



# **theramin 2020** thermal treatment of **conference** radioactive waste

**Mechanics' Institute, Manchester**

**Tuesday 4<sup>th</sup> and Wednesday 5<sup>th</sup> February 2020**

Plus optional visit to University of Sheffield on Thursday 6<sup>th</sup> February 2020

Editors: S.J. Scourfield, J.E. Kent, A.W. Banford and N.C. Hyatt



**UK Research  
and Innovation**

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## Welcome

The organising committee are pleased to welcome you to the EC THERAMIN (Thermal Treatment for Radioactive Waste Minimisation and Hazard Reduction) Project Conference. The conference will share the results of the THERAMIN project and other recent developments within the field of thermal treatment of radioactive waste.

THERAMIN aimed to identify which wastes 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, including:

- Significant volume reduction.
- Waste passivation.
- Destruction of organic materials.

These benefits reduce risks during waste storage and support development of safety cases for geological disposal.

The agenda is structured according to the main themes of the THERAMIN project:

- Strategic review of radioactive wastes that could potentially be thermally treated and available thermal treatment technologies (Work Package 2, WP2).
- Demonstration of thermal treatment technologies for selected waste stream/technology combinations (WP3).
- Characterisation and disposability of thermally treated products (WP4)

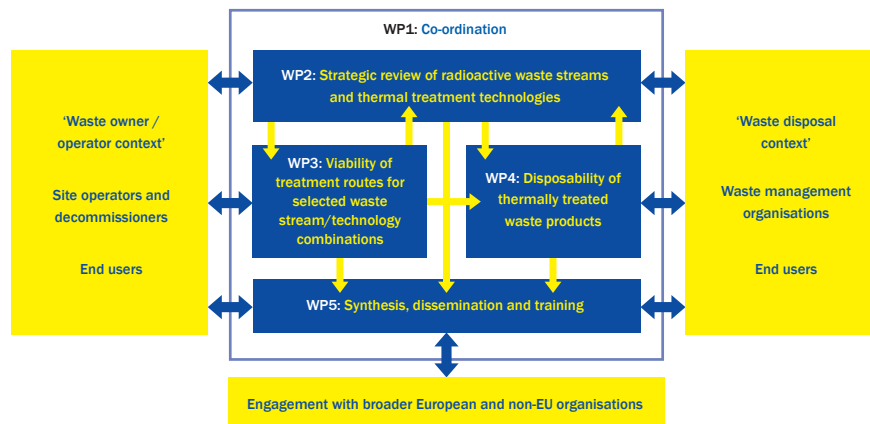


Figure 1: Structure of and interactions between the work packages in the THERAMIN project.

The THERAMIN project involved a consortium of 12 partners representing a European-wide community of experts on thermal treatment technologies and radioactive waste management and disposal. The project included an advisory group of waste producers and management organisations to provide an end-user view. This project has received funding from the Euratom research and training programme 2014-2018 under grant agreement No 755480. The work presented at the conference reflects the views of the authors and the European Commission is not responsible for any use that may be made of it.

The THERAMIN conference is being co-hosted by HADES Facility at the University of Sheffield. The HADES Facility was

established by investment of £1M from UK Research and Innovation / Engineering & Physical Sciences Research Council as a national user facility and centre of excellence for research in radioactive waste management and disposal (investment of £1M from UK Research and Innovation / Engineering & Physical Sciences Research Council as a national user facility and centre of excellence for research in radioactive waste management and disposal (under grant EP/T011424/1). The award, as part of the National Nuclear User Facility network, will leverage prior recent investment of ca. £8M in existing state of the art infrastructure for radiological materials science, from UK Government, the University of Sheffield, EPSRC, NDA, the Royal Academy of Engineering, and others.

## Organising committee

The conference organising committee consists of:

**Professor Neil Hyatt,**  
Department of Materials Science & Engineering, The University of Sheffield.  
**Dr Jenny Kent,**  
Galson Sciences Ltd, UK.  
**Dr Anthony Banford,**  
National Nuclear Laboratory, UK.

In addition, Dr Sally Scourfield (Galson Sciences Ltd.) and Dr Laura Gardner (The University of Sheffield), played a pivotal role in the organisation of the meeting. The organising committee is grateful to Dr Maxime Fournier (CEA), Dr Matti Nieminen (VTT), Dr Stephen Wickham (Galson Sciences Limited) and Charlie Scales (National Nuclear Laboratory) and other conference participants for peer review of papers submitted for inclusion within the conference proceedings.

## Agenda

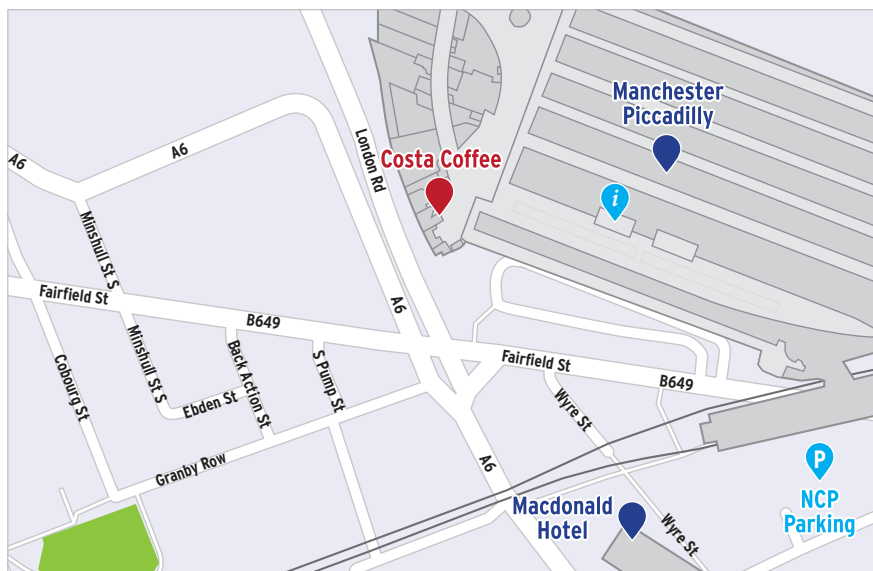
The THERAMIN conference is being held at the Mechanics' Institute, Manchester on 4<sup>th</sup> and 5<sup>th</sup> February 2020, followed by an optional visit to the Immobilisation Science Laboratory at the University of Sheffield on 6<sup>th</sup> February 2020.

Day One – Tuesday 4 <sup>th</sup> February 2020		
0900 – 1030	Registration (with refreshments)	
1030 – 1035	Start of conference	Chair
1035 – 1050	Welcome, overview of project – objectives, main work packages, outcomes	Matti Nieminen, Project Co-ordinator
	WP2 session	
1050 - 1055	Introduction to WP2 session	Steve Wickham, GSL
1055 – 1125	IAEA's perspective on the role of thermal technologies in processing radioactive waste	Willie Meyer, IAEA
1125 – 1155	The potential for thermal treatment of UK higher activity wastes	Mark Dowson, Sellafield Ltd.
1155 – 1225	From technology to acceptance criteria: defining thermal treatment's place in the waste management technology mix	Mike Moulin-Ramsden, Veolia Nuclear Solutions
1225 – 1245	Strategic study of thermal treatment of European radioactive wastes	Slimane Doudou, Galson Sciences Ltd.
1245 – 1330	Lunch	
1330 – 1350	Assessing the value of thermal treatment technologies	Adam Fuller, Galson Sciences Ltd.
	WP3 session	
1350 – 1355	Introduction to WP3 session	Sean Clarke, NNL
1355 – 1425	Thermal treatment of Idaho National Laboratory problematic reactive wastes using GeoMelt® In-Container Vitrification	Michael Connolly, INL
1425 – 1455	Active demonstration of the thermal treatment of surrogate sludge and surrogate sea dump drums using the Geomelt ICV melter installed in the NNL Central Laboratory	Charlie Scales, NNL
1455 – 1510	Tea break	
1510 – 1540	DEM&MELT in-can melting technology for the vitrification of D&D and remediation waste	Régis Didierlaurent, Orano
1540 – 1610	Vitrification of inactive incineration ashes in a full scale in can melter	Nicolas Massoni, CEA
1610 – 1640	Plasma technology to recondition radioactive waste: tests with simulated bitumen and concrete in a plasma test facility	Jan Deckers, Belgoprocess
1640 – 1710	Vitrification of Low and Intermediate Level Wastes using Joule Heated Ceramic Melter (JHCM) Technology	Brad Bowan, Atkins
	Close of Day 1	
1710 – 1830	Poster session and refreshments	Poster presenters
1930	Conference dinner at the Manchester Conference Centre	

