caesium) from liquid waste (e.g., irradiated fuel cooling pond water). Example inorganic resins include: zeolites, Ionsiv® and clays.
Metallic waste (pure or high content)	Waste containing pure metal or metal mixed with other materials.
Miscellaneous contaminated solid waste (including PVC)	Other miscellaneous solid waste that is non-metallic, e.g., maintenance wastes, decommissioning wastes, contaminated gravel, concrete, polyvinyl chloride (PVC), etc.
Polymer-conditioned waste	Wastes that have been conditioned in a polymer matrix. The nature of the original raw waste is varied.
Organic ion-exchange material	Ion-exchange materials composed of high-molecular-weight polymers. They are also used for the removal of soluble radionuclides from solution.
Other liquid waste (e.g. Chrompik)	Contaminated aqueous liquids which do not contain organics.
Organic liquids and oils	Contaminated liquid waste which contains organics such as oils or solvents.
Sludge and concentrates	Includes bulk sludge, residuals, and concentrates. Sludges arise in tanks, sumps and ponds, and typically comprise a mixture of particulate materials and water.

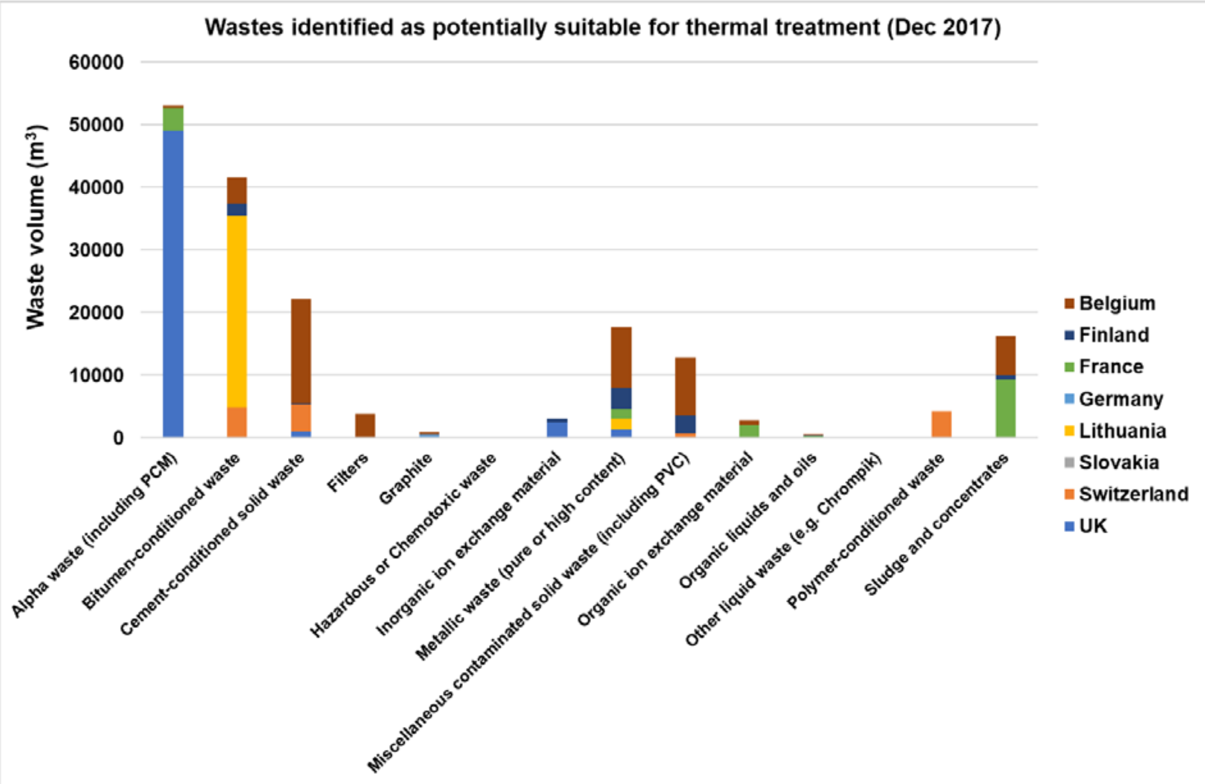

Figure 2.1 Waste volumes (existing and future arisings) identified for thermal treatment, grouped by generic waste group (excluding Ukrainian wastes). Note that the estimates provided correspond to waste streams identified as potentially suitable for thermal treatment and do not represent national inventories. Note also that if a particular waste has been mapped to more than one generic waste group (due to its heterogeneity for example), then its volume has been included in all of those waste groups

2.2 Availability and Planned Development of Thermal Treatment Technologies

Under Task 2.3, a report was produced describing potential thermal technologies available in Europe, and considering, at a high level, the range of wastes and materials that they have demonstrably treated, or are theoretically capable of treating [3]. The objective of Task 2.3 was to summarise the availability and maturity of thermal treatment technologies in Europe by:

- Providing a summary of the current status of each thermal technology with regard to treatment and processing of radioactive wastes.
- Evaluating the availability of thermal technologies to countries with significant waste arisings that could benefit from thermal treatment and processing.
- Collecting information to support production of a viability matrix and value assessment.

At a high level, the thermal technologies were first classified into three ***process categories*** according to the type of product produced as follows:

- Thermal processes that are expected to generate a product that requires additional conditioning (e.g., encapsulation) to allow it to become suitable for disposal. These technologies can be described as treatment processes that are employed to reduce chemical reactivity and/or potentially reduce volume.
- Thermal processes involving immobilisation of radioactive waste by encapsulation in a glass matrix or by direct incorporation into the glass matrix.
- Thermal processes involving immobilisation of radioactive waste by incorporation in a ceramic matrix or in a glass/ceramic composite matrix.

For each process category above, a sub-level of classification was used to sort existing technologies by heating mode and process wall material (e.g. refractory or cold wall, etc.). For each technology, examples of existing facilities (commercial or laboratory scale) in Europe were then provided. The example facilities were only used to illustrate the possible variations that may exist within a treatment technology, and therefore are not exhaustive. Table 2.1 provides a summary of the classification that was adopted.

Table 2.2 Classification of thermal treatment and processing technologies considered in THERAMIN.

<table>
<tr><th>High-level process</th><th>Technology</th><th>Facility</th><th>Country</th></tr>
<tr><td rowspan="13">Treatment for volume reduction and passivation</td><td rowspan="3">Incineration with burner and refractory walls</td><td>JÜV 50/2 - Jülich JEN</td><td>Germany</td></tr>
<tr><td>KTE incinerator</td><td>Germany</td></tr>
<tr><td>EDF CENTRACO</td><td>France</td></tr>
<tr><td>Rotary kiln incineration</td><td>IRIS</td><td>France</td></tr>
<tr><td>Pyrolysis</td><td>Belgoprocess (decommissioned)</td><td>Belgium</td></tr>
<tr><td>Thermal gasification</td><td>VTT gasification</td><td>Finland</td></tr>
<tr><td>Calcination</td><td>Widely used</td><td>France, UK</td></tr>
<tr><td>Underwater plasma incineration</td><td>ELIPSE</td><td>France</td></tr>
<tr><td>Hydrothermal Oxidation (HTO)</td><td>DELOS</td><td>France</td></tr>
<tr><td rowspan="3">Induction metal melter</td><td>CARLA</td><td>Germany</td></tr>
<tr><td>EDF CENTRACO</td><td>France</td></tr>
<tr><td>Cyclife (formerly Studsvik)</td><td>Sweden</td></tr>
<tr><td rowspan="2">Conditioning by immobilisation in glass</td><td rowspan="2">Joule-Heated In-Can Vitrification</td><td>In-Can Melter and DEM & MELT (metallic inner wall), CEA</td><td>France</td></tr>
<tr><td>GeoMelt® (ceramic inner wall), NNL</td><td>UK</td></tr>
</table>

High-level process	Technology	Facility	Country
	Joule-Heated Ceramic Melter (JHCM) [1]	VEK, PAMELA (both decommissioned)	Germany, Belgium
	Cold crucible induction melter (CCIM)	La Hague CCIM and Marcoule CCIM pilot	France
	Advanced CCIM (A-CCIM)	Marcoule A-CCIM pilot	France
	Indirect induction (metallic wall - hot metal pot)	VICHR	Slovakia
		La Hague and Sellafield	France, UK
	Coupled cold wall direct metal induction melting and plasma burner	PIVIC	France
	Coupled cold wall direct glass induction melting and plasma burner	SHIVA	France
	Refractory wall plasma burning and melting	Retech (ZWILAG)	Switzerland
		EUROPLASMA – Belgoprocess	Bulgaria
		Tetronics	UK
Conditioning by immobilisation in ceramic or glass-ceramic	HIP	NNL – Workington and University of Sheffield	UK

[1] Currently used and in development outside Europe for vitrification of legacy wastes, including ILW and LLW sludges in the United States (Savannah River and Hanford).

A process summary table was generated for each technology in order to provide information to support Tasks 2.4 (viability matrix) and 2.5 (value assessment). For each of the European facilities included in the technology survey, information was provided by THERAMIN Partners on the following topics:

- Waste compatibility with technology (solid, liquid, organic, inorganic, metal, level of activity, etc.).
- Mode of heating.
- Containment.
- Nature of product.
- Limitations of technology.
- Maximum volumetric throughput.
- Continuous or batch process.
- Secondary waste generation.
- Maturity of technology (TRL).
- Target date for active commissioning of full-scale industrial facility.
- Technological complexity.
- Flexibility to treat a wide range of waste types.
- For specific facilities, the size of furnace or crucible (or volume of waste that can be treated in one batch).
- Scaleability.
- Investment and operational costs.

The information collected under Task 2.1 (wastes) and Task 2.3 (technologies) was used to assess the suitability of different technologies to treat specific waste types. The waste types were organised in the same way as in the THERAMIN waste database so that they could be linked directly to specific countries and, if needed, to specific waste streams.

3 Work Package 3: Viability of Treatment Routes for Selected Waste Stream/Technology Combinations

The objective of WP3 was to demonstrate the suitability of a range of thermal treatment processes for the immobilisation of waste streams identified in WP2. The demonstrations that were carried out produced samples for the characterisation tests undertaken within WP4. WP3 was split into two tasks:

- Task 3.1: matching the generic waste streams identified in WP2 to appropriate technology demonstrators at facilities made available by project Partners. (Section 3.1 and THERAMIN project Deliverable D3.1 [6]).
- Task 3.2: demonstrating thermal treatment solutions; this task was split into six sub-tasks, each concerning one of the waste stream – demonstrator combinations that were matched in Task 3.1 (Section 3.2 and THERAMIN project Deliverable D3.2 [7]).

3.1 Matching of waste streams to appropriate technology demonstrators

WP2 developed a list of generic waste groups (Table 2.1), including pre-treated and raw waste materials suitable for thermal treatment demonstration, which were recommended for consideration within WP3.

Technology owners matched waste streams from this list with existing rigs and facilities on which demonstration trials could be carried out. It was recognised that no new facilities would be constructed specifically for this work, and that therefore the study would need to be undertaken using technology demonstrators or processes already in existence.

Table 3.1 gives details of the technology demonstrator/waste stream combinations selected.

Table 3.1 Technology demonstrator/waste stream combination selected for trials within WP3.

Facility	Organisation	Waste Stream	Waste Category	Product
SHIVA	CEA/Orano, France	Mixture of organic and inorganic ion-exchange material	Unconditioned waste	Glass
In-Can Melter	CEA/Orano, France	Ash	Unconditioned waste	Glass
GeoMelt®	NNL/VNS, UK	Cementitious waste	Conditioned waste	Glass
		Heterogenous sludges	Unconditioned waste	Glass
Gasification	VTT, Finland	Organic ion-exchange material	Unconditioned waste	Solid residue
Hot Isostatic Pressing (HIP)	USFD, UK	Corroded Magnox sludge containing uranium	Unconditioned waste	Glass-ceramic
	NNL, UK	Corroded Magnox sludge and clinoptilolite containing uranium surrogates	Unconditioned waste	Glass-ceramic
Vitrification	JAVYS, Inc./VUJE, Slovakia	Chrompik	Liquid wastes	Glass

3.2 Demonstration of thermal treatment solutions

The demonstrations carried out by project Partners in WP3, as detailed in Table 3.1, are summarised in this section. Full details for each of the six demonstrations can be found in the following THERAMIN Project deliverables:

- Deliverable D3.3 [8] – SHIVA and In-Can Melter demonstrations (Section 3.2.1 and Section 3.2.2, respectively).
- Deliverable D3.4 [9] – Gasification demonstration (Section 3.2.3).
- Deliverable D3.5 [10] – GeoMelt® demonstration (Section 3.2.4).
- Deliverable D3.6 [11] – HIP demonstrations (Section 3.2.5).
- Deliverable D3.7 [12] – Vitrification demonstration (Section 3.2.6).

3.2.1 SHIVA demonstration

The SHIVA process is an incineration–vitrification process that combines cold wall direct glass induction melting and plasma burner technologies. It has been used in the THERAMIN project by the CEA, France, to demonstrate the processing of a waste stream containing organic ion exchange resins (IER), chosen because of the capability of the plasma treatment facility to destroy the organic component. The resulting phases are subsequently vitrified by the induction heating system to generate a glass product. Full details of this demonstration can be found in the THERAMIN project Deliverable D3.3 [8].

The SHIVA process is shown in Figure 3.1. The set-up consists of a water-cooled, stainless-steel cylindrical reactor, equipped with a flat coil at the bottom and a transferred arc plasma system in the reactor chamber. The SHIVA bottom structure is built to be transparent to the electromagnetic field such that the glass is directly heated by the field while the cylindrical shell is not. The plasma torches were developed to minimize their maintenance requirements. The consumable electrodes are automatically fed. As oxygen is used and no secondary chamber is present, the off-gas treatment is simple - it consists of an electrostatic tubular filter and a gas scrubber. The dust in the filter is recovered in a bottom ashtray for recycling. The glass product is then drained from the cold crucible.

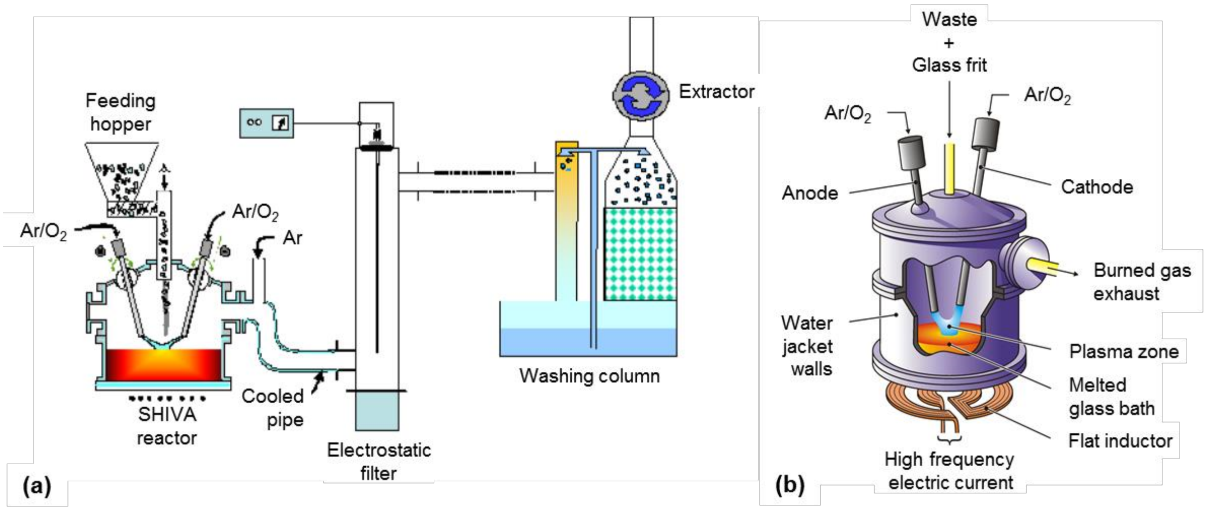

Figure 3.1 A Simplified diagram of the SHIVA process (a) and an artist's view of the reactor (b).

