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Thermal treatment for radioactive waste minimisation and hazard reduction: overview and summary of the EC THERAMIN project

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Abstract. The EC 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. Thermal pre-treatment or immobilisation processes result in significant volume reduction, waste passivation and destruction of organic materials, which reduces risks during waste storage and supports development of safety cases for geological disposal. This paper presents the key conclusions from the project. The potential for thermal treatment of European radioactive waste streams was evaluated and a "Value Assessment" framework was developed to assist in decision making, taking into account all stages of the waste management lifecycle. An overview is given of the strategy followed in performing demonstration trials and subsequent waste product characterisation for a range of waste groups. Case studies for select demonstrator-waste group combinations provide information about these processes in greater depth, including details about the treatment technique, resulting waste product and its characterisation, and disposability implications. Finally, the key conclusions from the project are summarised.

1. Introduction

The THErmal treatment for RAdioactive waste MINimisation and hazard reduction (THERAMIN) project was a European Commission (EC) programme of work jointly funded by the Horizon 2020 Euratom research and innovation programme and European nuclear waste management organisations (WMOs). THERAMIN ran from June 2017 to May 2020. Twelve European organisations (including WMOs, research institutes and consultancies), from seven European countries (Belgium, France, Germany, Lithuania, Slovakia, Finland and the UK), participated in THERAMIN.

The overall 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

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synthesis of project outcomes and their dissemination to interested parties. The links between these work packages are shown in Figure 1 and each is discussed in turn below.

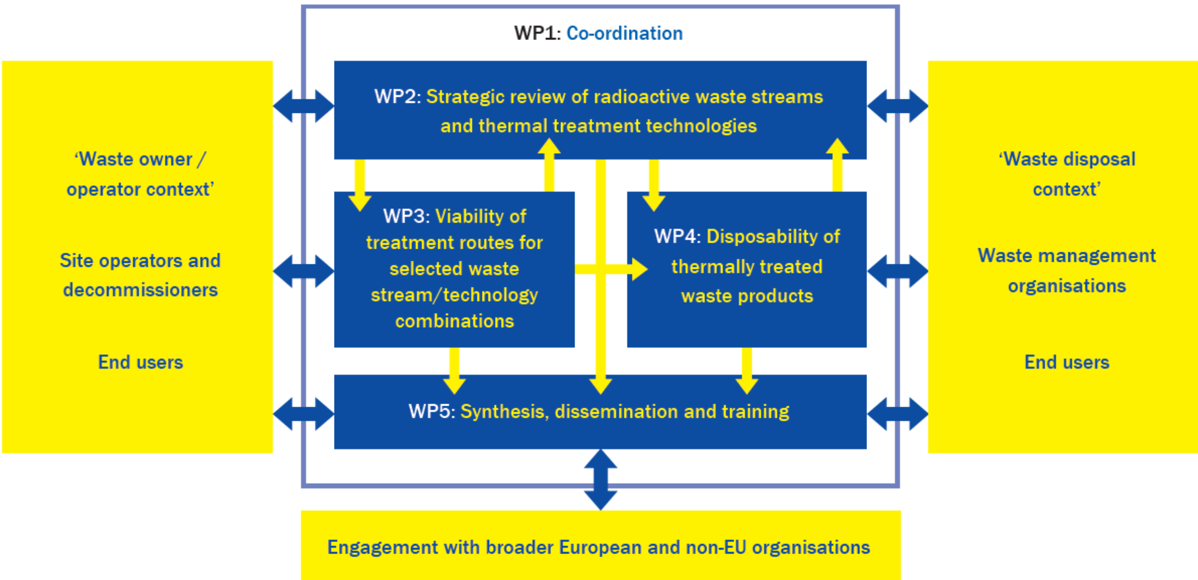

Figure 1. Structure of the THERAMIN project.

2. Work Package 2: Opportunities for thermal treatment: a strategic review

WP2 evaluated the potential for thermal treatment of particular waste streams across the EU and Ukraine, including consideration of where the greatest benefits from EU-wide collaboration may be realised, and development of a methodology to support decision-making on the potential use of thermal treatment technologies to treat various radioactive wastes [1]. To achieve this, wastes in Europe that are potentially suitable for thermal treatment, and potential thermal treatment technologies available in Europe, were identified. This information was used to derive a viability matrix in which the suitability of particular technologies to treat particular waste groups was shown [2].

Inventory information on radioactive waste streams that could benefit from thermal treatment, including any specific issues associated with treatment, processing, packaging or transport, was obtained by distributing a questionnaire to the seven THERAMIN-participating countries and Ukraine. The resulting database provided volumetric information about potential candidate wastes for thermal treatment; wastes occurring in several countries include alpha-contaminated waste, bitumen- or cement-conditioned solid waste, metallic waste, mixed waste, ion exchange materials and sludges/concentrates. It also enabled a strategic analysis of the benefits of thermal treatment for each country, by considering the drivers for thermal treatment and the risks and barriers to applying this treatment.