Day Two – Wednesday 5 <sup>th</sup> February		
0800 – 0830	Registration (with refreshments)	
0830 – 0840	Welcome	Chair
0840 – 0910	French innovative thermal treatment processes for the management of radioactive organic liquid waste	Hélène Nonnet, CEA
0910 – 0940	Hot Isostatic Pressing (HIP): Trials of active and inactive simulant UK ILW wastes	Laura Gardner, University of Sheffield
0940 – 1000	Pilot scale demonstration of the use of Hot Isostatic Pressing (HIP) for the immobilisation of sludge and cemented wastes	Charlie Scales, NNL
1000 – 1030	Gasification based thermal treatment of LILW	Matti Nieminen, VTT
1030 – 1100	Practical Applications for Plasma Vitrification – Treatment of PCM	Leigh Wakefield, Costain
1100 – 1130	Tea break	
	WP4 session	
1130 – 1135	Introduction to WP4 session	Benjamin Frasca, Andra
1135 – 1205	Development of generic criteria for evaluating the disposability of thermally treated wastes	Liz Harvey, Galson Sciences Ltd.
1205 – 1235	Characterisation of current and historic vitreous simulant wasteforms at the University of Sheffield	Samuel Walling, University of Sheffield
1235 – 1320	Lunch	
1320 – 1350	Characterization and stability of selected ash fractions from thermal treatment of radioactive wastes at the JÜV incineration facility	Natalia Daniels, Forschungszentrum-Jülich
1350 – 1410	Geopolymerisation of gasified ion-exchange resins	Tapio Vehmas, VTT
1410 – 1430	Synthesis and characterisation of high ceramic fraction brannerite (UTi <sub>2</sub> O <sub>6</sub> ) glass-ceramic composites	Malin Dixon Wilkins, University of Sheffield
1430 – 1500	Influence of thermal treatment on the disposability of ion exchange resins in deep geological repository: a French case	Lise Griffault, Andra
1500 – 1530	Plasma incineration of cemented concentrates and resins: preliminary evaluation of the disposability of the end product	Katrien Meert, NIRAS/ ONDRAF
1530 - 1600	Panel session - Q&A How has the THERAMIN project moved the state of the art for thermal treatment on? What should the next steps be?	Jenny Kent, GSL and panel of THERAMIN project participants
1600 - 1610	Close of conference	
	Chair	

Day Three (optional) – Thursday 6 <sup>th</sup> February	
Visit to the University of Sheffield Immobilisation Science Laboratories	
0820	Meet at Manchester Piccadilly - see map below for exact location
0845	Train to Sheffield departs (duration 50 mins)
1000 – 1400	Visit to University of Sheffield laboratories
1445	Train to Manchester for those who need it (duration 50 mins)

Map of Manchester Piccadilly and approach from the direction of the Pendulum Hotel. Meeting point outside Costa Coffee, opposite platform 12.



### Useful information: visit to University of Sheffield

When we arrive in Sheffield, the Immobilisation Science Laboratories are a short walk (approx. 20 minutes) from the train station, or you can take a taxi or the tram (Blue line, alight at West Street and walk):

Immobilisation Science Laboratories  
 Department of Materials Science and Engineering  
 University of Sheffield  
 Mappin Street  
 Sheffield  
 S1 3JD

### List of posters

The poster session will take place on Tuesday 4<sup>th</sup> February (Day One) between 17:10 – 18:30. Refreshments will be provided.

Lead Author	Institution	Title
Tom Fletcher	LLWR	Feasibility of thermal treatment techniques for UK problematic waste
Hélène Nonnet	CEA	French innovative thermal treatment processes for the management of radioactive organic liquid waste
Liam Harnett	University of Sheffield	Development of phosphate glass compositions for conditioning of irradiated nuclear fuel residues
Karine Ferrand	SCK-CEN	Characterisation of plasma slags produced from cemented resins and concentrates
Alexandra Mikusova	VUJE Inc.	Material characterisation of Chrompik III glass samples
Maxime Fournier	CEA	Characterisation of wasteforms produced by the SHIVA and In-Can Melter processes
Lucy Mottram	University of Sheffield	Developing laboratory-based X-ray Absorption Spectroscopy (XAS) for nuclear waste management
Hao Ding	University of Sheffield	Micro-XAS analysis of Chernobyl lava
Hao Ding	University of Sheffield	A Feasibility Study of Advanced Gas-cooled Reactor SIMFuel Fabricated by Hot Isostatic Pressing
Tapio Vehmas	VTT	Geopolymerisation of gasified ion-exchange resins, mechanical properties and short-term leaching studies
A. Boyer	CEA	Thermal-hydraulic modelling of a vitrification furnace – DEM&MELT project
Neil Hyatt	University of Sheffield	HADES: A National Nuclear User Facility for high activity decommissioning engineering science
Sally Scourfield	GSL	Thermal treatment for radioactive waste minimisation and hazard reduction: overview and summary of the THERAMIN project
Marianne De Vreese	CEA	Characterisation of calcined and vitrified simulants developed to represent a typical D&D sludge.
Lewis Blackburn	University of Sheffield	Hot isostatically pressed zirconolite wasteforms for actinide immobilisation
J. Hansen	Montair Process Technology	Pyrolysis of radioactive spent resins in the prime installation
Paul Heath	GRI Ltd	Development of HIP process for the treatment of sellafeld sludges and slurries

## Locations

Mechanics' Institute  
103 Princess St,  
Manchester,  
M1 6DD

Phone: +44 (0) 70 9330 1622

Pendulum Hotel and Conference Centre  
Sackville St,  
Manchester,  
M1 3BB

Phone: +44 (0) 161 955 8000

## Conference dinner



A conference dinner will be held at the Manchester Conference Centre (attached to the Pendulum Hotel) on Tuesday 4<sup>th</sup> February at 7:30 pm. Directions from the Mechanics' Centre are shown on the map below.

### Conference dinner menu

If you have dietary requirements and did not indicate these during registration for the conference, please contact a member of the organising committee to arrange alternative menu options.

#### Starter

Caesar salad

*Little gem lettuce with croutons, parmesan shavings  
and classic dressing. Crispy bacon optional.*

Vegetarian option: Vegetable soup  
*Black pepper and herb croutons.*

#### Main

Roast chicken

*Roasted with thyme and garlic and served with gratin potatoes,  
roast carrots and sugar snap peas and red wine jus.*

Vegetarian option: Tagine of Sweet Potato and Chickpeas  
*Served with mushroom and coriander rice.*

#### Dessert

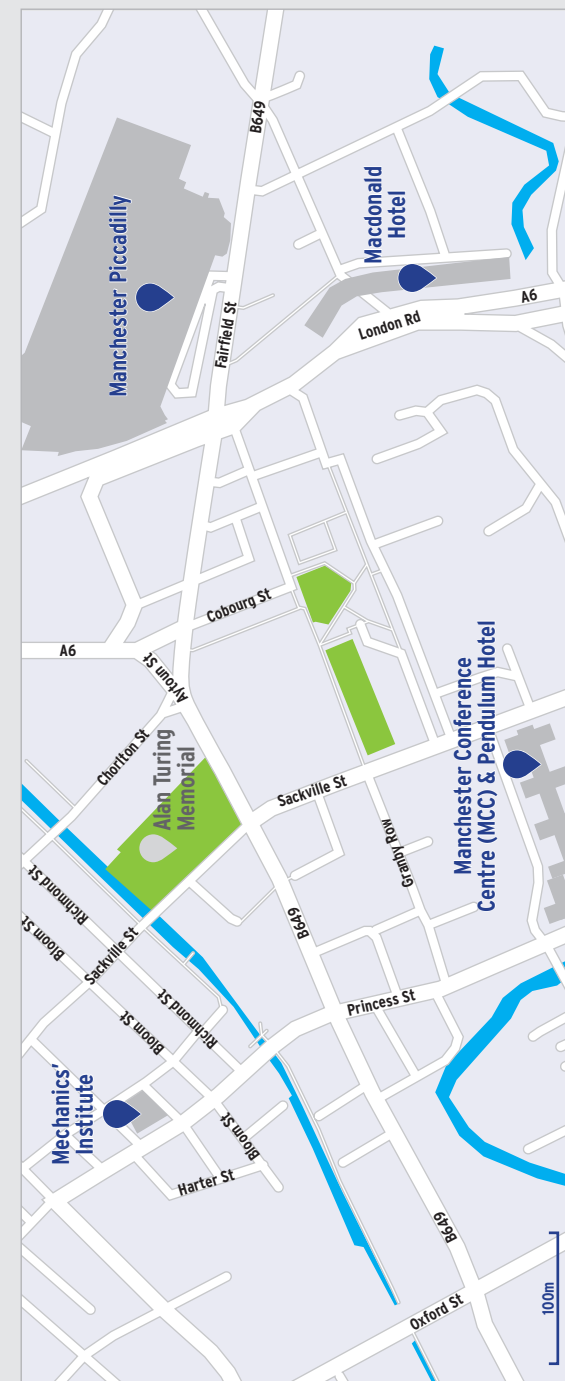
Chocolate truffle torte

*Served with clotted cream ice cream and raspberries.  
Vegetarian. May contain nuts/peanuts.*

## Map and useful transport links

The conference centre is approx. 8 minutes walk (0.4 miles) from the Mechanics' Institute. See map below for directions.

Manchester Taxi Service: +44(0) 1612 708 701





## IAEA's perspective on the role of thermal technologies in processing radioactive waste

W.C.M.H. Meyer

International Atomic Energy Agency (IAEA)  
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The International Atomic Energy Agency (IAEA) promotes the safe and peaceful uses of nuclear energy including the management of radioactive waste according to nationally and internationally agreed principles and standards with safe disposal as endpoint for all waste types.

A wide variety of low- and intermediate-level (LILW) streams require treatment/conditioning to meet the Waste Acceptance Criteria (WAC) for storage and / or disposal facilities. Many technologies, thermal and non-thermal, are operational world-wide to produce stable waste forms suitable for storage and disposal. In this regard, the processing of waste using thermal technologies is important for waste minimization (a key principle) resulting in the production of a stable waste form suitable for storage or disposal.

Although technologies are available for processing the majority of waste streams, there remain some waste streams for which either a suitable technology does not exist or cannot be accessed (for a number of reasons – cost, lack of technical capability, small volume, unknown characteristics). Considering the high overall costs for developing new individual treatment/conditioning processes to immobilize this waste into suitable waste forms for geological disposal, the benefits offered by thermal treatment technologies under development may be able to assist in processing these waste types. Modular or shared/international thermal facilities would facilitate processing

small volume of waste as encountered in many Member States.