The waste selected for use in the trial was a 25 kg mixture of inorganic and organic ion-exchange media composed of zeolites, diatoms, and IER. 40 kg of a pre-formed glass frit was preloaded into the reactor to provide a melt pool to vitrify the residue from the plasma treatment. Inputs to the process were therefore composed of 38.5% waste and 61.5% glass frit.

The waste feed was introduced to the upper part of the reactor using a feeding hopper and a worm screw in three separate 8-hour feed campaigns. This sequencing was used to allow daytime operation for the waste incineration. The induction heating was started using a titanium starter ring and the generator power was incrementally increased from 40 kW up to 90 kW.

The SHIVA trial demonstrated the successful thermal treatment of a mixture of organic and mineral waste composed of zeolites, diatoms and ion-exchange resins. The waste loading of 38 wt.% is high and could probably be increased in future. The vitrified product appeared from visual observation to be homogenous.

3.2.2 In-Can Melting demonstration

The In-Can Melter trial was designed to demonstrate the feasibility of confining by-products of existing incineration processes (in this case, inactive ash resulting from the incineration of technological waste) in a vitreous matrix. It has been used in the THERAMIN project by the CEA, France. Full detail of this demonstration can be found in the THERAMIN project Deliverable D3.3 [8].

The In-Can Melter consists of a metallic crucible heated in a refractory furnace using electrical resistors, allowing in-container vitrification (Figure 3.2). The process is simple, compact, and well suited to decommissioning waste, as liquid waste steams can be fed directly to the canister inside the furnace without a separate calcination step. The can is single use and forms the product container for disposal.

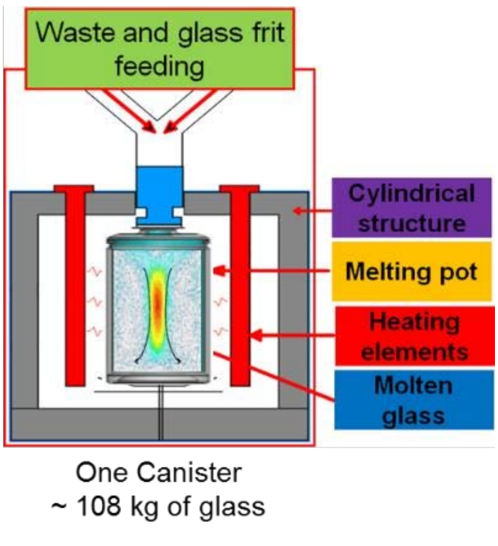

Figure 3.2 Schematic of the In-Can Melter system.

The waste selected for this vitrification trial was ash obtained from multiple incineration tests of surrogate technological waste (polyvinyl chloride, latex, neoprene, polyethylene, cotton, etc.) produced by the CEA IRIS process. IRIS is a research facility for the incineration of solids developed to treat organic waste from glove boxes in the nuclear industry, contaminated with alpha bearing actinides and containing high quantities of chlorine. The feed material was formed into pellets with 10% bentonite to avoid clogging of the feeding pipe and dust carry-over into the off-gas train.

Prior to the full-scale trial (≈50 kg), laboratory-scale (≈10 g) and bench-scale (≈1 kg) tests were carried out to determine the optimum waste to glass frit ratio. The full-scale test was carried out using the In-Can mock-up (DIVA), equipped with a resistive furnace (35 kW), a complete gas treatment system and an Inconel 601 can with an outer diameter of 400 mm, a height of 600 mm and a wall thickness of 10 mm.

The can was pre-loaded with glass frit and non-pelletized ash, then heated to 1100°C at a rate of $300°C \cdot h^{-1}$. Ash pellets were then introduced into the can via the solid feeding system, followed by 1 kg of glass frit. The mixture was soaked for two hours. Following this, recycling of the dust scrubber into the can took place over a period of two hours followed by a final period of soaking for two hours at 1100°C. The can was allowed to cool naturally to ambient temperature.

The trial demonstrated that the In-Can Melter process can successfully vitrify ash resulting from the incineration of organic waste from glove boxes, achieving a waste loading of 50 wt.%. The trial also involved the development of technical methods to form pellets to enable processing of powdery solids, avoiding excessive dust formation.

3.2.3 Gasification demonstration

Thermal gasification is a technology most often used to produce energy from carbon-containing fuels. The process is used to produce fuel gas for direct combustion in power plants, industrial kilns and gas turbines, etc. Gasification enables the production of a combustible gas from different wastes and, following gas clean-up, can be combusted as a clean gas. This technology can be applied to a variety of different wastes for energy production and can also be used to treat certain radioactive waste. In addition to the production of a clean combustible gas, the process generates by-products in the form of a bottom ash from the fluidised bed reactor, and filter dust from the off-gas abatement system.

The gasification test trials conducted as part of the THERAMIN project were carried out with an atmospheric pressure pilot-scale Circulating Fluidised-Bed (CFB) gasification test rig at Bioruukki, VTT's Piloting Center. Full details of these demonstrations can be found in the THERAMIN project Deliverable D3.4 [9].

The waste stream used during the gasification test trials was unused organic ion-exchange resin impregnated with CsCl to simulate radioactive Cs in a real spent IER from a nuclear power plant (NPP). Approximately 520 kg of IER was partially dried, reducing the moisture content from 50 wt.% to about 40 wt.%. A measured amount of CsCl solution was added to the pre-dried batch of IER, stirred for 1-2 hours before being was dried again, resulting in a final moisture content of approximately 30 wt.%. Cs concentration was targeted to be 4 ppm.

A total of 325 kg of organic IER was treated during three test trial days in October 2018. The total duration of the trials was 26.5 hours. The average gasification temperature was 885 to 915°C, and the filtering temperature varied between 415 and 450°C. Inert Al_2O_3 (particle size of 0.18-0.25 mm) was used as a bed material in the CFB gasification reactor.

The efficiency of removal of organic matter from the IER, i.e. carbon conversion, was calculated as conversion of feedstock carbon into the gaseous carbon compounds and tars in the product gas, which is conditioned further during the process. The verified carbon conversion to gas and tars was 92-96 wt.%, which confirms a successful conversion rate.

The main final residue is filter dust, which requires further processing in order to immobilise it and enable safe disposal. In these test trials, VTT immobilised these residues via geopolymerisation. Samples of the geopolymerised products were taken for characterisation within WP4, further details of which can be found in Section 4.2.

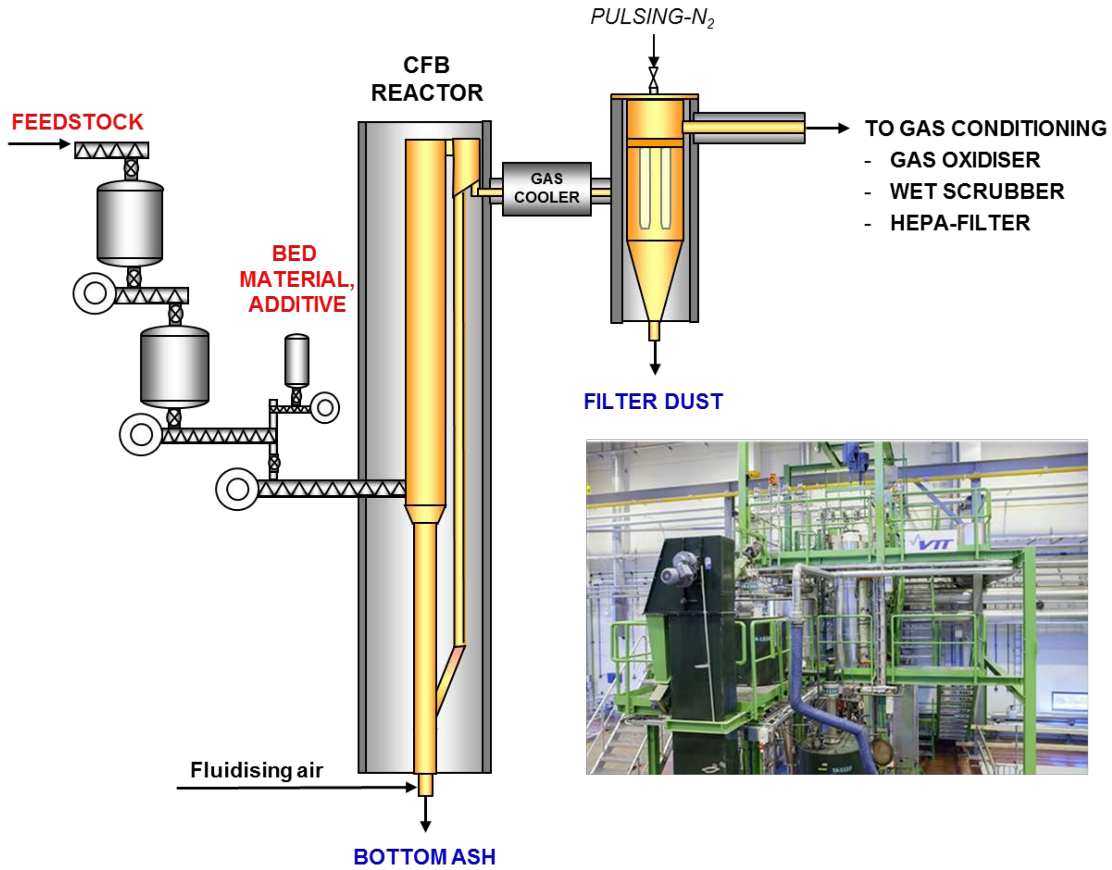

Figure 3.3 The pilot-scale Circulation Fluidised Bed (CFB) gasification test rig.

3.2.4 GeoMelt® in-container vitrification demonstration

NNL and Veolia Nuclear Services (VNS) have an operational In-Container Vitrification (ICV) system, GeoMelt®, situated in NNL's active rig hall in the Central Laboratory on the Sellafield site (Figure 3.4). The system is configured to take active feeds. GeoMelt® is a batch treatment process which uses Joule-heating to vitrify waste materials and glass-forming precursor chemicals (or frit) into a stable vitrified product. GeoMelt® treatment encompasses the following processes: immobilisation or encapsulation of non-volatile waste components into a glass matrix; thermal destruction of organic materials; and abatement of volatile materials in an off-gas system.

Two GeoMelt® trials were carried out under THERAMIN, using two different waste streams:

- TH-01: A cementitious stream representative of sea dump drums or failing cemented waste packages.
- TH-02: A sludge stream made up of a naturally occurring zeolite (clinoptilolite), sand, Magnox storage pond sludge and miscellaneous contaminants known to arise in a range of UK waste streams.

Trials TH-01 and TH-02 are summarised below; full details can be found in the THERAMIN project Deliverable D3.5 [10].

Figure 3.4 The GeoMelt® rig as installed in the NNL Central Lab.

Trial TH-01

Trial TH-01 demonstrated the thermal treatment via ICV of sea dump drums using the GeoMelt® facility. The waste surrogate comprised 36 metal tins loaded with grout, aluminium metal and PVC. 25 MBq of Cs-137 was added into the waste mix, and fluxed soil was the main glass-forming material. In total, 279 kg of waste materials were processed in 15 hours, at a rate of 18.6 kg h^{-1}. This included feeding an additional 61 kg of soil ("fluxed" with sodium and boron to reduce the working temperature of the glass component) via the feed. The maximum temperature and power were 1400°C and ~70 kW, respectively. On cooling, the product was removed and weighed. The product mass was approximately 800 kg, which included a glass monolith weighing 236 kg. Approximately 1 kg of particulate was recovered from the sintered metal filter.

Active analysis data indicated a Cs-137 retention rate in the glass product of 76%. This retention rate could be improved following optimisation of the formulation and operation. Some residual metal encountered when core drilling suggests that not all of the cans melted and incorporated. However, the residual un-melted metal was encapsulated. Radioactivity analysis and the chemical analysis showed that the product was well mixed and homogenous.

This was a successful test which produced a vitrified monolith which contained all the waste materials except for a small amount of particulate carry over which was within expected norms. A waste loading of 49% was achieved.

Trial TH-02

Trial TH-02 demonstrated in-container vitrification of a blend of corroded magnesium sludge (CMgS), used as a simulant for corroded Magnox sludge, and clinoptilolite ion-exchange material using the GeoMelt® facility. Contaminants, glass formers and radionuclides Cs-137 (25MBq) and Sr-85 (16MBq) were added into the waste mix. At the start of the trial, 190.55 kg of waste material was pre-loaded, and 12 kg was fed into the process during the melt.

A power application fault required the test to be terminated and the melter allowed to cool, before a restart procedure was put in place. This process test was re-started and 238 kg of waste materials were processed in 15 hours at a rate of 15.9 kg h^{-1}. The maximum temperature and power were 1260°C and ~70 kW, respectively. On cooling, the product was removed and weighed, and was found to have a mass of approximately 800 kg, which included a glass monolith weighing 197 kg. Less than 1 kg of particulate was recovered from the sintered metal filter.

Analysis indicated a Cs-137 retention rate of 76% in the glass product, similar to TH-01. Both the radioactivity analysis and the chemical analysis showed that the product was well mixed and homogenous.

This was also a successful test which produced a homogenous vitrified monolith containing all the waste materials, except for a small amount of particulate carry over, which was within expected norms. A waste loading of 72% was achieved.

3.2.5 Hot Isostatic Pressing demonstration

HIP trials were carried out at NNL and the University of Sheffield (USFD) to demonstrate the efficacy of the process for the immobilisation of corroded magnesium sludge (used as a surrogate for corroded Magnox sludge). The trials also demonstrated the potential of the process for co-immobilising clinoptilolite and ion-exchange media, which are used on the Sellafield site. Full details of the HIP trials carried out as part of the THERAMIN project can be found in the THERAMIN project Deliverable D3.6 [11].

Production of wasteforms using HIP requires a pre-treatment step in which water, organics and other volatiles are removed by calcination, after which glass/ceramic precursor is added. The mixture is then introduced into a HIP can, which is subsequently evacuated of air and sealed before placing into the HIP. Here, it is subjected to sufficient temperature and pressure to result in a consolidated wasteform that is suitable for ongoing storage and ultimate disposal.

Demonstrations of HIP were carried out in two facilities on two different scales:

1. The HIP installed at UFSD, which had a hot zone of 125 mm by 75 mm and the capacity to treat feeds containing uranium oxide/metal.

2. A larger HIP installed at the NNL Workington Laboratory, which had a hot zone of 400 mm by 250 mm and the capacity to process non-active feeds only.

The use of these facilities demonstrated the treatment of actinide-containing feeds and the scalability of the HIP process.

Figure 3.5 The HIP equipment at NNL (left) and USFD (right).

HIP trials at NNL

Two surrogates for corroded Magnox sludge and clinoptilolite were prepared at NNL: a formulation containing 33% CMgS, 57% clinoptilolite and 10% alkali borosilicate glass frit (later referred to as sample HIP-01 in the WP4 characterisation tests in Section 4.2.2.5), and a second formulation containing 45% CMgS, 45% clinoptilolite and 10% borax (referred to as sample HIP-02 in the characterisation tests in WP4, Section 4.2.2.5). The clinoptilolite was pre-loaded with stable Cs, and CeO_2 was added as a surrogate for actinides. Calcination was carried out at 950°C for 3 hours. Following this, some material was loaded into cans and prepared for HIPing at NNL, and a smaller amount was sent to USFD where it was used in the small-scale HIP, with the addition of uranium oxide to one of the batches.