The availability and maturity of thermal treatment technologies within Europe was also summarised within WP2. The survey identified a wide range of techniques that could be grouped into three high-level process types: thermal treatment for volume reduction and passivation (eight technologies), conditioning by immobilisation in glass (eight technologies), and conditioning by immobilisation in ceramic or glass-ceramic matrices (one technology). Potential thermal treatment technologies and example facilities were identified, and the range of wastes they had demonstrably treated, or are theoretically capable of treating, was considered.

The final task of WP2 was to develop a Value Assessment methodology, which was intended to assist stakeholders in assessing the 'value' of a treatment technology when used to treat a particular radioactive waste stream. Value, in this context, was defined as realisable benefit in safety, monetary and environmental outcomes from implementing an option at a specified time. This included benefits and challenges across all stages of the waste management lifecycle. The assessment framework was

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based on a range of attributes that considered all of these different aspects of value across the whole waste management lifecycle [1].

The Value Assessment methodology was developed through the THERAMIN project, before being tested and refined at a workshop involving THERAMIN project Partners and End Users in the project's final year. The methodology is a generic starting point that can be tailored to the needs and context in which the assessment is being completed and the stage in the decision-making process that it is intended to inform.

3. Work package 3: Demonstration of thermal treatment technologies

Work within WP3 successfully demonstrated the thermal treatment of a range of waste groups in demonstrations carried out at existing thermal treatment facilities across the EU – namely, the SHIVA, In-can Melter, Geomelt®, thermal gasification, vitrification and hot isostatic pressing (HIP) demonstrations. These trials are described in detail in [3] and are summarised below. Photographs of selected pilot and full-scale plants are shown in Figure 2.

SHIVA (Advanced Incineration-Vitrification Hybrid System): the CEA incinerated waste using a plasma burner and vitrified the resulting ashes in a cold-wall direct glass induction melting system. The process is well suited to treating organic and mineral waste with high alpha contamination and results in a homogeneous vitrified product.

In-can Melter: the CEA vitrified inactive ash (by-products of incineration of organic waste) within a metallic crucible, which was heated in a refractory furnace using electrical resistors. This allowed in-container vitrification and the production of a crystallised glass product.

GeoMelt® in-container vitrification: the National Nuclear Laboratory (NNL) treated two combinations of surrogate wastes, with added radioactive tracers: cemented products (surrogate sea disposal drums) and soil; and Magnox sludge and clinoptilolite ion exchange media. Both experiments successfully demonstrated co-processing and resulted in a vitrified waste product.

Thermal gasification: VTT treated organic ion exchange resins, reducing the volume significantly and producing a fine dust product requiring further immobilisation e.g. by geopolymer encapsulation, before disposal. The method can also be used for low-level operational waste containing organic matter, if crushed before treatment.

Vitrification: inorganic liquid (chrompik) waste was vitrified by heating with glass frit in an inductively heated melting crucible by VUJE and JAVYS. Following the evaporation of water and continued heating for 6 hours, the resulting vitrified product was poured into a storage container. Off-gas from the process was decontaminated via a sorption column.

HIP: NNL and the University of Sheffield used HIP to demonstrate treatment of surrogates for Magnox sludge and clinoptilolite. The University of Sheffield completed an active trial, demonstrating the use of an active furnace isolation chamber (AFIC) that allows processing of radioactive waste simulants without risk of contamination to the processing equipment [4].

These processes and the resulting products are illustrated in Figure 2.