The IAEA recognizes that thermal technologies has an important role and as such the Waste Technology Section (WTS) at the IAEA has initiated the International Predisposal Network (IPN) platform to share waste management information (including thermal processing technologies) from publications (predisposal handbooks), Co-ordinated Research Programs (CRP), Case studies, fellowship training and Wiki articles among its members.

The paper conveys the IAEA's perspective on the role of thermal technologies in waste minimization and processing of challenging waste streams and the synergies with the current EC THERAMIN project.

## The potential for thermal treatment of UK higher activity wastes

M. Dowson\*<sup>1</sup> and C.R. Scales<sup>2</sup>

<sup>1</sup>Sellafield Ltd

<sup>2</sup>National Nuclear Laboratory (NNL)

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Sellafield Ltd (SL) manages the decommissioning and clean-up of the Sellafield site in Cumbria, UK. One of the challenges is retrieval and subsequent treatment of higher activity wastes into a form suitable for safe storage and ultimate disposal. SL has an established process for the vitrification of high-level wastes from the reprocessing of spent fuel but aside from operational wastes which undergo cementation, legacy wastes remain which do not have a defined disposition route.

One of the possibilities is to thermally treat the wastes. As well as passivating reactive metals and removing organics which could be an impediment to disposal, a big driver for SL is the reduction in overall waste package volumes leading to large cost savings for on-site storage while awaiting disposal in a geological disposal facility (GDF).

While Sellafield is by far the largest nuclear site in the NDA estate, similar wastes exist at other locations and, to support an estate wide assessment NDA has established a Thermal Treatment Integrated Project Team.

In reviewing the Sellafield Inventory, the use of a Data Quality Objectives (DQO) process has established the potential for thermal treatment and current and historic trials using existing facilities on the Sellafield site and in the supply chain have demonstrated credibility. The use of a commercial engagement process with the supply chain has established a basis on which to make a business case for thermal treatment.

For some wastes such as sludges, ion exchange materials and plutonium contaminated materials (PCM), the need to make decisions in the near term requires action by the individual projects. Alongside that, a site-wide business case is being explored which, if successful, may lead to thermal treatment being deployed on a far wider range of feeds covering for example legacy ponds and silos and decommissioning wastes.

The results of decisions being made in the coming months could result in a strategic shift in how SL and the wider NDA estate treat their waste arisings and bring significant opportunities for the supply chain.

## From technology to acceptance criteria: defining thermal treatment's place in the waste management technology mix

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Veolia Nuclear Solutions has been working with the UK's National Nuclear Laboratory and key UK site license companies to understand the role that advanced thermal treatment could play in the processing and disposition of wastes in the UK. Early in the process it became clear that specific thermal treatment technology capabilities were not the only consideration necessary to evaluate the applicability of this class of techniques to waste challenges. Route constraints such as host site classification & discharge permits, transport considerations, and disposal site acceptance criteria define the envelope within which thermal treatment technology can operate.

The objective of this work was to map these constraints so that key stakeholders could quantify and evaluate the impact thermal treatment could have on their waste inventories from a route, rather than technology, perspective.

The work was carried out by experts with experience in environmental permitting, waste management and site permit arrangements. The result was the creation of a parametric model that can be used to evaluate candidate wastes for a specific waste management route. The technique used also offers a framework for assessing the capabilities of other waste management route options (alternative sites, disposal facilities). This powerful tool brings advanced thermal treatment a step closer to delivering on its promises by helping strategy makers and potential end users quantify capability and applicability, hence informing key decisions.

## Strategic study of thermal treatment of European radioactive wastes

S. Doudou<sup>1</sup>, Emily Swain-Phipps<sup>1</sup>, A. Fuller<sup>1</sup>, S. Wickham<sup>1</sup>, N. Daniels<sup>2</sup>, E. Fourcy<sup>3</sup>, M. Fournier<sup>4</sup>, B. Frasca<sup>5</sup>, K. Meert<sup>6</sup>, A. Mikusova<sup>7</sup>, M. Olin<sup>8</sup>, R. Poskas<sup>9</sup>, C. Scales<sup>10</sup>

<sup>1</sup> Galson Sciences Ltd (UK), <sup>2</sup> Juelich (Germany), <sup>3</sup> Orano (France), <sup>4</sup> CEA (France), <sup>5</sup> Andra (France), <sup>6</sup> ONDRAF/NIRAS (Belgium), <sup>7</sup> VUJE (Slovakia), <sup>8</sup> VTT (Finland), <sup>9</sup> LEI (Lithuania), <sup>10</sup> NNL (UK)  
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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. This paper presents the outcomes of an international review of inventory information concerning European wastes potentially suitable for thermal treatment, a review of thermal treatment technologies, and a strategic gap analysis to identify countries where there are significant waste arisings with potential benefits for thermal treatment, but little prospect of these countries developing thermal treatment facilities independently.

Inventory information was gathered by means of a questionnaire. The waste streams identified by project partners were included in a database and categorised into 14 different generic waste groups. 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 groups of common waste that occur in several countries, such as sludges, ion exchange materials, cement-conditioned wastes, and bitumen-conditioned waste, whilst others may be present in fewer countries (e.g. polymer-conditioned waste and non-organic liquid wastes).

The survey of European thermal treatment technologies 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). The technical details of the technologies, and of specific facilities implementing each technology, were captured including information on waste acceptance criteria and the technical readiness of the facility (e.g. experimental vs full-scale industrial facility). This information was then utilised to identify the specific advantages and limitations of each of the treatment technologies as represented by specific facilities. This allowed the creation of a viability matrix that compares the generic waste groups to thermal treatment technologies that could potentially treat the identified wastes.

A strategic gap analysis was undertaken to identify those countries with wastes identified as suitable for thermal treatment but with no suitable domestic thermal treatment facility. Facilities with the technological capability to treat those wastes in other European countries were identified. Although this analysis does not account for non-technical limitations on movement of waste across international borders, it provides preliminary inputs for future decision making and further analysis taking into account wider considerations on specific waste characteristics and other facility constraints.



## Assessing the value of thermal treatment technologies

A.J. Fuller\*, S. Doudou, J.E. Kent, E.J. Harvey and S.M. Wickham

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Work Package 2 (WP2) of the THERAMIN project provides a framework to support decision making on the potential use of thermal technologies to treat various radioactive wastes. To do this, a methodology based on multi-attribute decision analysis has been devised to identify the benefits of applying thermal treatment techniques to a range of radioactive waste streams.

The value assessment methodology used in THERAMIN follows an attribute-based approach with the potential benefits of the treatment technology assessed against different criteria, considering performance over the whole waste management lifecycle, from pre-treatment through to final disposal.

The attribute groups considered are: operational safety, environmental impact, impact of disposability/long term safety, implementation, timescale, technical readiness, and cost impact. To guide the decision maker, each attribute is split into data categories (sub-criteria) and a summary of topics to be considered under each attribute is provided.

The assessment attributes have been compiled by drawing on knowledge from across the THERAMIN partner organisations to make them as comprehensive as possible. Consideration is given to the whole of the waste life-cycle and to different European regulatory contexts.

## Thermal treatment of Idaho National Laboratory problematic reactive wastes using GeoMelt® in-container vitrification

M. Connolly<sup>\*1</sup>, R. Miklos<sup>1</sup> and B. Campbell<sup>2</sup>

<sup>1</sup>Idaho National Laboratory, <sup>2</sup>Veolia Nuclear Solutions

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Previous liquid metal fast reactor programs, Experimental Breeder Reactor-II (EBR-II), and associated research and development activities at the Idaho National Laboratory (INL) generated a number of reactive metal waste configurations that are problematic to treat and typically lack cost effective treatment methods and disposal options. These problematic wastes include contact handled (<2mSv/hr), remote handled (>2mSv/hr), and materials currently managed as spent nuclear fuel. Handling and treatment of these wastes is further complicated by the complex configurations and reactive nature of the sodium contained within the waste items. Collectively these materials represent a \$1.5B liability for the INL.

In 2016, INL contracted with Veolia Nuclear Solutions to initiate a phased demonstration of the application of GeoMelt® In-Container Vitrification technology to safely convert sodium metal to a non-reactive vitrified oxide form as a potential treatment technology for a large fraction of its \$1.5B reactive contaminated waste liability. Reactive metal wastes at INL require treatment in order to remove the Resource Conservation and Recovery Act (RCRA) reactivity and ignitability characteristics in order to comply with land disposal restrictions. A phased approach including crucible, bench-scale, and engineering-scale studies were conducted on a range of surrogate waste configurations with varying ratios of sodium metal and glass formers. The objective of the phased demonstrations included determination of sodium loading factors and demonstration of

the effectiveness of GeoMelt® In-Container Vitrification technology in deactivation of reactive sodium metal.

Results of the initial studies demonstrated that GeoMelt® In-Container Vitrification technology results in complete sodium deactivation in all of the surrogate configurations evaluated. Due to the success of the initial demonstrations, Veolia Nuclear Solutions invested in deploying a 10-metric ton full-scale GeoMelt® In-Container Vitrification system at Perma-Fix Northwest in Richland, Washington for treatment of reactive metal contaminated waste. As of September 2019, over 900 55-gallon drums containing a total of around 3,500 lb of sodium with low levels of radioactivity have been treated at GeoMelt® Richland, with resulting glass monoliths disposed at the Nevada National Security Site (NNSS). Additional GeoMelt® demonstration tests, focused on more complex shapes and other reactive metals (mocked up EBR-II subassembly, sodium filled heat exchanger, and a can containing sodium potassium alloy), were performed at engineering-scale. A full-scale radiological demonstration melt on an actual EBR-II subassembly will be performed in the fall of 2019.