The large HIP cans were subjected to a simultaneous application of pressure (>75MPa) and temperature (1250°C) for two hours. Both HIP trials were successfully consolidated, the shrinkage being evidence that the can had retained its seal throughout the HIPing process. The HIP cans were then sampled for characterisation in THERAMIN WP4. A waste loading of 90% was achieved.

HIP trials at USFD

Three different types of sample, all simulating corroded Magnox sludge (CMgS), were used in seven USFD HIP trials. These samples were:

- A magnesium sludge/clinoptilolite mixed sample, consisting of material batched at the NNL HIP trial, as mentioned previously. This sample was used in two trials, one with and one without the equimolar replacement of CeO_2 for U_3O_8, which was used to simulate the actinide oxides present in the waste. The inactive components were pre-calcined at 950°C for 3 hours by NNL, therefore the U_3O_8 was also calcined at USFD at 950°C for 3 hours prior to batching.

- A magnesium borosilicate glass sample containing U_3O_8, as an actinide oxide simulant. Trials were run using high (42.22 wt.%) and low (6.76 wt.%) waste loadings. Both samples were pre-calcined at 600°C for 12 hours in a general muffle furnace (air atmosphere) prior to canister packing.
- An alkali borosilicate glass sample. Three trials were run using this sample, two prepared with U_3O_8 at waste loadings of 44.43% and 6.67%, and one without U_3O_8. All formulations were pre-calcined at 600°C for 12 hours in a general muffle furnace (air atmosphere) prior to canister packing.

Following pre-calcination, each sample was packed into a straight walled stainless-steel HIP canister (15 cm^3 volume) with in-built metal sintered filters. Canisters were then processed under evacuation and bake-out steps to remove organic volatiles and excess water and ensure that welds can withstand a high-temperature and high-pressure environment, before being hermetically sealed ready for the HIP process.

For all samples discussed, the target peak temperature and pressure for each HIP cycle was 1250°C and 100 MPa, with a 2-hour dwell at peak temperature and pressure. All canisters were successfully HIPed and visually confirmed to have been successfully consolidated. No loss of containment was observed and the canisters remained hermetically sealed (i.e. there were no weld failures). Characterisation of the resulting wasteforms was carried out in THERAMIN WP4 (Section 4).

3.2.6 Vitrification process demonstration

As part of THERAMIN WP3, vitrification technology was chosen to demonstrate the treatment and conditioning of a liquid radioactive waste material known as Chrompik III, the main contaminant of which is Cs-137. The owner and operator of the vitrification VICHR facility that conducted these demonstrations (shown in Figure 3.6) is JAVYS, Inc., Slovakia. Full details of these demonstrations can be found in the THERAMIN project Deliverable D3.7 [12].

Figure 3.6 A model of the JAVYS vitrification facility, VICHR.

The VICHR vitrification line is a batch process. Processing one batch of 50 dm^3 of Chrompik takes 24 hours. Dilute liquid waste is initially concentrated before being added to glass frit in an inductively heated melting crucible. Water is evaporated at approximately 130°C before being heated to a maximum temperature of 1050°C over a period of approximately 6 hours, producing a vitrified product. Chromium salts are reduced to Chromium III Oxide (Cr_2O_3), which is soluble in the glass matrix. The resultant vitrified product is poured into a storage container by opening a pour valve using a plunger mechanism. Two pours are added to one container. Off-gas from the vitrification process is drawn into an off-gas treatment system producing a condensate which is decontaminated via a sorption column prior to further treatment.

Between 1996 and 2001, the entire volume of Chrompik I (18.5 m^3) was conditioned by vitrification in the VICHR facility, resulting in the manufacture of 211 glass products with total volume of 1.53 m^3. Due to the higher Cs-137 contamination levels associated with Chrompik III, the VICHR vitrification line required several modifications to improve the vitrification process.

The scope of work undertaken by VUJE and JAVYS, Inc. within the THERAMIN project was to report on the following ongoing activities:

- Laboratory-scale studies to optimise the vitrification conditions, glass chemistry and to minimise Cs-137 volatilisation for processing Chrompik III.
- Technological modifications to the vitrification line prior to processing Chrompik III.
- Full-scale glass making trials with low active Chrompik surrogates followed by full-scale active trials using real Chrompik waste.

Vitrification of Chrompik III

The treatment of the Chrompik III commenced on the modified VICHR facility after trials in 2016. Chemical analyses of Chrompik III showed that the level of soluble Cr in Chrompik III is 1% of that in the original solution and much lower than in Chrompik I. This suggests that a significant amount of chromate was reduced into insoluble compounds of Cr (III), which subsequently settled in a sludge phase at the bottom of the Chrompik storage tank. Owing to this, reduction of Cr (IV) to Cr (III) was not required, so no difficulties were expected in using LKU glass frit during vitrification.

The use of chemical additives to absorb Cs and thereby minimise Cs losses from the melter crucible during drying and melting was investigated. Work was required to minimise the separation of chromium salts from the glass frit during drying in the crucible. Small-scale laboratory melts led to a modified drying and melting temperature profile. Glass chemistry was modified to achieve a glass viscosity to facilitate pouring.

During full-scale active trials, Cs-absorption additives were seen to reduce the Cs-137 activity in the off-gas scrubber system five-fold.

The outcome of lab-scale studies and full-scale trials resulted in the following temperature regime for drying and melting:

- Drying phase - heat to 95°C at a rate of 10°C/min in order to evaporate water content.
- Chrompik decomposition phase - heat to 650°C at a rate of 10°C/min and hold temperature constant for two hours.
- CO_2 release phase – heat to 800°C at a rate of 10°C/min and hold temperature constant for two hours.
- Vitrification phase – heat to 960 - 990°C at a rate of 10°C/min and hold temperature constant for two hours.
- After the above temperature regime steps have elapsed, the melting process is complete, and the melt can be drained from the melting crucible.

The glass manufactured during the trials poured from the melter without difficulty and was considered typical of a good vitrified product through visual inspection. The vitrified product met the local quality requirements for storage and disposal. The glass samples produced from Chrompik III surrogate solution were characterised in THERAMIN WP4 (Section 4.2).

In summary, thermal treatment of Chrompik III via vitrification was successfully demonstrated using the VICHR facility. Additionally, work undertaken to reduce Cs-137 volatility was successful when tested at full-scale. Modifications to the glass chemistry and to the heating profile resulted in a satisfactory glass product and reduced Cs-137 doses in the off-gas system.

3.3 Summary

WP3 of the THERAMIN project successfully demonstrated the applicability of six different thermal treatment technologies (SHIVA, In-Can Melter, GeoMelt®, thermal gasification, vitrification and HIP) to a range of waste groups, as shown in Table 3.1.

Common advantages of all the thermal treatment technologies demonstrated in THERAMIN WP3 include significant volume reduction (although this is highly dependent on the composition of the waste and need for further conditioning) and reduced voidage. The following generic thermal treatment challenges were identified:

- Feeding the waste into the thermal treatment process, which in some cases required pre-treatment (e.g. size reduction, compaction or pelletisation of powders, calcination).
- Optimisation of feedstocks and operational parameters (e.g. melt viscosity, presence of a cold cap) to improve waste loading and radionuclide retention.
- Further conditioning is required for the products of some thermal treatment processes (e.g. gasification, incineration, pyrolysis) to meet the WAC for some disposal facilities.

To determine whether the thermally treated products generated through WP3 activities meet the WAC for disposal, characterisation tests are required. A selection of samples from thermally treated products generated in WP3 were selected for characterisation in WP4 of the THERAMIN project (described in Section 4, below); details of the samples selected can be found in Table 3.2.

Information from WP3, including waste loadings for most of the trials described above, also informed the development and testing of the THERAMIN value assessment process completed in WP5 and described in Section 5.2.

Table 3.2 Samples produced during the treatment technology demonstrations in WP3 that were selected for characterisation in WP4.

Treatment technology demonstrated in WP3	THERAMIN Partner	Sample identification	Initial waste
SHIVA	CEA	THERAMIN-SHIVA-VDM1 sample	Mixture of zeolites, diatoms and IER
In-Can Melting	CEA	THERAMIN-INCAN-BST sample	Ashes from technological waste incineration
Gasification	VTT	Thermal gasification sample	Organic IER
GeoMelt®	NNL	TH 01 GeoMelt® ICV sample	Simulated cemented package representing conditioned waste such as failing cemented packages and sea dump drums
	NNL	TH 02 GeoMelt® ICV sample	Heterogenous sludge
	USFD	Glass 6 – GeoMelt® sample	PCM/Magnox sludge simulants
	USFD	Glass 12 – GeoMelt® sample	Pile fuel cladding/SIXEP
HIP	NNL	HIP-NNL-1 sample	Simulants for Magnox sludge and clinoptilolite
	NNL	HIP-NNL-2 sample	
	USFD	HIP-Ce sample	Magnox sludge simulant
	USFD	HIP-U sample	Magnox sludge simulant
Vitrification process	VUJE	Chrompik glass sample	Chrompik liquors

4 Work Package 4: Disposability of Waste Products

THERAMIN WP4 aimed to carry out an evaluation of the disposability of thermally treated waste products and of the manageability of the resulting secondary waste, depending on the waste stream/treatment process combinations, and on the disposal concepts in each participating country. WP4 is divided into three tasks, described below, the outputs of which feed the overall value assessment carried out in WP2 (Section 5.2).

- Task 4.1 involves the **identification of generic Waste Acceptance Criteria (WAC) for the products of thermal treatment**. These WAC identify the characteristics required in a waste product in order to ensure that the waste cannot have a significant detrimental impact on the long-term safety provided by a disposal facility. Based on the identification of WAC by each participating country, a set of generic criteria were prepared that can be used to evaluate products from thermal treatment for disposal regardless of the political, regulatory or socio-economic context. These WAC were required to inform the subsequent phases of WP4. Characterisation approaches were also defined. This work is summarised in Section 4.1 of this report, and full details can be found in the THERAMIN project Deliverable D4.1 [13].
- In order to be disposed of, radioactive waste must comply with the requirements of disposal, i.e. WAC. Therefore, the second task of WP4, Task 4.2, was dedicated to conducting **characterisation tests** on the thermal products resulting from WP3, in addition to some existing samples provided by THERAMIN Partners. This work is summarised in Section 4.2 of this report, and full details can be found in the THERAMIN project Deliverable D4.2 [14].
- Based on the WAC identified during the project for thermal treatment products (Task 4.1) and the characterisation test results (Task 4.2), Task 4.3 aimed to **evaluate the impact of thermal treatment on the disposability of the waste** by means of a safety case implication study. For a selection of waste stream/treatment process combinations studied in the project, and based on the disposal context of participating countries, Partners and End Users investigated the ways in which thermal treatment can provide benefits and disadvantages in terms of meeting WAC, performance, long-term behaviour or demonstration of safety. This work is summarised in Section 4.3 of this report, and full details can be found in the THERAMIN project Deliverable D4.3 [15].

A synthesis report summarising the work conducted within WP4 and its key conclusions was also produced (THERAMIN project Deliverable D4.4 [16]).

4.1 Waste Acceptance Criteria and requirements in terms of characterisation

Generic disposability criteria for thermally treated products were proposed for consideration alongside established WAC, where available, in determining whether the products may be suitable for disposal in a surface, near-surface or geological disposal facility, depending on the national context.

This section summarises the review of the national WAC and the derived generic WAC for thermal treatment products. Full details of this work can be found in the THERAMIN project Deliverable D4.1 [13].

4.1.1 Approach to deriving generic disposability criteria

The disposability of thermally treated waste products and management of the resulting secondary wastes was examined by reviewing national WAC and other disposability requirements applicable in individual countries (collectively referred to here as 'national disposability criteria') and using these to derive a set of generic disposability criteria that can be applied more generally. Inputs were received from each of the eight countries participating in THERAMIN.

For each country, a short summary was prepared, setting out factors relevant to the management of radioactive waste and application of thermal treatment in that country, such as the approach to classify radioactive waste, a summary of the inventory, the status of disposal facilities, planned development activities and known issues. Existing criteria applicable to the disposability of thermally treated waste products were then compiled for each participating country. Table 4.1 summarises the scope of national disposability criteria inputs.

Table 4.1. Summary of the scope of national disposability criteria inputs.

Country	Scope of inputs	Type of disposal	Packaging concept	Formal WAC?
Belgium	Planned disposal of short-lived LLW/ILW near Dessel in north-east Belgium	Surface	Storage packages grouted in concrete monoliths	No – preliminary – formal WAC will be finalised once a licence for disposal is obtained
Finland	LLW/ILW (reactor operating waste) disposal	Intermediate-depth (Olkiluoto VLJ, Loviisa VLJ)	Concrete boxes	Yes
France	Long-lived ILW disposal in clay	Geological (in a clay host rock)	Storage packages grouted in concrete boxes / direct disposal	No – preliminary WAC submitted to the French Safety Authority in 2016
Germany	Long-lived LLW/ILW disposal at Konrad in northern Germany	Geological (host rock to be determined)	Cast iron / steel / concrete boxes	Yes
Lithuania	Planned disposal of ILW	Geological (host rock to be determined)	Cement-grouted metallic waste	No – preliminary WAC
Slovakia	LLW disposal at Mochovce	Surface	Fibre-reinforced concrete containers	Yes
Switzerland	Planned disposal in clay (long-lived LLW/ILW and spent fuel)	Geological (clay)	Steel canisters / storage drums in concrete boxes	No – preliminary WAC
United Kingdom	Planned disposal of ILW (also applies to high-level waste/spent fuel disposal)	Geological (salt / clay / crystalline – host rock to be determined)	Waste grouted in steel / cast iron / concrete containers	Not WAC, but formal requirements – specifications to be met for issue of a Letter of Compliance (LoC)

It is important to note that existing national disposability criteria tend to be more generally applicable to all wastes of a certain classification or destined for a particular disposal route, rather than being specific to the disposal of thermally treated wastes. This reflects the relative novelty of thermal treatment as a management route, particularly for LLW and ILW, compared to more conventional waste conditioning approaches such as cement encapsulation, and underpins the need for explicit consideration of criteria for thermally treated wastes. Also, the national inputs provided do not reflect all applicable WAC for radioactive waste disposal in the participating countries. Generally, those provided reflect the wastes that are currently being considered for thermal treatment. However, this is not the case in Belgium, where WAC depend on both the disposal route and the waste type, and new WAC would need to be developed for thermally treated waste.

4.1.2 Suite of generic disposability criteria

Generic disposability criteria have been derived that can be used to evaluate the products from any form of thermal treatment. These generic disposability criteria highlight factors that are relevant for waste product disposability and the ways in which thermal treatment can impact on these factors (both positively and negatively). They have been developed to be applicable to any packaging or disposal concept, for any thermally treated waste, regardless of the engineered barriers that are present, and in any disposal environment, regardless of its characteristics and the nature of the host rock / geology. They are intended to provide a starting point or point of reference for WMOs to tailor their own national disposability criteria to thermally treated waste in a manner that gives confidence that relevant factors are being considered.

The generic disposability criteria have been defined at a relatively high level, focusing on additional requirements and/or clarifications relating specifically to thermal treatment that can build on more widely applicable disposability criteria defined within national programmes. Qualitative, rather than quantitative, metrics are set out, against which disposability can be assessed. This is because numerical requirements identified within national disposability criteria tend to be strongly linked to the national context (e.g. activity limits for different waste classifications), so have limited transferability for wider use outside the country of origin.