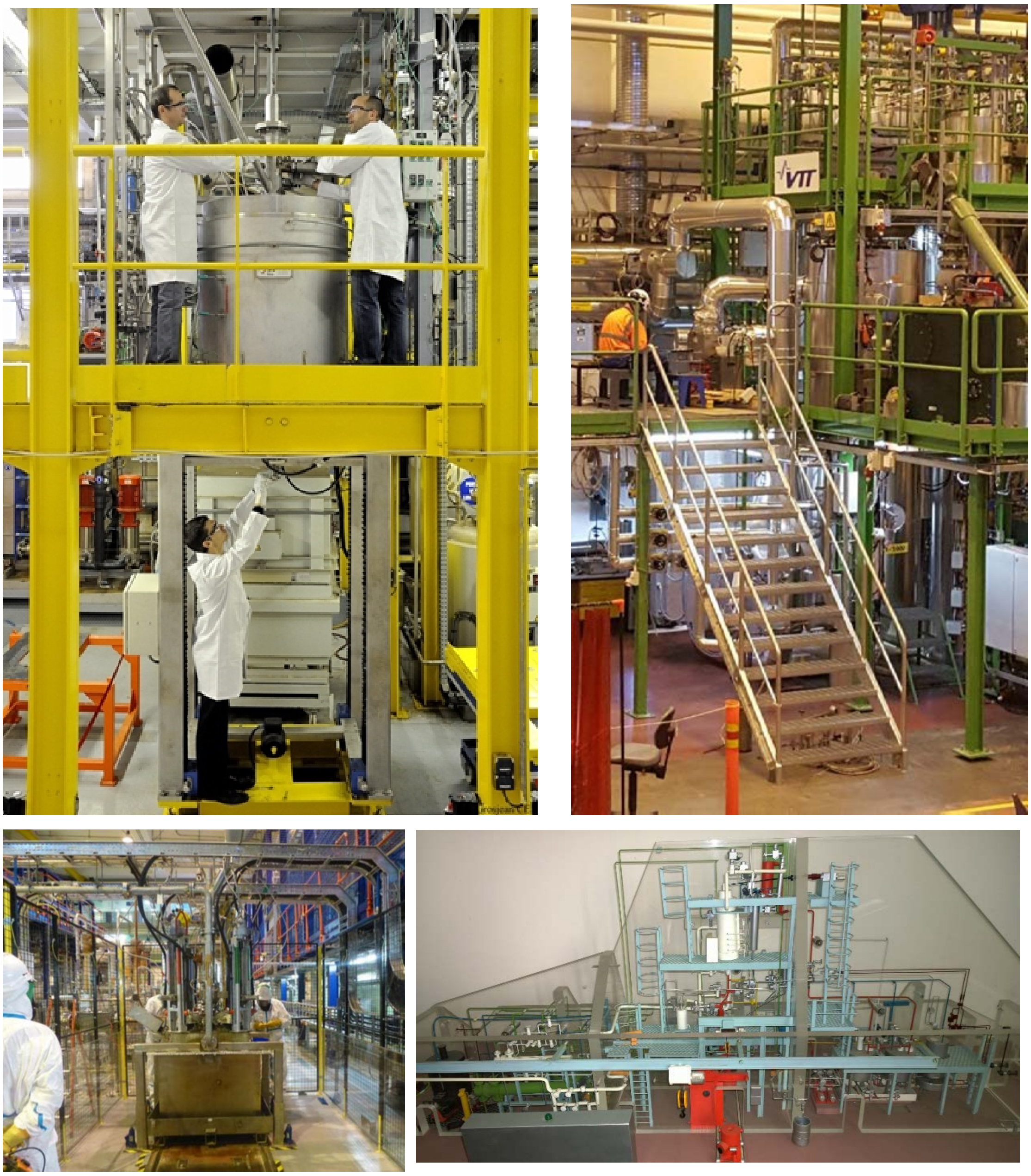

Figure 2. Thermal treatment facilities (from top left, clockwise: In-can melting pilot plant, CEA/Orano; VTT's pilot scale thermal gasification test facility; model of the vitrification facility, VUJE/Javys; Geomelt rig, NNL).

4. Work Package 4: Characterisation and disposability of thermally treated waste products

In order to be disposed of, radioactive waste products must comply with the Waste Acceptance Criteria (WAC) for a disposal facility. The 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 the disposal facility. Compliance with the WAC is confirmed by characterisation of the thermally treated products.

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4.1. Identification and review of criteria and requirements for the disposability of thermally treated waste products

WP4 involved the identification and review of criteria and requirements for the disposability of thermally treated waste products; the characterisation of thermally treated waste products and secondary waste; and consideration of the downstream / safety case implications for disposability of the waste products. This also required consideration of the differing disposal concepts and national contexts of the countries participating in THERAMIN [5].

A set of generic disposability criteria were derived following review of national WAC and other disposability requirements applicable in individual countries (collectively referred to here as 'national disposability criteria') [5]. To this end, information relevant to the management of radioactive waste and application of thermal treatment, such as the approach to classifying radioactive waste, a summary of the inventory, the status of disposal facilities, planned development activities and known issues, was obtained from each of the seven countries participating in THERAMIN.

The generic disposability criteria were intended to be applicable to any packaging or disposal concept, any thermally treated waste, and any disposal environment. These criteria were [6]: no free liquid or gas; permeability and/diffusivity of the waste; no or limited hazardous material content; immobilisation of radionuclides; limited voids / limited porosity; homogeneity of the thermally treated product; leaching behaviour of the waste product; mechanical resistance of the waste product; no metal with a redox lower than 0.84 V Standard Hydrogen Electrode (SHE); and the thermal behaviour of the waste.

The criteria were also designed to highlight the impact thermal treatment can have on waste disposability. Characterisation requirements for thermally treated products in order to test compliance against each criteria, and suitable analytical techniques that could enable this, were then identified [7].