This paper will present the results of the phased demonstration, discuss the design of the GeoMelt® Richland system, and strategic value of the GeoMelt® technology in addressing INL's reactive metal waste liability.

## Active demonstration of the thermal treatment of surrogate sludge and surrogate sea dump drums using the Geomelt ICV melter installed in the NNL Central Laboratory

C.R. Scales\*, S. Clarke, J. Roe, N. Patel

National Nuclear Laboratory (NNL)

\*charlie.r.scales@uknnl.com

Under the auspices of the THERAMIN project, the Geomelt in-container vitrification (ICV) melter system installed in the NNL Central Lab has been used to demonstrate the immobilisation of two surrogate feeds.

The streams selected for demonstration are a cementitious stream representative of legacy sea disposal drums or failing cemented wastes packages and a sludge stream made up of a surrogate for Magnox storage pond sludge combined with another surrogate waste clinoptilolite, an ion exchanger, as a way of demonstrating co-immobilisation.

Two key fission products of interest in intermediate-level waste (ILW) streams are radioactive caesium and strontium. The cementitious stream was dosed with 25MBq of Cs-137 and the sludge stream, 25MBq of Cs-137 and 16MBq of Sr-85, both sufficient to assess the decontamination factors (DFs) across the GeoMelt process.

Geomelt is a batch process in which a cast refractory box (CRB) is used both as melt vessel and disposal containment. Batches of the waste surrogate and glass formers were introduced into the CRB and positioned in the Geomelt rig. A starter path was used to

begin the melt and the power was gradually increased until the target temperature was reached. Following time held at the target temperature, power to the melt was cut and it was allowed to cool. The CRB was then removed from the rig and the block was drilled to take cores for analysis and characterisation.

Gamma scans were carried out on the samples as well as conventional analysis. The distribution of waste species throughout the block were then assessed to determine the efficacy of the mixing on the melt. Analysis of samples from the off-gas system was carried out to establish DFs across the process.

The presentation will outline the process set up, provide operational data from the melt and discuss the analysis results, concluding with an assessment of the applicability of the trial results to thermal treatment of these and similar wastes.

## DEM&MELT In-Can melting technology for the vitrification of D&D and remediation waste

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Some waste streams from decommissioning and dismantling operations are identified as an issue in terms of handling, transportation, adapted conditioning matrix or evacuation outlet within the existing regulations. In this context, the DEM&MELT project consortium, which gathers Orano, the CEA, ECM technologies and Andra, is currently qualifying a robust, simple and versatile In-Can vitrification process dedicated to D&D and remediation waste. This project is supported by the French government program "Programme d'Investissements d'Avenir". The In-Can melting process has already been developed by the CEA for an internal application (alpha liquid waste vitrification) and can be rated TRL 7.

The DEM&MELT In-Can vitrification process is designed for high or intermediate level waste and is compact enough to be implemented in a decommissioned cell or close to the waste to be treated. It is developed to treat liquid and solid waste, to produce a small amount of secondary waste and to minimize investment and operating costs. This In-Can vitrification process is also developed with a modular design and can be adapted regarding to nuclear operators' needs and requirements; it is flexible enough to accommodate uncertainties on waste composition.

This paper presents the DEM&MELT In-Can vitrification process and its performance for a typical D&D and remediation waste that has a high Cs loading. The vitrification tests results will be particularly discussed in terms of process parameters, glass properties and caesium volatility.

## In-Can vitrification of ash

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The In-Can Melter is a metallic crucible heated in a refractory furnace using electrical resistors allowing in-container vitrification. The In-Can Melter trial aims to demonstrate the feasibility of the confinement in a glassy matrix of ash coming from existing incineration processes. The ash was pelletised to allow

its introduction into the can without dust emissions and then incorporated in a 50 wt.% waste loading confinement matrix. The full-scale trial was preceded by laboratory- and bench-scale tests. The microstructure and chemical durability of the wasteform were characterised.

## Vitrification of Low and Intermediate Level Wastes using Joule Heated Ceramic Melter (JHCM) Technology

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JHCM technology has been deployed in numerous locations across the globe to vitrify and stabilize high level radioactive wastes. The technology has been proven to be robust and flexible for a wide range of HLW characteristics: acidic/caustic, liquid/slurry, and high/low fuel burnup waste origin.

With introduction of bubbling technology to JHCMs in the 1990's, JHCMs have been evaluated and deployed as an economic and safe alternative to stabilize low and intermediate level radioactive wastes. This paper reviews JHCM technology applications for lower activity wastes, where volume reduction, waste form quality and project

economics were key attributes for technology adoption. These applications include the Savannah River M-Area waste stabilization project, the Low Activity Waste vitrification facility at Hanford's Waste Treatment and Immobilization Plant, and recent testing with JHCM technology for the potential treatment of ILW sludges at the Sellafield Site.

## Plasma technology to recondition radioactive waste: tests with simulated bitumen and concrete in a plasma test facility

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The operation and maintenance of nuclear power plants, the non-nuclear fuel cycle, etc generate low-level radioactive waste which, along with the historical radioactive waste from past nuclear activities, needs to be treated and stored, awaiting final disposal. Plasma technology offers a very effective way of treating this waste with a high-volume reduction factor (VRF), free from organics, liquids and moisture, and meets without a doubt the acceptance criteria for safe storage and disposal. By means of a plasma beam of approximately 5000°C, the inorganic materials are melted into a glassy slag, containing most of the radioactive isotopes while the organic material is gasified, oxidized and purified in an off-gas cleaning system.

First the paper describes the new full-scale Plasma Melting Facility (PMF) at the Kozloduy Nuclear Power Plant in Bulgaria which was taken in nuclear operation in May 2018. The plant has a capacity of 250 tons per year and the maximum contact dose rates of the incoming waste is 2mSv/h. Different mixtures of radioactive waste packed in 200l drums were successfully treated resulting in a glassy slag free from liquids and organic material with an important volume reduction factor (VRF). The Project was co-financed through a grant by Kozloduy International Decommissioning fund (KIDSF) administrated by the EBRD through Bulgarian national funding.

Plasma is a suitable technology for treatment of problematic waste or even reconditioning waste so Belgoprocess was contracted to do plasma tests with simulated conditioned waste types. One can do tests on a laboratory scale on smaller samples and torch capacities of e.g. 50kW but Belgoprocess wanted to do more realistic and reliable tests. So Belgoprocess contracted Phoenix Solutions Co who has a full-scope test facility equipped with a 1200kW plasma torch for full-scope treatment of simulated conditioned waste.

For a first confidential contract simulated 200l (55 gallon) bitumen drums were treated. The drums contained different pucks of compacted waste such as rags, used filters, granulates, etc. The pucks were stacked in the 200l drums and subsequently embedded with bitumen. A total of 6 drums were treated in the plasma facility. For a second contract simulated homogeneous 200l (55 gallon) concrete drums with on the one hand concentrates and on the other hand spent resins were selected. A total of 6 drums with concrete and spent resins were treated and melted in the plasma testing facility. The paper describes the test facility, volume reduction factor (VRF) of different waste streams and most important parameters.

## French innovative thermal treatment processes for the management of radioactive organic liquid waste

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The nuclear industry, as well as hospitals and some research laboratories, produce Radioactive Organic Liquid Waste (ROLW) such as scintillation cocktails for medical imaging, radiolabelled molecules for medical research or liquids from former activities. In order to be compatible with disposal facilities' requirements, this waste must be inerted and stabilized in a solid matrix.

The route generally used is incineration in the Centraco facilities, with the ashes produced being cemented downstream. However, liquids whose chemical or radiological characteristics are not compatible with the incinerator operating rules (high level of <sup>14</sup>C or halogen content, corrosive gas production etc.) cannot follow this path.

The purpose of the MILOR project is to propose alternative paths to treat all kinds of ROLW. Initiated in September 2017, this project has the technical objective of developing, over a four-year period, two pilot-scale mineralization processes. Two variants of complementary plasma technologies are evaluated: aerial plasma and submerged plasma processes. Compared with other methods, thermal plasma has unique characteristics such as rapid decomposition with high throughput, fast startup and shutdown, and high-energy delivery.

The aerial process consists of an induction plasma into which the liquids are directly

introduced. It has been designed for treatment rates less than 0.5 l/h. Specific gas treatment systems have been developed to direct the gas produced to a dry or wet gas treatment system. One aim of the project is to subsequently install the process in a radioactive zone at CEA Fundamental Research Division.

The second process involves a submerged plasma at the heart of which organic liquids are introduced. Current research has demonstrated that such a technique may enable the instantaneous and complete destruction of liquids with a wide variety of challenging constituents, such as chlorine, fluorine, or phosphorus. The submersion solution offers many advantages: quenching and cleaning of combustion gases, filtering of the particles they contain and cooling of the entire process, which guarantees excellent corrosion control. An advantage of this type of design is that the gas treatment system can be reduced to a demister-condenser followed by a simple safety filter, thereby offering the additional advantage of an extremely compact treatment system.

The MILOR project, financed by the French State through an investment programme is a tripartite project bringing together Andra, in charge of radioactive waste management, the CEA in charge of leading scientific and technological researches and the company A3i whose role is to ensure the subsequent industrial development.