Generic disposability criteria and associated considerations are set out in Table 4.2, based on WAC affected by thermal treatment. Further detail on the underpinning rationale for these criteria is provided in THERAMIN project Deliverable D4.1 [13]. If used, these criteria and considerations would need to be applied in conjunction with existing criteria applicable to other wastes that are planned for disposal in a particular facility. They do not stand alone as a complete set of requirements that could underpin WAC for thermally treated waste.

Table 4.2. Generic disposability criteria specific to thermally treated waste products.

Topic / Category	Generic disposability criterion	Considerations applicable to measure compliance
Dimensions / mass of packages	The dimensions and mass of containers used to package thermally treated waste (and other aspects of the container design) should be compatible with (i) the thermal processing route being employed and (ii) relevant safety functions for storage and disposal, and with all applicable constraints on waste classification, handling, transport and disposal, taking account of the processed waste characteristics.	None.
Package integrity and required lifetime	Apply existing criteria for the disposal context in question. Any additional criteria on package integrity defined for thermally treated waste should be linked to safety functions applied to such waste.	The characteristics of thermally treated waste should be considered as part of demonstrating compliance with existing requirements.

Topic / Category	Generic disposability criterion	Considerations applicable to measure compliance
Activity content	The disposal concept for thermally treated waste should take account of the potential for activity to be concentrated during thermal treatment (as a result of waste volume reduction), which could have implications for the waste classification, waste package dose rates (and associated handling requirements) and the likelihood of nuclear criticality.	None.
Thermal output	The thermal output of thermally treated waste should not have a detrimental impact on performance of the engineered and natural barriers that make up the disposal system, taking account of the potential for activity to be concentrated during thermal treatment.	None.
Voids	Void space within packages of thermally treated waste should be minimised wherever practicable; this may influence aspects of how thermal treatment is implemented.	None.
Chemical content	Apply existing criteria for the disposal context in question. The choice of thermal treatment route and the design of the associated disposal facility should ensure the chemical compatibility of thermally treated waste with other disposal system components.	None.
Chemical durability	Existing requirements on chemical durability for the applicable disposal route should be applied to thermally treated waste. No additional generic disposability criteria for thermally treated waste are considered necessary, although requirements relating to the containment provided by a wasteform may be justified, depending on the post-closure safety case.	If criteria relating to the durability of a thermally treated wasteform are deemed to be required for application in a particular context, then it is recommended that these should be linked to a required containment lifetime (as assumed in the relevant post-closure safety case), rather than to a threshold dissolution rate.
Data management	Data management requirements for the relevant disposal route should be applied to thermally treated waste. In addition, records of the thermal treatment regime applied to the waste should be kept.	None.
Secondary waste	Secondary waste associated with thermal treatment should be minimised to the extent that is practicable.	None.

In a number of areas, no additional disposability criteria relating specifically to thermally treated wastes are proposed, but a product from thermal treatment may be better able to meet existing requirements / WAC as a result of the processes taking place during thermal treatment. Such requirements include (but are not limited to):

- No free liquid (water) or gas present in the waste – volatiles are driven off.
- No hazardous material present / inert product – reactive waste components often consumed.

- Minimal gas generation.
- Robust wasteform – the product of thermal treatment is often (although not always) monolithic.
- Homogeneity / no localised accumulations of radioactivity.
- Mechanical resistance to stresses imposed during transport, handling and disposal operations.

However, additional considerations relating to the impacts of thermal treatment and the characteristics of the resulting product are often relevant when determining whether existing, more generally applicable criteria have been met. Such considerations include:

- The potential for thermal treatment to concentrate radioactivity and fissile material in the waste product (and thereby to increase heat generation per unit volume).
- The impacts of generating a relatively high density, low voidage wasteform in many cases, which could affect waste package handling.
- The potential for thermal treatment to introduce new mechanisms for contamination of equipment and/or waste packages (e.g. splashing, particulate generation and/or carry-over to the off-gas system).
- The chemistry and mechanical properties of thermally treated wastes, which may behave differently during handling, storage and disposal, thereby introducing uncertainties, e.g. relating to chemical compatibility in a disposal environment.
- Safety functions applicable to thermally treated wastes may or may not be the same as those applicable to other wastes to be disposed of in the same facility, depending on the drivers for implementing thermal treatment. Evaluation of thermally treated waste in the post-closure safety case for a disposal facility may therefore differ from that for other wasteforms.

4.1.3 Identification of characterisation requirements

Having collated national disposability criteria and derived generic disposability criteria, an exercise was conducted to identify WAC requiring characterisation of thermally treated waste products in order to test compliance, along with identification of suitable analytical techniques that could meet these characterisation requirements.

Table 4.3 summarises WAC requiring characterisation, physico-chemical properties that can be measured to evaluate whether each of these criteria have been met, along with some examples of applicable, commonly used, measurement techniques. It can be seen that most criteria can be verified through the use of electron microscopy, possibly combined with analyses of the chemical and radiological composition of the sample. However, some WAC require more specialist techniques to measure compliance.

Table 4.3. Measurable properties and techniques applicable for verifying whether WAC have been met.

Waste Acceptance Criterion	Properties	Examples of Applicable Techniques
No, or limited, free liquid or gas	Homogeneity of the waste	Thermogravimetric analysis (TGA), X-ray fluorescence (XRF), electron microscopy
Permeability and/or diffusivity of the waste sufficient to evacuate gas or other products	Permeability + diffusivity	XRF, electron microscopy
No or limited content of hazardous materials (combustible, pyrophoric, reactive, etc.)	Homogeneity of the waste (no untreated area) + identification of chemical species in the waste	XRF, X-ray diffraction (XRD), Inductively coupled plasma (ICP) analysis after dissolution
Immobilisation of radionuclides	Distribution of radionuclides in the waste	α-spectrometry, autoradiography, Raman spectroscopy
Limited voids / limited porosity	Porosity	Wide-angle X-ray scattering (WAXS), surface area measurement by gas physisorption, porosimetry
No localised accumulations of radioactive material	Homogeneity of the waste / microstructure	XRF, electron microscopy
Leaching behaviour of the waste product	Chemical durability	Leaching tests, ICP, ion chromatography, UV-Visible spectroscopy, α-spectrometry
Mechanical resistance of the waste product (mechanical constraint in disposal, impacts, etc.)	Mechanical behaviour	Mechanical resistance test methods (compression, tension,)
Limited or no metal	Homogeneity of the waste / microstructure	XRF, electron microscopy
Thermal conductivity of the waste product (especially for self-heating waste)	Thermal conductivity / thermal behaviour	Thermal conductivity measurement

4.2 Characterisation of thermally treated waste products

The second task of WP4, Task 4.2, was dedicated to characterisation tests carried out to comply with the WAC previously identified in Table 4.3. These characterisation tests were performed on the products from the thermal treatment demonstrations carried out within THERAMIN WP3 and on other existing samples provided by THERAMIN partners. The results are summarised in this section and full details of this work can be found in the THERAMIN project Deliverable D4.2 [14].

4.2.1 Adaptation of characterisation tests for THERAMIN project

In order to be disposed of, radioactive waste must comply with the relevant Waste Acceptance Criteria (WAC). This compliance can be checked by conducting characterisation tests, as identified in Task 4.1 and listed in Table 4.3, above. Time constraints and financial limitations meant that not all of these tests could be carried out for the characterisation of thermally treated products during the THERAMIN project; therefore, a smaller selection was chosen to form a common basis for the characterisation of solid products. As a minimum, the techniques selected needed to provide information on:

- The degree of homogeneity of the sample and the absence of free liquid of gas.
- The overall chemical composition of a homogeneous sample or the local compositions of a heterogeneous sample.
- The amorphous or crystalline nature of a sample and the structure of the crystals present in a crystalline sample.
- The chemical durability of the samples against the hydrolysis process (leaching tests).

The analytical techniques that constitute the common basis of characterisation tests, which were accessible to all THERAMIN partner laboratories, are listed below.

- Scanning electron microscopy (SEM): A technique that produces high resolution images of a sample surface using electron–matter interactions. It can be associated with X-ray energy dispersive microanalysis to study the chemical composition of the sample by using the characteristic X-ray emission stimulated by the electron beam.
- X-ray fluorescence (XRF) spectrometry: A technique for analysis and determination of the chemical composition of a material.
- And/or electron microprobe analysis: A non-destructive technique used to determine the chemical composition of materials, with spatial resolution, at small scale.
- And/or inductively coupled plasma analysis after dissolution of the solid: A destructive method of chemical analysis that allows for the quantification of almost all dissolved elements simultaneously.
- X-ray diffraction (XRD): Provides information on the crystalline phases present in the sample and any amorphous phase in sufficient concentration.

Depending on the nature of the samples and the national radioactive waste management context, partners may also be required to use other techniques, e.g.:

- Total organic and inorganic carbon analyses.
- Gas physisorption to determine the specific surface area of a powder sample.
- Thermal conductivity.
- Transmission electron microscopy.

Finally, the chemical durability of the samples against the hydrolysis process was estimated by leaching tests based on the ASTM Standard Test Method C 1285 – 14 "Standard Test Methods for Determining Chemical Durability of Nuclear, Hazardous, and Mixed Waste Glasses and Multiphase Glass Ceramics: The Product Consistency Test (PCT)" [24].

4.2.2 Characterisation results of thermally treated waste products

Characterisation tests were performed on the products from thermal treatment demonstrators conducted under THERAMIN WP3 and from other existing samples provided by THERAMIN Partners. A full list of the samples characterised is given in Table 4.4.

Table 4.4 Samples characterised in WP4.

THERAMIN Partner	Sample identification	Initial waste	Thermal treatment applied
CEA	THERAMIN-SHIVA-VDM1 sample	Mixture of zeolites, diatoms and IER	SHIVA
CEA	THERAMIN-INCAN-BST sample	Ashes from technological waste incineration	In Can melting
FZJ	Sample from JÜV 50/2	Mixed radioactive waste from German research reactor	Incineration
NNL	TH 01 GeoMelt® ICV sample	Simulated cemented package representing conditioned waste such as failing cemented packages and sea dump drums	GeoMelt®
NNL	TH 02 GeoMelt® ICV sample	Heterogenous sludge	GeoMelt®
NNL	HIP-NNL-1 sample	Simulants for Magnox sludge and clinoptilolite	HIP
NNL	HIP-NNL-2 sample		
USFD	Glass 6 – GeoMelt® sample	PCM/Magnox sludge simulants	GeoMelt®

THERAMIN Partner	Sample identification	Initial waste	Thermal treatment applied
USFD	Glass 12 – GeoMelt® sample	Pile fuel cladding/SIXEP	GeoMelt®
USFD	Plasma vitrified PCM – cold crucible	PCM	Plasma vitrification
USFD	HIP-Ce sample	Magnox sludge simulant	HIP
USFD	HIP-U sample	Magnox sludge simulant	HIP
VTT	Thermal gasification sample	Organic IER	Thermal gasification
VUJE	Chrompik glass sample	Chrompik liquors	Vitrification process
SCK.CEN	Concentrate slag – Simuli-2	Cemented concentrates	Plasma incineration
SCK.CEN	Concentrate slag – Simuli-3A	Cemented concentrates	Plasma incineration
SCK.CEN	Resin slag – R2 IRN-78	Cemented anionic resins	Plasma incineration

The sections below summarise the results of the individual characterisation tests carried out by project Partners in WP4. The detailed characterisation results can be found in THERAMIN Project Deliverable D4.2 [14].

4.2.2.1 SHIVA samples

The matrices produced during the tests conducted by CEA using the SHIVA process (see Section 3.2.1) were characterised by scanning electron microscopy (SEM, Figure 4.1), X-ray diffraction and PCT-type leaching tests.

The wasteform produced using the SHIVA process consists of an amorphous glass, exhibiting no crystallisation visible in SEM or identifiable in XRD. It is mainly composed of SiO_2, B_2O_3, Nd_2O_3 and Al_2O_3.

Leaching tests show that the hydrolysis rate of the SHIVA glass is significantly lower than that of the ISG (International Simple Glass) reference glass. Further investigations would be required to draw conclusions about the long-term behaviour of the wasteform but these first results are encouraging, considering the objective of demonstration of good leaching behaviour for longer durations.

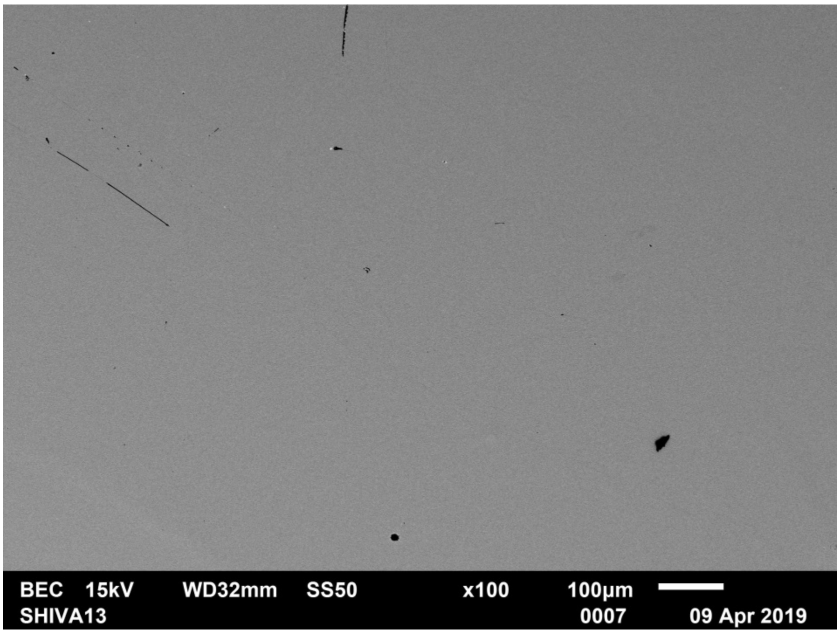

Figure 4.1 SEM observation of SHIVA glass surface with x100 magnifications.

4.2.2.2 In-Can Melter samples

The matrix produced during the tests conducted by CEA using the In-Can Melter process (see Section 3.2.2) was characterised by scanning electron microscopy, X-ray diffraction and PCT-type leaching tests.

The wasteform produced using the In-Can Melter process consists in a crystallised glass mainly composed of SiO_2, Na_2O, B_2O_3, Al_2O_3, and CaO. The term “crystallised glass” refers to a vitreous matrix including crystals of apatite, zincochromite and bismuth alloy.

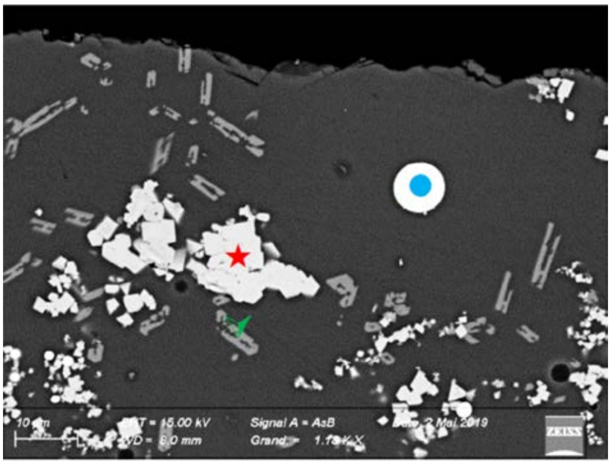

Figure 4.2 Scanning electron microscopy image of the wasteform produced using the In-Can Melter, showing crystals of apatites (★) and zincochromites (✓), and bismuth alloys (●) embedded in a borosilicate glass matrix.