4.2. Characterisation of thermally treated waste products

Characterisation tests were completed for samples of thermally treated products from all the demonstrations carried out within WP3 (Section 3) and some additional samples from thermal treatment processes not tested in the project (shown in Table 1). A common basis for solid characterisations was chosen to test [7]:

- the degree of homogeneity of the sample and to verify the absence of free liquid or gas;
- the overall chemical composition of a homogeneous sample of the local compositions of a heterogeneous sample;
- and the amorphous or crystalline nature of a sample and the structure of the crystals present in the crystallised sample.

The analytical techniques available to all THERAMIN partner laboratories which constitute this common basis were scanning electron microscopy, X-ray fluorescence spectrometry, electron microprobe, inductively coupled plasma analysis after dissolution of the solid and X-ray diffraction. The chemical durability of the samples 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)" [8]. Additionally, depending on the nature of the samples and the national waste management context, other techniques, such as total organic and inorganic carbon analyses, gas physisorption to determine the specific surface area of a powder sample, thermal conductivity or transmission electron microscopy were used if available.

The characterisation results for 17 samples, including glass, ceramic, ashes and geopolymerised products following thermal treatment are presented in [7], including a description of the sample macrostructure and microstructure, chemical composition and chemical durability.

Figure 3. Thermal treatment products (left, HIP containers, NNL; right, glass sample from the SHIVA trial, CEA/Orano).

4.3. Disposability and safety case implications

The disposability of thermally treated waste products and the resulting secondary wastes was evaluated for a subset of the characterised samples, based on the disposability criteria of the country in question or the generic WAC derived in [6]. These assessments have been completed for both near-surface and geological disposal facilities by the WMOs within the THERAMIN End User Group.

As an example, the disposability of ion exchange resins that have been incinerated and vitrified using the SHIVA process (as described in Section 3) has been qualitatively evaluated by Andra [9]. Based on characterisation performed by CEA [7], the final product consists of an amorphous alumina-borosilicate glass with radionuclide incorporation in the glass matrix. The characterisation results indicate that organic matter has been removed by thermal treatment, leading to favourable characteristics for geological disposal due to the absence of organic complexing compounds and potential sources of hydrogen gas release. The rate of radionuclide release will be directly dependent on the chemical durability of the glass product; the long-term behaviour of the final product needs to be assessed in more detail but is considered to compare favourably to the instant release model associated with the baseline cemented ion exchange resin wasteform.

5. WP5: Dissemination of project results and outcomes

WP5 aimed to disseminate knowledge and outcomes of the project within the technical community and more widely. This was achieved through a variety of activities, including training, organisation of a project conference and development of a project synthesis report.

Training placements were hosted by the University of Sheffield, VTT and the CEA in conjunction with the thermal treatment trials conducted as part of WP3. These placements were attended by students and researchers either early in their career or from countries with less well-developed thermal treatment technologies. A technical training school, hosted by the CEA, was held in Marcoule in June 2019. The course, which included site visits to the CEA's thermal treatment rig hall and the neighbouring EDF-Cyclife incinerator and metal melting facility, was attended by 20 people. Presentations from the course, which aimed to share the key learning points of the project with a wider audience of non-project participants, were also made available on the THERAMIN website.

In February 2020, the THERAMIN Conference was held in Manchester which brought together project participants, researchers and WMO representatives worldwide, and for which this publication constitutes the conference proceedings. The EC also funded attendance at the conference for three students, as part of the training activities described above.

Finally, WP5 will deliver a project synthesis report, which will be publicly available from end May 2020 and will contain an overview of the main research, activities and outcomes of the project.

6. Conclusions from the THERAMIN project

The THERAMIN project successfully demonstrated the applicability of six different thermal treatment technologies to a range of waste groups (labelled WP3 in Table 1) and provided tools to compare these technologies with each other and with established baselines, where present. The thermally treated products of these and other trials have also been characterised (labelled WP4 in Table 1) and these data used to undertake preliminary disposability assessments. Although further studies of long-term leaching behaviour are required, the destruction of organic species by thermal treatment has been demonstrated to provide benefits in terms of reduced waste volume, reduced gas generation and removal of complexants that could increase the rate of radionuclide transport within the disposal facility.

A community of thermal treatment specialists has been developed through the THERAMIN project, which provides a forum for sharing experience, understanding challenges and discussion and identification of potential solutions. At the THERAMIN conference, opportunities to continue to develop this community were identified, including related work in the new EC PREDIS (Predisposal waste management) project and annual meetings in the UK supported by the University of Sheffield HADES facility [10].

Table 1. Waste-technology combinations tested in THERAMIN WP3 [3] and WP4 [7].

<table>
<tr><th>Technology</th><th>Sludge</th><th>Cement conditioned wastes</th><th>Organic ion-exchange material</th><th>Inorganic ion-exchange material</th><th>Ash</th><th>Inorganic liquor</th><th>Mixed solid waste</th><th>Uranium</th></tr>
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