## Hot isostatic pressing: thermal treatment trials of simulant inactive and radioactive UK ILW

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Hot Isostatic Pressing (HIPing) is a batch process thermal treatment technology where wastes are heated and compressed within a sealed stainless-steel canister; typically resulting in durable, high density ceramics or glasses with minimal loss of volatile elements, and accountability of active inventories. The University of Sheffield has a small-scale research HIP with capability to process simulant wasteforms containing radioactive materials, to help underpin larger-scale industrial applications of this technology. It was under this remit that a series of trials were undertaken, to produce small simulant radioactive wasteforms incorporating problematic UK waste streams such as Magnox sludges and clinoptilolite ion-exchange material.

Each trial was successfully batched, sealed, and HIPed at 1250°C, resulting in solidified products entirely contained within the steel HIP canisters. The ability to safely produce active wasteforms within the same facility validates the active furnace isolation chamber (AFIC) system. Overall, the success of these trials demonstrates the ability of smaller research HIP facilities to build up the scientific and technical case for further implementation of HIP technology as a viable waste treatment option.

## Pilot-scale demonstration of the use of hot isostatic pressing (HIP) for the immobilisation of sludge and cemented wastes

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Under the EU THERAMIN project, the NNL hot isostatic press (HIP) at the Workington laboratory has been used to demonstrate the immobilisation of sludge wastes. This demonstration was carried out at 5 litre scale and was carried out in collaboration with the University of Sheffield who are conducting lab scale trials. Those trials are being presented in an accompanying presentation. The combined output will demonstrate that data obtained in the laboratory can be replicated at a larger scale.

Two trials using HIP were carried out with surrogate Magnox sludge and ion exchange material (clinoptilolite); the aim to demonstrate the immobilisation of the wastes in a glass ceramic composite matrix. Variations in the recipe between trials were made to explore the effect of additional glass formers on wasteform development.

In order to prepare material for HIP-ping, the feeds were first calcined at 950°C for 3 hours to remove water and any other volatiles. Following can filling and bake out, the cans were sealed, and each can be loaded into the HIP and subject to a target pressure of 100MPa at a peak temperature of 1250°C for 2 hours in two separate HIP runs.

Following the HIP cycle, the HIP was unloaded, and the HIP cans removed. A visual observation verified that the cans had consolidated as expected. The cans were sectioned and the material removed for analysis.

This presentation will outline the process set up, provide operational data from the HIP trials and an assessment of the applicability of the trials results to the thermal treatment of these and similar wastes.

## Gasification-based thermal treatment of Low and Intermediate Level Waste

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VTT has developed a gasification-based technology for processing low and intermediate level waste (LILW) rich in organic matter prior disposal. VTT's process is based on well-controlled thermal fluidised-bed gasification followed by efficient gas cleaning, gas conditioning/oxidation, wet scrubbing of the oxidised gas (flue gas), and finally a high efficiency particulate air (HEPA) filter, which acts as a backup cleaning system.

In the case of an organic ion exchange resin (IXR) the primary product from thermal gasification is fine filter dust. In addition, the process produces some bottom ash, which consists mainly of the bed material. The wet scrubber liquid may also contain some activity. Filter dust and bottom ash are powdery and therefore they have to be immobilised before final disposal. VTT has previously developed an advanced immobilisation technology based on geopolymerisation and this is applied also for the samples from the THERAMIN test trials.

Demonstration trials were carried out with unspent organic IXR containing a small amount of added stable Cs in order to simulate Cs content in spent IXR. Gasification tests confirmed the capability of the process to remove organic matter from the IXR and clean the resulting off-gas as required. In THERAMIN test trials simulated waste IXR was reduced to about 1 wt-% of its original mass before immobilisation. VTT's technology has been designed as a compact process, which can be operated at the nuclear power plant site. Until now, all the development and verification test trials have been done using simulated waste materials. Next step of the development is the demonstration of the process with real radioactive waste.

## Development of generic criteria for evaluating the disposability of thermally treated wastes

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The potential to generate wasteforms with enhanced properties that support safe storage and disposal is one of the key factors driving thermal treatment of radioactive waste. Depending on the specific treatment method and waste substrate, thermal treatment can greatly reduce chemical reactivity, yielding a primary product that is more durable than wasteforms produced via non-thermal routes and with reduced potential for gas generation and other detrimental behaviour in a storage / disposal environment. On the other hand, thermal treatment can concentrate radioactivity into a smaller volume, potentially affecting waste handling and / or classification. It also generates secondary wastes whose management requires consideration as part of a holistic evaluation of thermal treatment. Moreover, some thermal treatment routes do not generate a primary product that is directly disposable without further conditioning.

Generic disposability criteria have been derived that can be used to evaluate the primary products from any form of thermal treatment. These generic disposability criteria highlight the 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 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. They could aid waste management organisations in developing their own disposability criteria, tailored to a particular context, for application in national waste management programmes.

## Characterisation of co-mixed HIP wasteforms for magnesium rich material

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Co-mixed simulant wasteforms consisting of calcined Magnox sludge simulant and clinoptilolite, with additions of a glass forming frit and  $\text{CeO}_2$  or  $\text{U}_3\text{O}_8$  were processed using Hot Isostatic Pressing (HIP). This enabled the production of high waste loaded materials, with the successful incorporation of both simulant and active material. These formed heterogeneous glass-ceramic products, with decomposition of raw materials and some vitreous phase formation. The aqueous durability of these materials was assessed over a 28-day period using a modified PCT test, and favourably compared to the durability of an international glass. Overall this verifies the potential for HIP technology for usage in wasteform production, with potential large reductions in waste volume, especially if co-mixed wastes are considered



## Characterization and stability of selected ash fractions from thermal treatment of radioactive wastes at the JÜV incineration facility

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Decommissioning of nuclear facilities is associated with the generation of various types of radioactive waste, including low and intermediate level waste (LILW), whose volume significantly exceeds that of high-level waste. A number of thermal treatment technologies that could contribute to a safe and cost-efficient management of LILW were considered in the framework of a project on "Thermal treatment for radioactive waste minimisation and hazard reduction" (THERAMIN). The objective of the THERAMIN project is to provide an EU-wide strategic review and assessment of the added value of thermal treatment technologies in optimisation of radioactive waste management programmes across Europe. Within this scope, the Forschungszentrum Jülich performed a structural characterization and stability studies of selected ash fractions originating from the incineration facility JÜV50/2, operated by JEN mbH.

For an in-depth characterization 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. Moreover, the characterisation included the 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.

Results demonstrate the complex phase composition of the selected ash fractions, including quartz, hematite, lime, corundum, and some mixed oxides (Si,Al,Ca)O<sub>x</sub>. Besides, 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 <sup>137</sup>Cs and <sup>60</sup>Co, along with traces of <sup>241</sup>Am 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 product consistency test ASTM C1285-14, revealed a fast release of <sup>137</sup>Cs irrespective of leaching conditions, solution composition or temperature. In contrast, no significant release of <sup>60</sup>Co was found. No further radionuclides were found in the leaching solutions, indicating fixation of the remaining activity in the solids.

In this contribution we present and discuss the applied methodology and recent results of the ongoing work, contributing to an improved understanding of the properties of the thermally treated waste product and supporting the understanding of its long-term performance under generic disposal conditions.

## Geopolymerisation of gasified ion-exchange resins, mechanical properties and short-term leaching studies

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A combination of gasification of low and intermediate level radioactive waste (LILW) and conditioning of the resulting product within a geopolymer matrix is a potential alternative for vitrification technologies as an immobilisation method. Geopolymer matrixes have demonstrated good retention capability for radionuclides in multiple studies and the technology has been implemented in an industrial scale in Slovakia and Czech Republic. However, the practical waste loading has been limited by the mechanical properties of the encapsulated matrix.

Even a small amount of ion exchange resin (IXR) decreases the strength of the matrix and cohesion of the matrix is lost when the fraction of resin exceeds 15-20%.

In the study, potential to combine gasification as thermal treatment and various inorganic binders as encapsulation matrixes was evaluated. After gasification, the mechanical properties were not similarly sensitive to the encapsulation of IXR. Gasification enabled substantially higher loading of IXR into the sample. Also, gasification of the IXR decreased matrix apparent diffusion. Very low apparent diffusion coefficients were observed when gasified resin was encapsulated in metakaolin matrix. Theoretically, the amount of encapsulated material could be increased by 980 times.

## Synthesis and characterisation of high ceramic fraction brannerite ( $\text{UTi}_2\text{O}_6$ ) glass-ceramic composites

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Brannerite,  $\text{UTi}_2\text{O}_6$ , glass-ceramic composites have been prepared, using  $\text{UO}_2$  and  $\text{TiO}_2$  as the ceramic phase precursors. A range of cold-press and sinter samples with varying glass:ceramic ratios have been prepared under argon at 1200°C to investigate the effect of glass content on formation of brannerite. Ceramic brannerite formed well in all compositions, even at low (10% by weight) glass fractions, with  $\text{UO}_2$  as a minor product. Three further brannerite glass-ceramics have been formed by HIP to investigate the compatibility of this system to HIPing. The samples HIPed at 1200°C form brannerite with  $\text{UO}_2$  as a minor phase, with slightly higher abundance than the cold-press and sinter samples.

## Influence of thermal treatment on the disposability of spent ion exchange resins in deep geological repository: a French case

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The filtration of contaminated aqueous waste streams from nuclear applications produces spent ion exchange resins (IER) which can be classified either as low level (LLW) or as intermediate level (ILW). For the purpose of the work conducted in the framework of the European THERAMIN project, studies considered spent IER waste form to be routed to the French deep geological disposal in the Callovo-Oxfordian formation (Cigéo).

This form of waste is known to release hydrogen by radiolysis degradation, reactive species and complexing compounds. Today, for disposability in Cigéo, direct cementation of IER is the main immobilization process. Production of hydrogen is enhanced in case of cementation.