The crystals are distributed homogeneously in the characterised sample. The crystalline phases being durable, the durability of the wasteform is controlled by that of the vitreous matrix. The hydrolysis rate of this vitreous part is relatively high because of its high contents of B_2O_3 and Na_2O. However, based on the leaching test results and longer-term experiments with similar glass samples, it is expected that, in the long term, an alteration layer will form, leading to a significant decrease in the alteration rate, but further work is required to confirm this.

4.2.2.3 Thermal gasification sample

The product of thermal gasification of IER, conducted by VTT (see Section 3.2.3), is a fine powder, which has to be immobilised. VTT selected geopolymerisation as the method of immobilisation. The characterisation methods that were selected by VTT are not the same as those selected in the case of vitrified products because the nature of the geopolymerised product is different.

Combining the gasification of low-level and intermediate-level radioactive waste with immobilisation of the product in a geopolymer matrix is a potential alternative to using vitrification technologies. Geopolymer matrices have demonstrated a high retention capability of radionuclides in multiple studies, and industrial-scale encapsulation has been presented in Slovakia and the Czech Republic [25]. However, the practical encapsulation amount has been limited by the mechanical properties of the encapsulated matrix. Even a small amount of resin decreases the strength of the matrix and cohesion of the matrix is lost when the fraction of resin exceeds 15-20%. After gasification, practical waste (IER) loading is not limited by the mechanical properties of the encapsulation matrix.

Suitability of the geopolymer/gasification technique for low- and intermediate-level radioactive waste encapsulation was evaluated using a standardised method (ANSI/ANS-16.1.2003): “Measurement of the leachability of the solidified low-level radioactive wastes by a short-term procedure” [26]. The apparent diffusion coefficient measured was 10^{-12} to 10^{-13} cm^2/s for Cs. For comparison, apparent diffusion coefficients for Portland cement encapsulated pristine resin, gasified resin encapsulated with alkali-activated material and are also presented in Figure 4.3.

According to results, the geopolymer/gasification technique was able to produce a high strength encapsulated product with an apparent diffusion coefficient significantly lower than that for currently used methods.

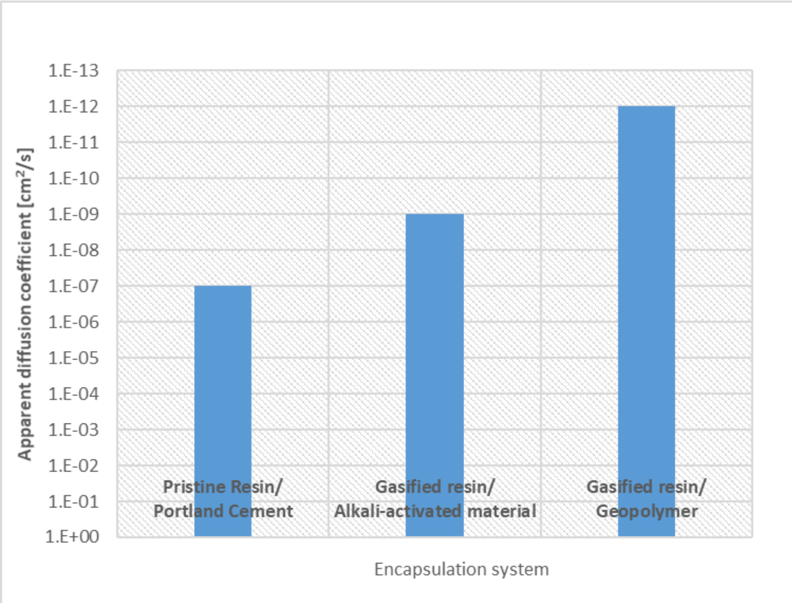

Figure 4.3 Apparent diffusion coefficients for Cs in pristine resin/Portland cement system, gasified resin/alkali-activated matrix and gasified resin/geopolymer matrix.

4.2.2.4 GeoMelt® samples

Two samples from the in-container vitrification trials conducted in WP3 by NNL using the GeoMelt® facility (see Section 3.2.4) were characterised in WP4:

- TH-01 GeoMelt® ICV of simulant sea dump drums (Figure 4.4).
- TH-02 GeoMelt® ICV of simulant sludge and clinoptilolite.

Figure 4.4 TH-01 GeoMelt® ICV sample.

Homogeneity across the vitrified block was examined through use of XRF on the major glass-forming elements. Analyses of inactive elements and active tracers (with gamma scanning) showed a homogeneous wasteform at a macroscopic level and thus indicated good mixing of the feed components during processing.

PCT leach tests on vitrified sea dump drums showed a superior durability to ISG glass under the conditions applied, while the leaching behaviour exhibiting by vitrified sludge showed equivalent durability to ISG glass under the conditions applied. It should also be noted that as a one-off demonstration melt, optimisation of the product was not attempted. Under current geological disposal facility (GDF) requirements in the UK for disposal of ILW, no credit is taken for durability of the wasteform in the disposal system safety case and as such this is not currently a discriminator for disposal.

In addition to the characterisation of the THERAMIN samples described above, two historic GeoMelt® samples were characterised by USFD:

- GeoMelt-Glass 6 sample (PCM simulant, Figure 4.5).
- GeoMelt-Glass 12 sample (Magnox simulant).

The products consisted of a glassy material with noticeable heterogeneity. XRD and SEM analysis showed substituted magnesium silicate crystals within a glass matrix.

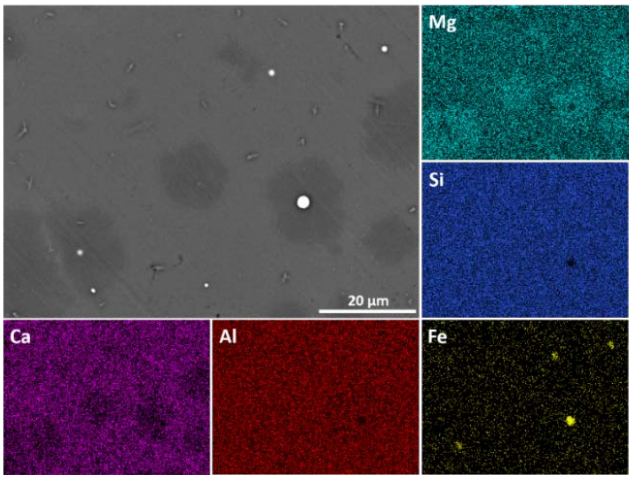

Figure 4.5 SEM/EDS of GeoMelt-Glass 6, Phase 1.

XRF analysis carried out on vitrified PCM simulant determined that a combined SiO_2 and Al_2O_3 content greater than 70 wt.%, combined with a lower alkali content, could help impart durability to these samples. Compared to ISG glass, vitrified PCM simulant had a higher Al_2O_3 content, and a very high MgO component. For the Magnox simulant sample, XRF analysis determined a combined SiO_2 and Al_2O_3 content of approximately 64%, with high contents of MgO and Fe_2O_3 and a low overall alkali content. This composition was therefore quite different from ISG and likely to perform differently under aqueous leaching conditions.

Chemical durability over a 28-day timeframe showed superior performance to ISG, although durability was more difficult to assess given the absence of boron within these samples. Data suggested that dissolution was continuing to occur, though longer-term dissolution data would be required to confirm this.

4.2.2.5 HIP samples

Four samples from the HIP tests carried out in THERAMIN WP3 were characterised by NNL and USFD:

- HIP-NNL-01: Calcined Magnox sludge and Cs-exchanged clinoptilolite with borosilicate glass.
- HIP-NNL-02: Calcined Magnox sludge and Cs-exchanged clinoptilolite with borax.
- HIP-USFD-Ce: Calcined Magnox sludge and Cs-exchanged clinoptilolite with Ce surrogate (Figure 4.6).
- HIP-USFD-U: Calcined Magnox sludge and Cs-exchanged clinoptilolite with U_3O_8.

For the four thermally treated products, SEM and XRD analyses showed that they had a heterogeneous, glass-ceramic microstructure in which added CeO_2 (or U for the HIP-USFD-U sample) has been encapsulated within the wasteform. Simulant waste streams (calcined Magnox sludge and clinoptilolite) had partially or fully reacted to form constituent phases (either glassy or crystalline). As such, a high waste loading appears possible with this thermal treatment.

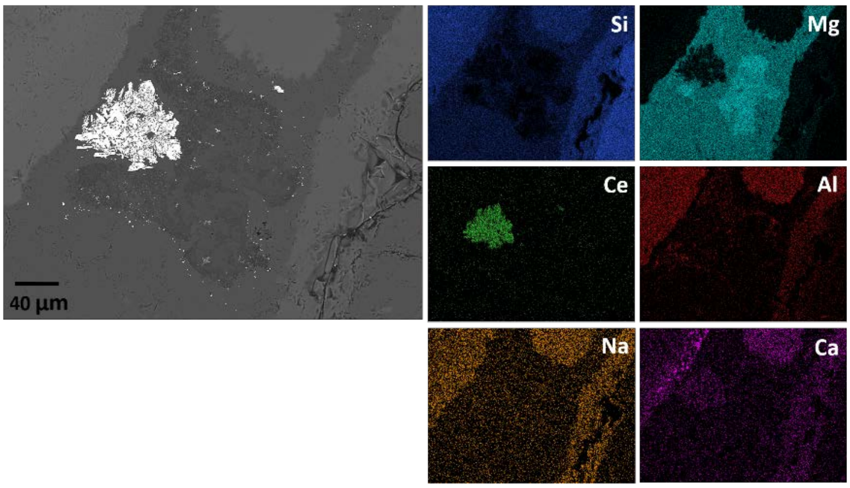

Figure 4.6 SEM/EDS micrograph of HIP-USFD-Ce (x500 magnification).

Chemical durability assessments of these products were difficult to compare to ISG due to the difference in dissolution mechanisms between a homogeneous glass and this multi-phase material. The results obtained for the overall boron and silicon normalised mass losses were dependent on the sample. It was noted that these mass losses depended on the existence or formation of any secondary precipitation products during dissolution.

4.2.2.6 Plasma vitrified PCM sample

A sample from a historic demonstration of plasma vitrification for plutonium contaminated material, which contained quantities of masonry, steel, aluminium and plastic materials, was also characterised by USFD (Figure 4.7).

This plasma vitrified material presents a largely glassy structure, with some crystalline features. Simulant plutonium material was added as CeO_2 partitioned into the glass. A high waste loading (54.1 wt.%) was achieved, which would translate to high volume reduction for this waste stream.

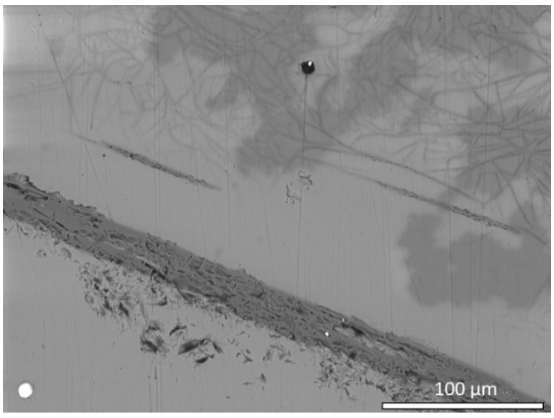

Figure 4.7 Backscattered electrons **(**BSE) micrograph of PCM 54.1 wt.% glass.

The chemical durability of this material appeared to be good, resulting in lower normalised mass losses for silicon and aluminium than for ISG. Due to the lack of boron in this material, and the low levels of sodium, there were fewer elements to compare between these samples to assess durability. Overall, however, this material performed well, with longer-term leaching studies recommended in order to determine whether these dissolution rates continue at a low level, or change over a longer timeframe.

4.2.2.7 Sample from JÜV 50/2 facility

FZJ performed structural characterisation and stability studies of selected ash fractions originating from the incineration facility JÜV 50/2.

For an in-depth characterisation of selected ash fractions, various radio and microanalytical techniques were applied to determine radionuclide content and speciation, as well as phase assemblage and microstructure of different grains (Figure 4.8). Moreover, the characterisation included examination of the leaching behaviour of selected ash fractions under typical conditions for cementitious disposal environment, as well as post-leaching examinations of the solids to evaluate the evolution of microstructural properties and phase composition, when subjected to different disposal relevant conditions.

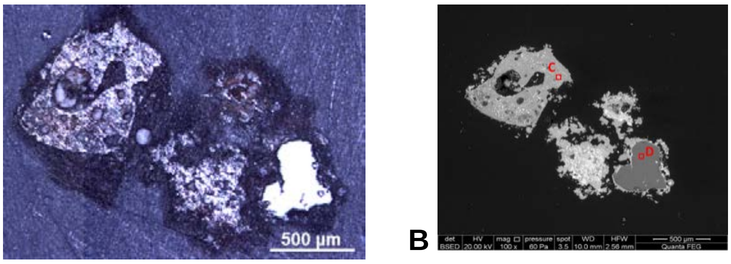

Figure 4.8 Microscopic analysis of a highly radioactive particle from specimen F2-D. An image produced using optical microscopy (A) and a SEM image in back-scattered electron (BSE) mode (B).

Results demonstrate a complex phase composition of the selected ash fractions, including quartz, hematite, lime, corundum, and some mixed oxides $(Si,Al,Ca)O_x$. Minor amounts of chlorides, like halite (NaCl) and sylvite (KCl), and inclusions of elementary Al were found. Radioanalytical investigations revealed in particular the presence of Cs-137 and Co-60, along with traces of Am-241 and Eu isotopes, distributed with a certain degree of heterogeneity on microscopic scale.

Complementary microanalytical investigations showed that the radioactivity is mainly associated with oxide phases. Investigations of the selected fractions´ stability, based on the PCT ASTM C1285-14 [24], revealed a fast release of Cs-137 irrespective of leaching conditions, solution composition or temperature. In contrast, no significant release of Co-60 was found. No further radionuclides were found in the leaching solutions, indicating fixation of the remaining activity in the solids.

4.2.2.8 Vitrification of inorganic liquid Chrompik

Chrompik non-active glass samples were prepared from glass, additives and Chrompik surrogate solution in the ratio 40/17/43 in VUJE´s laboratory. Chrompik III- mainly contains of K^+, HCO_3^-, CO_3^{2-}, and the Cr(VI)-content in the soluble form is 1%. Samples were prepared by pouring into cuboid monoliths of approx. 1.5 cm × 1.5 cm × 1.5 cm.

The chemical composition of glass samples was analysed using two techniques, SEM/EDS and XRF. The tests of chemical durability of Chrompik III glass samples were carried out by leaching in water according to the modified ASTM C1220 using three methods:

1. Leaching in demineralised water to the boiling point.
2. Leaching at 90°C in demineralised water in an oven without renewal of the specimen surface.
3. Leaching at 90°C in demineralised water in an oven with renewal of the specimen surface.

There were only small differences observed between theoretical composition and XRF results. The final Chrompik III glass product consisted of an amorphous glass (Figure 4.9), mainly composed of SiO_2, Na_2O, B_2O_3, and K_2O; these compounds comprised 84.6% of the glass composition, with no evidence of any crystalline phase assemblages or other inhomogeneity.