This work aims to evaluate the interest of the incineration-vitrification with plasma process, SHIVA on a mixture of zeolites, diatoms, and spent IER in regards to the reference immobilization process by cementation. Production of an alumina-borosilicate type glass using SHIVA incineration-vitrification process was considered in order to examine the impact of this process for managing the specificities of this IER waste form with the additional advantage of minimizing the disposal volume.

The work consists of a preliminary approach, based on the physical characterizations and leaching experiments carried out by CEA on the resulting vitrified product, to evaluate the potential benefits that a thermal treatment can provide in the context of Cigéo for such spent IER waste form.

The influence of thermal treatment on several characteristics in line with a generic list of WAC defined within the THERAMIN project (including the radiological inventory, the waste volume and stability of the waste form, thermal output, criticality...) have been qualitatively evaluated considering their potential influences on operational and long-term safety.

The characterization results indicate that organic matter has been removed during the thermal treatment, leading to favourable characteristics for the deep geological disposal due to the absence of organic complexing compounds and hydrogen gas release. The final product consists of an amorphous alumina-borosilicate glass with radionuclides incorporation in the glass matrix. It means that the radionuclide release directly depends on the chemical durability of the produced glass.

## Plasma incineration of cemented concentrates and resins: preliminary evaluation of the disposability of the end product

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In Belgium, the national policy for waste that is categorised as category A, being low and medium level short lived waste, is surface disposal in a dedicated facility. The disposal concept for this facility envisages placing waste in concrete containers (called caissons), which are then filled with mortar to produce a monolith. These monoliths are placed in large concrete structures called disposal modules. Once the modules are filled with monoliths, they are closed with a concrete plate and covered by a layer of soil, which ensures the functions of inclusion, protection and sealing. A licence for the construction and operation of the category A waste disposal facility has been submitted.

Waste eligible for surface disposal must meet strict requirements and criteria with regard to its radiological and physicochemical content. Currently, there are several waste streams for which the compliance with the conformity criteria of this disposal concept and thus its disposability is not yet guaranteed. In most cases, optimisation of the current management route(s) is possible. Examples of conditioned waste showing non-conformities with final disposal are the cemented concentrates and resins from some of the NPPs showing gel formation. Different management scenarios are considered for these wastes. One of the scenarios is to demonstrate that gel formation can be or has stopped for a sufficiently long time. If this cannot be proven, the next scenario is to show that the ASR impact on the engineered barrier system is limited. If it is shown that all these attempts are not able

to guarantee the disposability of the waste product, the reprocessing of the affected drums, by for example thermal treatment, will be considered.

In this paper, the scenario of plasma incineration of the cemented concentrates and resins will be evaluated. For this evaluation, the whole management route will be considered, from acceptance to the plasma facility to disposal of the end-product, with its advantages and limitations. To date no conformity criteria for this specific waste type, the plasma slag exist, as only cemented waste is considered in the development of the conformity criteria of the surface disposal. Therefore, the generic disposability criteria as developed in THERAMIN will be used to assess the disposability of the end product, limiting it to a preliminary assessment.

## Feasibility of thermal treatment techniques for UK problematic waste

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To support delivery of the Nuclear Decommissioning Authority's (NDA's) 2016 Strategy, a Problematic Waste Integrated Project Team was established in May 2016. Its objective is to develop a co-ordinated and improved approach to the industry-wide management of problematic radioactive waste. The IPT is being led by LLW Repository Ltd and Radioactive Waste Management (RWM) Ltd on behalf of the NDA and its activities include delivery of a technical work programme and engagement with a range of stakeholders.

Problematic waste is defined as radioactive waste for which an existing waste management route is either not available or not planned in detail or is considered significantly sub-optimal. It incorporates "orphan wastes" and "wastes requiring additional treatment". These are wastes from across the radiological spectrum, including Low Level Waste (LLW) and Higher Activity Waste (HAW). Wastes may be problematic for a variety of reasons including: technical issues with characterising the waste, a lack of availability of transport packaging, complexity of implementing treatment solutions, or the disposability of treated or conditioned waste products.

The NDA has also established a Thermal Treatment IPT (TT IPT), led by Sellafield Ltd in collaboration with National Nuclear Laboratory (NNL), to investigate thermal

treatment and its applicability to HAW. Thermal treatment techniques use heat and associated process steps to transform wastes into a range of materials, which can range from ashes to vitrified and stable products suitable for long-term storage and disposal. There are various technologies available that can achieve this available around the world. The intent of the TT IPT is to develop thermal treatment technologies to the degree that they can be considered a technically credible option for the treatment of ILW at Sellafield, the NDA estate and potentially across the rest of the UK Nuclear industry.

The PW IPT and TT IPT are working together on a collaborative project to demonstrate the applicability of thermal treatment technology for the management of specific problematic wastestreams. Initial work has focused on:

- identification of suitable problematic waste streams (asbestos, historically conditioned drums and oily sludges);
- development of problem statements and the wider relevance of potential solutions to other UK waste streams; and
- development of wiring diagrams that highlight the issues and questions that need to be addressed to demonstrate the feasibility of thermal treatment for these problematic waste streams.

## Development of phosphate glass and multi-phase titanate ceramic compositions for thermal treatment of irradiated nuclear fuel residues

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The highly heterogeneous nature of UK legacy damaged and degraded spent nuclear fuels and so called, 'orphan fuels', prohibits the use of principal waste treatment methodologies. An inventory of UK residual fuels yielded an account for three main fuel types: Magnox, AGR (advanced gas-cooled reactor) and MOx (mixed oxides). A series of glass and ceramic type host systems have been investigated for potential conditioning of these high uranium content fuel wastes. Electron microscopy and powder X-ray diffraction techniques were used to characterise the prototypical wasteforms.

Two sets of low-melt temperature phosphate glass compositions were trialled with additions of CeO<sub>2</sub> to simulate the fluorite structure and large ionic radius of U in oxide fuels. Evolution of monazite-type phases at simulant oxide waste loadings above 15 wt.% highlighted a potential development into a glass-ceramic hybrid assemblage. Investigation into the use of an alkoxide nitrate synthesis route for SYNROC-F type ceramic precursors has allowed for the demonstration of a sintered host pyrochlore phase containing up to ~40 wt.% waste simulant ceria. Gas evolution has led to increased porosity at higher temperatures and longer sintering times, this may be mitigated by higher pre-calcination temperatures.

## Dissolution of plasma treated non-radioactive surrogate waste forms in KOH solution at 40°C

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In Belgium, plasma incineration is a possible option to (re-)condition low and intermediate level short lived waste streams in order to obtain end products which can fulfill the criteria developed for a near surface disposal. In this study, the chemical durability of plasma treated non-radioactive surrogate waste forms (or plasma slags) was investigated by performing MCC-1 leach tests in a KOH solution at pH 13.5 and 40°C. Results showed that initial dissolution rates of plasma slags were similar to that of SON68-I glass, and final dissolution rates around one order of magnitude lower. SEM analysis revealed the presence of alteration layers rich in K, Ca and Fe, which probably act as diffusion barriers and, associated to the increase of Si and Al concentration in solution, lead to the decrease of the plasma slag dissolution rates.

## Characterisation of chropik III glass samples

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This work within the THERAMIN project determined the leachability of a glass product produced using vitrification of inorganic liquid radioactive waste (RAW) - chropik III. The aim of this study is the chemical characterisation of chropik glass and comparison with basalt glass.

Chropik was used as coolant medium within NPP A1 Jaslovské Bohunice. Chropik is inorganic liquid radioactive waste with dissolved salts up to 3 w%, main contaminant is Cs-137 with activity level E10 Bq/dm<sup>3</sup>.

Chropik non-radioactive glass samples were prepared from glass, additives and chropik surrogate solution in ratio 40/17/43 in VUJE's laboratory. The chropik glass composition consists of: additives on aluminosilicates/geopolymer basis, surrogate solution of chropik III (K<sup>+</sup>, HCO<sub>3</sub><sup>-</sup>, CO<sub>3</sub><sup>2-</sup>), and the Cr (VI) and glass frit (SiO<sub>2</sub>, TiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, B<sub>2</sub>O<sub>3</sub>, Na<sub>2</sub>O, Li<sub>2</sub>O and Fe<sub>2</sub>O<sub>3</sub>). Samples were prepared by moulding into cuboid monoliths of approx. 1.5 cm × 1.5 cm × 1.5 cm.

Chemical composition of glass samples was analyzed by two techniques: SEM/EDX (Scanning Electron Microscopy/Energy Dispersive X-rays Spectroscopy and XRF (X-ray fluorescence spectrometry). The tests of chemical durability of chropik III glass samples and basalts were carried out by leaching in water according to a protocol adapted from the ASTM C1220 using three methods:

- Leaching in demineralized water to the boiling point.

- Leaching at 90 °C in demineralized water in oven without renewal the specimen surface
- Leaching at 90 °C in demineralized water in oven with renewal of the specimen surface

There were only small differences observed between theoretical composition and XRF results. The final chropik III glass product consists of an amorphous glass mainly composed of SiO<sub>2</sub>, Na<sub>2</sub>O, B<sub>2</sub>O<sub>3</sub>, and K<sub>2</sub>O and these compounds represent 84.6 % of the glass composition, with no evidence of any crystalline phase assemblages or other inhomogeneity. For the basalt glass, the main compounds are SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>, Fe<sub>2</sub>O<sub>3</sub>, MgO and CaO and these compounds represent 80 % of the glass composition.