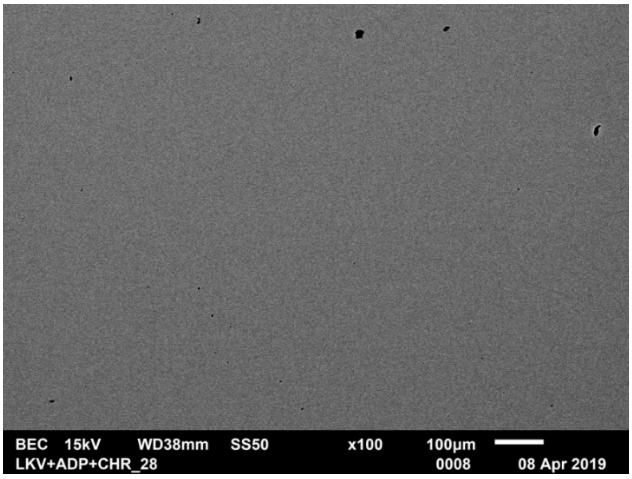

Figure 4.9 SEM observation of Chrompik III glass with x100 magnifications.

The chemical durability was assessed and all three methods were compared for Chrompik III glass samples: weight loss for method 1 was approximately double that of leaching using method 2. To compare method 2 and method 3 the weight loss rate is twice higher for method 2 than using method 3. Leaching tests conducted at 90°C show a similar evolution of the pH value, and chemical durability assessment seems to be better for method 3.

4.2.2.9 Plasma slags

Characterisation tests were conducted by SCK-CEN on three plasma slags:

- Simuli 2 (Cemented concentrates).
- Simuli 3-A (Cemented concentrates).
- R2 IRN 78 (Cemented anionic resins).

SEM-EDX (Scanning Electron Microscope/Energy Dispersive Using X-Ray), XRD and XRF showed that the plasma slags are mainly amorphous and composed of silicon, iron, calcium and aluminium oxides. Simuli 2 is porous and contained some magnetite and clinopyroxene (Figure 4.10).

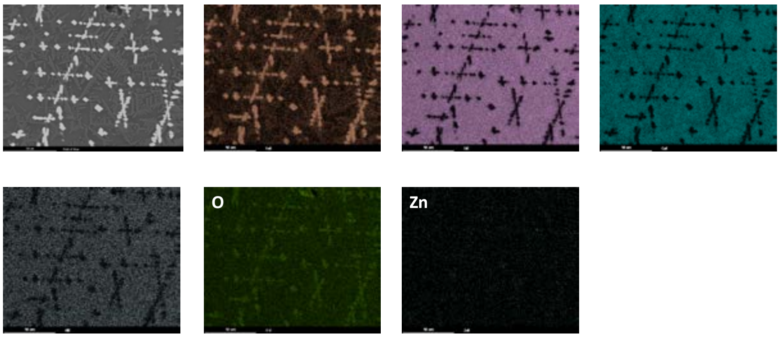

Figure 4.10 SEM micrograph and elemental mappings for the Simuli 2 sample.

The chemical durability of these samples was evaluated by performing static leaching tests based on standard procedures (MCC-1 or ASTM C1285). The experimental conditions were either those relevant for the planned near-surface disposal of radioactive waste in Belgium, i.e. KOH solution at $pH_{(25°C)}$ = 13.5 and 40°C, or those selected by the THERAMIN partner laboratories, i.e. ultrapure water and 90°C. In order to make a comparison with reference nuclear glasses, tests were also done with inactive SON68 glass or the ISG, which is designed as an analogue for SON68.

In the tests carried out in ultrapure water and at 90°C, the porous plasma slags containing crystalline phases and ISG dissolved incongruently as compared to the other amorphous plasma slags. The dissolution rates established after 28 days of alteration for the plasma slags and ISG were in the range of 0.060 - 0.2 $g \cdot m^{-2} \cdot d^{-1}$.

The leaching tests suggested that plasma slags have a short-term chemical durability as good as that of nuclear glasses at near neutral pH or even better at very alkaline pH. Further investigations to study the long-term behaviour are still needed.

4.3 Disposability evaluations

The main objective of Task 4.3 was to evaluate the impact of thermal treatment on the disposability of waste by means of a safety case implication study. For a selection of waste stream/treatment process combinations studied in the project (in WP3 and Task 4.2 of WP4), and based on the disposal context of participating countries, partners investigated the ways in which thermal treatment can provide benefits and disadvantages in terms of compliance with WAC, wasteform performance, long-term behaviour or demonstration of safety. The outputs from Task 4.3 fed into the overall value assessment carried out in WP2 (Section 5.2).

The evaluations were conducted for products characterised under Task 4.2, except for LEI, which evaluated a thermally treated metallic waste from Ignalina NPP based on a review of literature data.

It is noted that only a selection of products characterised under Task 4.2 (identified in Table 4.5) were evaluated due to time constraints. However, the majority of the thermal treatment technologies studied under the THERAMIN project benefitted from at least one safety case implication study[2].

Table 4.5 Samples characterised in Task 4.2 and studied in Task 4.3 by THERAMIN Partners and End Users.

Sample identification	Initial waste	Type of treatment	Safety case implication study
SHIVA sample	Mixture of zeolites, diatoms and IER	Incineration-vitrification	Andra
In-Can sample	Ashes from technological waste incineration	Vitrification	Andra
Sample from JÜV 50/2	Mixed radioactive waste from German research reactor	Incineration	FZJ
GeoMelt® ICV sample	Simulated cemented package representing failing cemented packages and sea dump drums	Vitrification	ONDRAF/ NIRAS
Glass 6 - GeoMelt® sample	PCM/Magnox sludge simulants	Vitrification	RWM
HIP sample	Magnox sludge simulant	HIP	RWM
Vitrification	Non-active surrogate solution of Chrompik	Vitrification	VUJE
Concentrate slag - Simuli-2	Concentrates mixed with concrete	Plasma incineration	ONDRAF/ NIRAS

[2] VTT had no authority to perform a safety case implication study for the geopolymerised products from thermal gasification treatment; however, observations relevant to disposability were drawn from other THERAMIN deliverables.

Sample identification	Initial waste	Type of treatment	Safety case implication study
Thermally treated metallic waste	Metallic waste from Ignalina NPP	Metal melting (process study based on literature).	LEI

4.3.1 Safety case implication studies

The sections below summarise the individual safety case evaluations completed by project partners in WP4. The detailed evaluations can be found in THERAMIN Deliverable D4.3 [15].

4.3.1.1 SHIVA sample – French context

The thermal treatment of ILW-LL IER, mixed with zeolites and diatoms, via the SHIVA process produces an ILW-LL glass. In the French context, the usual baseline for these mixtures is cementation in concrete packaging.

The SHIVA thermal treatment process should result in a less heterogeneous waste and a significant reduction in the packaging volume. However, the overall treatment may influence the waste category according to Andra's classification, which could influence the disposal strategy. For ILW-SL, this could influence whether the waste is disposed of in an existing surface disposal facility (CSA) or the deep geological disposal project (Cigéo). Because glassy wasteforms are already taken into account in the Cigéo project, this analysis focused on disposal of the final product in Cigéo.

Most of the current preliminary WAC seem to be respected with the benefit of the absence of H_2 release, thanks to the destruction of organic matter and the absence of water in the final wasteform, which simplifies the ventilation constraints in a disposal cell. However, due to concentration of activity by volume reduction, some criteria need to be checked, such as heat generation and criticality.

The final product is an amorphous alumino-borosilicate glass with radionuclides immobilised in a glassy matrix which allows analogies with vitrified HLW to be disposed of via deep geological disposal. Disposal of vitrified HLW in Cigéo will necessitate the application of operational safety provisions that are expected to be more than sufficient to cover the vitrified products from the SHIVA process, applied to ILW. The long-term safety approach should be similar to the one already considered. The release of radionuclides induced by the glassy wasteform of the final product depends on the chemical durability of the vitreous matrix. Even if long-term behaviour of the final product needs to be assessed in more detail, this appears more favourable that the instant release model associated with the baseline for this type of waste.

4.3.1.2 In-Can Melter sample – French context

The thermal treatment of ash via the In-Can Melter process produces an ILW glass. The hypothesis adopted for this safety case implication study is the combined presence of organic matter and alpha radionuclides. The baseline for such alpha surrogate technological ILW-LL is cementation in metallic packaging of waste, which may or may not have been previously compacted.

The thermal treatment should induce a significant reduction of the packaging volume. However, the overall treatment may influence the waste category according to Andra's classification (i.e. ILW-SL which could become ILW-LL) which could influence the disposal strategy and concept. For ILW-SL, this could influence whether the waste is disposed of in an existing surface disposal facility (CSA) or the deep geological disposal (Cigéo). Because glassy wasteforms are already taken into account in the Cigéo concept, this analysis focused on disposal of the final product in Cigéo.

Most of the current preliminary WAC seem to be respected with the benefit of the absence of H_2 release, thanks to the destruction of organic matter and despite the presence of alpha radionuclides. This simplifies the ventilation constraints in the disposal cell. However, due to the concentration of activity due to volume reduction, the criticality risk must be checked.

The final product is a vitreous matrix including crystals. The distribution of these crystals in the matrix is homogeneous with the exception of the contact zone with the can.

An analogy with vitrified waste to be disposed of in the deep geological disposal is considered, with a similar long-term safety approach. Disposal of vitrified HLW in Cigéo will necessitate the application of operational safety provisions that are expected to be more than sufficient to cover the vitrified products from the In Can melter process, applied to ILW. The release of radionuclides induced by the glassy wasteform of the final product depends on the chemical durability of the vitreous matrix. Even if long-term behaviour of the final product needs to be assessed in more detail, this appears more favourable that the instant release model associated to the baseline for this type of waste.

4.3.1.3 Sample from JÜV 50/2 – German context

WAC for geological disposal of wastes with negligible heat generation are well formulated in Germany. For instance, WAC define criteria regarding the stability of the thermally treated product, the radionuclide inventory and the chemical composition. The existing thermal treatment process (incineration in JÜV/50) was established in order to yield a product which meets these requirements. The improvement due to thermal treatment for these waste streams is that it enables the disposability of the (treated) wastes, which would not be possible otherwise. The product of thermal treatment is much easier to declare (or characterise) and this can be a significant advantage in terms of the cost of disposal of the ashes.

For disposal in Schacht Konrad, the release behaviour of some volatile radionuclides (e.g. H-3 or C-14) during the operational phase of the repository is an issue, in order to prevent uncontrolled incorporation of these radionuclides into the human body through inhalation. It is therefore essential to provide release prognoses over the operational time of 40 years, when personnel working in the repository could come into direct contact with released radionuclides, in order to assure that respective personnel exposure limits are not exceeded. Use of different packaging for different waste products is considered, depending on the radionuclide inventory and release rate of some volatile radionuclides. Thermal treatment allows elimination of the sources (e.g. organics) of potentially critical volatile radionuclides H-3, C-14 or I-129. This allows packaging of treated wastes in simpler containers (e.g. without specified gas tightness), such as Type IV containers.

The long-term safety of Schacht Konrad is associated with stable wasteforms, i.e. non-degradable waste, preventing unwanted gas generation. The latter may compromise the stability of the waste packages and promote early uncontrolled release of radionuclides. By applying thermal treatment, a waste product (ashes) with high stability in the environment relevant to disposal is obtained, which enables waste disposal. The actual release behaviour of radionuclides from the wastes in the long-term is less decisive for Schacht Konrad, as the activity limits in the WAC are formulated based on the (conservative) assumption of a practically spontaneous water saturation of the repository in the post closure phase, leading to an instantaneous contact of the emplaced waste containers with groundwater and a rather fast release of the radionuclides from the waste packages (i.e. complete release of the total RN inventory in less than 600 years). The resulting radionuclide concentrations in (usable) groundwater in this stylised scenario have been shown to meet the legal requirements.

The main added value of thermal treatment of LLW/ILW in JÜV/50 is transformation of mixed wastes into a wasteform that is compliant with the WAC for Schacht Konrad and can be disposed of without requiring encapsulation. Volume reduction of LLW/ILW is also beneficial for reducing the costs of storage and disposal of the final product. Secondary wastes from the off-gas treatment systems are treated by evaporation to create a final product (granulate) that is packed into waste drums following a well-established methodology to yield a product that also meets the WAC for Schacht Konrad.

4.3.1.4 TH-01 GeoMelt® ICV sample ⁻ Belgian context

The thermal treatment of a cementitious waste stream representative of sea dump drums or failing cement waste packages via the GeoMelt® process produces an amorphous glass which would be intended for surface disposal in a Belgian context. In Belgium, the usual baseline for these types of drums would be surface disposal.

Passivation of the waste and the decomposition of its organic content is considered to be one of the main advantages for this waste stream / thermal treatment combination.

If the historic waste drums show non-conformities such as corrosion at the welding, treatment by GeoMelt® and the incorporation of the end product within an intact disposable container might increase its operational safety.

The long-term safety of this waste product should be investigated. In the Belgian surface disposal concept, the sorption on the cementitious matrix contributes to the limitation of the release of the radionuclides. Further investigation is required to confirm whether the GeoMelt® end product can fulfil this safety function.

Limited knowledge of the waste characteristics is one of the main challenges for its disposal. Thermal treatment can at least partially overcome this problem, as all organics are decomposed and the waste is passivated. However, the inclusion of the radionuclides within the matrix and the durability of the matrix need further investigation to guarantee long-term safety.

4.3.1.5 GeoMelt® Glass 6 sample – UK context

The thermal treatment of PCM/Magnox sludge with GeoMelt® process produces an ILW glass which would be intended for a GDF with a suitable package. The expected baseline for this waste is grout encapsulation and disposal to a GDF.

The benefits of thermal treatment for this waste are primarily volume reduction and reduced package numbers. The baseline will involve significant capital investment in a new grouting plant and a decision is due in the next few years.

The UK is currently engaged with a siting process inviting host communities to participate in finding a site for a GDF for its higher activity waste. RWM currently assesses against three generic disposal concepts in three illustrative geological environments. Therefore, there are no formal WAC in place. RWM assesses against waste package specifications for Low- and High-Heat Generating Wastes using its Disposability Assessment Process.

Long-term and operational safety have not been assessed but they are expected to be no worse than the baseline. In conclusion, additional studies are required to assess the disposability of this product if the baseline changes.

4.3.1.6 Magnox HIP sample – UK context

The thermal treatment of calcined Magnox sludge / Cs-exchanged clinoptilolite with the HIP process produces an ILW glassy ceramic that would be intended for a GDF within a suitable package. The expected baseline for this waste is grout encapsulation and disposal to a GDF.

The interest of thermal treatment for this waste are primarily volume reduction and reduced package numbers. The baseline will involve significant capital investment in a new grouting plant at the SIXEP plant and a decision is due in the next few years.

The UK context is as noted in Section 4.3.1.5 above. As for the GeoMelt® sample, the long-term and operational safety have not been assessed but they are expected to be no worse than the baseline. In conclusion, additional studies are required to assess the disposability of this product if the baseline changes.

4.3.1.7 Vitrification of Chrompik inorganic liquid – Slovakian context

The original waste is not intended for near-surface disposal due to its radiological content and volume. The baseline for this wastestream is vitrification and disposal in a deep geological repository.

The vitrified waste is not compliant with the radiological WAC for the Slovak surface repository in Mochovce. At the moment only VLLW and LLW types of waste are referred to in the existing WAC for this facility.

The specific activity of the original waste and vitrified product affects the radiation protection in the operational period, therefore all operations with vitrified products are performed remotely.

In the long-term safety of the GDF (the considered destination for Chrompik III) a safety function is assigned to the vitrified wasteform. All aspects mentioned above need to be investigated and WAC for this thermally treated wasteform should be developed.

4.3.1.8 Concentrate slags - Belgian context

The original waste (cemented concentrates) is already cemented and intended for surface disposal. The thermally-treated waste product, via a plasma incineration process, would also be intended for surface disposal.

The original waste risks alkali-silica reactions (ASR), resulting in the formation of a swelling gel which can have an impact on the physical integrity of the systems, structures and components of the disposal system. In addition, there is a risk that the WAC could be exceeded with respect to chlorides. Plasma incineration might result in the passivation of the ASR affected packages. The exclusion of ASR under disposal conditions for the plasma treated packages should be demonstrated. The chlorides might partially evaporate from the waste package during the thermal treatment. The carry-over fraction of Cl^- during plasma incineration should be analysed. In addition, the chloride limit is determined specifically for cemented waste packages as it can have an impact on cement sorption. As such, other chloride limits might be valid for thermally treated waste that do not rely on a cementitious conditioning matrix. In general, the properties and the stability of the end product with respect to the disposal conditions should be evaluated.

The specific activity of the original waste packages is rather high which might affect the radiation protection measures that should be incorporated in the operational period. Characterisation of the waste before thermal treatment might be necessary to ensure its compliance with the WAC of the plasma treatment facility itself.

In the long-term safety case for the Belgian surface disposal facility (the destination for the considered original wasteform) a safety function is assigned to the cementitious conditioned matrix. Therefore, it should be demonstrated that the lifetime of the plasma slag is such that it can ensure a sufficiently slow release of the radionuclides in accordance with the safety function. In addition, the chemical stability of the plasma slag shall be investigated to ensure its compliance with the wider disposal system.

The main added value of plasma incineration of the cemented concentrate would be its potential ability to exclude the ASR reaction and the resulting swelling gel formation. In addition, the (partial) evaporation of the chloride content could be such that the resulting waste would comply with the existing WAC. All these aspects need to be investigated and specific WAC should be developed for this thermally treated wasteform.

4.3.1.9 Thermally treated metallic waste (LEI) – Lithuanian context

A desk-based study of the products generated by the thermal treatment of activated metallic waste (ILW-LL) generated after dismantling radioactive carbon steel constructions at Ignalina NPP was chosen by LEI.

The baseline is the incorporation of metallic waste in a disposal container, subsequent grouting with a cement-based material and disposal to a GDF.

From the comparison of activity limits in the case of the non-treated waste and in the case of thermally treated waste disposal it can be concluded that thermal treatment allows disposal of higher activity waste. However, this conclusion is drawn only from the leaching scenario. The benefits and drawbacks of thermal treatment should be evaluated taking into account other scenarios, operational limitations and considering economic issues.

For disposal of the thermally treated waste, the peaks of the dose are observed at later times and all radionuclides are lower than in the case of disposal of the non-treated waste. The later appearance of the peak dose is related to the slow corrosion of ingots, accumulation of the radionuclides released from the ingots in the container and sudden radionuclide release after the containers' breach. The highest dose in the case of disposal of the thermally treated waste is from Zr-93 and corresponds to about 10^{-5} mSv/y. Other radionuclides give rise to a dose that is lower by a few orders of magnitude than that of Zr-93.

4.3.2 Consideration of Disposability within the THERAMIN Value Assessment

A value assessment approach has been developed under THERAMIN WP2, which is described in further detail in Section 5.2. It provides a structured methodology for evaluating the potential application of a thermal treatment technology to treat a waste stream of interest. 'Value' in this context is defined as realisable benefit in safety, monetary and/or environmental outcomes from implementing an option at a specified time. This includes benefits and challenges across all stages of the waste management lifecycle. The value assessment methodology aims to integrate learning from across the THERAMIN project.

The methodology developed employs a multi-attribute assessment approach that builds on the UK Nuclear Decommissioning Authority's (NDA's) Value Framework [27]. The following assessment attributes are included in the THERAMIN value assessment methodology:

- Operational and transport safety
- Environment impact
- Impact on disposability and long-term safety
- Factors affecting the implementation of a thermal treatment technology
- Timescales
- Technical readiness of the technology
- Strategic cost impacts

Each of these attributes is sub-divided into a series of ‘data categories’, accompanied by a set of assessment considerations that provide guidance to the user on factors to consider when conducting a value assessment.

From a disposability perspective, important drivers for thermal treatment include:

- The generation of a robust, durable, monolithic wasteform that shows good radionuclide retention.
- The homogeneous distribution of waste components / radionuclides across the wasteform.
- The destruction, or passivation (through encapsulation or immobilisation) of hazardous and reactive waste components.
- The production of a wasteform where free liquids and gases are absent.
- A reduction in the packaged waste volume, particularly if this enables disposal to a facility where space or volumetric capacity is limited (wider consideration of volume reduction is relevant from a safety, environmental and cost perspective).

These drivers can be linked back to safety functions applicable to the wasteform and waste package during long-term safety assessment. The wording and scope of safety functions differs from country to country and between different disposal facilities, but typical requirements on the wasteform include the ability to:

- Contain radioactivity over long timescales, and retard releases into the surrounding environment when the wasteform comes into contact with water and slowly degrades.
- Minimise the potential for the wasteform to interact detrimentally with other components of the disposal system.
- Manage the distribution of fissile material and reduce the likelihood of criticality events occurring.
- Limit releases of gaseous species into the surrounding environment.

The disposability attributes developed as part of the THERAMIN value assessment methodology aim to reflect these drivers, translating them into typical requirements on the characteristics of the product from thermal treatment. Further information on disposability inputs to the THERAMIN Value Assessment are provided in THERAMIN project Deliverable D4.3 [15].

5 Work Package 2: Viability Matrix and Value Assessment

5.1 Viability and Gap Analysis

The information collected under Task 2.1 (wastes) and Task 2.3 (technologies) was used to consider the suitability of particular technologies to treat particular waste types. The waste types were organised in the same way as in the THERAMIN waste database so that they could be linked directly to specific countries and, if needed, to specific waste streams.

For each waste type – technology combination, the key factors determining whether the combination is viable or not were considered. Viability was considered in two distinct ways:

- Wastes that can be processed in principle by a particular technology.
- Wastes that have been demonstrably processed by a particular technology.

To assess the potential to apply each of the thermal treatment technologies to the identified waste groups, key properties of the wastes and the compatibility of the technologies were reviewed. The D2.3 and D2.4 reports [3], [4] focus on a number of specific attributes, including whether the technologies can treat solids, liquids, or both solids and liquids. It also considered whether they could treat organic or metallic wastes. In addition, the maximum accepted levels of activity that each facility can accept were highlighted, where this information was available. Finally, the flexibility of each technology was also evaluated to assess whether it could be used to treat a wide range of wastes, and/or highly heterogeneous waste streams.

To determine accurately the viability of each treatment technology, it is important not just to assess their beneficial attributes, but also the potential limitations that may render treatment of particular waste streams challenging or impossible. The technical limitations inherent in each technology were therefore also considered (e.g., some technologies might be challenged by the presence of a significant metal fraction in the waste feed). Additionally, each of the facilities are bound by logistical constraints including their maximum treatment capacity and throughput, and whether they are already in operation at an industrial scale. These strategic aspects are also likely to inform decision making (e.g., a low capacity method may not be ideal for treating large waste volumes). Finally, the TRL of the technique was considered as some technologies, although theoretically able to treat a waste with the identified properties, may still only be at the experimental or pilot stage.

Considering all of the above information, the suitability of the thermal treatment technologies for treating each of the generic waste groups was considered. Each technology was categorised as either having already been tested to treat that waste, potentially having applicability, having only limited applicability, or not being applicable. A number of the technologies have been tested in THERAMIN WP3 on a range of waste groups (Table 5.1).

Table 5.1 A summary of the waste groups which were treated by various thermal treatment technologies in THERAMIN WP3.

Technology	Sludge	Cement conditioned wastes	Organic ion-exchange material	Inorganic ion-exchange material	Ash	Inorganic liquor
GeoMelt®	✓	✓				
Hot Isostatic Pressing	✓			✓		
SHIVA			✓	✓		
In-Can Melter					✓	
VICHR Vitrification						✓
Thermal Gasification			✓			

Gap Analysis

The assessment of viability and the THERAMIN waste database were used to make a strategic gap analysis, identifying countries where there are significant waste arisings with potential to benefit from thermal treatment using technologies available within the country or in other European countries. Where gaps in domestic technologies and facilities were identified, suggestions were made of resources available to process the wastes in other countries.

It is noted that the gap analysis is based on the assessment of viability, where the applicability of the treatment technology to specific generic waste types takes into account the properties of the wastes and technical aspects of the technology. It does not account for non-technical constraints and limitations on waste treatment, such as constraints on moving wastes across international borders for treatment, regulatory barriers, or stakeholder implications. In addition, the mapping does not consider detailed characterisation data and specific properties of the waste streams within a generic waste group. Therefore, the information in the strategic analysis provides only a preliminary input and a starting point to aid decision making, rather than a definitive or optimised options appraisal for treating a particular waste stream.

The analysis concluded that although a few European countries may have the resources to thermally treat their own wastes, many other countries could benefit from cross-country collaboration and treatment of wastes outside their borders. This could provide a cost-effective option for treating challenging and problematic wastes. However, the non-technical constraints listed above may prevent the transfer of wastes across international frontiers for treatment. This means that, in practice, it may be the technology itself that is transferred (though knowledge-sharing exercises or through the construction of additional facilities) rather than the waste. Further information and country-by-country analysis are provided in the D2.3 report [3].

5.2 Value Assessment

The THERAMIN value assessment exercise builds on the work undertaken in Task 1 of THERAMIN WP4 (THERAMIN project Deliverable D4.1 [13]) and the UK NDA's Value Framework [27], and is designed to provide a structured methodology to assess the 'value' of a chosen waste treatment technology when used to treat a waste stream of interest. Value, in this context, is defined as realisable benefit in safety, monetary and environmental outcomes from implementing an option at a specified time. This includes benefits and challenges across all stages of the waste management lifecycle. However, it should be recognised that value varies somewhat between stakeholders, with different stakeholders assigning greater or lesser importance to different attributes. Therefore, a value assessment, like the one outlined in this report, is a multi-attribute assessment that considers all of these different aspects of value across the whole lifecycle of waste management, and of the treatment facility. A methodology for assessing value was developed based on the outputs of a value assessment workshop in December 2019 attended by the THERAMIN partners, where discussions focused on the approach that should be taken to assess the value of thermal treatment technologies, particularly those trialled within the project. It aimed to provide guidance to support stakeholders who wish to make an assessment of the advantages and disadvantages of different thermal treatment technologies.

When conducting a value assessment, it is important to follow a structured approach that focuses the assessment before it is undertaken. As outlined in Figure 5.1, the first stage in this process is to choose the specific waste(s) and technologies that will be assessed. Once the candidate waste(s) and technologies have been chosen, the assessment scope and approach can be clearly defined. This involves choosing the attributes that are to be considered and the lifecycle stages that are to be assessed.

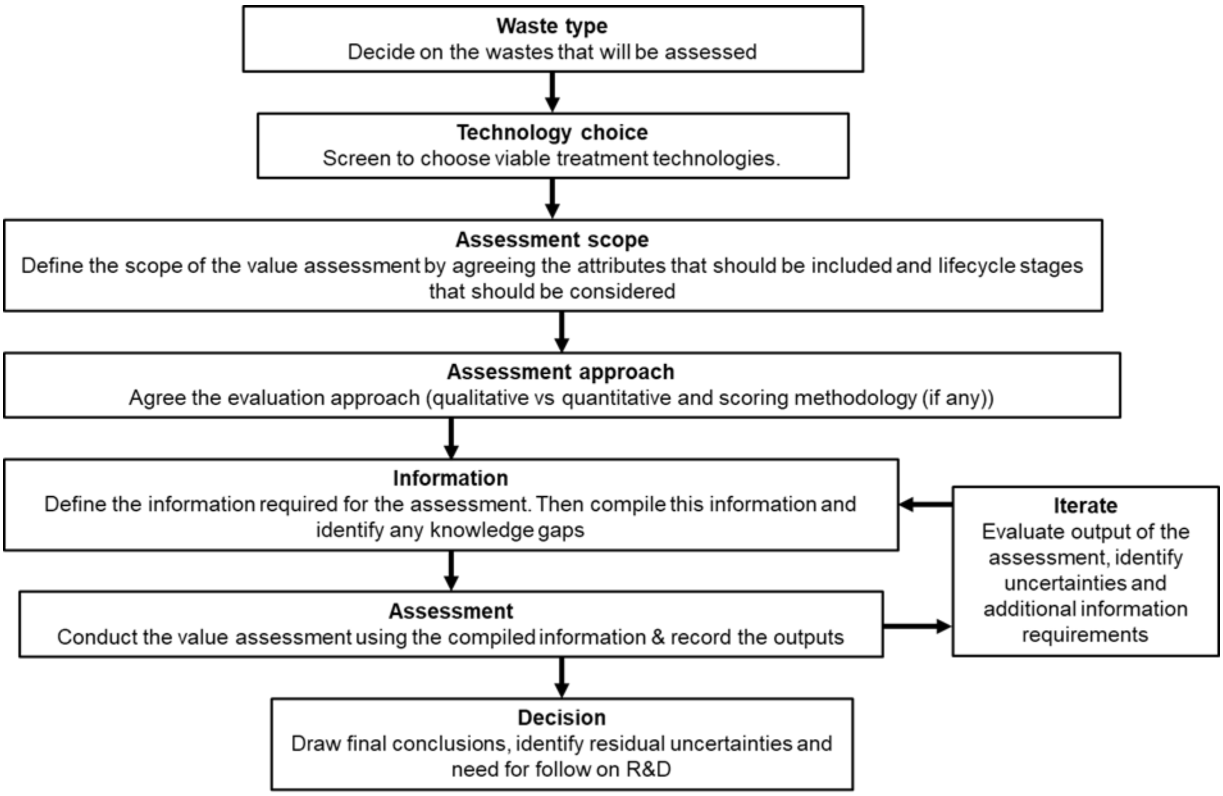

Figure 5.1 Flow chart summary of a structured value assessment process.

The THERAMIN value assessment is based around a series of attributes that are sub-divided into data categories. The attributes chosen in the THERAMIN project are outlined in Table 5.2. The assessment attributes have been developed so that they address all the lifecycle stages and the priorities of the different partners (technology owners, waste owners / operators and WMOs).

Table 5.2 Summary of assessment attributes chosen to assess thermal treatment technologies in the value assessment methodology developed in THERAMIN WP2.

Attribute	Data Category
Operational and Transport Safety	Facility construction and decommissioning
	Waste pre-treatment requirements (conventional and radiological safety implications)
	Waste post-treatment requirements (conventional and radiological safety implications)
	Waste operational safety issues (e.g., ease of providing shielding during operation)
	Transport safety issues
Environmental Impact	Material requirements
	Energy requirements
	Secondary waste and gaseous/liquid discharges generated
	Nuisance (visual and noise pollution, reduction in local air quality)
Impact on disposability/ long-term safety	Ability to meet waste acceptance criteria
	Disposability of secondary waste
Implementation	Indicative lifetime feed
	Ease of achieving required throughput for process (full-scale facility)
	Potential to treat a wide range of waste groups (flexibility) including problematic and orphan wastes
	Impact on waste management strategy
Timescale	Design, construction and active commissioning timescale
	Lifetime operating timescale
	Decommissioning timescale

Attribute	Data Category
Technical Readiness	Maturity of the technology
Strategic Cost Impact	Costs of construction, operation and decommissioning
	Impact on disposal costs (total packaged waste volume and required storage and disposal capacity)

Some attributes are likely to apply over discrete lifecycle stages. Others, such as technological readiness and timescales, cover the whole lifecycle and may not need to be fully differentiated. The lifecycle stages of relevance to the value assessment are:

- Facility design and construction.
- Waste pre-treatment (such as size reduction).
- Treatment operations.
- Waste post-treatment processes (such as conditioning).
- Storage and disposal of the treated waste product.
- Decommissioning of the facility at end of life.