Chemical durability assessment was made and a comparison of the three methods mentioned above was performed for chropik III glass samples: weight loss for method 1 is approximately twice as fast than leaching using method 2. To compare method 2 and method 3 the weight loss rate is twice as fast for method 2 than method 3. Leaching tests conducted at 90 °C show a similar evolution of the pH values, and the best method for chemical durability assessment seems to be method 3.

The comparison of the results obtained from chropik III glass and basalt for chemical durability according to the modified ASTM C1220 shown that weight loss is approximately two to six times better in the case of chropik III glass.

## Incineration-vitrification of a mixture of zeolites, diatoms and ion exchange resins using the SHIVA process

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The Advanced Incineration-Vitrification Hybrid System (SHIVA) process is well suited to treat organic and mineral waste with high alpha contamination management capabilities. It allows, in a single reactor, waste incineration by a plasma burner and ash vitrification in a cold wall direct glass induction melting system. The SHIVA trial demonstrates the successful processing of a waste stream that contains a mixture of mineral and organic ion exchange media. The load rate attains a promising value of 38 wt.% and a glass wasteform is obtained. Further, the microstructure, chemical composition, and chemical durability of the wasteform are characterized.



## Developing laboratory-based X-ray Absorption Spectroscopy for nuclear waste management

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X-ray absorption spectroscopy (XAS) provides a unique and sensitive probe of element speciation and local environment in materials relevant to the nuclear fuel cycle and security. Hitherto, this technique has primarily required access to a synchrotron radiation facility as a broadband X-ray source of high brilliance, which has limited its application for routine and high throughput studies. This is of particular challenge for the field of nuclear materials where scientific opportunity may be constrained by the absence of an accessible synchrotron source or sample containment requirements and associated inventory limits. Here, we report our exploitation of a newly available commercial XAS spectrometer, utilising

spherically bent crystal analysers to acquire XAS data in the range 5 – 18 keV resolution from actinide, nuclear and radiological materials. We show that XAS data may be acquired in a few hours, or less in favourable circumstances, from moderately dilute to concentrated absorbers to address routine questions of element speciation and co-ordination of particular relevance to radioactive waste immobilisation. These data and analyses are compared with counterpart synchrotron studies, on an identical sample suite, to highlight both the potential opportunities and limitations of laboratory XAS, and to demonstrate the feasibility of routine application.

## Micro-XAS analysis of Chernobyl lava

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To understand the chemical and physical properties of Chernobyl lava, a suite of simulant samples has been synthesised and characterized at lab scale with the same morphology and mineralogy as real lava. In order to investigate the uranium behaviour in solid solutions and the glass matrix, microfocus synchrotron X-ray analysis including X-ray fluorescent, X-ray diffraction pattern and X-ray absorption near edge structure ( $\mu$ -XRF,  $\mu$ -XRD and  $\mu$ -XANES) were applied to selected regions of the simulant Chernobyl samples.

XRD of scanned area were interpreted by matching with ICSD-PDF database under the monochromatic wave length. XRF was conducted by collecting the counts of specific elements' fluorescent emission of interest. With the contribution of both data sets, the tomography of phases distribution was

reconstructed by using an in-house MATLAB model. Uranium LIII edge XANES spectra of representative spots were obtained and analysed based on the linear relationship set up by the whiteline points of standard spectra. A range of crystallites including uranium zircon, Zr-U-O,  $\text{UO}_2$  and  $\text{ZrO}_2$  were found in total diffraction pattern of brown lava (shown as Fig.1). Uranium is present mainly as Zr-U-O solid solutions located at the fused morphology area mixed in three crystal structures: monoclinic phase (yellow), tetragonal phase (green) and cubic phase (blue). The major part of zircon (cyan) was found to be located at the triangular shaped particle. The estimated oxidation state values of these selected spots are between 4 and 4.5 with an average of 4.2 and could be classified into three phases based on their distribution (shown as Fig.2).

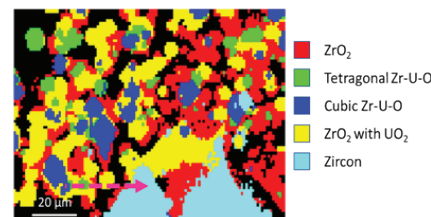


Fig.1 Reconstructed tomography of X-ray diffraction pattern

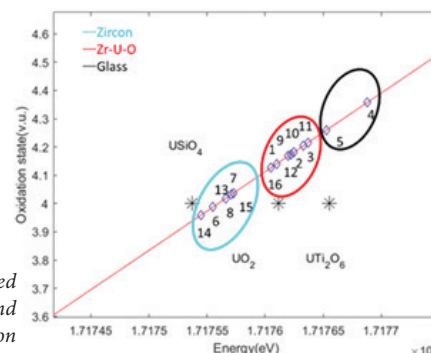


Fig.2 Estimated oxidation states and their distribution

The ratio of uranium and zirconium concentrations agrees well with the phase identifications of Zr-U-O and their distribution. The trend of uranium oxidation states increases from the centre to the edge and further to glass indicating the increase of solubility for higher oxidation states. Spots

in the zircon area has the lowest value, close to the coffinite standard, demonstrating uranium zircon formation. Understanding these behaviours and interactions is a critical support to decommissioning the severely damage nuclear fuels and products.



# A Feasibility Study of Advanced Gas-cooled Reactor SIMFuel Fabricated by Hot Isostatic Pressing

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To properly manage the geological disposal of high-level activity waste (HLW) and spent nuclear fuel (SNF) several studies have investigated the properties of simulated spent fuels (SIMFuel) for Advanced Gas-cooled Reactors (AGR). The microstructural characterisation and Raman analysis for low and high burnup SIMFuels has previously been conducted, showing the different behaviours of fission products (FPs) in surrogate fuel pellets. Some FPs have been found dissolved in the  $\text{UO}_2$  matrix leading to an oxygen-defect-induced lattice distortion; while others have been observed to form metallic and oxide precipitates contributing to changes in pellet density and grain size. However, conventional pressureless sintering has limited ability to simulate the behaviours of certain fission products, especially volatile species such as caesium. Hot Isostatic Pressing (HIP) is a consolidating technique that applies a combination of isostatic pressure and high temperature to samples packed into a closed stainless-steel canister. As volatile fission products are retained within the canister, it results in a better simulation of real AGR spent fuels contained in cladding with similar composition to the HIP canister.

$\text{UO}_2$  and ten fission products based on calculated burnups of 25 GWd/t U and 43 GWd/t U were milled together to achieve homogeneity. The blends were packed into the stainless tube and calcined at 500°C (below decomposition temperature of  $\text{Cs}_2\text{CO}_3$ ). After hermetically sealing, tube samples were HIPed (consolidated at 1250°C under 200 MPa). The area percentage porosity (Fig 1) of SIMFuels were close to those of real spent fuels ( $2.19 \pm 0.84\%$ ). Larger amount of dissolved

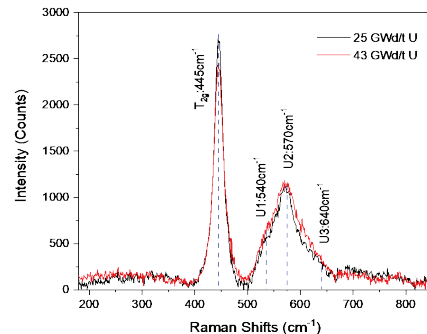


Fig.1 SEM images of porosity in low-doped SIMFuel

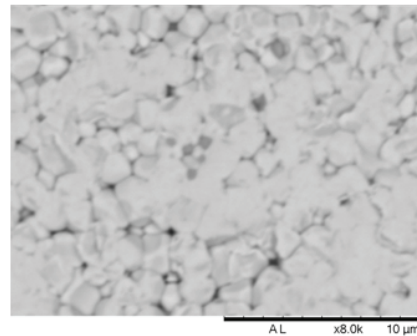


Fig.2 Raman spectra of SIMFuel samples

FPs in higher burnup SIMFuel introduced more defects in the  $\text{UO}_2$  matrix resulting in an increase of the lattice parameters. The change is confirmed by increased intensity of the defect peak (U1) and decrease intensity of U-O stretching mode band deconvoluted by Raman spectrum, shown as Fig 2. The formation of oxide precipitates (such as  $\text{BaZrO}_3$ ) and metallic precipitations (such as Ru-Mo) were observed in SEM-EDX images, which agrees well with the secondary phases in real spent fuels.

## Thermal-hydraulic modelling of a vitrification furnace – DEM&MELT project

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Based on CEA's experience of the In-Can Vitrification process, the DEM&MELT consortium, which gathers ANDRA, Orano, CEA and ECM technologies is currently developing a robust, simple and versatile industrial scale In-Can vitrification prototype, dedicated to treatment of Decontamination and Decommissioning (D&D) and remediation waste. In this context, a thermo-hydraulic model has been developed to optimize its design and operation.

This poster is focused on the simulation carried out to optimize this three-zone resistive In-Can melter furnace. Based on Ansys FLUENT tool, we performed a 2D axisymmetric thermo-hydraulic modelling for various heating configurations (multi-zone, chimney effect, etc).

This model allows operational parameters during the feeding period to be determined in order to optimize glass temperature as well as its thermal convection. It also helps to manage the cooling phase of the Can, switching off of the lower zone power, towards the end of the feeding period, to anticipate the cooling time of the Can. Moreover, taking into account the properties of a given liquid waste, the model provided a relation between the Can filling ratio and waste throughput rate.

The model was first validated with preliminary heating tests; the DEM&MELT prototype will be commissioned during 2020, which will give the opportunity to compare it to the experimental results obtained.

## The HADES facility for high activity decommissioning engineering & science: part of the UK National Nuclear User Facility

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Research and innovation is key to delivering UK Government's civil nuclear energy policy, in particular to accelerate reduction in the hazard, timescale and cost of legacy decommissioning and geological disposal of radioactive wastes. To address this challenge, a national centre of excellence for research and innovation, the HADES Facility, has been established to support research and innovation in High Activity Decommissioning Engineering and Science, as part of the wider network of UK National Nuclear User Facilities. Herein, we describe the development of this user facility, the current status of its capability, and functional equipment specifications.

The unique capabilities of the HADES Facility, in the UK academic landscape, are emphasised, including: handling of weighable quantities of  $^{99}\text{Tc}$  and transuranics; quantitative electron probe microanalysis of radioactive materials; hot isostatic pressing of radioactive materials; and laboratory-based X-ray absorption and emission spectroscopy.

An example case study of the need for and application of the HADES capability is described, involving thermal treatment of a real radioactive ion exchange resin waste to produce a passively safe vitrified waste form.

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

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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 poster presents the key conclusions from the project. A summary of European radioactive waste streams that may be suitable for thermal treatment, and available thermal treatment technologies, is presented. 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.

## Characterisation of calcined simulants developed to represent a typical D&D sludge

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The Vitrification and High Temperature Processes Unit of The French Alternative Energies and Atomic Energy Commission (CEA) is mainly dedicated to the management of radioactive waste. Among the waste streams considered, the waste coming from Decontamination and Decommissioning (D&D) operations will represent increasingly large volumes. As such, prospective studies are being conducted to adapt the methodology of conditioning matrices development (upscaling, waste surrogates, etc.) to these new wastes whose natures are extremely varied and sometimes poorly characterised.

This work deals with thermal treatment by calcination of a typical D&D sludge, mainly composed of sulphated and nitrated compounds. As a first step, the sludge composition was simulated by mixing oxide powders and chemical reagents, according to reasonable hypothesis. Then a calcination step in a muffle-furnace made it possible to obtain a non-reactive waste. The calcination behaviour of the surrogate sludge was evaluated — with or without calcination additives — by microscopy, X-ray diffraction and thermogravimetry.

The decomposition of water and nitrate compounds occurs below 650°C and is accompanied by densification of the calcined product. Little evolution of the waste was then observed between 650°C and 800°C. This behaviour was compared to that of a non-radioactive sludge calcined under the same conditions. The objective is to evaluate the representativeness of the sludge simulated by a mix of chemical powders. This method, which is easy to implement, can be a powerful tool to effectively adapt laboratory-scale experiments to the compositional variability of D&D waste.

Ongoing tests will validate and identify the limitations of using such surrogates in the study of the thermal treatment of typical D&D sludge. Future developments of the study will focus on conditioning the residues obtained at the end of the calcination step.

## Hot isostatically pressed zirconolite wasteforms for actinide immobilisation

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In order to demonstrate the deployment of Hot Isostatic Pressing (HIP) for the immobilisation of Pu stocks and residues, a series of active and inactive zirconolite formulations have been processed and characterised. In this instance, Ce, U, and Th have been applied as chemical surrogates for Pu<sup>4+</sup>. A range of formulations targeting isovalent B-site substitution (i.e. CaZr<sub>1-x</sub>Pu<sub>x</sub>Ti<sub>2</sub>O<sub>7</sub>) have been processed by HIP and characterised by powder X-ray diffraction, and scanning electron microscopy, in order to determine surrogate partitioning between the host zirconolite phase, and accessory phases that may have formed during the HIP process.

## Pyrolysis of radioactive spent resins in the PRIME Installation

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Montair and Belgoprocess developed the pilot PRIME (Pyrolysis Resins In Mobile Electric) Installation for treatment of spent resins in one single reactor without pre-treatment and without intermixing of other waste, allowing for easy traceability and isotopic characterization of the end product. Both low level waste (LLW, < 2 mSv/h) and medium level waste (MLW, > 2 mSv/h) can be treated in the installation.

The PRIME pilot installation is designed to treat 2 batches of 100 litres of saturated ion exchange resins over a period of 24 hours. A dosing unit with screw feeder is used for the dosing of the saturated ion exchange resins to the pyrolysis reactor. The pyrolysis reactor contains a special design mechanical stirrer and can operate at a temperature of up to 600°C under inert conditions. As a result, the organic molecules will decompose at an accelerated rate until only the mineralized material remains. The lower temperature and the absence of oxygen also prevent the breakdown of ion exchange resins into (powdery) ash, which is a considerable advantage during further handling.

Gases that are released during drying and pyrolysis are treated in a thermal oxidizer, which uses an innovative, electrically heated design instead of a traditional natural gas- or fuel oil secondary combustion chamber design. After passing through the thermal oxidizer, the gases are cooled down to saturation temperature and treated in the wet scrubbing system. An induced draft fan keeps the system under negative pressure to prevent the possibility of any off-gas leakage out of the system.

A series of tests were performed successfully with wet cationic resins, anionic resins and a mixture of both. The results of these tests are described in the presentation.

For intellectual property protection, the system and process of pyrolysis of organic waste and the system of thermal oxidation of pyrolysis gases described in this paper are patent pending.

## Development of HIP process for the treatment of Sellafield sludges and slurries

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The poster summarises GRI Ltd's efforts to mature the Hot-Isostatic Pressing (HIP) process option for the treatment of Sellafield sludges and slurries. This work culminated in the production of the largest scale demonstration of HIP technology for radioactive waste immobilisation performed to date (>100 L canister).

The poster summarises the development of: representative simulant materials; custom silicate and titanate wasteforms; investigations into the viable process envelope (focused on the impact of feedstock outliers on the wasteform and process, including variations in organic content, particle size distribution, metallic content and carbonated material); the development of customised HIP canister designs; the at scale demonstration of a commercially viable front-end; the scale-up of waste-packages from ~30 ml to > 100 L; the characterisation of wasteform products, including at those produced at 100 % waste-loading; and the production of lab-scale uranium-doped samples.

HIP has been shown to be a technically viable option for treating Sellafield ILW sludges and slurries. The technology has been shown to tolerate a range of waste-loadings using various wasteform system concepts (titanate/ silicate/ mixed titanate silicate). HIP is amenable to processing both Magnox and SIXEP sludges at 100 wt% waste-loading. Volume reduction ranges from a factor of 3.3 (100 % SIXEP sludges) to a factor of 8.9 (100 % Magnox sludges) with a targeted wasteform co-processing these materials typically giving 6-fold reduction in waste volume. This compares favourably with vitrification where both the achievable waste-loading and product density are lower and cementation where an approximately 3-fold increase in volume is typical.

## Ce and U speciation in ceramic and vitrified wasteforms for thermal treatment of plutonium residues and contaminated material wastes probed by L<sub>3</sub> edge XANES.

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X-ray absorption spectroscopy was applied to understand the speciation of elements relevant to the immobilisation and disposal of radioactive plutonium bearing wastes, utilizing Ce as a Pu surrogate. Ce L<sub>3</sub> XANES (X-ray Absorption Near Edge Structure) characterization of a crystallised glass material produced by cold crucible plasma vitrification, at demonstration scale, evidenced incorporation as Ce<sup>3+</sup> within the glass phase, providing an important validation of laboratory scale studies. U and Ce L<sub>3</sub> XANES investigation of brannerite ceramics, U<sub>0.9</sub>Ce<sub>0.1</sub>Ti<sub>2</sub>O<sub>6</sub>, synthesized under oxidizing, neutral and reducing conditions, established the charge compensation mechanism as incorporation of Ce<sup>3+</sup> through formation of U<sup>5+</sup>. In each of these examples, X-ray Absorption Spectroscopy has provided a pivotal understanding of element speciation in relation to the mechanism of incorporation within the host waste form intended for geological disposal

## Notes





## Project participants

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The THERAMIN project benefits from participation from waste management organisations, waste producers, and research organisations, with input from technology specialists. It therefore offers a “joined-up” perspective of the advantages and disadvantages of using thermal treatment within the waste management lifecycle.

## Project partners

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- VTT Technical Research Centre of Finland (VTT), Finland
- Agence Nationale pour la Gestion des Déchets Radioactifs (Andra), France
- Orano, France
- Commissariat à l'énergie Atomique et aux énergies Alternatives (CEA), France
- Galson Sciences Limited (GSL), United Kingdom
- Forschungszentrum Jülich GmbH (FZJ), Germany
- Lithuanian Energy Institute (LEI), Lithuania
- National Nuclear Laboratory Limited (NNL), United Kingdom
- Belgian Agency for Radioactive Waste and Enriched Fissile Materials (ONDRAF/NIRAS), Belgium
- Studiecentrum voor Kernenergie/Centre d'Etude de l'Energie Nucléaire (SCK•CEN), Belgium
- University of Sheffield (USFD), United Kingdom
- VUJE a.s. (VUJ), Slovakia

## End users

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- National Cooperative for the Disposal of Radioactive Waste (Nagra), Switzerland
- Radioactive Waste Management (RWM), UK
- Sellafield Limited, UK
- Implementing Geological Disposal of Radioactive Waste Technology Platform (IGD-TP)
- Électricité de France (EDF), France
- Fortum, Finland
- Teollisuuden Voima Oy (TVO), Finland
- Agence Nationale pour la Gestion des Déchets Radioactifs (Andra), France - Project Partner as well
- Commissariat à l'Énergie Atomique et aux Énergies Alternatives (CEA), France - Project Partner as well
- Belgian Agency for Radioactive Waste and Enriched Fissile Materials (ONDRAF/NIRAS), Belgium - Project Partner as well
- AWE, UK
- Idaho National Laboratory (INL), USA