When performing an assessment, it is easiest to evaluate the chosen waste – technology combinations on a comparative basis, so that each thermal processing option may be compared against a non-thermal waste management baseline, such as grout encapsulation. Such a comparative evaluation allows the advantages and challenges of each waste – technology combination to be clearly identified across the full waste lifecycle (i.e., from retrieval of raw waste through pre-treatment, conditioning, packaging, storage and disposal). A comparative evaluation also allows exclusion of management steps for which there is no differentiation between thermal treatment and the non-thermal baseline, thus simplifying the value assessment.

It should be noted that:

- The baseline is typically chosen to be the current reference approach being considered for treatment of a waste stream in one or more country.
- The baseline does not necessarily need to be the same for each waste – technology combination being considered.

Some thought may be needed where a particular waste stream does not have an alternative baseline non-thermal management route. In some cases, the baseline may simply be “do nothing”, or alternatively, a hypothetical baseline management route may need to be constructed. In fact, a “do nothing” approach may not be suitable for comparison because it does not represent a full lifecycle, as no disposable product is produced.

Assessors must decide whether they wish to quantitatively rank and score the different technologies being assessed. However, even if no scoring is applied, the assessment should still highlight the advantages and challenges of the combination being considered. The value assessment methodology developed in THERAMIN considers the strengths of a waste stream/thermal technology combination versus the non-thermal baseline management route. The assessment is qualitative, but structured by attribute, so that it becomes clear what are the key differences, and what are the key strategic reasons why the use of thermal technology may be a more appropriate management strategy than the current baseline. Should assessors wish to apply some quantitative scoring, this should be carefully calibrated so that no technology is unduly favoured. Some guidance on possible scoring criteria are provided for each attribute in the THERAMIN D2.5 report [5].

When scoring the cost impacts of different thermal treatment technologies, one of the most important attributes is likely to be the volume reduction, as this will have a significant impact on lifecycle storage and disposal costs. The two most commonly used metrics to judge this are volume reduction factor (VRF) and waste loading (%).

The volume reduction factor is a measure of the change in volume of the final product with respect to the original waste. It can be calculated as shown below and when VRF > 1, the waste volume was reduced by the process, whereas if VRF < 1, the waste volume was increased by the process.

$$VRF = \frac{Original\ volume\ of\ waste\ (m^3)}{Final\ volume\ of\ treated\ waste\ (m^3)}$$

Alternatively, the efficiency of the treatment process can be assessed using the waste loading, representing the mass of the final product that is waste (rather than additives). Here a higher % waste loading indicates a more efficient process (and a lower total number of final waste packages).

$$Loading(\%) = \frac{mass\ of\ waste\ (kg)}{total\ mass\ of\ product\ (kg)} \times 100$$

Once these values have been estimated, it may be possible to translate the reduction in volume (and waste packages) into an overall cost reduction, but this is reliant on the availability of information and cost may only be approximate.

This synthesis report provides a short overview of the value assessment methodology that was developed during the THERAMIN project as the final output of WP2. It is designed to encourage structured decision making about the implications of utilising thermal treatment technologies for radioactive waste management. Further details can be found in THERAMIN project Deliverable D2.5 [5].

The methodology provides a generic starting point that can be tailored to the needs of the assessor. It is recommended that before conducting any assessment, the required objectives should be clearly defined and the assessment approach tailored to achieve these. For example, a decision should be made on whether the output should be a qualitative narrative (simply listing the advantages and challenges associated with each technology) or include a quantitative scoring. Additionally, the level of detail required in the output should also be decided, so that sufficient information can be gathered to support the analysis. The level of information required is likely to depend on the stage in the decision process that the assessment is being done. For a scoping assessment (on a wide range of options) the relatively small amount of information available in the public domain may be sufficient to allow a high-level judgement to be made. However, at later stages in the process where more robust and fully supported arguments are required, a greater level of detail would be necessary.

6 Summary

The THERAMIN project aimed to identify which wastes could benefit from thermal treatment, which treatment technologies are under development in participating countries, and how these could be combined to deliver a wide range of benefits. This section presents the key conclusions from the project (Section 6.1) and identifies remaining uncertainties, R&D requirements, and next steps for the development of thermal treatment technologies (Section 6.2).

6.1 Conclusions

In WP2, a summary-level inventory of European radioactive wastes potentially suitable for thermal treatment was collated, and a strategic analysis of the drivers and benefits of applying thermal treatment techniques to these wastes was conducted. The availability and level of maturity of thermal treatment technologies in European countries was also summarised; over 25 facilities were identified throughout Europe. Waste groups identified within several countries' inventories that could potentially be suitable for thermal treatment include alpha waste (including PCM), bitumen-conditioned waste, cement-conditioned solid waste, metallic wastes, IER and sludge and concentrates. A strategic gap analysis concluded that, although a few European countries may have the resources to thermally treat their own wastes, many other countries could benefit from cross-country collaboration and treatment of wastes outside their borders.

In WP3, the THERAMIN project successfully demonstrated the applicability of six different thermal treatment technologies (SHIVA, In-Can Melter, GeoMelt®, thermal gasification, vitrification and HIP) to a range of waste groups (labelled WP3 in Table 6.1). Advantages of thermal treatment demonstrated by WP3 include significant volume reduction (this is highly dependent on the composition of the waste and any need for further conditioning) and reduced voidage. The following generic thermal treatment challenges were identified:

- Feeding the waste into the thermal treatment process, which in some cases required pre-treatment (e.g. size reduction, compaction, calcination).
- Optimisation of feedstocks and operational parameters (e.g. melt viscosity, presence of a cold cap) to improve waste loading and radionuclide retention.
- Further conditioning is required for the products of some thermal treatment processes (e.g. gasification, incineration, pyrolysis) to meet the WAC for some disposal facilities.

In WP4, the thermally treated products of these and other trials have been characterised (labelled WP4 in Table 6.1) and these data used to undertake preliminary disposability assessments. Although further studies of long-term behaviour are required to fully assess the suitability of the product for safe disposal, characterisation has demonstrated the removal of volatile components, organic complexants and water, which has benefits in terms of reducing the potential for gas generation, corrosion of storage and disposal containers and radionuclide transport rates within the disposal facility. Additionally, characterisation demonstrated that, where present in the original waste, organic species were destroyed by thermal treatment.

A Value Assessment methodology was also developed and trialled in WP2. The methodology is intended to assist stakeholders in assessing the 'value' (defined as realisable benefit in safety, monetary and environmental outcomes from implementing an option at a specified time) of a treatment technology when used to treat a particular radioactive waste stream. This trial highlighted the need for a more systematic approach to the determination of waste loading and volume reduction factors.

A community of thermal treatment specialists has been developed through the THERAMIN project, which provides a forum for sharing experience, understanding challenges, and discussing and identifying potential solutions.

Table 6.1 Waste-technology combinations tested in THERAMIN WP3 and WP4.

Technology	Sludge	Cement conditioned wastes	Organic ion-exchange material	Inorganic ion-exchange material	Ash	Inorganic liquor	Mixed solid waste	Uranium
GeoMelt®	WP3 WP4	WP3 WP4		WP4				
Hot Isostatic Pressing	WP3 WP4			WP3 WP4				WP4
SHIVA			WP3 WP4	WP3 WP4				
In-Can Melter					WP3 WP4			
VICHR Vitrification						WP3 WP4		
Thermal Gasification			WP3 WP4					
Plasma vitrification		WP4	WP4				WP4	
Incineration					WP4		WP4	

6.2 Next Steps

Remaining uncertainties, R&D requirements and next steps for the development of thermal treatment technologies were discussed during a panel session of WP Leads at the THERAMIN conference in Manchester in February 2020, and at a THERAMIN General Assembly meeting in May 2020. The following areas of further work were identified:

- Maintenance and development of the community of thermal treatment specialists developed through the THERAMIN project.

- Gathering of further information to support comparison of thermal treatment technologies against the baseline.
- Optimisation of thermally treated product composition to increase waste loadings and/or improve wasteform performance.
- Development of WAC for thermally treated products.
- Understanding of long-term behaviour and chemical durability of thermally treated products.

Each of these topics is discussed in turn below.

6.2.1 Maintenance and development of the community of thermal treatment specialists

The need to maintain the community of thermal treatment specialists was highlighted by attendees at the THERAMIN conference. The following opportunities to continue to develop this community were identified:

- Related work in the new EC PREDIS (Predisposal waste management) project, described in Section 6.2.2 below.
- The IAEA has an ongoing programme of meetings and workshops, including a meeting on establishing standard tests in November 2020, which would benefit from the experience of THERAMIN WP4 participants.
- Regular thermal treatment meetings organised by the University of Sheffield HADES facility, a national centre of excellence to support research and innovation in High Activity Decommissioning Engineering & Science, as part of a wider network of UK National Nuclear User Facilities. These meetings will be approximately annual, free to attend and open to all THERAMIN participants and other interested stakeholders.
- A future Sustainable Nuclear Energy Technology Platform (SNETP) – Nugenia forum session on thermal treatment.

6.2.2 Gathering of further information to support comparison of thermal treatment technologies against the baseline

To support further consideration and implementation of thermal treatment options for radioactive waste, further information is likely to be needed to support baseline change on a waste group and waste-stream specific basis.

At the THERAMIN conference, it was noted that thermal technologies are routinely implemented for the treatment of certain LLW/ILW groups in the USA (as described by INL and VNS) and that continued sharing of operational experience would increase confidence and support further implementation in Europe. Gathering of further information on thermal treatment processes and products will be needed to support comparison of thermal treatment technologies against the baseline for specific waste streams, as identified at the value assessment workshop. Specifically, this includes reducing the uncertainties in data such as operational throughput/scalability, secondary waste generation, carbon footprint, waste loading, volume reduction factors, costs and disposability of the product (discussed in Section 6.2.5).

The THERAMIN project also identified how countries with waste could be put in contact with countries that had technologies/facilities available to treat those wastes. Possible ways of enabling the implementation of these opportunities were also identified, and it was noted that further consideration of transport, container availability, requirements of the IAEA Transport Regulations and inter-Governmental agreements to enable waste transfers for treatment to be arranged, and potential licensing of technologies would need to be considered. Additionally, for co-operation such as this to occur, confidence in the maturity and availability of waste treatment routes is needed to encourage waste owners / operators to consider thermal treatment options within their decision-making processes.

The EC project “PREDIS” (Pre-disposal Management of Radioactive Waste) will run for four years from September 2020. The objectives of PREDIS include developing new solutions for treatment and conditioning of waste for which no industrially mature or adequate solution is currently available, and improving existing solutions with safer, cheaper or more effective processes. The PREDIS project, while building on the understanding developed within THERAMIN, covers a broader range of technologies and management approaches, and includes four waste group-focused WPs identified through discussions with End Users:

- Organic liquids.
- Organic solids.
- Metallic waste.
- Cemented wastes (focusing on long-term behaviour, including automation and monitoring of stores).

The PREDIS project will also consider the wider strategic implementation of treatment and conditioning solutions, and the work proposed will support the comparison of different management options for the four waste groups.

6.2.3 Optimisation of thermally treated product composition

While the thermal treatment technologies trialled in WP3 were successfully demonstrated, they have not yet been optimised for the specific composition of the waste that will be treated, and therefore higher waste loadings and improved wasteform performance are likely to be achievable with further work, as has been completed for HLW in France [28] and the UK [29]. This is likely to involve:

- Laboratory-scale and bench-scale tests for specific waste stream – treatment technology combinations to optimise ratios of e.g. glass-formers, additives, different waste groups, and to understand the implications for product composition.
- Characterisation of the samples from these tests, as described in Section 4.2.

- Systematic pilot-scale optimisation trials (not necessarily active) for both the process and treatment equipment, in order to upscale and adapt the process design to the specific characteristics of the waste stream (e.g., feeding system, off-gas treatment system).
- Active trials, as many of the THERAMIN tests were completed with inactive surrogates.
- Development of data to demonstrate product compliance with WAC as well as mechanistic understanding and prediction of product evolution in the disposal environment, to evidence the post closure safety case for the disposal facility.

6.2.4 Development of WAC for thermally treated products

The THERAMIN project developed generic disposability criteria that can be used to evaluate the primary products from any form of thermal treatment, which are equally applicable to any packaging or disposal concept, regardless of the engineered barriers that are present, and in any disposal environment, regardless of its characteristics and the nature of the host rock / geology. However, it is recognised that waste management organisations will also need to develop their own disposability criteria, tailored to a particular context, for application in national waste management programmes. Differences have also been identified between criteria for surface and geological disposal facilities and between waste containing short-lived and long-lived radionuclides.

Several other collaborative EC projects are considering, or have recently considered, the development and application of WAC within their work programmes:

- The EC Horizon 2020 project “CHANCE” (Characterization of Nuclear Waste for its Safe Disposal in Europe) aims to address issues associated with the characterisation of conditioned wastes. It aims to establish a comprehensive understanding of current conditioned radioactive waste characterisation and quality control schemes across European national radioactive waste management programmes. Knowledge of applicable WAC is an important input to CHANCE in order to identify characterisation requirements [30].
- WP9 (“ROUTES”) of the European Joint Research Programme on Radioactive Waste Management (EURAD), which commenced in June 2019 and runs for five years, focuses on waste management routes in Europe from cradle to grave. Task 4 involves identification of WAC used in Member States for different disposal alternatives in order to inform development of WAC in countries without WAC / disposal facilities.
- The EC PREDIS project also includes scope for the wider investigation of generic WAC.

There are opportunities for synergy across projects and a need to ensure future project activities learn from recent developments, as well as opportunities for individual countries to develop specific WAC for vitrified LLW/ILW building on these outputs.

6.2.5 Understanding of the long-term behaviour and chemical durability of thermally treated products

HLW glass wasteforms are homogeneous and their leaching behaviour is well understood. However, LLW/ILW wasteforms produced by thermal treatment may be more heterogeneous (depending on the waste, the thermal process – for example, thermal gasification and incineration produce ashes – and any additives or glass formers, as discussed in Section 4.2.2) and so their leaching behaviour may be more complex. A range of observations were made at the THERAMIN conference on the need and scope for further work in this area:

- In terms of chemical durability, the benefits provided by thermal treatment for vitrified ILW should be evaluated based on other types of ILW (e.g. cement-encapsulated waste) rather than an HLW glass.
- The standardised conditions of the characterisation tests, to allow a generic evaluation, were needed to allow a broader use of the results. However, the safety case implication studies showed that choice also limits the interpretation of these results. Therefore, these characterisation tests should be more specific and integrate repository conditions, with at least, a distinction between surface and geological disposal.
- It may be beneficial to develop a new leaching test, or new standard sample (i.e. ISG or SON68) for ILW glasses; however, care is needed in their use as they are not directly comparable to the thermally treated products studied in WP4.
- The durability evaluation of geopolymerisation for LLW/ILW may still require further work to understand the long-term behaviour of waste conditioned with this process.

If improved long-term behaviour of vitrified LLW/ILW is substantiated, compared to the baseline, this may be beneficial in supporting the development of a safety case for a disposal facility, by reducing some areas of uncertainty.

There will be a need for continued characterisation of the products of the optimisation trials identified in Section 6.2.3 and further development of understanding of the disposability implications in conjunction with the development of both thermal treatment technologies and surface, near-surface and geological disposal concepts, as applicable in each national context.

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